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REVIEW PAPER

Groundwater Arsenic and Fluoride and Associated Arsenicosis and Fluorosis in China: Occurrence, Distribution and Management

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Abstract
Arsenic and fluoride are two natural components affecting human health. Long-term exposure to arsenic and fluoride, mainly through drinking water intake, can lead to arsenicosis and fluorosis. In this paper, we summarized the distribution and hydrochemical characteristics of high-arsenic and high-fluoride groundwater, and reviewed the arsenicosis and fluorosis distribution due to consumption of high-arsenic and high-fluoride groundwater in China. The results show that there are 20 major provinces//autonomous regions (about 60%) in China suffering from high-arsenic groundwater, and these high-arsenic groundwater provinces are mainly located in the fluvial/alluvial-lacustrine plains and basins located in arid/semi-arid regions and alluvial plains/basins and river deltas in humid/semi-humid regions. Drinking water arsenicosis has been found in 13 provinces/autonomous regions, with Shanxi and Inner Mongolia being the two most seriously affected areas. High-fluoride groundwater is widely distributed in north, northeast and northwest parts of China, occurring mainly in shallow groundwater. Fluorosis has been found in 29 provinces/autonomous regions. With the continuous implementation of water quality improvement projects, drinking water fluorosis and arsenic poisoning have been effectively controlled. However, the long-term maintenance and operation of the water quality improvement project need to be further strengthened.

Keywords High-arsenic groundwater · High-fluoride groundwater · Arsenicosis · Fluorosis · Groundwater quality

Introduction

Drinking water quality is closely related to human health and the sustainable development of human society (Li and Wu 2019a, b; Wu et al. 2020). Arsenic and fluoride are two most commonly found elements affecting the suitability of water for drinking purpose. Arsenic is a natural component of the earth's crust, existing often as arsenates, sulfides or metal arsenides (Selinus et al. 2005). It is widely distributed in the environment, and is highly toxic in its inorganic form. In groundwater, it is mostly present as arsenate (+5), but in anaerobic conditions, it is likely to be present as arsenite (+3) which has higher toxicity (WHO 2017). Long-term exposure to arsenic, mainly through drinking water and consumption of food, can lead to chronic arsenic poisoning, including skin lesions, peripheral neuropathy, diabetes, cardiovascular diseases and cancer (Cheng et al. 2016; Morales et al. 2000). Natural arsenic contamination is a global phenomenon, with known levels and regions increasing rapidly in recent years. However, its distribution is almost certainly incompletely mapped. High-arsenic groundwater is the largest source of arsenic exposure, which has caused direct harm to human health in many parts of the world (Guo et al. 2014; Hodges 2017; Ravenscroft 2007; Ravenscroft et al. 2009; Selinus et al. 2005; Yu et al. 2007). Arsenicosis is a serious environmental chemical disease in the world due to exposure to over-load arsenic. Drinking water arsenicosis has been reported in more than 20 countries, including India, Bangladesh, Thailand, China, Chile, Argentina, the United States, Canada and Mexico (Chung et al. 2014;

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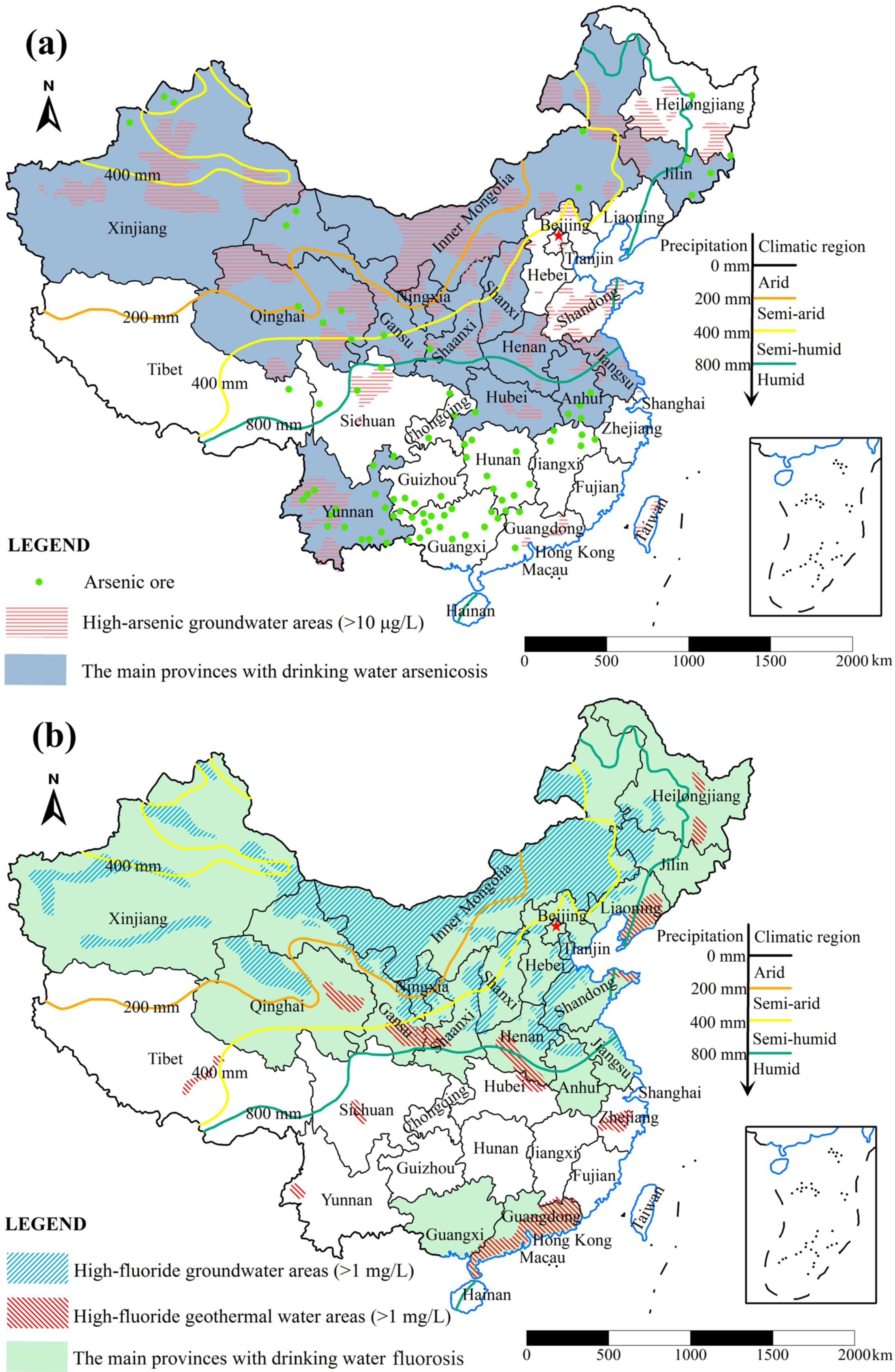
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Jiangxi
Fujian
Guangdong
Hong Kong
Macau
Taiwan
Zhejiang
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Precipitation
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0 mm
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LEGEND
Arsenic ore
High-arsenic groundwater areas (>10 μg/L)
The main provinces with drinking water arsenicosis
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(b)
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Xinjiang
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Tibet
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Sichuan
Yunnan
Guizhou
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Hainan
Inner Mongolia
Ningxia
Gansu
Shaanxi
Shanxi
Henan
Hubei
Chongqing
Hunan
Jiangxi
Fujian
Guangdong
Hong Kong
Macau
Taiwan
Zhejiang
Shanghai
Anhui
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0 mm
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Semi-humid
800 mm
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LEGEND
High-fluoride groundwater areas (>1 mg/L)
High-fluoride geothermal water areas (>1 mg/L)
The main provinces with drinking water fluorosis
0 500 1000 1500 2000km

◂**Fig. 1** Distribution of **a** high-arsenic groundwater and **b** high-fluoride groundwater [modified from Wang et al. (2018), Wen et al. (2013) and Zhang et al. (2017)] and the main provinces of drinking water **a** arsenicosis and **b** fluorosis in China

He et al. 2020; Smedley and Kinniburgh 2002). China is one of the countries with serious drinking water arsenicosis. Since 1980, large-scale outbreaks of drinking water arsenicosis have been reported in Xinjiang, Inner Mongolia, Shanxi and other regions. Jin et al. (2003) reported an investigation of drinking water arsenicosis and revealed the occurrence of drinking water arsenicosis in 8 Chinese provinces and 40 counties. Rodríguez-Lado et al. (2013) developed a statistical risk model, and they estimated that 19.6 million individuals are at risk of being affected by consumption of arsenic-contaminated groundwater in China.

Fluoride is an essential element for humans, which is the 13th most abundant trace element in the crust. However, the optimal intake is within a very narrow range. Low concentration of F^- (< 0.5 mg/L) in drinking water may induce dental decay, while high F^- may cause dental fluorosis (1.5–4.0 mg/L) and skeletal fluorosis (> 4 mg/L; Li et al. 2019; Selinus et al. 2005). Consumption of high-fluoride groundwater is the major exposure pathway of fluoride. Fluorosis is a worldwide health problem, and usually presents as endemic fluorosis, which is quite widespread in many areas. The major types of fluorosis include drinking water fluorosis, burning coal fluorosis and drinking tea fluorosis, among which the drinking water fluorosis is the most common (Sun et al. 2013; Vithanage and Bhattacharya 2015). Long-term intake of high-fluoride groundwater can lead to drinking water fluorosis, which has been reported in many countries, especially in low-income countries, such as India, China, Tanzania, Mexico, Argentina, and South Africa (Adimalla and Li 2019; Adimalla et al. 2019; Ayoob and Gupta 2006; Li et al. 2014; Selinus et al. 2005; Sun et al. 2013; Subba Rao 2017; Vithanage and Bhattacharya 2015; Wang et al. 2012). In China, drinking water fluorosis has been found in 29 provinces/autonomous regions (Wang et al. 2012; Wen et al. 2013).

To give a clear picture of the occurrence and distribution of arsenic and fluoride associated arsenicosis and fluorosis in China and to provide necessary information for researchers and decision makers to take necessary actions against the diseases, this paper summarizes the distribution and hydrochemical characteristics of high-arsenic and high-fluoride groundwater in China, and reviews the arsenicosis and fluorosis distribution caused by long-term intake of high-arsenic and high-fluoride groundwater in China. This study will be helpful for groundwater quality protection and management in China, and will also benefit the international scientific community by providing them with a lot of useful information of groundwater arsenic and fluoride.

Methodology

In this study, the data of drinking water arsenicosis and fluorosis in China were mainly collected from the *Health Statistics Yearbook of China*, edited by National Health Commission of the P. R. China, which is an annual publication reflecting the development of China's health and family planning and the health status of residents (https://www.nhc.gov.cn/zwgkzt/tjnj/list.shtml). This book contains the statistical data of the health care development and the current health level in 31 provinces/autonomous regions. The data of arsenicosis and fluorosis were collected annually by the health departments and research centers for endemic disease control of provinces//autonomous regions. For this study, the data of patients, villages affected by drinking water arsenicosis and fluorosis, and villages undergoing water quality improvement were collected from the *Health Statistics Yearbook of China*, and the data from 2002 to 2018 were used. In addition, data of arsenic trioxide production in China and the world were obtained from USGS (https://www.usgs.gov/centers/nmic/arsenic-statistics-and-information).

Results and Discussion

Occurrence and Distribution of High-Arsenic Groundwater

China is a typical high-arsenic groundwater region. High-arsenic groundwater has been mainly discovered in 20 out of 34 provinces/autonomous regions, and they are Anhui, Beijing, Gansu, Guangdong, Hebei, Henan, Hubei, Inner Mongolia, Jilin, Jiangsu, Liaoning, Ningxia, Qinghai, Shandong, Shanxi, Shaanxi, Sichuan, Taiwan, Xinjiang, and Yunnan (Fig. 1a). In these regions, high-arsenic groundwater mainly occurs in the fluvial/alluvial-lacustrine plains and basins located in arid/semi-arid regions, such as Datong Basin, Junggar Basin, Huhhot Basin, Guide Basin, Hetao Plain and Yinchuan Plain, and alluvial plains/basins and river deltas in humid/semi-humid and tropical regions such as Songnen Plain, Taiyuan Basin, Yuncheng Basin, the Yellow River alluvial plain, the Huai River alluvial plain, Yangtze River Delta, Yellow River Delta and Pearl River Delta (Fig. 2a; Guo et al. 2014; Wen et al. 2013). In arid and semi-arid basins and plains, high-arsenic groundwater mainly occurs in shallow Quaternary sedimentary aquifers with thick fluvial/alluvial-lacustrine deposits, and groundwater is usually a characterized with reducing condition, high concentrations of Fe, Mn, F^-, HCO_3^-, Na^+, and TDS, neutral-alkaline pH,

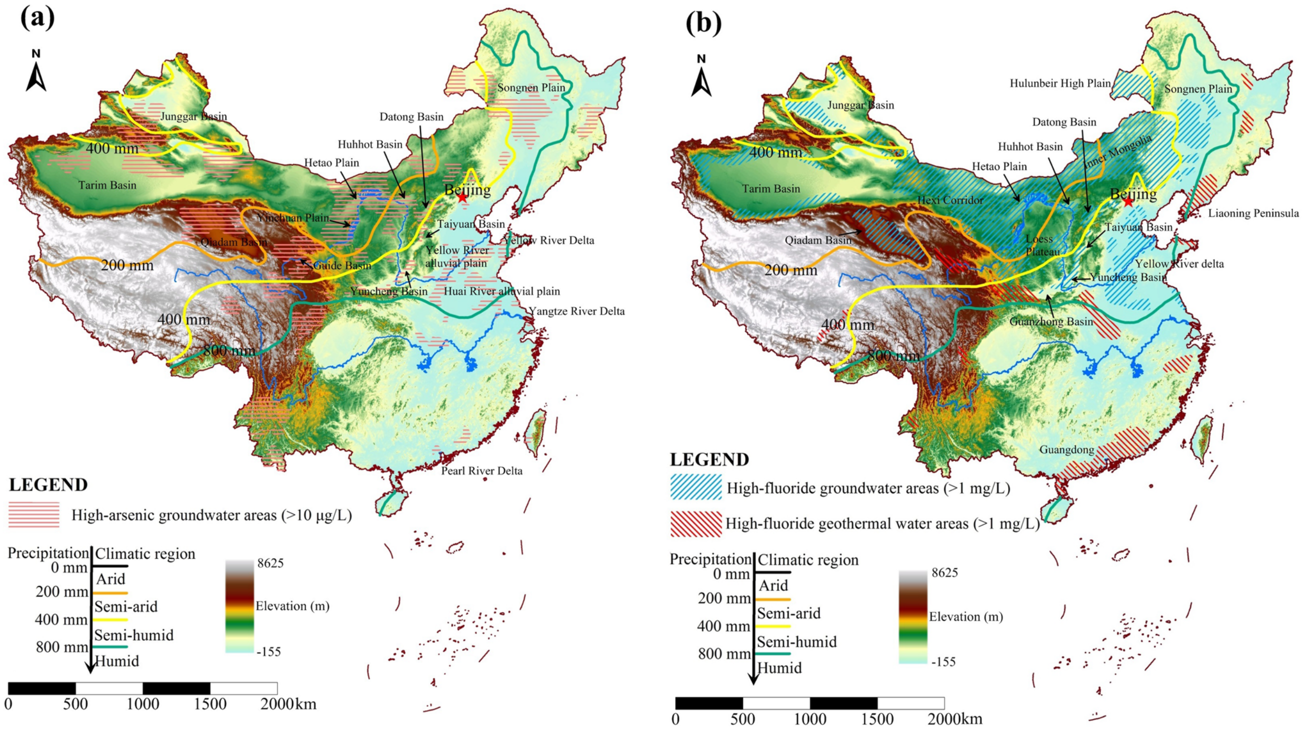

Fig. 2 Distribution of **a** high-arsenic groundwater and **b** high-fluoride groundwater in the major basins/plains in China

low NO_3^-. In alluvial plains and river deltas in humid/semi-humid and tropical regions, high-arsenic groundwater is characterized by reducing conditions, high concentrations of Fe, Mn, F^- and TDS (Guo et al. 2007a, 2014).

Furthermore, the high-arsenic groundwater discovered in Guide Basin located in the northeastern Qinghai-Tibet plateau and Tengchong area in Yunnan Province is related to geothermal water (Wang et al. 2018). Host rock leaching and magma degassing are identified as the major source for the high concentrations of arsenic, and the arsenic levels were found to be positively associated with water level depth and temperature of the geothermal water. Geothermal water with high arsenic concentration can contaminate other aquifer systems by naturally upward movement of geothermal fluids and anthropogenic activities (Guo et al. 2007b, 2015; He and Charlet 2013). In the development and utilization of high-arsenic geothermal water, some aquifers may be contaminated and become potential high-arsenic aquifers.

Most arsenic environmental problems are the result of mobilization under natural conditions such as weathering, minerals-water interaction, biological activities, geothermal activities and volcanic emissions (Guo et al. 2014; Smedley and Kinniburgh 2002). The main geochemical mechanisms of natural arsenic mobilization can be summarized as follows: (1) Oxidation of sulfide minerals: In oxidizing environment, oxygen triggers the oxidization and dissolution of sulfide minerals (arsenopyrite, pyrite and pyrrhotite), releasing arsenic into groundwater. It requires abundant oxygen and water, so the process occurs in localized regions where groundwater level is rising and mining activity is occurring (Lin and Wang 2017; Smedley and Kinniburgh 2002). (2) Dissolution of metal oxides containing arsenic in reducing conditions: Iron/manganese oxides adsorbing arsenic are reduced and dissolved under the influence of organic matter decomposition, and then As(III) is released into groundwater. High-arsenic groundwater is closely associated with reducing conditions, particularly in alluvial and deltaic environments (Ravenscroft 2007; Wen et al. 2013). (3) Desorption in the alkaline conditions: The adsorption capacity of most iron oxides decreases with the increase of pH values. In the condition of high pH (>8.5), arsenic adsorbed on the surface of oxides is easily desorbed (Guo et al. 2007a; Selinus et al. 2005). (4) Anion competitive adsorption: Arsenic suffers competitive adsorption with anions co-existing with arsenic such as PO_4^{3-}, HCO_3^-, SO_4^{2-}, and NO_3^- (Gao et al. 2011; Guo et al. 2014). (5) Geothermal arsenic mixed with groundwater: Geothermal arsenic is formed by high temperature leaching of silicate rocks. The mixing of high-arsenic geothermal water and underground fresh water causes arsenic contamination (Ravenscroft et al. 2009; Smedley and Kinniburgh 2002). (6) Microbial metabolisms: microorganisms play an important role in the form and release of arsenic, and they can be important catalysts in the oxidation of arsenite, reduction of arsenite, methylation and volatilization of different forms of arsenic. Microbes are fueled by organic matter in the aquifers and catalyze the redox reaction (Guo et al. 2014; Selinus et al. 2005). (7) Irrigation and evaporation: irrigation water can carry considerable oxidants and fresh organic matter from the surface into the groundwater. Evaporation can increase arsenic concentrations. It usually occurs in the shallow groundwater (Selinus et al. 2005; Xie et al. 2015). The mechanisms of arsenic mobilization are intricate and the exact mechanisms are still poorly understood. A mechanism is difficult to explain arsenic mobilization, and the release mechanism of arsenic in groundwater can be explained more reasonably by various methods.

Except the geogenic reasons for the high-arsenic groundwater, human activities can be another important factors responsible for groundwater arsenic contamination. China is rich in arsenic mine resources, accounting for around 70% of total arsenic storage in the world (Fig. 1a; Zhang et al. 2017). China is also the leading producer of arsenic metal in the world (USGS 2019). In 1994, arsenic trioxide production was 13,000 metric tons, accounted for around 30% of the world production. The percentage continues to rise every year. In 2018, arsenic production reached 24,000 metric tons, accounted 68.6% of the world production (Fig. 3). High arsenic production is supported by large-scale mining activities. Arsenic pollutants generated by arsenic mining can contaminate groundwater, surface water and soil easily. Arsenic ores drainage is a common type of pollution in mining districts, and is also one of the most important anthropogenic sources (Han et al. 2003; Lefticariu et al. 2019; Zhang et al. 2014). Lots of arsenic ores are being mined in southwest China where karst aquifers are widely developed. As a result of human arsenic mining activities, karst groundwater had been reported to be seriously contaminated with arsenic, in particular in areas of Guizhou Province and Guangxi Province (Tao et al. 2012; Zhang et al. 2014, 2017). The acute arsenic poisoning with increasing of mining, smelters and chemical industries has been repeatedly reported in recent years (He and Charlet 2013). One of the most serious incidents occurred in Yangzonghai Lake in Yunnan Province, China in 2008. Yangzonghai Lake, a large plateau lake with an area of 31 km^2, was contaminated due to the illegal arsenic discharge by chemical company (Liu et al. 2014). The concentration of arsenic in lake reached 0.128 mg/L, making it impossible to use the lake water for drinking (Zhu et al. 2016). The high-arsenic groundwater caused by human activities poses a serious threat to the health of local residents.

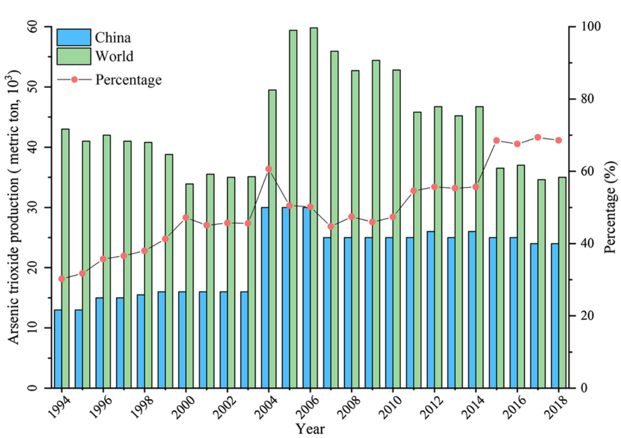

Fig. 3 Arsenic trioxide production from 1994 to 2018: China versus the World (Data from USGS)

Occurrence and Distribution of High-Fluoride Groundwater

High-fluoride groundwater is widely distributed in China. More than 68 million population are exposed in the environment with high-fluoride groundwater, particularly in the provinces/autonomous regions in north, northeast and northwest parts of China (Table 1, Fig. 1b). In China, high-fluoride groundwater is widely discovered in arid and semi-arid regions with shallow depth (< 50 m, Li et al. 2018a). Similar to arsenic, fluoride in groundwater generally has two sources: geogenic sources and anthropogenic sources (Li et al. 2019). High-fluoride groundwater pollution caused by human activities is mainly related to waste discharge from steel industries and chemical fertilizer and pesticide in agricultural activities (Fuge 2019). Geogenic sources, however, are the main sources of high-fluoride groundwater, and high-fluoride groundwater can be classified into three types according to the occurrence conditions: shallow high-fluoride groundwater; deep high-fluoride groundwater and high-fluoride geothermal water (Wang et al. 2018; Wen et al. 2013). Shallow high-fluoride groundwater is the most widely distributed, mainly distributed in the northeastern China (Central and western parts of Songnen Plain, the western Songliao Plain, eastern Inner Mongolia and Hulunbeir High Plain), northwestern China (Junggar Basin, Tarim Basin, Qiadam Basin, Hexi Corridor and Guanzhong Basin), northern China (Datong Basin, Taiyuan Basin, Yuncheng Basin, Huhhot Basin, Hetao Plain and the Yellow River delta, Fig. 2b, Ren and Jiao 1988; He et al. 2010; Wang et al. 2018). In some regions of the Chinese Loess Plateau, high-fluoride groundwater is also found in loess aquifers (Jia et al. 2019; Li et al. 2018b, 2019). Arid and semi-arid climate, strong evaporation, ion exchange and slow groundwater flow are important factors responsible for fluoride enrichment in shallow groundwater. Deep high-fluoride groundwater is mainly found in coastal plains of Bohai Sea and Yellow Sea, and it is also found in alluvial plains/basins where shallow high-fluoride groundwater is distributed such as Yuncheng Basin, Junggar Basin and Tarim Basin (Currell

Table 1 Overview of arsenic (As) and fluoride (F^-) in groundwater in China

Provinces	Area	Population	Population exposed (10^4)[b]		Max concentration detected[c]		Climate	Groundwater supply ratio (%)[d]
	(10^4 km^2)	(10^4)[a]	As	F^-	As (μg/L)	F^- (mg/L)		
Beijing	1.68	2154		56.7	143	8.0	Semi-humid	41.48
Tianjin	1.13	1560		272		4.0	Semi-humid	15.49
Hebei	18.77	7556		873.3		7.0	Semi-arid and semi-humid	58.17
Shanxi	15.63	3718	22.3	457.6	1300	12.5	Semi-arid and semi-humid	40.38
Inner Mongolia	118.3	2534	38.2	490.6	1932	15.5	Arid and semi-arid	46.17
Liaoning	14.59	4359		169.5	25	16.0	Semi-humid	40.91
Jilin	18.74	2704	15.3	190.3	360	10.0	Semi-humid and humid	35.56
Heilongjiang	47.3	3773		106.3	360	10.0	Semi-humid and humid	44.43
Jiangsu	10.26	8051	7.3	440.1	324	7.0	Humid	1.33
Zhejiang	10.2	5737		17.1	125	3.0	Humid	0.46
Anhui	13.97	6324	13.1	550	1150	4.1	Humid	10.43
Fujian	12.13	3941		10.3		13.5	Humid	2.35
Jiangxi	16.7	4648		5.6		8.2	Humid	3.19
Shandong	15.38	10,047		987	440	11.0	Semi-humid	36.81
Henan	16.7	9605	3.3	1287.6	258	4.0	Semi-humid	49.45
Hubei	18.59	5917	7.7	34.2	175	3.7	Humid	2.63
Hunan	21.18	6899		3.4		8.4	Humid	4.24
Guangdong	18	11,346		77.3	161	25.1	Humid	2.99
Guangxi	23.6	4926		10		5.7	Humid	3.47
Chongqing	8.23	934		2.5		2.2	Humid	1.42
Sichuan	48.14	8341		18.8	287	13.6	Humid	3.98
Yunnan	38.33	4830	5.1	8.9	887	6.9	Humid	2.18
Tibet	122.8	344		1.8		19.6	Semi-humid	11.67
Shaanxi	20.56	3864	1.4	400.7		11.8	Semi-arid and semi-humid	33.83
Gansu	45.44	2637	4.6	142.2	2160	10.0	Arid	22.08
Qinghai	72.23	603	0.1	34.3	1070	3.6	Arid and semi-arid	19.16
Ningxia	6.64	688	3.7	105.6	200	4.9	Arid and semi-arid	9.21
Xinjing	166	2487	35	58.8	850	21.5	Arid	18.46

[a]National Statistics Bureau (2019)

[b]National Health Commission of the P. R China (2019)

[c]Compiled from Guo et al. (2014),Wen et al. (2013)

[d]Ministry of Water Resources of the P. R China (2019)

et al. 2010; He et al. 2010; Wang et al. 2018). Deep high-fluoride groundwater in these plains/basins may be attributed to palaeo-seawater and palaeo-brine (Wang et al. 2018; Wen et al. 2013). High-fluoride geothermal water is mainly found in and around the Liaodong Mountains, Liaoning Peninsula, southeastern China, southern Tibet, Longzhong in the Chinese Loess Plateau and Guanzhong Basin (Fig. 2b; Fan et al. 2008; He et al. 2010; Wang et al. 2018). High temperature of geothermal water can enhance water–rock interactions, and promote the dissolution of fluorine-bearing minerals.

In some areas, high-arsenic and high-fluoride groundwater can be found in aquifers simultaneously. Such areas include Datong Basin, Taiyuan Basin, Yuncheng Basin, Huhhot Basin, Junggar Basin, Songnen Plain, Hetao Plain (Fig. 2). In Datong Basin, for example, the arsenic and fluoride concentrations in groundwater can reach up to 2680 μg/L and 80.89 mg/L, respectively (Guo et al. 2003; Pi et al. 2015). In summary, arsenic-bearing minerals (such as pyrite) and fluorine-bearing minerals (such as fluorite, biotite, amphiboles and apatite) in rocks and sediments are the geogenic sources of high-arsenic and high-fluoride groundwater (Selinus et al. 2005), while unique climate conditions and special geological environments are important factors leading to the enrichment of arsenic and fluoride in groundwater, with high-arsenic and high-fluoride groundwater usually occurring in the arid/semi-arid basins and plains

(Fig. 2). In addition, mining activities, fertilizers and pesticides also add up to arsenic and fluoride concentrations in soil and groundwater in some regions (Brindha and Elango 2011; Han et al. 2003).

Drinking Water Arsenicosis

Intake of high-arsenic and high-fluoride groundwater over a long period can cause arsenicosis and fluorosis. Many researches have verified that there is an obvious relationship between the arsenic (fluoride) content in drinking water and the arsenicosis (fluorosis), i.e., where there is higher contents of arsenic and fluoride in the drinking water, there may be more serious arsenicosis and fluorosis (Chung et al. 2014; Hou et al. 2002; Tang et al. 2017; Yu et al. 2007; Wang et al. 2012). As shown in Table 1, in regions with high groundwater supply ratio and high concentrations of arsenic and fluoride, such as Shanxi, Inner Mongolia, Hebei and Henan, large populations are exposed to high-arsenic and high-fluoride groundwater.

In China, cases of Blackfoot disease (a disease caused by exposure to arsenic) were reported in 1950s in Taiwan, and researchers discovered that artesian well water with high arsenic concentration was the cause leading to this disease (Cheng et al. 2016). In Mainland China, drinking water arsenicosis was first reported in Kuitun, Xinjiang Autonomous Region in 1980. Then, in 1989, severe arsenicosis broke out in the Inner Mongolia Autonomous Region (Sun 2004; Yu et al. 2007). The Chinese government declared it as an endemic disease in 1992 (Sun 2004). Since then, endemic arsenicosis has been found in other provinces. The drinking water arsenicosis data from 2002 to 2018 were collected for this study, and the data cover 13 provinces/autonomous regions including Anhui, Gansu, Henan, Hubei, Inner Mongolia, Jilin, Jiangsu, Ningxia, Qinghai, Shanxi, Shaanxi, Xinjiang, and Yunnan (Fig. 1a). Based on the data collected, the total patients of arsenicosis in these regions were described in Fig. 4a. The number of arsenicosis presents shows a peak in 2010 with 20,114 cases. Before the year 2010, the number of arsenicosis presents shows an increase and after 2010 it decreases. This can be explained as follows: With the water quality improvement projects implemented continuously, high-arsenic groundwater has been treated before consumption in more and more arsenic-poisoned areas. Therefore, the number of patients decreased, especially after 2010. However, arsenic poisoning usually has a certain incubation period, and although people in some regions have been stopped from drinking high-arsenic groundwater, patients will still appear. In addition, as the scope of the high-arsenic groundwater investigation expanded, some new endemic arsenicosis areas are identified, and more people are diagnosed with arsenicosis. That is why the patients still exist even after many years of implementation of water improvement projects.

The number of arsenicosis patients in 2018 is marked in Fig. 5. North and northwest regions are the main affected areas because of long-term intake of high-arsenic groundwater. Shanxi and Inner Mongolia are the two most seriously affected areas, where the numbers of arsenicosis patients are 1221 and 1914, respectively. Groundwater is the main source for water supply in Shanxi and Inner Mongolia, accounting for 40.38% and 46.17% of the total water supply, respectively. The arsenic concentrations are also very high in the two provinces, causing around 223,000 and 382,000 population exposed in high-arsenic groundwater, respectively (Table 1). Figure 4b shows the temporal variation of

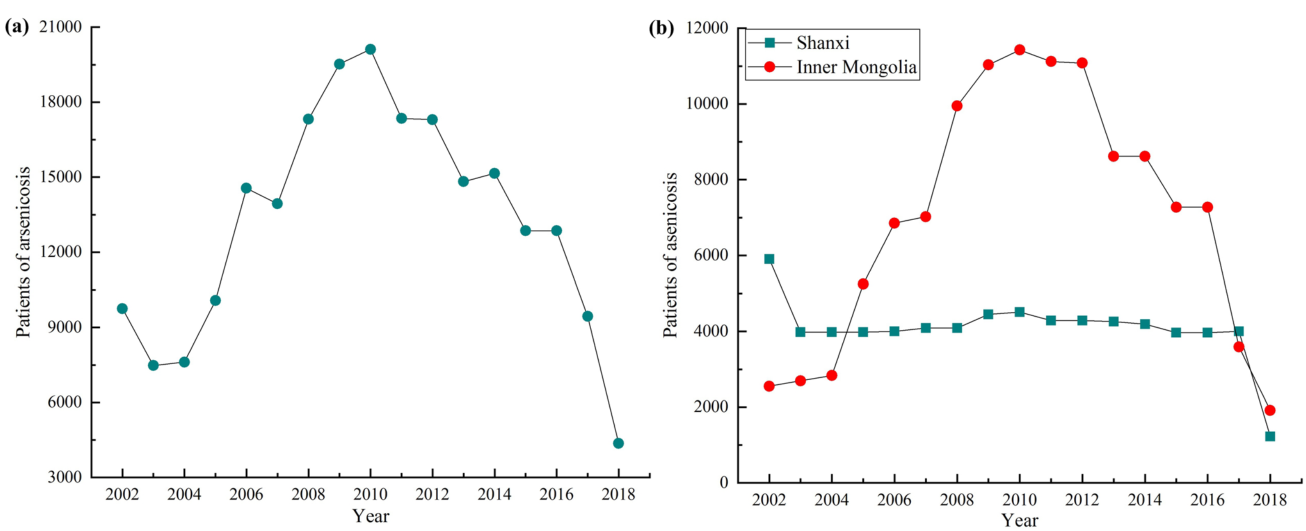

Fig. 4 Variation of patients of arsenicosis from 2002 to 2018: **a** China; **b** Shanxi and Inner Mongolia

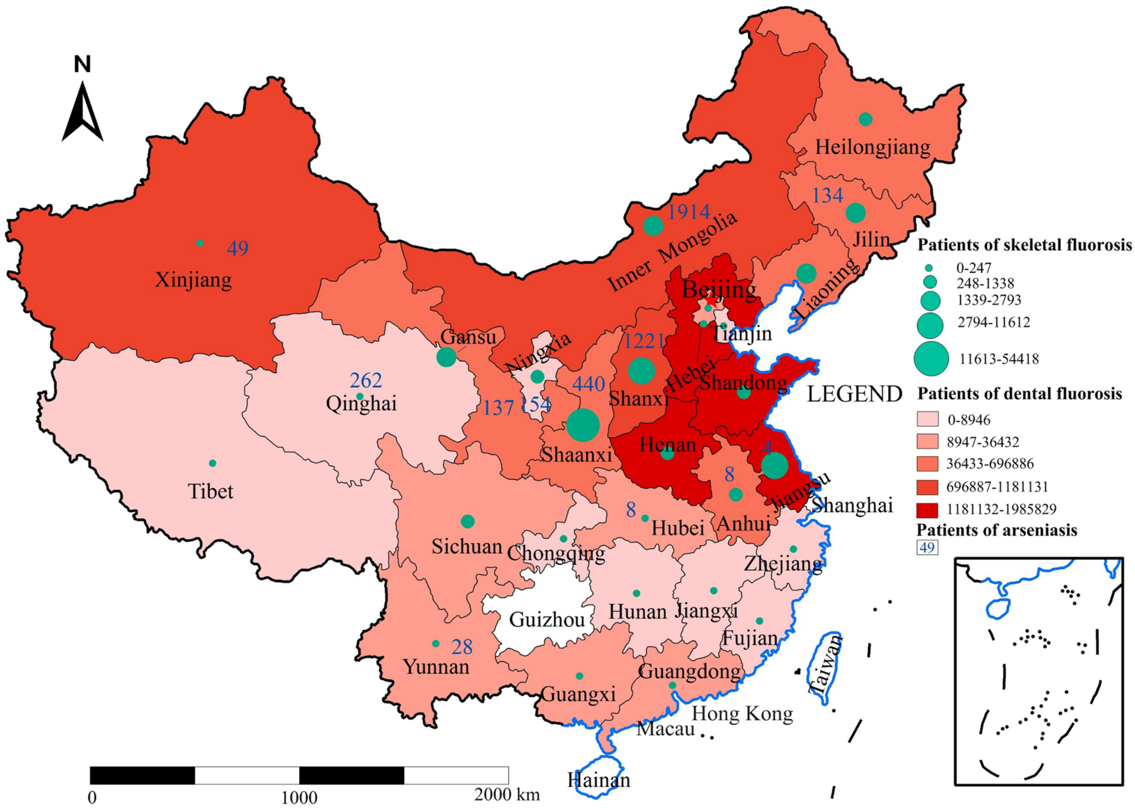

Fig. 5 Distribution of patients of arsenicosis and fluorosis in different provinces/autonomous regions in 2018

arsenicosis patients in Shanxi and Inner Mongolia, respectively. As can be seen from Fig. 4b, the number of endemic arsenic poisoning patients in Shanxi Province has stabilized at around 4,000 since 2003. However, for Inner Mongolia, the number of arsenicosis patients shows an increase from 2002 to 2010 and then a decrease after 2010. Shanxi is a small province in area (Table 1) and endemic arsenic poisoning occurs only in some large basins such as Datong basin and Yuncheng basin. Inner Monglia, on the contrary, is a bigger autonomous region (Table 1), and many new areas have been identified as the disease affected areas as the scope of the investigation expanded. In spite of the implementation of more and more water quality improvement projects and advancement of medical technology, the problem of asenicosis cannot be solved in a short term. In some areas, arsenic poisoning may even be lifelong problem.

Drinking Water Fluorosis

In China, the endemic fluorosis can be divided into drinking water fluorosis, burning coal fluorosis and drinking tea fluorosis (National Health Commission of the P. R. China 2019). Burning coal fluorosis is also a special fluorosis in China, because Chinese residents, especially those in the north part of China, burn fossil coal which contains high fluoride for cooking and heating during the winter (Dai et al. 2007). Drinking tea fluorosis is prevalent in the minority concentration areas, where the residents are accustomed to drinking pure brick tea or brick tea with milk and/or butter (Selinus et al. 2005). Generally, fluorosis has been reported in 29 provinces and/or autonomous regions except Shanghai. Anhui, Beijing, Gansu, Guangdong, Hebei, Heilongjiang, Henan, Hubei, Inner Mongolia, Jilin, Jiangsu, Liaoning, Ningxia, Qinghai, Shandong, Shanxi, Shaanxi, and Xinjiang are the main provinces and/or autonomous regions of drinking water fluorosis (Fig. 1b). The number of fluorosis patients in 2018 is also marked in Fig. 5. As showed in Fig. 5, north China (Hebei, Henan, Inner Mongolia, Shanxi), northwest China (Xinjiang, Shaanxi), and eastern coastal regions (Shandong and Jiangsu) are the main disease affected areas. Among these areas, Jiangsu Province has the highest number of dental fluorosis patients with 1,985,829 and the highest number of skeletal fluorosis patients is found in Shaanxi Province with 54,418. In southern China, the number of fluorosis is lower, because in the southern China areas, surface water which usually has low fluoride concentration accounts for a greater proportion of drinking water (over 90%, Table 1). In Shanghai, almost all water supply comes from surface water and no patients of fluorosis have been found (Ministry of Water Resources of the P. R.

China 2019; National Health Commission of the P. R. China 2019). High level fluoride in south China is mainly found in geothermal water that is used for heating and bathing. The temporal variations of total number of dental fluorosis and skeletal fluorosis patients in these regions are presented in Fig. 6a, b. They both show an increase trend from 2003 to 2008 and decrease after 2008. The number of skeletal fluorosis decreased significantly in 2018 and more and more patients getting rid of their diseases. The number of patients with dental fluorosis and skeletal fluorosis was compared in this study in Shanxi and Inner Mongolia, which are two representative disease areas (Fig. 6c, d). An interesting phenomenon can be found that there are more patients of dental fluorosis in Shanxi but more skeletal fluorosis in Inner Mongolia, which may be attributed to the higher fluoride levels and groundwater supply ratio in Inner Mongolia (Table 1). In addition, diet habits in the two areas may also contribute to the difference. For example, the average rate of drinking water intake for adults is 1.5 L/day in Inner Mongolia, which is higher than Shanxi with an average rate of drinking water intake of 1.125 L/day (Ministry of Environmental Protection of the P. R. China 2013).

Since the arsenicosis and fluorosis are found, the local and central governments as well as the medical and health institutions have attached great importance to this problem. In many provinces, institutes of endemic diseases have been set up. Many researchers have also carried out a lot of research work on arsenicosis and fluorosis (Jin et al. 2003; Sun 2004; Yu et al. 2007). With the continuous implementation of water quality improvement projects and therapies

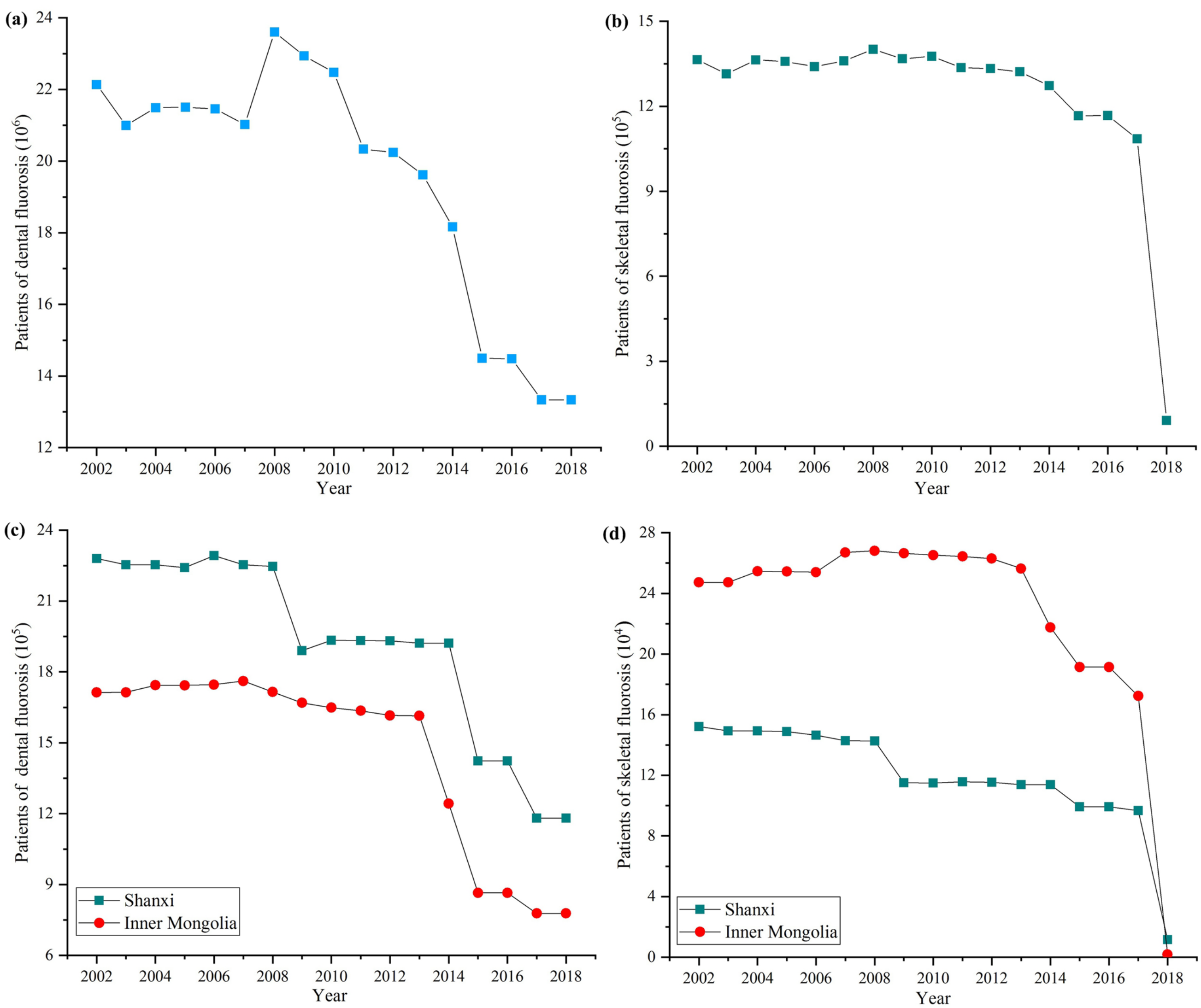

Fig. 6 Variation of patients of fluorosis from 2002 to 2018: **a** dental fluorosis in China; **b** skeletal fluorosis in China; **c** dental fluorosis in Shanxi and Inner Mongolia; **d** skeletal fluorosis in Shanxi and Inner Mongolia

to patients, drinking water arsenicosis and fluorosis have been effectively controlled. The number of patients has been declining since around 2010. Recent years, the number of patients of arsenicosis and skeletal fluorosis are declining quickly, but dental fluorosis is not easy to control and the number of patients is difficult to be reduced rapidly (Figs. 4 and 6).

Arsenicosis and Fluorosis Management

Drinking safe water is a critical prerequisite for human health (Li and Wu 2019a). However, in China, water resources are inadequate and unevenly distributed (Zhang et al. 2010; Wang et al. 2018; He et al. 2019). Poor quality groundwater which contains high concentrations of nitrate, As, F, I, TDS, Fe, and Mn is widely distributed over China. Consumption of poor quality groundwater has negatively affected the safety of drinking water supply and caused severe threats to human health (Wu and Sun 2016; Jia et al. 2018; Wang et al. 2018; He and Wu 2019). Sustainable groundwater management and utilization to ensure safe drinking water supply are extremely important (Li and Qian 2018; He and Li 2020).

The most effective and direct way of reducing the water arsenic-fluorosis is to cut off the source of exposure. Many techniques have been developed in the past decades to remove arsenic and fluoride from drinking water. The mostly applied techniques include coagulation/precipitation methods, membrane processes (reverse osmosis and nanofiltration, electrodialysis), ion-exchange processes, and adsorption processes (Lata and Samadder 2016; Singh et al. 2016; Kabir and Chowdhury 2017). Different materials and techniques can be selected for the removal of contaminants in drinking water. Each of these techniques has its own merits, limitations, influencing factors and work efficiency. Some of these technologies can be suitable for simultaneous removal of arsenic and fluoride (Singh et al. 2016). However, these techniques are now mainly used in centralized water supply plants, and the application of them in household treatment of high-arsenic and high-fluoride drinking water should be enhanced.

Water quality improvement projects are the fundamental measures to prevent drinking water arsenicosis and fluorosis. Since the discovery of drinking water arsenicosis and fluorosis, the local and national governments have attached great importance to this problem and taken measures to reduce arsenic and fluoride concentrations in drinking water. Wang et al. (2012) conducted a nationwide study on the effects of fluoride on safe water supply, involving 81,786 children aged from 8 to 12 and 594,698 adults aged over 16 from 27 provinces over China. They found the rate of fluorosis prevalence is significantly higher in the areas with threat of high-fluoride drinking water than those in the safe water supply areas. Another fact that makes the problem even worse is that the endemic areas are mostly remote and poor areas. The number of villages affected by arsenicosis and fluorosis and the number of villages implementing water quality improvement from 2002 to 2018 are portrayed in Fig. 7. As shown in Fig. 7a, the number of villages with arsenicosis and the number of villages with water quality improvement show the same pattern. It reveals that if an arsenicosis affected village is found, the water quality will soon be improved. However, due to economic and technical constraints, the number of villages undergoing water quality improvement is obviously lower than that of disease affected villages. Both numbers increased sharply in 2015, because large-scale surveys carried out in this year revealed more disease affected areas. In Fig. 7b, the number of villages with water quality

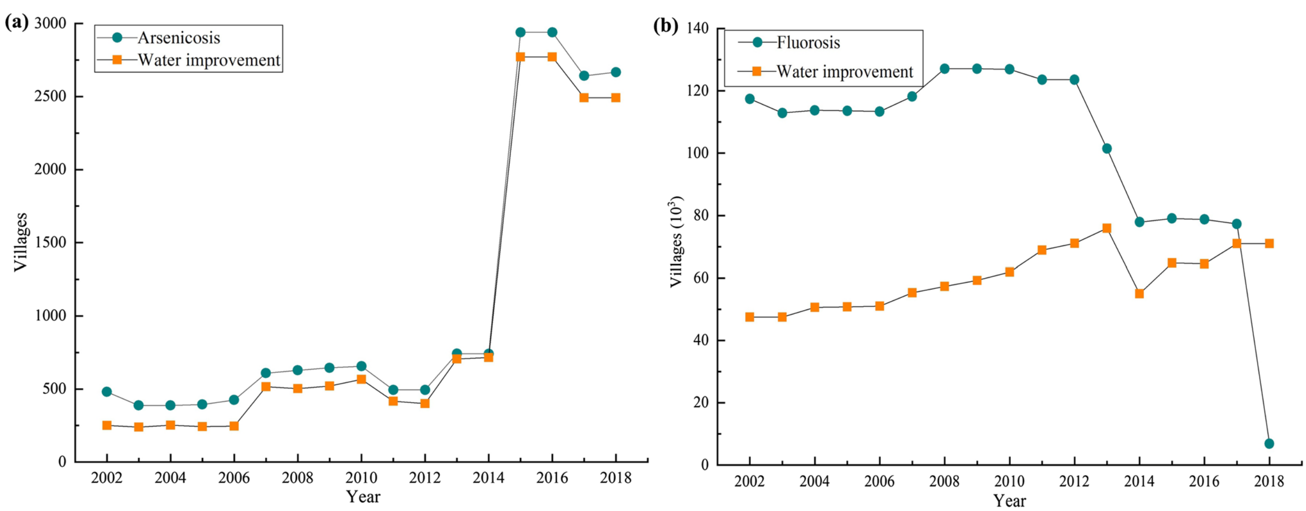

Fig. 7 Number of villages with arsenicosis and fluorosis versus the number of villages with water quality improvement

improvement is apparently less than the number of fluorosis affected villages. However, due to economic and technical constraints, the present situation of high-fluoride and high-arsenic groundwater treatment is not adequate. Li et al (2018a) investigated 39 water quality improvement projects in five provinces (Anhui, Hebei, Henan, Shanxi and Tianjin). They found that there were only 21 projects in normal operation, among which only 14 projects yielded acceptable treatment performance. During the period from 2012 to 2013, 45 counties in Shanxi Province were selected to check the running condition and performance of water quality improvement projects (Li et al. 2015). A total of 1804 water quality improvement projects were investigated for drinking water fluorosis, and 1673 (92.7%) of the projects were in normal operation, and 1328 yield eligible performance ($F^- < 1$ mg/L). A total of 145 water quality improvement projects were investigated for drinking water arsenicosis, and 138 were in normal operation, accounting for 95.2%. Among the normally operated projects, 143 projects produced acceptable arsenic reduction performance (arsenic concentration < 10 μg/L).

Furthermore, there are a number of options to reduce the consumption of high-arsenic and high-fluoride water. Rainwater, treated surface water and spring water with low arsenic and fluoride can be alternative sources of water for drinking, cooking, bathing and washing clothes (Wang et al. 2019; Wu et al. 2019). It is highly suggested that groundwater and surface water are conjunctively used in some areas for domestic uses to achieve a sustainable goal (Li et al. 2018c). Moreover, mixing low-arsenic (low-fluoride) water with higher-arsenic (high-fluoride) water can be an option to reduce the arsenic and fluoride concentration to an acceptable level before consumed (WHO 2018; Li et al. 2018b, 2019). However, due to the lack of appropriate treatment technology and equipment, and/or the improper running of the existing treatment facilities, above mentioned options are impossible to be adopted in some areas to really solve the problem of high-arsenic and high-fluoride water consumption. Therefore, it is mandatory to continuously strengthen the centralized water supply with low-arsenic and low-fluoride water to communities, and to check the operation status of existing water treatment equipment and facilities.

Conclusions

In this paper, we summarized the distribution and hydrochemical characteristics of high-arsenic and high-fluoride groundwater in China, and reviews the arsenicosis and fluorosis associated with drinking high high-arsenic and high-fluoride groundwater in China. The following conclusions were drawn:

(1) China is a country widely affected by high-arsenic and high-fluoride groundwater. There are 20 major provinces/autonomous regions in China facing the problem of high-arsenic groundwater, which is mainly found in the fluvial/alluvial-lacustrine plains and basins located in arid and semi-arid regions and alluvial plains/basins and river deltas in humid/semi-humid regions. High-fluoride groundwater is widely distributed in mainly in shallow aquifers and geothermal aquifers in north, northeast and northwest parts of China. Arsenic-bearing and fluoride-bearing minerals in rocks and sediments are the main sources of high-arsenic and high-fluoride groundwater, and special climate conditions, geological environments and human activities are important factors leading to the occurrence of high levels of arsenic and fluoride in groundwater.

(2) Drinking water arsenicosis has mainly been found in 13 provinces and/or autonomous regions, among which Shanxi and Inner Mongolia are most seriously affected areas. The number of arsenicosis presents two variation periods: increase from 2003 to 2010 and decrease after 2010 with the peak in 2010. Fluorosis has been found in 29 provinces and/or autonomous regions except Shanghai. North China (Inner Mongolia, Shanxi, Hebei), northwest China (Xinjiang, Shaanxi), and eastern coastal regions (Shandong and Jiangsu) are the main prevalent areas.

(3) With the continuous implementation of water improvement projects, drinking water fluorosis and arsenicosis have been effectively controlled in China. However, due to the lack of appropriate treatment technology and equipment, or the existing treatment facilities cannot operate effectively, some people are still facing the threat of high-arsenic and high-fluoride groundwater. Long-term implementation of the water improvement project needs further attention.

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Compliance with Ethical Standards

Conflict of interest The authors declare that they have no conflicts of interest.

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