Review on Health Impacts from Domestic Coal Burning: Emphasis on Endemic Fluorosis in Guizhou Province, Southwest China

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Contents

Abstract Endemic fluorosis in Guizhou Province, Southwest China was firstly reported by Lyth in 1946 and was extensively concerned since the early 1980s. Initially, the pathological cause of endemic fluorosis in Guizhou Province was

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instinctively ascribed to the drinking water. However, increasing evidences pointed that the major exposure route of fluorine for the local residents is via the roasted foodstuffs, especially the roasted pepper and corn. Source of fluorine in roasted foodstuffs was once blamed on the local coal and subsequently imputed to clay mixed in the coal. In fact, both are probably the source. Geogenic fluorine concentration in soil and clay is indeed high in Guizhou Province, but is not likely to be the direct cause for endemic fluorosis. The real culprit for endemic fluorosis in Guizhou Province is the unhealthy lifestyle of the local residents, who usually roasted their foodstuffs using local coal or briquettes (a mixture of coal and clay), resulting in the elevated fluorine in roasted foodstuffs. Nowadays, endemic fluorosis in Guizhou Province has substantially mitigated. Nevertheless, millions of confirmed cases of dental fluorosis remain left. In addition to endemic fluorosis, other health problems associated with domestic coal burning may also exist, because of the enrichment of toxic/harmful elements in the local coal. It is necessary to determine how serious the situation is and find out the possible solution. As people in other developing countries may suffer from similar health issues, same health issues around the world deserve more attention.

Keywords Epidemiology · Fluorine · Roasted foodstuffs · Source · Toxic trace elements

1 Introduction

Coal plays an important role in fueling the world industrialization and remains an important energy source, especially in the developing countries such as China and India (Finkelman et al. 2002). Taking China as an example, although proportion of coal in the energy mix has declined, consumption rate of coal has continued increasing (Dai et al. 2012; You and Xu 2010). During the formation of coal, potentially harmful or toxic elements can be incorporated into the coal, such as fluorine (F), arsenic (As), antimony (Sb), selenium (Se), mercury (Hg), chromium (Cr), and cadmium (Cd) (Dai et al. 2006a, b, 2012; Li et al. 2006). These elements can be released into the surrounding environments during the mining, storage, and combustion of coal, resulting in a variety of environmental and health problems (Finkelman et al. 1999, 2002; Tian et al. 2010). Considering the widespread utilization and the huge consumption of coal, pollution caused by the coal burning is not only a local or regional issue, but also a global issue.

China is the largest producer and consumer of coal in the world (Zhao and Luo 2018). In China, coal is extensively used for domestic purposes such as house heating and cooking, due to its cost-effectiveness and easy-accessibility. This situation is quite common in Guizhou Province, Southwest China where the winter is cold and damp. Although health problem associated with coal used for electric utility is less reported, health issues associated with domestic coal burning have been frequently reported, especially in Guizhou Province (Finkelman et al. 1999, 2002). The most prominent health issue is endemic fluorosis, with millions of cases of dental fluorosis being confirmed (Dai et al. 2004).

Fluorine (F) is the 13th most abundant element in the Earth's crust and the lightest member of the halogens. As the most electronegative and reactive member of all elements, F is naturally occurred as fluoride-bearing minerals in rocks and dissolved fluoride in water (Ali et al. 2016; Schafer et al. 2018, 2020). Similar to many other trace elements, F is beneficial to human health in trace amounts, but can be harmful in excess (Fordyce et al. 2007). Dental protection benefitted from low intake of F is well documented (Ayoob and Gupta 2006), while dental fluorosis or skeletal fluorosis caused by excessive intake of F is also found worldwide, because of the powerful calcium-seeking property of F (Fordyce et al. 2007). The narrow margin between the desired and the harmful dose of F makes it difficult to keep a balance between the dental protection and the fluorosis. This is probably the main reason for the ubiquitous incidents of fluorosis (Ali et al. 2016). Due to the double-sided nature of F, it is critical to understand the geological and chemical provenance of F in different environmental settings, from a public health perspective.

Endemic fluorosis in Guizhou Province has been extensively investigated within the last several decades. Studies conducted so far, however, were mostly confined to local or regional scale and were inclined to derive descriptive conclusions. It is desired to get a comprehensive understanding on endemic fluorosis in Guizhou Province. In addition to endemic fluorosis, other health problems have also been reported in Guizhou Province associated with the domestic coal burning. Therefore, in the present work, a full description on endemic fluorosis in Guizhou Province associated with the domestic coal burning was provided at first, including the origin of the health issue, the possible exposure routes of F for the local residents, possible sources of F in the roasted foodstuffs, and the real culprits. Secondly, other health problems hidden behind the endemic fluorosis were presented briefly. Lastly, possible research aspects associated with the domestic coal burning were proposed.

2 Endemic Fluorosis in Guizhou Province

2.1 Origin of the Health Issue

Fluorosis was firstly recognized at the beginning of the last century by McKay and Black (1916). They found that enamel developmental imperfection was prevalent in Colorado, a phenomenon confirmed to be related to elevated F^- in the local drinking water. After that, fluorosis was found in various countries/regions, especially in China and India (Sun 2017).

As to Guizhou Province, it can retrospect to 1934 when endemic fluorosis was firstly realized in Southwest Guizhou Province and Northeast Yunnan Province, an area covering approximately 2×10^4 km^2 in Southwest China (Kilborn et al. 1950). This phenomenon was firstly reported by Lyth at Kweichow, a small village in

Guizhou Province. In this work, 134 cases of dental fluorosis were investigated and four cases of skeletal fluorosis were described in detail (Lyth 1946), while the cause of this health issue was not carefully explored. Based on the high contents of F^{-} in a little stream running out of a coal-mine (Lyth 1946), the pathological cause of endemic fluorosis was instinctively ascribed to F^{-} in the local water, a viewpoint proven to be wrong subsequently.

Prior to 1979, endemic fluorosis is less concerned by the Chinese central government. Early tentative efforts on endemic fluorosis from the central government were mainly paid to the dental fluorosis in Northeast/Northwest China, where elevated F^{-} was usually found in the local drinking water. Meanwhile, endemic fluorosis in Guizhou Province was rarely mentioned in the documents and scientific literature. In early 1980s, endemic fluorosis in Guizhou Province was confirmed to be caused by the domestic coal burning rather than the drinking water (Guiyang Epidemic Prevention Station et al. 1981). It was subsequently named as "coal-burning type of endemic fluorosis," a special type of endemic fluorosis found to be prevalent in Southwest China, with millions of cases of dental fluorosis being confirmed (Sun 2017). With the accumulation of epidemiological data, endemic fluorosis in Guizhou Province was extensively concerned.

2.2 Epidemiological Data

Epidemiological survey on endemic fluorosis in Guizhou Province can track back to 1979 when a small survey was conducted at a heavily polluted village in Zhijin, western Guizhou Province, in which 192 volunteers were involved (He et al. 2007). The results indicated that 98.9% of the volunteers were the confirmed cases of dental fluorosis and 77.6% of the adult volunteers were the confirmed cases of skeletal fluorosis. Same serious situation was also found in another heavily polluted village in Jinsha, northwestern Guizhou Province, with all volunteers being the confirmed cases of dental fluorosis and 94% of adult volunteers being the confirmed cases of skeletal fluorosis (He et al. 2007). In fact, the prevalence of endemic fluorosis in Guizhou Province is far beyond a few small villages, but is widespread in the whole province (Zhang et al. 2017a, b). After that, more extensive surveys on endemic fluorosis have been conducted. The results were mostly published in Chinese and were seldom available for foreigners (An et al. 2009; Gao et al. 2015; Li et al. 2003, 2005a, b; Wang et al. 2013). Some of which have been summarized in a recent work (Zhang et al. 2017a, b) and were schematically shown in Fig. 1.

For the adolescent, the latest survey was conducted in 2014 (17,962 volunteers were involved), covering 23 administrative regions of Guizhou Province (Zhang et al. 2017a, b). The results indicated that the average prevalence rate of dental fluorosis was 32.3% (Fig. 1). According to the survey conducted at the same regions in 2007 (502,457 volunteers were involved), the confirmed cases of dental fluorosis was 229,943, with the average prevalence rate of 45.8%. As to the survey conducted in 2000 (188,642 volunteers were involved), the confirmed cases of dental fluorosis

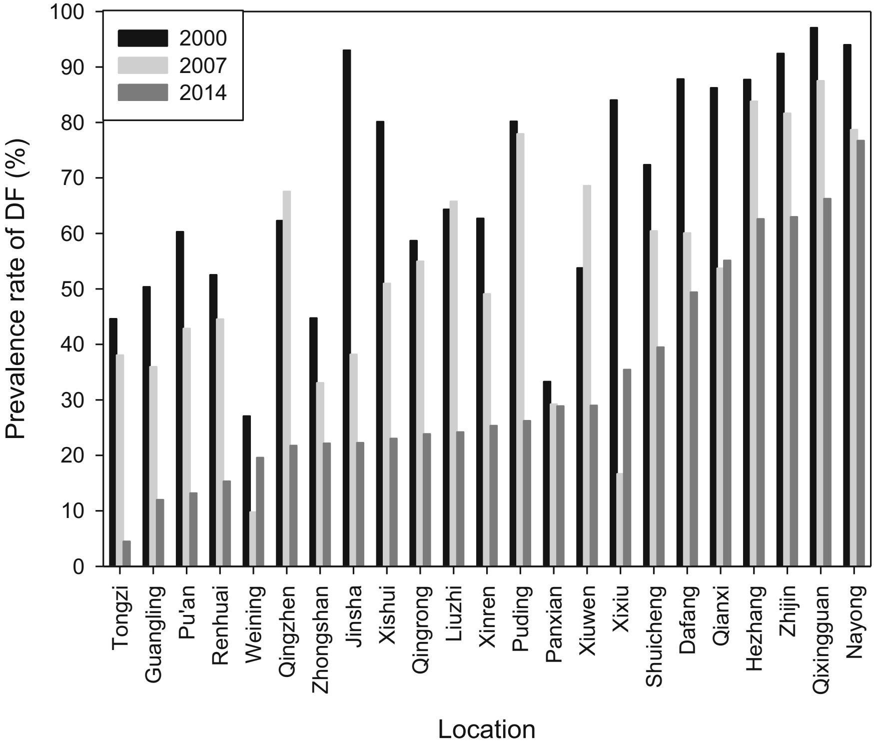

Fig. 1 Prevalence rate of dental fluorosis in 2000, 2007, and 2014 in Guizhou Province (Data source: Zhang et al. 2017a, b)

were 138,256, with the average prevalence rate of 73.3% (Zhang et al. 2017a, b). This indicated that the prevalence rate of dental fluorosis in Guizhou Province was substantially declined since 2000, owing to the great efforts from the local governments (Sun 2017). However, based on the latest statistics, there still have 8.79 million cases of dental fluorosis in Guizhou Province by the end of 2018 (Chinese Health Statistics Yearbook 2019). In addition, significantly high prevalence rates of dental fluorosis were still found in some administrative regions, such as Dafang, Qianxi, Zhijin, Qixingguan, and Nayong, according to the latest survey (Zhang et al. 2017a, b).

As to the adults, based on the survey conducted in seven administrative regions of Guizhou Province during 2001–2003 (122,275 volunteers were involved), the suspected cases of skeletal fluorosis were 33,074, with a suspected rate of 27.03% (Wang et al. 2013). Among the suspected cases, 62.54% of which was confirmed (Li et al. 2005a, b). Among different administrative regions, Qianxinan has the highest suspected rate of skeletal fluorosis (50.76%), followed by Liupanshui, Bijie, and Anshun. Combined with the confirmed rates of skeletal fluorosis, the highest prevalence rate of skeletal fluorosis was found in Liupanshui (30.7%),

Table 1 Results of epidemiological survey conducted in 2001–2003 in Guizhou Province (Data source: Wang et al. 2013)

	Volunteers	Suspected cases	Suspected rate (%)	Confirmed rate (%)	Prevalence rate of skeletal fluorosis (%)
Liupanshui	14,992	4,735	31.58	97.15	30.7
Qianxinan	5,875	2,982	50.76	52.98	26.9
Anshun	19,250	5,970	31.01	74.77	23.2
Bijie	28,304	8,845	31.25	58.12	18.2
Zunyi	21,096	4,755	22.54	43.59	9.83
Qiannan	14,392	3,104	21.57	45.33	9.78
Guiyang	18,366	2,656	14.46	52.76	7.63
Total	122,275	33,047	27.03	62.54	16.9

followed by Qianxinan, Anshun, and Bijie. In comparison, situation is much better in Guiyang, Qiannan, and Zunyi. The detailed information was shown in Table 1.

Historically, endemic fluorosis is very serious in Southwest China, with millions of cases of dental fluorosis being found (Sun et al. 2001), especially in Guizhou Province and Yunnan Province. Fortunately, the confirmed cases of skeletal fluorosis in Guizhou Province have greatly declined and only 2,592 cases were left by the end of 2018 (Chinese Health Statistics Yearbook 2019). During the control and prevention of endemic diseases in China (including the endemic fluorosis in Guizhou Province), great efforts have been done and have been summarized in a recent book (Sun 2017). With regard to endemic fluorosis in Guizhou Province, the possible exposure routes of F for the local residents, the possible sources of F in the foodstuffs and the real culprits have been carefully investigated (Dai et al. 2004, 2007; Luo et al. 2010; Finkelman et al. 1999). This makes the whole story of endemic fluorosis in Guizhou Province becoming clear.

2.3 Possible Exposure Routes

Generally speaking, there are three main routes for human exposure to pollutants, i.e., dietary intake, respiratory inhalation, and dermal exposure. Among them, dietary intake is the major route for most pollutants entering into the human body, although respiratory inhalation is also very important for volatile or semi-volatile organic pollutants. Drinking water and foodstuffs are the major ways of dietary intake of pollutants.

2.3.1 Drinking Water

As far as F^{-} is concerned, drinking water is the most common way for human exposure in most regions, including China, India, Africa, Australia, Europe, and the USA (Ali et al. 2016; Arif et al. 2012), while this is not the case at all in

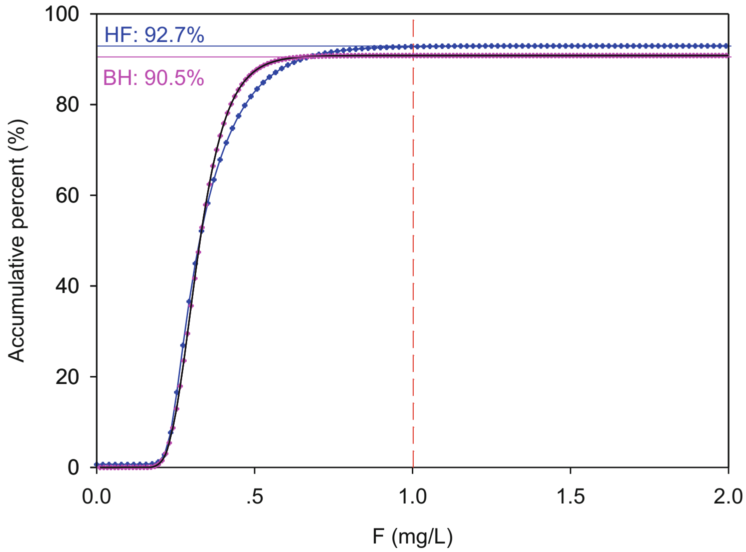

Fig. 2 Contents of F in drinking water sources (Blue line: HF; Pink line: BH) in Guizhou Province

Guizhou Province. Although Guizhou Province is one of the most serious regions suffered from endemic fluorosis, F^- in the local drinking water is seldom to be a problem. The first evidence comes from the long-term monitoring of F^- at two drinking water sources adjacent to Guiyang, central Guizhou Province (5,155 samples were analyzed). The results indicated that content of F^- in more than 90% of the samples was less than the permissible limit proposed by the WHO (1.5 mg/L) and content of F^- in more than 80% of the samples was less than the optimal value (0.5–1.0 mg/L) recommended by the WHO (Fig. 2). The second evidence comes from an extensive survey on F^- in groundwater (1,023 samples were analyzed) (Pu et al. 2013).The results indicated that in Guizhou Province, contents of F^- in the groundwater were within the range of 0.002–3.72 mg/L (mean: 0.313 mg/L), most of which were <1.0 mg/L (Table 2). If these evidences are not strong enough, data on F^- in surface water, groundwater, and drinking water from the heavily polluted areas can further support that endemic fluorosis in Guizhou Province cannot put the blame on the drinking water. As summarized in Table 2, contents of F^- in water are quite low in Guizhou Province, even in the heavily polluted regions. This suggests that F^- in drinking water is not the major cause for endemic fluorosis in Guizhou Province. In the early 1980s, local organizations have realized that endemic fluorosis in Guizhou Province has nothing to do with F^- in the drinking water (Guiyang Medical College et al. 1981). Therefore, people' concerns were shifted to the foodstuffs.

Table 2 Fluorine in groundwater, drinking water, and surface water collected from Guizhou Province

Water type	Location	No. of sample	Content of F (mg/L)	Description	Reference
Ground water	Guizhou province	1,023	96.7% <1.0	Non-specific	Pu et al. (2013)
Drinking water	Liupanshui	354	96.6% <0.5	Heavy polluted area	Li and Yan (1994)
Surface water	Guiyang	7	0.36–0.91 (0.49)	Polluted area	Yu et al. (1994)
Drinking water	Zhijin	5	0.09–0.42 (0.19)	Heavy polluted area	Dai et al. (2004)
Drinking water	Zhijin	13	0.05–0.14	Heavy polluted area	Wu et al. (2004)
Drinking water	Zhijin, Nayong, and Pingba	76	<0.02–0.44 (0.09)	Heavy polluted area	Xie et al. (2010)
Ground water	Dafang	31	0.09–0.37 (0.15)	Heavy polluted area	Zhang et al. (2016)
Drinking water	Shuicheng	10	0.03–0.14	Heavy polluted area	Xiao et al. (2016)
Surface water	Zhijin	15	0.05–0.38 (0.19)	Heavy polluted area	Li et al. (2016)
Ground water	Zhijin	20	0.01–0.20 (0.08)	Heavy polluted area	Li et al. (2016)

2.3.2 Foodstuffs

It has long been suspected that endemic fluorosis in Guizhou Province may associate with F^- in the foodstuffs. The earliest data on F^- in the local foodstuffs were reported in 1981, in which ten kinds of food items were analyzed and contents of F^- in the foodstuffs were in the range of 0.5–9.3 mg/kg (Guiyang Medical College et al. 1981). However, to date, there is no extensive survey on F^- in the foodstuffs and most reported data were associated with the roasted pepper and corn. The available data were summarized in Table 3.

In 1990s, An and his co-authors (1996) conducted a survey on F^- in foodstuffs collected from Puding, Zhijin, and Guiyang, in which seven villages were involved and 20 families were randomly selected in each village. The results indicated that extremely high level of F^- was found in roasted pepper (Table 3). In the following case studies conducted in Zhijin, western Guizhou Province, extremely high level of F^- in roasted pepper was also found at Chengguan Township, Qimo Township, and Sanjia Township (Li et al. 2004), as well as some small villages (such as Hualuo, Majiazhuang, and Pianpozhai) (Dai et al. 2007; Wu et al. 2004). Even very recently, extremely high level of F^- (574 ± 297 mg/kg) was still reported in roasted pepper collected from Nayong, western Guizhou Province (Liu et al. 2013). These data are two or three orders of magnitude higher than the permitted level proposed by the Chinese government (1.0 mg/kg, GB 2762–2005).

Table 3 Contents of fluorine in foodstuffs collected from Guizhou Province

Location	Date	Food type	No. of sample	F (mg/kg)	Reference
Liupanshui	1994	Roasted pepper	17	167–1,267	Li and Yan (1994)
Xinzhai, Zhijin	1996	Roasted pepper	20	1,126 ± 604	An et al. (1996)
Daga, Zhijin	1996	Roasted pepper	20	674 ± 513	An et al. (1996)
Hehua, Zhijin	1996	Roasted pepper	20	1,136 ± 727	An et al. (1996)
Haoyun, Puding	1996	Roasted pepper	20	349 ± 714	An et al. (1996)
Xiayan, Guiyang	1996	Roasted pepper	20	593 ± 424	An et al. (1996)
Erguai, Guiyang	1996	Roasted pepper	20	459 ± 471	An et al. (1996)
Chengguan, Zhijin	2002	Roasted pepper	5	222 ± 175	Li et al. (2004)
Qimo, Zhijin	2002	Roasted pepper	5	408 ± 305	Li et al. (2004)
Sanjia, Zhijin	2002	Roasted pepper	5	362 ± 306	Li et al. (2004)
Hualuo, Zhijin	2003	Roasted pepper	10	513 ± 389	Wu et al. (2004)
Majiazhuang, Zhijin	2003	Roasted pepper	7	343 ± 238	Wu et al. (2004)
Pianpozhai, Zhijin	2003	Roasted pepper	13	281 ± 225	Wu et al. (2004)
Zhijin	NA[a]	Roasted pepper	9	Ave: 1419	Dai et al. (2007)
Four villages, Nayong	NA	Roasted pepper	11	574 ± 297	Liu et al. (2013)
Liupanshui	1994	Roasted corn	88	4.2–300	Li and Yan (1994)
Xinzhai, Zhijin	1996	Roasted corn	20	69.8 ± 24.2	An et al. (1996)
Daga, Zhijin	1996	Roasted corn	20	34.2 ± 14.3	An et al. (1996)
Hehua, Zhijin	1996	Roasted corn	20	23.9 ± 11.9	An et al. (1996)
Chengguan, Zhijin	2002	Roasted corn	5	8.33 ± 5.33	Li et al. (2004)
Qimo, Zhijin	2002	Roasted corn	5	25.0 ± 22.8	Li et al. (2004)
Sanjia, Zhijin	2002	Roasted corn	5	18.9 ± 6.38	Li et al. (2004)
Hualuo, Zhijin	2003	Roasted corn	10	30.6 ± 7.6	Wu et al. (2004)
Majiazhuang, Zhijin	2003	Roasted corn	7	30.4 ± 13.2	Wu et al. (2004)
Pianpozhai, Zhijin	2003	Roasted corn	13	49.8 ± 29.0	Wu et al. (2004)
Zhijin	NA	Roasted corn	9	Ave: 110	Dai et al. (2007)
Weining, Bijie	2006, 2008	Roasted corn	9	9.50–28.9	Luo et al. (2011a)

(continued)

Table 3 (continued)

Location	Date	Food type	No. of sample	F (mg/kg)	Reference
Four villages, Nayong	NA	Roasted corn	11	23.2 ± 12.8	Liu et al. (2013)
Haoyun, Puding	1996	Rice	20	0.92 ± 0.30	An et al. (1996)
Xiayan, Guiyang	1996	Rice	20	4.13 ± 2.85	An et al. (1996)
Erguai, Guiyang	1996	Rice	20	5.51 ± 3.22	An et al. (1996)
Liupanshui	1994	Fresh pepper	10	17.7–18.5	Li and Yan (1994)
Liupanshui	1994	Fresh corn	10	1.2–1.6	Li and Yan (1994)
Weining, Bijie	2006, 2008	Fresh corn	9	0.24–2.21	Luo et al. (2011a)

[a]NA: Not available

In addition to roasted pepper, high content of F^- was also found in roasted corn. As displayed in Table 3, contents of F^- in roasted corn collected from Xinzhai village, Daga village, and Hehua village were 69.8 ± 24.2 mg/kg, 34.2 ± 14.3 mg/kg, and 23.9 ± 11.9 mg/kg, respectively (An et al. 1996). These data were close to the results reported at Hualuo village (30.6 ± 7.6 mg/kg), Majiazhuang village (30.4 ± 13.2 mg/kg), and Pianpozhai village (49.8 ± 29.0 mg/kg) (Wu et al. 2004), and were slightly higher than the reported data at Chengguan Township (8.33 ± 5.33 mg/kg), Qimo Township (25.0 ± 22.8 mg/kg), and Sanjia Township (18.9 ± 6.38 mg/kg) (Li et al. 2004). Although contents of F^- in roasted corn varied among different regions, most were dozens of the permitted level proposed by the Chinese government (1.5 mg/kg, GB 2762–2005). Fortunately, contents of F^- in roasted pepper and corn have substantially decreased, based on the survey conducted in 2013 in the heavily polluted area (covering 23 administrative regions of Guizhou Province) (Zhang et al. 2017a, b).

As to F^- in other foodstuffs, related data were limited and scatted in different literature. The earliest data collected from Zhijin indicated that mean contents of F^- in wheat and rice were 9.3 mg/kg and 5.4 mg/kg, while they were within the range of 0.5–5.5 mg/kg in different kinds of vegetables (Guiyang Medical College et al. 1981). Contents of F^- in rice were also reported at two villages in Guiyang, with the mean values of 4.13 ± 2.85 mg/kg and 5.51 ± 3.22 mg/kg, respectively (An et al. 1996). They were clearly less than that in roasted pepper and corn, but still several times that of the permitted level proposed by the Chinese government (GB 2762–2005).

Pepper is the favorite food item for the local residents and is usually roasted before consumption. Extremely high F^- level in roasted pepper implied that high F^- exposure risk is expected via the consumption of roasted pepper, and things may be worse if the roasted corn is the staple food for the local residents.

2.3.3 Other Exposure Routes

In addition to dietary intake, other possible routes for human exposure to F^- include respiratory inhalation and dermal exposure. Dermal exposure of F^- for the local residents is seldom mentioned, while respiratory inhalation of F^- is occasionally reported. Based on the collected data, respiratory intakes of F^- for the local residents were very limited, although they were varied among different surveys (Table 4). For example, respiratory intake of F^- estimated for the local residents in Zhijin was 0.2 mg/day/capita (Guiyang Medical College et al. 1981). This is similar to that of a recent survey conducted in Nayong, western Guizhou Province, with the respiratory intake of F^- of 0.16 mg/day/capita during the house-heating season (Liu et al. 2013). Based on the survey conducted in 2006 in Zhijin, Jinsha, and Guiyang (Li et al. 2011), the respiratory intake of F^- was even less, within the range of 0.03–0.12 mg/day/capita. Compared with the diary intake of F^-, the respiratory intake of F^- is quite low (Guiyang Medical College et al. 1981; Li et al. 2011; Liu et al. 2013). For example, respiratory intake of F^- for the local residents in Zhijin is only 0.2 mg/day/capita, accounting for 3.2–7.1% of the total intakes of F^- (Guiyang Medical College et al. 1981). While in Nayong, respiratory intake of F^- is only 1.4% of the total intakes of F^- (Liu et al. 2013). If more food items are included as estimated by Li and his co-authors (2011), the proportion of respiratory intake of F^- to the total exposure of F^- would further decrease to <1%. Therefore, the exposure of F^- for the local residents is mostly via the roasted foodstuffs (usually more than 90%), rather than the air and drinking water (Table 4).

Different organizations have set different safe doses for human exposure to F^-, most of which were within the range of 1.5–4.0 mg/day/capita. For example, the guideline for daily intake of F^- recommended by WHO is 2 mg/day/capita for children and 4 mg/day/capita for adults (WHO 2002). As to China, the recommended daily allowances (RDAs) in coal burning fluorosis area were set to be 2.0 mg/day/capita for children aged 8–15 years and 3.0 mg/day/capita for adults; While in drinking water fluorosis area, the RDAs were set to be 2.4 mg/day/capita for children aged 8–15 years and 3.5 mg/day/capita for adults (Li et al. 2015). Guizhou Province is the typical coal burning fluorosis area, and most of the actual exposure levels (as listed in Table 4) are much higher than the proposed RDAs for the local residents.

2.4 *Possible Sources of Fluorine*

Because human exposure of F^- in Guizhou Province is mainly via the food consumption, it is easy to link endemic fluorosis to F in soil. After all, high content of F^- has been found in vegetables (6.64–10.4 mg/kg) cultivated in soil with high-F background in Xuzhou, East China (Zhu et al. 2000), and elevated F^- in crops (ranging from 3.3–9.8 mg/kg) has been reported in Zhijin, western Guizhou

Table 4 Human exposure of F (mg/day/capita) for local residents in Guizhou Province

Location	Year	Pepper	Corn	Rice	Vegetables	Food[a]	Drinking water	Air	Total intake	Reference
Zhijin	2006	15.4	2.35	1.03	0.50	19.3	0.28	0.03	19.6	Li et al. (2011)
	1979	16.3	31.6	0.23	0.50	48.6	0.28	0.34	49.3	Li et al. (2011)
Jinsha	2006	13.9	16.2	0.26	0.51	30.9	0.24	0.08	31.1	Li et al. (2011)
	1986	15.0	40.2	0.23	0.50	55.9	0.24	0.45	56.6	Li et al. (2011)
Bijie	2006	24.7	7.29	1.03	0.50	33.5	0.22	0.07	33.9	Li et al. (2011)
Bijie	2006	22.0	3.51	1.03	0.50	27.0	0.14	0.06	27.2	Li et al. (2011)
Zhijin	2006	7.97	---	1.29	0.50	9.76	0.09	0.12	9.96	Li et al. (2011)
Jinsha	2006	0.32	---	1.29	0.50	2.11	0.18	0.11	2.39	Li et al. (2011)
Zhijin	NA[a]	---	---	---	---	4.3	0.20	0.37	4.87	Guiyang Medical College et al. (1981)
Zhijin	NA[a]	---	---	---	---	6.3	0.20	0.37	6.87	Guiyang Medical College et al. (1981)
Zhijin	NA[a]	---	---	---	---	8.6	0.20	0.37	9.17	Guiyang Medical College et al. (1981)
Zhijin	NA[a]	---	---	---	---	9.8	0.20	0.37	10.4	Guiyang Medical College et al. (1981)
Zhijin	NA[a]	---	---	---	---	10.9	0.20	0.37	11.5	Guiyang Medical College et al. (1981)
Nayong	NA[a]	8.60	2.30		---	10.9	---	0.16	11.1	Liu et al. (2013)

[a]Food: sum of all the food items included in the survey. *GMC* Guiyang Medical College, *NA* Not available

Province (Guiyang Medical College et al. 1981; Li and Chen 1997). Secondly, extremely high level of F^- is usually found in roasted foodstuffs, especially the roasted pepper and corn. This implied that endemic fluorosis in Guizhou Province is more likely associated with F^- in the coal, an energy source used for food roasting. In addition, during the source seeking of F^- in roasted foodstuffs, clay mixed in the coal is also the possible source.

2.4.1 Fluorine in Soil

Based on the pervious investigation (Fig. 3), geological background of F^- in Guizhou Province is highest nationwide, falling in the range of 800–899 mg/kg. This is approximately twice that of F^- in surface soil in China (440 mg/kg) and at least four times that of F^- in soil worldwide (200 mg/kg) (Wu et al. 2008). According to the survey conducted in 2007–2008 (more than 40,000 soil samples were involved), the arithmetic mean value of F^- was 929 mg/kg (Zheng et al. 2009), slightly higher than the previously reported data. If different strata were taken into account, high contents of F^- were usually found in soil developed in Triassic, Permian, Silurian, Ordovician, and Cambrian strata (Zheng et al. 2009). Recently, a survey conducted in Guiyang, central Guizhou Province indicated that contents of

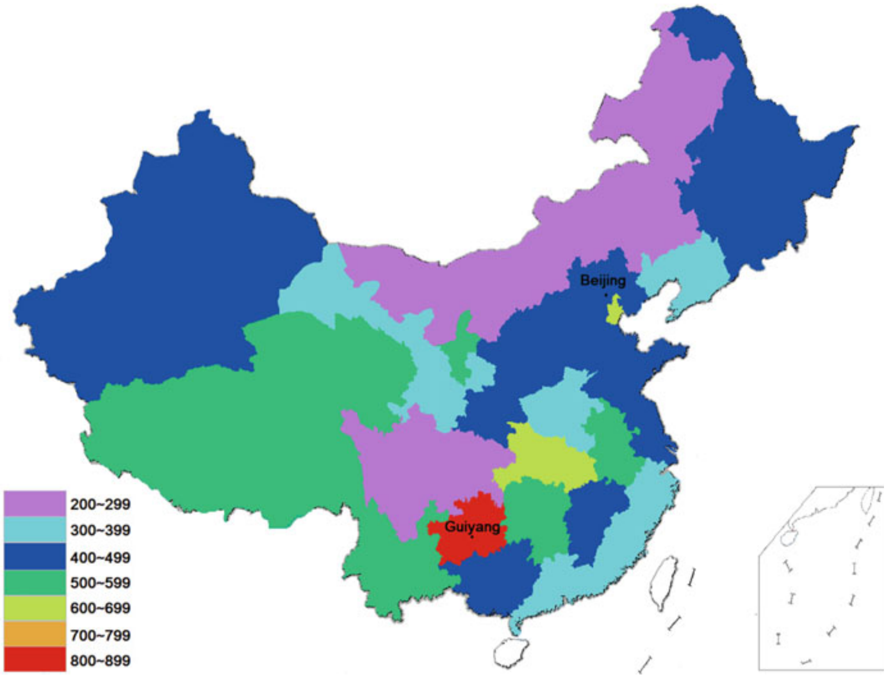

Fig. 3 Background levels of F (mg/kg) in soils in China

F^- in surface and deep soil were in the range of 274–3,663 mg/kg (mean: 1103 mg/kg) and 246–3,695 mg/kg (mean: 1382 mg/kg), respectively (Pan et al. 2018). All the results suggested that geological background of F^- is indeed high in Guizhou Province and could be the fundamental cause for endemic fluorosis.

Our concerns about the geological background of F^- stemmed from the hypothesis that high geological background of F^- may lead to elevated F^- in crops or vegetables, and further resulting in endemic fluorosis via the food consumption. Fortunately, the capability of crops or vegetables to accumulate F from soil is limited (Wu et al. 2008; Zhu et al. 2000). Although extremely high content of F^- was found in the roasted pepper and corn, content of F^- in the fresh pepper and corn is fairly low, even in samples collected from the heavily polluted areas (Table 3). For example, mean contents of F^- in fresh pepper and corn collected from Zhijin were 1.64 mg/kg and 0.79 mg/kg (Dai et al. 2007). This is close to the pervious reported data with mean value of 1.7 mg/kg in fresh pepper and 0.65 mg/kg in fresh corn (An et al. 1995). Similar results can also be found in Liupanshui and Weining, where contents of F^- in fresh corn were in the range of 1.2–1.6 mg/kg and 0.24–2.24 mg/kg, respectively (Li and Yan 1994; Luo et al. 2010). Therefore, high geological background of F^- is not necessary resulting in high content of F^- in the foodstuffs and is not necessary the direct cause for endemic fluorosis in Guizhou Province. High content of F^- in roasted foodstuffs should blame on the roasting processes, rather than the foodstuffs themselves. This understanding is helpful to find the real source of F^- in foodstuffs.

2.4.2 Fluorine in Coal

Because the foodstuffs themselves is seldom to be a problem, high content of F^- in roasted foodstuffs is more likely associated with coal used in the roasting processes. This is the main reason that endemic fluorosis in Guizhou Province is defined as the coal-burning type of fluorosis, a typical endemic fluorosis found in Southwest China.

Early on, it was assumed that the local coal could be abundant in F^-, resulting in elevated F^- in roasted foodstuffs. Because some coal samples from Guizhou Province is do abundant in F^- (Table 5), such as coal samples collected from Anlong, Southwest Guizhou Province (Liu et al. 2008a, b), domestic coal samples from Zhijin (Dai et al. 2007), and coal samples from Zhijin, Nayong and Pingba, western Guizhou Province (Xie et al. 2010). However, as the accumulation of data on F^- in coal, we realized that content of F^- in coals from Guizhou province is modest and is not so high as expected, even in coal samples collected from the heavily polluted areas (Dai et al. 2004; Hong et al. 2016; Liang et al. 2011). For example, in anthracite coal samples collected from Pu'an and Puding, western Guizhou Province, contents of F^- were in the range of 90.2–149 mg/kg (Liang et al. 2011). In 38 outcrop coal samples from western and northwestern Guizhou Province, contents of F^- were between 44 mg/kg and 382 mg/kg, with mean value of 125 mg/kg (Hong et al. 2016). While in seam channel coals from western Guizhou Province, contents of F^- were in the range of 16.6–500 mg/kg, with mean value of

Table 5 Fluorine in coal and clay of Guizhou Province

Sample	Location	No. of sample	Content of F (mg/kg)	Reference
Coal	Anlong	7	271–1,416 (925)[a]	Liu et al. (2008a, b)
Domestic coal	Zhijin	9	79.5–599 (237)	Dai et al. (2007)
Anthracite coal	Zhijin, Nayong, and Pingba	39	62–535 (202)	Xie et al. (2010)
Anthracite coal	Pu'an and Puding	21	90.2–149	Liang et al. (2011)
Outcrop coal	Xishui, Dafang, and Jinsha	38	44–382 (125)	Hong et al. (2016)
Seam channel coal	Zhijin	19	16.6–500 (89.3)	Dai et al. (2004)
Seam channel coal	Western Guizhou Province	50	16.6–500 (83.1)	Dai et al. (2004)
Anthracite coal	Guizhou Province	140	143	Zhong and Luo (2017)
Bituminous coal	Guizhou Province	215	89.0	Zhong and Luo (2017)
Domestic coal	Zhijin	15	89.8 ± 37.6	Li et al. (2004)
Coal and clay	Zhijin, Nayong, and Pingba	9	286–1,190 (507)	Xie et al. (2010)
Coal and clay	Zhijin	9	172–1,498 (828)	Dai et al. (2007)
Coal and clay	Zhijin	45	621–2,816 (1284)	Hong et al. (2015)
Clay	Zhijin	11	452–4,955 (2262)	Dai et al. (2007)
Clay	Zhijin	14	710–2,500 (1415)	Liu and Wang (2005)

[a]Data in the parentheses are the mean value

83.1 mg/kg (Dai et al. 2004). These data were close to the background level of F^- in Chinese coals (130 mg/kg, Dai et al. 2012) and US coals (98 mg/kg, Finkelman 1993), as well as that of the world average (88 mg/kg, Ketris and Yudovich 2009). Recently, a survey on F^- in coals from 90% of the coal-producing areas of Guizhou Province was conducted (more than 1,000 samples were analyzed) (Zhong and Luo 2017). The results indicated that contents of F^- in coal were mostly in the range of 50–180 mg/kg, with mean value of 143 mg/kg in anthracite coal and 89.0 mg/kg in bituminous coal, respectively (Zhong and Luo 2017).

Although content of F^- in local coal is modest, we cannot deny that elevated F^- in roasted foodstuffs comes from domestic coal burning. Because content of F^- in roasted foodstuffs depends not only on the content of F^- in coal, but also on the roasting time and the volatility of F^- from coal. If the roasting time is long enough and F^- in the coal is easy to be volatilized, extremely high level of F^- in roasted foodstuffs can still be expected. Unfortunately, both factors are common in Guizhou Province. Due to the damp weather in cold season, the local residents have to roast their foodstuffs (such as pepper and corn) for a long time, in order to keep their foodstuffs being preserved better. In the meantime, Coal in Guizhou Province is usually abundant in sulfur (Dai et al. 2015; Hong et al. 2015). This facilitates the release of F from coal during the combusting processes (Hong et al. 2018).

2.4.3 Fluorine in Clay

Endemic fluorosis in Guizhou Province was once blamed on high-F coal in early research works (Watanabe et al. 2000; Zhang and Cao 1996). One of the important facts is that extremely high contents of F^{-} were observed in briquettes collected from the heavily polluted areas (Zhang and Cao 1996). In fact, briquettes used for domestic purpose is actually a mixture of coal and clay (Dai et al. 2004, 2007; Wu et al. 2004). Considering the modest content of F^{-} in most coal samples from Guizhou Province (Table 5), extremely high level of F^{-} in briquettes is more likely stemmed from the clay, rather than the coal itself. As displayed in Table 5, clay is true abundant in F^{-} (Dai et al. 2007; Liu and Wang 2005) and if coal mixed with clay to produce the briquettes, content of F^{-} would increase sharply in briquette (Xie et al. 2010; Dai et al. 2007; Hong et al. 2015). As early as 1985, investigation on 137 briquette samples has proven that extremely high content of F^{-} in briquettes do come from the clay (Zhou et al. 1991, and the reference therein), instead of the coal. This is supported by the works conducted in heavily polluted regions (Dai et al. 2004; Wu et al. 2004) and confirmed by the works conducted in the whole province (Hong et al. 2015; Liu and Wang 2005; Luo et al. 2010; Xie et al. 2010).

Based on these information, Sun (2005) has concluded that endemic fluorosis in Guizhou Province is mainly caused by the clay rather than the coal, a viewpoint still being debatable. As mentioned above, if the roasting time is long enough, serious food contamination by F^{-} can still be possible, even though the content of F^{-} in coal is modest. In addition, F in the local coal is more easily to be volatilized during the roasting processes (Hong et al. 2018; Liang et al. 2011), because the local coal is usually rich in sulfur (Dai et al. 2015; Duan et al. 2018; Hong et al. 2018). Thus, it can be deduced that even there is no clay; long roasting time may also lead to heavy contamination of F^{-} in roasted foodstuffs. The mixing of clay in coal is more likely to make things worse, but is not necessary the primary cause for endemic fluorosis in Guizhou Province. Nowadays, it is widely accepted that both coal and clay are the major sources of F in roasted foodstuffs and consumption of roasted foodstuffs is the major cause for endemic fluorosis in Guizhou Province and the adjacent area (Li et al. 2012; Luo et al. 2011b).

2.5 *Real Culprit of the Endemic Fluorosis in Guizhou Province*

Based on the discussion above, it can be concluded that: (1) endemic fluorosis in Guizhou Province is nothing to do with F^{-} in the local drinking water; (2) geological background of F is indeed high in Guizhou Province, but is not necessary resulting in elevated content of F^{-} in the fresh food items; (3) consumption of roasted foodstuffs is the major route for the local residents' exposure to F^{-}; (4) coal is an important source of F in roasted foodstuffs, but content of F in local coal is not so high as expected; and (5) clay mixed in the coal is another important source of F in roasted foodstuffs. Mixing F-rich clay with the coal to produce briquettes can lead to higher exposure risk for the local residents.

Although geological background of F is indeed high in Guizhou Province, it is seldom to be a problem via the natural processes, such as leaching into water and bioaccumulating into crops or vegetables. It is certain that the major exposure route of F^- for local residents is the consumption of roasted foodstuffs and sources of F^- in roasted foodstuffs are coal or briquettes used for domestic purposes. However, there is no direct causal relationship between the endemic fluorosis and the domestic coal burning. Because domestic coal burning activities are common in Chinese rural area, especially in North China (Zhi et al. 2017), while coal-burning type of endemic fluorosis is mainly found in Southwest China and is seldom found in North China. Therefore, it seems unfair to blame the local coal for endemic fluorosis in Guizhou Province.

In fact, the important link between contaminated foodstuffs and F^- in coal/clay briquettes is the roasting processes, an unhealthy way to deal with the foodstuffs. This provides F in coal or briquettes a chance to get into the food chain, resulting in endemic fluorosis via consumption of contaminated foodstuffs. This behavior is usually happened in Southwest China, but is rarely found in other rural areas of China. This is why coal-burning type of endemic fluorosis is mainly happened in Southwest China, but is rarely found in other rural areas of China. Therefore, the real culprit of endemic fluorosis in Guizhou Province should be blamed on the unhealthy lifestyle of the local residents and more precisely, endemic fluorosis in Guizhou Province should be defined as the lifestyle-related endemic disease (Sun 2017). This becomes the theoretical foundation for the control of the endemic fluorosis in Guizhou Province and the adjacent area (Luo et al. 2010; Xu and Xu 2008).

2.6 *Control of the Endemic Fluorosis in Guizhou Province*

Based on the research works, local governments have taken many effective measures to control the endemic fluorosis in Guizhou Province. The specific measures are to: (1) encourage the use of cleaner fuels; (2) help the local residents to ameliorate their household stoves; (3) change the local residents' diet structure; and (4) enhance the related health awareness of the local residents.

Energy source previously used in the rural houses is usually the high-F briquettes, a mixture of local coal and clay. To control the endemic fluorosis, the first step is to encourage the use of cleaner energy. The early engineering design is to mix lime into the briquettes to inhibit the release of F from the briquettes (Sun 2005; Dai et al. 2006a). Nowadays, the use of outcrop coal and briquettes has been banned and seldom found in the rural houses, while commercial coal was still frequently found in rural area of Guizhou Province. The good news is that the coal used for domestic purposes is usually cleaner than it used to be. This can substantially reduce the emission of F into the surrounding environments and mitigate the exposure risk for the local residents. Previously, stoves used in the rural houses are simple and crude and often used in open environments. The second step is to ameliorate the stoves (e.g., adding exhaust pipes to improve the stove's ventilative capacity). Presently, most of the coal-burning stoves used in rural houses have been ameliorated and used in relatively isolated places (Hao et al. 2002; Liu et al. 2016). This can further reduce the residents' exposure risk to F. Formerly, local residents often took roasted corn (usually with elevated F^{-}) as their staple food. Nowadays, rice has become the staple food for local residents in most rural areas, though consumption of roasted corn still happened occasionally. This is also helpful for reducing the exposure risk to F for the local residents. The last step is to enhance the health awareness of local residents. The key point is to let the local residents known that how the endemic fluorosis happened and how to preserve their foodstuffs free of F contamination (Li et al. 2005a, b; Zhang et al. 2012). These measures have been summarized in the book of "Endemic Disease in China" (Sun 2017).

Since 2005, great efforts have been paid to the control of endemic fluorosis in Guizhou Province. As time passed, prevalence rate of endemic fluorosis in Guizhou Province has declined significantly compared with the historical data. However, prevalence rate of dental fluorosis (32.3%) in Guizhou Province was still high till very recently (Zhang et al. 2017a, b) and 8.79 million cases of dental fluorosis still left by the end of 2018 (Chinese Health Statistics Yearbook 2019). Although most existing cases of dental fluorosis are historically left, how many new cases developed remain unclear. Some administrative regions of Guizhou Province still deserve more attention, including Dafang, Qianxi, Zhijin, Qixingguan, and Nayong, where the prevalence rates of dental fluorosis were still higher than 50% till 2014 (Table 1).

So far, coal-burning type of endemic fluorosis is mainly found in Southwest China, similar health issue may also occur in other parts of the world. The findings on this issue are of great importance to the countries/regions where coal-burning type of endemic fluorosis could develop.

3 Other Health Problems

In addition to endemic fluorosis, other health problems associated with domestic coal burning may also exist in Guizhou Province, such as arsenosis, selenosis, and mercurialism (Finkelman et al. 1999, 2002). Our concerns about the health problems caused by domestic coal burning in Guizhou Province are based on the following facts: (1) commercial coal for domestic purpose is still common in the rural area of Guizhou Province, especially in cold season; (2) stove used for domestic coal burning in rural area is still simple, usually without any decontamination devices (Cheng et al. 2017); and (3) domestic coal used in rural households is usually without any legal or environmental requirements (Zhi et al. 2017). Based on the historical data, consumption rate of domestic coal in China has increased from approximately 50 million tons in 1992 to more than 90 million tons in 2015, and Guizhou Province is the main region of domestic coal utilization, second only to

Hebei Province and Shanxi Province (Zhao and Luo 2018). Although the use of outcrop coal and the briquettes have been banned, commercial coal for domestic purpose may still be widespread in the rural area of Guizhou Province. Coupled with the backward burning mode in the rural houses, local residents may face other health risks via indoor air pollution, which depends largely on the cleanliness of domestic coal.

Unfortunately, coals in Guizhou Province often contain high levels of toxic/harmful elements. Because Guizhou Province is one of the largest low-temperature metallogenic regions in the world and the low-temperature metallogenic processes usually lead to an enrichment of toxic/harmful elements in the coal (Dai et al. 2006b; Li et al. 2006; Song et al. 2007). For example, enrichment of As, Se, Hg, Cd, and Cr in coal is frequently found in Guizhou Province and the adjacent area (Li et al. 2006; Tang et al. 2009). High contents of As (7.6–230 mg/kg), Hg (0.34–19.1 mg/kg), Cr (15.1–155 mg/kg) and Cd (0.9–7.1 mg/kg) were also reported in coal collected from the major coal-producing areas of Guizhou Province (Song et al. 2007). In addition, extremely high levels of As (up to 35,000 mg/kg), Sb (up to 3,860 mg/kg) and Hg (up to 12.1 mg/kg) were observed in coals from Southwest Guizhou Province (Ding et al. 2001; Dai et al. 2006b). As summarized in Table 6, the toxic/harmful elements enriched in coal usually include As, Se, Hg, Cd, and Cr. Therefore, domestic coal burning in Guizhou Province is likely accompanied by the release of toxic/harmful elements and the local residents would face the exposure risk of toxic/harmful elements (especially to As, Se, and Hg), in addition to the common pollutants (such as $PM_{2.5}$ and PAHs).

Table 6 Toxic and harmful elements enriched in coals of Guizhou Province

Coal type	Location	Enriched elements	Reference
Late Permian coal	Xingren and Anlong	As, Sb, and Hg	Ding et al. (2001)
Late Permian/ Triassic Coal	Qianxi	F, As, Se, Hg, V, Mo, and U	Zhang et al. (2004)
Late Permian/ Triassic Coal	Guizhou Province and adjacent area	As, Se, Hg, Cd, and U	Li et al. (2006)
Late Permian coal	Xingren	As, Sb, Hg, and Tl	Dai et al. (2006b)
Late Permian coal	Pu'an	As, Se, Hg, Cd, Cr, Pb, Mo, and U	Yang (2006)
Anthracite coal	Southwest Guizhou Province	S, F, As, Hg, Cd, Cr, and Mo	Song et al. (2007)
Late Permian coal	Guiding	S, Se, Cd, Cr, Ni, V, Mo, and U	Dai et al. (2015)
Late Permian coal	Panxian	As, Hg, Se, Cd, Tl, and Mo.	Xie et al. (2017)
Late Permian coal	Xingren	S, Se, Cr, Cu, Co, V, U, and Mo	Duan et al. (2018)

Over the past few decades, with regard to the health problems caused by domestic coal burning, much attention has been paid to endemic fluorosis. Other health problems were seldom concerned, except that of arseniasis (An and Li 2015; Liu et al. 2007). Nevertheless, clues on other health problems can still be found in early research works (Finkelman et al. 1999, 2002; Liu et al. 2007). Associated health problems include endemic arseniasis, endemic selenosis, and mercury poisoning, as well as the pollution of organic compounds such as PAHs (Liu et al. 2008a, b). Because the health problems mentioned above usually have a long incubation period and strong concealment, it is necessary to carry out systematic research works on the suspected population.

4 Conclusion

Unlike endemic fluorosis in other parts of the world, endemic fluorosis in Guizhou Province is mainly caused by the roasted foodstuffs (especially the roasted pepper and corn), rather than the drinking water. Although geological background of F is high in Guizhou Province, it is not necessary the direct cause for endemic fluorosis. Both coal and clay are the probable sources of F in roasted foodstuffs, but the real culprit for endemic fluorosis is the unhealthy lifestyle of the local residents. The important link between F in coal/clay and endemic fluorosis is the roasting processes, which provides F a chance to get into the food chain.

As the primary energy source in China, coal will continue dominating in the near future (Zhao and Luo 2018). Currently, domestic coal burning is still widespread in rural area of China and can still be a serious environmental and health problem. Because domestic coal used in rural houses is usually without any legal or environmental requirements and coal-burning stoves used in rural area are usually without any decontamination devices. As to Guizhou Province and the adjacent areas, things may be worse owing to the enrichment of toxic/harmful elements in the local coal. It is necessary to determine how serious the situation is and find out the possible solution. To say the least, safe exposure thresholds related to the toxic/harmful elements should be established as soon as possible and effective engineering design should be developed to ensure the exposure doses being safe.

Nowadays, great efforts have been devoted to eradicating poverty in China. One of the main causes of poverty is the health problem. Ensuring the people' health is of great importance for the China's current battle against poverty. As people in other developing countries may also suffer from similar health issues related to the domestic coal burning, same environmental and social issues around the world deserve more attention.

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Conflict of Interest On behalf of my co-authors and myself, we declare no conflict of interest to this work.

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