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ORIGINAL ARTICLE

Environmental geochemistry and health of fluorine in Chinese coals

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Abstract Fluorine is one of the potential hazardous trace elements in coal. Fluorine may be released into atmosphere mostly during coal combustion process. When the coal is burning indoors without any controlling methods (chimney), the fluorine will pollute the room and is absorbed by food fired (dried) over stoves. Now many people have suffered from fluorosis due to coal burning indoors in Southwest China. In this paper, the concentration, distribution, mode of occurrences and impact factors of fluorine in Chinese coals are analyzed. The environmental impacts and typical features of fluorosis are studied. It is concluded that the value of fluorine in Chinese coals ranges from 20 to 300 mg/kg, and with average value of 122 mg/kg from 5,603 coal samples. It is higher than the average value of the world coals (80 mg/kg). In provincial coalfields of the SW China, the content of F is highest and many people have been affected due to coal combustion indoors.

Keywords Fluorine · Coal · Environmental geochemistry · Health

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Introduction

China is the largest coal producer and consumer in the world (Finkelman 1995). In contrast to the most developed countries, such as the US, where residential coal use constitutes a small fraction of 1% of coal consumption, a substantial portion of China's coal is used for domestic energy needs. It has been estimated that more than 75% of the energy production in China is based on coal (Chen et al. 2004), and more than 75% of China's primary energy needs are supplied by residential coal use (Florig 1997). Coal stoves and small coal boilers provide more than 50% of the energy for urban households and 22% of rural households in China (Florig 1997). Considering that 70% of the population in China resides in rural areas, these data indicate that about 400 million people in China rely primarily on coal for their residential energy needs. Due to the limited petroleum and natural gas reserves, and significant coal reserves (1,000 billion tons) in China, it is likely that this coal-based, relatively cheap energy structure will continue for the foreseeable future (Ni 2000; Xu et al. 2000; Zhong and Yang 2000)

Coal is a complex heterogeneous mixture of organic and inorganic constituents of allothigenic or authigenic origin. Besides major (>1%) and minor (0.1–1%) elements in coal, elements such as As, Se, Hg, F occur commonly as trace elements (<1,000 ppm) associated with both organic and inorganic matters. Trace elements in coals are the major pollution sources (Swaine 2000) and they may become easily accessible not only during coal mining, but also during the storage, transport, cleaning, combustion and other coal preparation processes (Ni 2000; Liu et al. 2002a).

It is estimated that in 2000, global mortality due to indoor air pollution from solid fuels (such as wood, charcoal, crop residues, but mainly coal), was more than 1.6 million (Ezzati et al. 2002). This rate of severe global mortality together with combustion-induced diseases such as pneumoconiosis, dermatosis and other relevant diseases indicate an urgent need to take immediate action against indoor air pollution, especially in developing regions. This is particularly a valid concern in China where elemental toxicosis is epidemic (Zheng et al. 1999; Finkelman et al. 1999, 2002; Liu et al. 2006). The World Bank (1992) estimated that between 400 and 700 million people, particularly women and children, are exposed to severe indoor air pollution in China. A substantial proportion of these people relies on coal for domestic cooking and heating and metalions, gases and organic compounds. In particular, significant numbers of people in SW China have been exposed to high level of toxic trace elements emitted from coal combustion for many years. This chronic poisoning due to burning of high toxic elements bearing coals has been further exacerbated by the consumption of foodstuffs prepared by drying directly over the coal fires (Zheng 1992; Zheng and Huang 1985, 1986).

It is well known that F is an essential trace element for plants, animals and humans. Although F is an essential trace element, it is toxic if taken in excess. Exceeding the tolerable upper intake level per day can lead to fluorosis. In China, there are many people suffering from fluorosis due to coal combustion indoors. F is one of most volatility trace elements during coal combustion and identified as hazardous air pollutants (HAPs) (US National Committee for Geochemistry 1990). Mostly F is emitted from coal during coal combustion (Liu et al. 1999a). Detailed knowledge of toxic trace elements, F, in coals, therefore, is required to understand its behavior during coal processing and utilization, and consequent environmental impacts. In this paper we will focus on the geochemistry of F in Chinese coals, document their concentrations, distributions, modes of occurrence and volatility during coal combustion, and then discuss F toxicosis in Southwest China.

Geochemistry

Content and distribution of fluorine in Chinese coals

The average content of fluorine in Chinese coals

The distribution of F in Chinese coals has been studied (Zheng and Huang 1988; Lu 1996; Liu et al. 1999b, c, 2000; Ren et al. 1999; Luo et al. 2001, 2004; Chen and Tang 2002; Tang and Huang 2004). Zheng and Huang (1988) first collected 337 coal samples, and reported the average F content in Chinese coals, as 200 mg/kg, much more than the world average of 80 mg/kg (Swaine 1990). Ren et al. (1999) reported that the F content was 100–3,600 mg/kg in eight coal samples mainly collected from Xishan coal mine in Shanxi Province; Luo et al. (2004) collected about 300 Chinese coal samples to study the F content and distribution. The results show that the F concentration in most Chinese coals is less than 200 mg/kg, but it is higher than the global average concentration of 80 mg/kg, especially in Guizhou, Hubei, Yunan, Hunan provinces where F content in coal is as high as 3,000 mg/kg. Tang and Huang (2004) studied F content in Chinese coals and summarized the results of other scientists; the results concluded that most Chinese coals are low F coals (20–300 mg/kg). The average F content in Chinese coals is about 186 mg/kg (collected from 1,069 coal samples), much higher than that in the world (80 mg/kg). Liu et al. (1999a) collected over 3,000 samples from Yanzhou coal, and the average F content is 115 mg/kg, lower than the 186 mg/kg and slightly higher than 80 mg/kg. According to all the studied data, the F content in Chinese coal commonly ranges from 20 to 300 mg/kg; it may be much higher in Guizhou, Hubei, Chongqi, Sichuan, Yunnan and Shaanxi coals, and about average value of 100 mg/kg, in other coalfields.

In this study, the majority of data of F contents was collected the studies reported in open published literatures, combined with F contents tested in authors Lab, 5,603 coal samples were calculated according to the average value of F in different provincial coals. And the minimum, maximum and average values in Chinese coals were analyzed in this study. The concentrations in different provincial coalfields (Fig. 1), different accumulating-coal ages (Table 1) and coal ranks (Table 2) were studied. In the study, the average value of fluorine in 5,603 Chinese coal samples is 122 mg/kg; it is higher than the average value of the world coal. However, it is the lowest value than previous values calculated. The impact factor that there are 3,710 coal samples collected from Shandong was analyzed. If 100 coal samples in Shandong are used to calculate the average F value, the value will be 141 mg/kg. Hence, it is a considerable and reasonable average value for Chinese coals.

The content and distribution of fluorine in different provincial coals

According to previous studies (Zheng and Huang 1988; Zheng et al. 1999; Luo et al. 2004; Liu et al. 1999a;

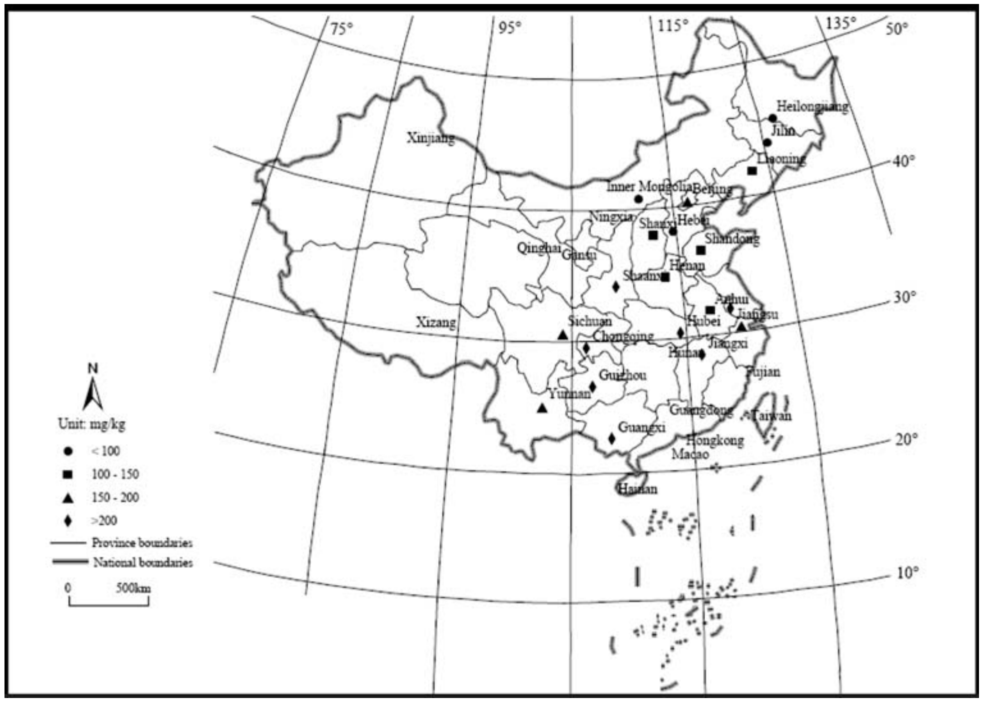

Fig. 1 The content and distribution of fluorine in Chinese coals

Table 1 Distribution of mercury concentrations in Chinese coals from different coal-forming periods (μg/g)

Geological period	Range (μg/g)	Arithmetic mean (μg/g)	Number of samples
Carboniferous and Early Permian (C–P_{1-2})	2–3,600	129	2,800
Permian (P_{1-2})	2–4,000	187	680
Late Triassic (T_3)	67–123	83	5
Jurassic (J_{1-2})	10–1,176	224	102
Late Devonian (D_3)	130–2,200	370	26

C–P_1 represents coal ages in North of China. P_{1-2} represents coal ages in South of China. T_3, J_{1-2} and J_{1-2} represent coal ages in all over China

Table 2 Distribution of mercury concentrations in Chinese coals from different coal ranks (μg/g)

Coal ranks	Range (μg/g)	Arithmetic mean (μg/g)	Number of samples
Stone coals	130–2,200	370	26
Lignite	83–1,176	268	70
Bituminous	2–3,600	150	4,305
Anthracite	13–4,000	97	202

The stone coal, distributed in Guizhou, Hubei and Yunnan, is easy to be mined for burning indoors because it is overlaid by shallow rocks or opened in air

Tang and Huang 2004), low-F coals are mainly distributed in stable platforms in North China and Northwest China. F content in all platform areas, where magmatic activity was less active, is comparatively low, about 20–300 mg/kg. For example, F content of most Permo–Carboniferous coals is less than 200 mg/kg in North China Plate, in the Northwest China and Yangzi Plate, mostly about 20–100 mg/kg. Luo et al. (2004) had studied the distribution patterns in Chinese coals, the results showed that medium-to-high F coals are mainly in Guizhou–Yunnan, where there was more volcanic activity and magma intrusion during and after coal-forming period than the North China Plate. Super high-F coals including stone coal in igneous rock in geosynclines region South Qinling Mountain and Late Permian coals of Longtan Formation occur in the southwest of Yunnan and Guizhou, but not all Late Permian coals in Yunnan and Guizhou are high-F coals. And super high F coals have resulted in serious indoor air pollution and human health problems in Daba Area of South Qinling Mountain and Yunnan–Guizhou. Previous studies (Luo et al. 2004; Zheng et al. 1999; Tang and Huang 2004; Ren et al. 1999; Liu et al. 1999a) have concluded that in geosyncline areas and on the edge of platform areas (e.g., Southwest China), there have been numerous tectonic movements, especially since Cambrian. Volcanism brought abundant F into coal basins and coal seams. F is more enriched by absorption in organic matter and clay in coal-bearing strata. It is very clear that volcanic activity and magma intrusion are the primary reasons for F-rich coals in Daba Area, Dabie Mountain Area and Southwest China, where there were more volcanic activity and magmatic intrusion during and after coal-forming periods than the on North China Plate. The F content in coal correlates negatively with the distance from igneous rock in geosyncline areas and on the edge of platform areas (Luo et al. 2004; Zheng et al. 1999).

The average content of fluorine in different geological age coals

F content in Chinese coals is also related with coal-forming periods and circumstances (Zheng et al. 1999; Luo et al. 2004; Liu et al. 1999a). According to main geological ages in Chinese coal, the coal samples were classified into several geological periods (Table 1): Carboniferous and Permian (C–P), Permian (P_{1-2}), Late Triassic (T_3), Jurassic (J_{1-2}) and Late Devonian (D_3). And the C–P_1, J_{1-2} coalfields mainly distribute in northern China (such as Shangdong, Hebei, Anhui), the P_{1-2} and T_3 coalfields mainly in southern China (such as Jiangsu, Zhejiang, Yunnan, sichuang, Guizhou), and Coals of D_3 period are mainly stone coals (Hubei, Guizhou).

In some literatures, authors do not explain that from which geological ages the samples were collected; we only use the samples confirmed their ages and coal ranks to calculate the average value (Tables 1, 2). It can be seen from Table 1, remarkable varieties of F contents occur in different coal-forming ages in Chinese coals. The highest average F concentration is in D_3 coals (370 μg/g), and the lowest in T_3 (83 μg/g), and the average F concentrations of different coal-forming periods increases in the following order: D_3 (370 μg/g) > J_{1-2} (224 μg/g) > P_{1-2} (187 μg/g) > C-P_{1-2} (129 μg/g) > T_3 (83 μg/g).

The average content of fluorine in different coal ranks

The F content is impacted by the rank of coalification. F content in coals in the platform may decrease with their increasing metamorphic degree. According to the Table 2, it is not obvious that the ranges of F in different coal ranks change, and it can been concluded that F content of anthracites, generally speaking, is from 13 to 4,000 mg/kg, with a average value of 97 mg/kg and the average value of high rank bituminous coal is about 150 mg/kg. Highest average value is stone coals with an average value of 370 mg/kg. However, the ranges may not be obvious in different coal ranks, the order of average value in different coal ranks is very obvious and the order is stone coal > lignite > bituminous > anthracite. It can be concluded that the average value of F in coals increases with the coal rank decreasing.

The occurrence of fluorine in coals

The mode of occurrence of an element is a description of the manner in which an element is chemically bound in the host material. In coal, elements can be associated with the inorganic constituents (minerals) occurring as or in silicates, sulfides, oxides, carbonates, phosphates, sulfates, etc., or with the organic constituents in a variety of less well-defined ways. The element's modes of occurrence can strongly influence its behavior during coal cleaning, weathering, leaching, combustion and conversion (Finkelman 1989, 2004). Some elements such as arsenic, chromium and nickel are present in coal in both organic and inorganic forms. These different modes of occurrence will cause the element to behave differently during coal cleaning and coal combustion and thus will have different environmental and human health impacts (Finkelman et al. 2002; Finkelman 2004).

The mode of occurrence of F in coals varies significantly. F may occur as inorganic as well as organic compounds (Godbeer and Swaine 1987; Swaine 1990; Finkelman 1994; Liu et al. 1999c, 2000, 2002b; Zheng et al. 1999). Several previous discussions (Swaine 1975; Durie and Schafer 1964) on F in coal suggested that F may occur as fluorapatite. Based on the relative scarcity of apatite in coal, Finkelman (1981) expressed doubt that the mode of occurrence could account for most of the F in coal. F can substitute readily for OH^- in clays and mica (Wedepokl 1972) and may be associated with these minerals in coal (Finkelman 1981). According to the study of Beising and Kirsch (1974), much of the F in coals is commonly associated with illite. Bethell (1962) suggested that some F in coal might be ionically bound or occurred as fluorite. Finkelman (1981) concluded that the problem of the mode of occurrence of F remains unresolved. It is possible that this element has a very complex mode, occurring in apatites, fluorites, amphiboles, clays and mica. In individual coals in form may dominate over the others. And he estimated level of F confidence is 5 in coal. Laboratory float-sink for F was carried out (Liu et al. 1999a; Miller and Given 1978). The results showed the apparent organic association of F in the samples. However, Zubovic (1976) pointed out that the low ionic potential (<1) of F argues against an organic association.

The mode of occurrences of F in coal has been studied by Liu et al. (1999a) in the recent years; according to our results, F in Chinese coals is primarily associated with the inorganic constituents, some of F in coal samples is associated with organic matter.

Environmental behavior and health

Volatility and emission

F is one of the most toxic and volatilized elements present in coal. During combustion, F is emitted as HF, SiF_4 and CF_4 (Lu 1996; Jeng et al. 1998; Yan et al. 1999; Liu et al. 1999b, c; Luo et al. 2002; Notcutt and Davies 2001). HF is one of the most serious pollutions adversely affecting both plants and animals, the toxicity being about 10–100 times higher than that of SO_2 (Jeng et al. 1998; Piekos and Paslawska 1999; Notcutt and Davies 2001).

The US National Environmental Trust reported that the fluorine emission is about 8% of total power plant pollutant emissions, following HCl (69%) and H_2SO_4 (21%) and becomes the third important pollutant. It has been shown that about 85–90% of the F present in coal is emitted as HF, either as vapor or absorbed on fine ash particles when the temperature is above 850°C (Guo et al. 1994; Liu et al. 1999b, c, 2006).

The air, water and soil around the Hubei coal-burning power plant have all been affected by F pollution to varying degrees, the power plant having been identified as the source of the emission (Tang et al. 1999a, b). Luo et al. (2002) have measured the annual coal consumption in China for power plants and domestic heating as about 8×10^9 t. If the lower range of F concentration (100 g/t) is used, in atmosphere it is about 66,000 t, which is more than twice that of the US.

Environmental impact on human health in China

The health problems caused by F released during coal combustion are more extensive than those caused by arsenic. More than 14 provinces (including the northwest part of Guizhou, the south part of Sichuan and northeast of Yunnan, the southwest part of Hubei, the southeast of Sichuan, and the northwest part of Hubei) and more than 30 million people in China suffer from various forms of fluorosis, and about 15 million people have been diagnosed as having fluorosis (Zheng et al. 1999; Finkelman et al. 1999, 2002; Ando et al. 1998).

According to the data reported (Zheng et al. 1999), more specifically, around 10 million people in Guizhou Province and surrounding areas suffer from various forms of fluorosis. About 720,000 of 740,000 residents of Zhijin County of Guizhou Province have fluorosis. In Beimen Zheng and Hua chun in Guizhou, about 78% of the inhabitants are diagnosed as having skeletal fluorosis including osteosclerosis. Almost every family in this district has members suffering. In Xiaotang Zheng, Pengshui, Sichuan province, among 5,633 residents, there are about 98% of the residents suffering from tooth enamel mottling (dental fluorosis). Zheng et al. (1999), Finkelman et al. (1999) documented that 97% of people older than 8 years suffer from the devastating dental fluorosis.

In most of the fluorosis areas, the main symptom of this disease is dental fluorosis, which has little impairment to other parts of the body, for example, skeletal fluorosis with limited movement of the joints, and outward manifestations such as knock-knees, bow legs and spinal curvature. With more severe exposure, however, the effects of fluorosis impairment are quite serious.

The authors investigated a series of fluorosis in the Zhijin County, Guizhou and the Enshi, Hubei. In a survey from one elementary school, among 57 students, only 1 student was found to be free of dental fluorosis, while 99% of the student population was diagnosed as having dental fluorosis. During this investigating, the authors also found that the youngest patient was about 1 year old. In adults of 50 years old, most of them have suffered from not only dental fluorosis, but also from skeletal fluorosis. Some of the residents in the areas have not been able to walk without help due to various serious forms of skeletal fluorosis.

The cause of fluorosis in SW China is that mostly residents cook by burning high F coal without furnace. A substantial proportion of these people relies on coal for domestic cooking and heating and thereby exposed to particulates, metalions, gases and organic compounds. Significant numbers of people in the studied area have been particularly exposed to high level of toxic trace elements F emitted from coal combustion for many years. This chronic poisoning due to burning of high toxic element-bearing coals has been further exacerbated by consuming the crops dried directly over the coal fires (Zheng and Huang 1986, 1988; Zheng et al. 1999). Because of their small grain size, these toxic elements easily enter into the human bodies through respiratory tract or food chain, and accumulate in the bodies. When the concentrations of trace elements are higher than the standard value that people need, they will affect human's normal metabolism and destroy some physiological functions, which produce pathological phenomenon for human health. These health problems are unfortunately severe (leading to death) and widespread (affecting many millions of people).

Conclusion

(1) F is one of toxic elements in coal and F has high volatility during coal combustion. In SW China, coal is burning without chimney indoors, and

room air and food will be polluted by fluoride and this chronic poisoning due to burning of high toxic elements bearing coals has been further exacerbated by consuming the crops dried directly over the coal fires high-F coal combustion, and fluorosis have been occurred and many people have been suffering from the fluorosis for many years.

(2) Compared with F contents of coal of other countries, the content of F in Chinese coals ranges from 20 to 300 ppm, particularity in SW China, and the F content of Chinese coal is higher. The reason of this observation is that during the period of coal formation in southwestern China, crustal movement was active and volcanic activity occurred frequently, allowing a large amount of F in the gaseous state to be absorbed by plants or deposited with plants as F-rich volcanic ash.

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