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Pim de Voogt *Editor*

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Foreword

International concern in scientific, industrial, and governmental communities over traces of xenobiotics in foods and in both abiotic and biotic environments has justified the present triumvirate of specialized publications in this field: comprehensive reviews, rapidly published research papers and progress reports, and archival documentations These three international publications are integrated and scheduled to provide the coherency essential for nonduplicative and current progress in a field as dynamic and complex as environmental contamination and toxicology. This series is reserved exclusively for the diversified literature on "toxic" chemicals in our food, our feeds, our homes, recreational and working surroundings, our domestic animals, our wildlife, and ourselves. Tremendous efforts worldwide have been mobilized to evaluate the nature, presence, magnitude, fate, and toxicology of the chemicals loosed upon the Earth. Among the sequelae of this broad new emphasis is an undeniable need for an articulated set of authoritative publications, where one can find the latest important world literature produced by these emerging areas of science together with documentation of pertinent ancillary legislation.

Research directors and legislative or administrative advisers do not have the time to scan the escalating number of technical publications that may contain articles important to current responsibility. Rather, these individuals need the background provided by detailed reviews and the assurance that the latest information is made available to them, all with minimal literature searching. Similarly, the scientist assigned or attracted to a new problem is required to glean all literature pertinent to the task, to publish new developments or important new experimental details quickly, to inform others of findings that might alter their own efforts, and eventually to publish all his/her supporting data and conclusions for archival purposes.

In the fields of environmental contamination and toxicology, the sum of these concerns and responsibilities is decisively addressed by the uniform, encompassing, and timely publication format of the Springer triumvirate:

Reviews of Environmental Contamination and Toxicology [Vol. 1 through 97 (1962–1986) as Residue Reviews] for detailed review articles concerned with any aspects of chemical contaminants, including pesticides, in the total environment with toxicological considerations and consequences.

Bulletin of Environmental Contamination and Toxicology (Vol. 1 in 1966) for rapid publication of short reports of significant advances and discoveries in the fields of air, soil, water, and food contamination and pollution as well as methodology and other disciplines concerned with the introduction, presence, and effects of toxicants in the total environment.

Archives of Environmental Contamination and Toxicology (Vol. 1 in 1973) for important complete articles emphasizing and describing original experimental or theoretical research work pertaining to the scientific aspects of chemical contaminants in the environment.

The individual editors of these three publications comprise the joint Coordinating Board of Editors with referral within the board of manuscripts submitted to one publication but deemed by major emphasis or length more suitable for one of the others.

Coordinating Board of Editors

Preface

The role of *Reviews* is to publish detailed scientific review articles on all aspects of environmental contamination and associated (eco)toxicological consequences. Such articles facilitate the often complex task of accessing and interpreting cogent scientific data within the confines of one or more closely related research fields.

In the 50+ years since *Reviews of Environmental Contamination and Toxicology* (formerly *Residue Reviews)* was first published, the number, scope, and complexity of environmental pollution incidents have grown unabated. During this entire period, the emphasis has been on publishing articles that address the presence and toxicity of environmental contaminants. New research is published each year on a myriad of environmental pollution issues facing people worldwide. This fact, and the routine discovery and reporting of emerging contaminants and new environmental contamination cases, creates an increasingly important function for *Reviews.* The staggering volume of scientific literature demands remedy by which data can be synthesized and made available to readers in an abridged form. *Reviews* addresses this need and provides detailed reviews worldwide to key scientists and science or policy administrators, whether employed by government, universities, nongovernmental organizations, or the private sector.

There is a panoply of environmental issues and concerns on which many scientists have focused their research in past years. The scope of this list is quite broad, encompassing environmental events globally that affect marine and terrestrial ecosystems; biotic and abiotic environments; impacts on plants, humans, and wildlife; and pollutants, both chemical and radioactive; as well as the ravages of environmental disease in virtually all environmental media (soil, water, air). New or enhanced safety and environmental concerns have emerged in the last decade to be added to incidents covered by the media, studied by scientists, and addressed by governmental and private institutions. Among these are events so striking that they are creating a paradigm shift. Two in particular are at the center of ever increasing media as well as scientific attention: bioterrorism and global warming. Unfortunately, these very worrisome issues are now superimposed on the already extensive list of ongoing environmental challenges.

The ultimate role of publishing scientific environmental research is to enhance understanding of the environment in ways that allow the public to be better informed or, in other words, to enable the public to have access to sufficient information. Because the public gets most of its information on science and technology from internet, TV news, and reports, the role for scientists as interpreters and brokers of scientific information to the public will grow rather than diminish. Environmentalism is an important global political force, resulting in the emergence of multinational consortia to control pollution and the evolution of the environmental ethic. Will the new politics of the twenty-first century involve a consortium of technologists and environmentalists, or a progressive confrontation? These matters are of genuine concern to governmental agencies and legislative bodies around the world.

For those who make the decisions about how our planet is managed, there is an ongoing need for continual surveillance and intelligent controls to avoid endangering the environment, public health, and wildlife. Ensuring safety-in-use of the many chemicals involved in our highly industrialized culture is a dynamic challenge, because the old, established materials are continually being displaced by newly developed molecules more acceptable to federal and state regulatory agencies, public health officials, and environmentalists. New legislation that will deal in an appropriate manner with this challenge is currently in the making or has been implemented recently, such as the REACH legislation in Europe. These regulations demand scientifically sound and documented dossiers on new chemicals.

Reviews publishes synoptic articles designed to treat the presence, fate, and, if possible, the safety of xenobiotics in any segment of the environment. These reviews can be either general or specific, but properly lie in the domains of analytical chemistry and its methodology, biochemistry, human and animal medicine, legislation, pharmacology, physiology, (eco)toxicology, and regulation. Certain affairs in food technology concerned specifically with pesticide and other food-additive problems may also be appropriate.

Because manuscripts are published in the order in which they are received in final form, it may seem that some important aspects have been neglected at times. However, these apparent omissions are recognized, and pertinent manuscripts are likely in preparation or planned. The field is so very large and the interests in it are so varied that the editor and the editorial board earnestly solicit authors and suggestions of underrepresented topics to make this international book series yet more useful and worthwhile.

Justification for the preparation of any review for this book series is that it deals with some aspect of the many real problems arising from the presence of anthropogenic chemicals in our surroundings. Thus, manuscripts may encompass case studies from any country. Additionally, chemical contamination in any manner of air, water, soil, or plant or animal life is within these objectives and their scope.

Manuscripts are often contributed by invitation. However, nominations for new topics or topics in areas that are rapidly advancing are welcome. Preliminary communication with the Editor-in-Chief is recommended before volunteered review manuscripts are submitted. *Reviews* is registered in WebofScience™.

Inclusion in the Science Citation Index serves to encourage scientists in academia to contribute to the series. The impact factor in recent years has increased from 2.5 in 2009 to 7.0 in 2017. The Editor-in-Chief and the Editorial Board strive for a further increase of the journal impact factor by actively inviting authors to submit manuscripts.

Amsterdam, The Netherlands Pim de Voogt
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Review on Health Impacts from Domestic Coal Burning: Emphasis on Endemic Fluorosis in Guizhou Province, Southwest China

Jianyang Guo, Hongchen Wu, Zhiqi Zhao, Jingfu Wang, and Haiqing Liao

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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

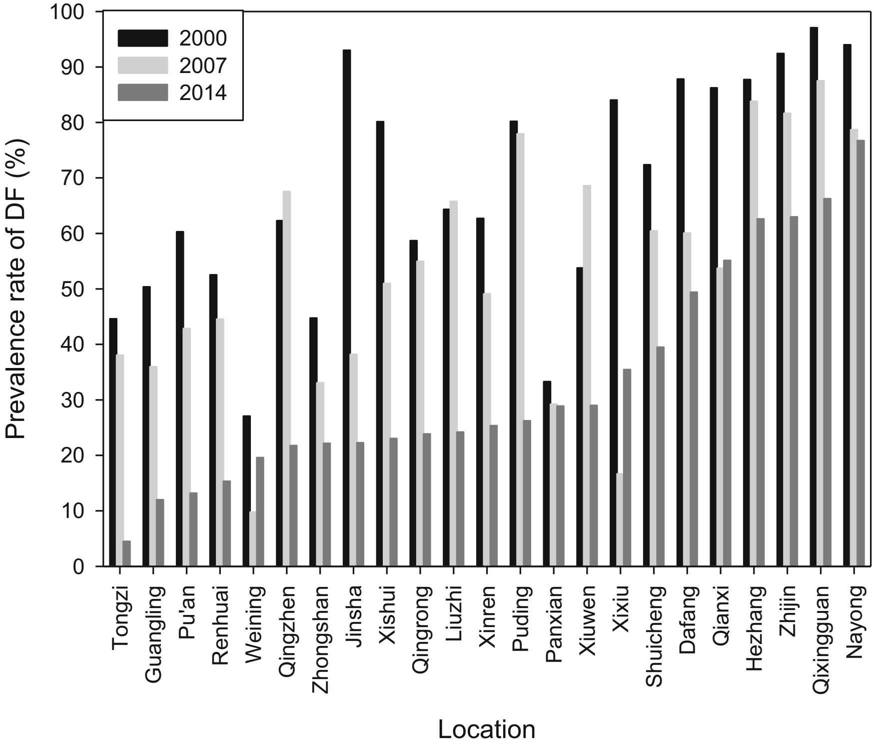

Fig. 1 Prevalence rate of dental fluorosis in 2000, 2007, and 2014 in Guizhou Province (Data source: Zhang et al. 2017a, b)

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

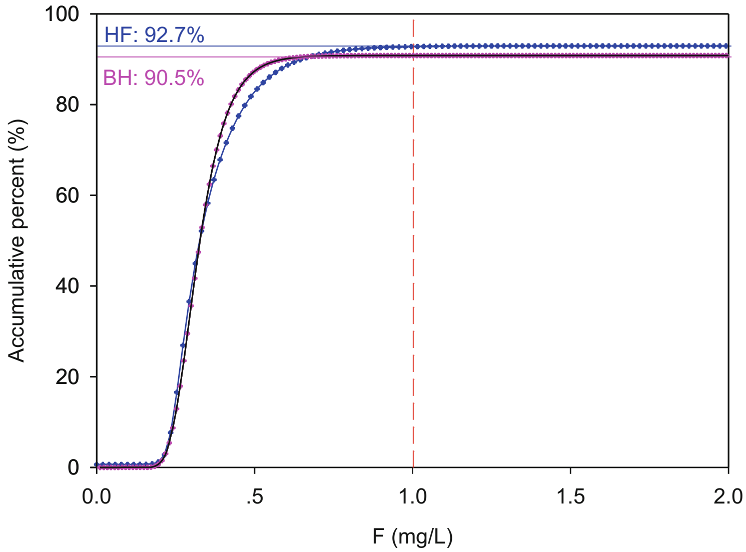

Fig. 2 Contents of F in drinking water sources (Blue line: HF; Pink line: BH) in Guizhou Province

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