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Review papers

A review of arsenic presence in China drinking water

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SUMMARY

Chronic endemic arsenicosis areas have been discovered in China since 1960s. Up to 2012, 19 provinces had been found to have As concentration in drinking water exceeding the standard level (0.05 mg/L). Inner Mongolia, Xinjiang and Shanxi Province are historical well-known "hotspots" of geogenic As-contaminated drinking water. The goal of this review is to examine, summarize and discuss the information of As in drinking water for all provinces and territories in China. Possible natural As sources for elevating As level in drinking water, were documented. Geogenic As-contaminated drinking water examples were taken to introduce typical environmental conditions where the problems occurred: closed basins in arid or semi-arid areas and reducing aquifers under high pH conditions. Geothermal water or mineral water in mountains areas can be high-As water as well. For undiscovered areas, prediction of potential As-affected groundwater has been carried out by some research groups by use of logistic regression. Modeled maps of probability of geogenic As contamination in groundwater are promising to be used as references to discover unknown areas. Furthermore, anthropogenic As contaminations were summarized and mining, smelters and chemical industries were found to be major sources for As pollution in China.

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1. Introduction

Arsenic is a naturally-occurring element that can enter drinking water through natural and anthropogenic sources. Interaction of arsenic with both organic and mineral colloids can elevate As concentration in natural waters. It is likely that in most aquifers, biogeo interactions are likely to play the dominant role in resulting in high-As level in groundwater. Furthermore, inorganic As species are significantly more harmful than organic As species. Hence, public health and environmental concerns drive more attention on geogenic inorganic As level in the nature. At the same time, many freshwater aquifers are being contaminated by human activities as well, as a result of mining and smelting activities, pesticide use, chemical production and so on.

Acute arsenic poisoning is associated initially with nausea, vomiting, abdominal pain, and severe diarrhea (Ratnaike, 2003). Long-term drinking water exposure of arsenic may cause skin lesions (Tondel et al., 1999), peripheral vascular disease (Engel et al., 1994), hypertension (Chen et al., 1995), black-foot disease (Chen, 1990), and high risk of cancers (Bates et al., 1992). High drinking water As concentration was found to elevate late fetal mortality and neonatal/postneonatal mortality (Hopenhayn-Rich et al., 2000). As exposure was also observed to impair cognitive development in school children and lead to children DNA damage and immunodeficiency (Yanez et al., 2003; Vega et al., 2008). Recent findings from Chile link in utero and early life As exposure to cardiovascular, respiratory and lung cancer later in adult life (Smith et al., 2006; Yuan et al., 2007).

WHO's norms for drinking-water quality go back to 1958. The International Standards for Drinking-Water established allowable level arsenic in drinking water as 0.20 mg/L. In 1963 the standard was re-evaluated and reduced to 0.05 mg/L. The guideline value was provisionally reduced in 1993 from 0.05 mg/L to 0.01 mg/L. Many countries including China had kept the old guideline value 0.05 mg/L as water quality standard for several years. In 2006, Ministry of Health PRC gave a revision of the *Standards of drinking water quality* with allowable arsenic concentration as 0.01 mg/L instead of 0.05 mg/L, as used in the past decades (Ministry of Health of China and Standardization Administration of China, 2006).

Chronic endemic arsenicosis was found in Taiwan in 1968 and reported in Xinjiang Province in Mainland China in the 1970s. In the 1980s, more areas were reported to be affected by arsenicosis via drinking water. Up to year 2012, endemic arsenicosis distributed over 45 counties in nine provinces, while 19 provinces had the problem that arsenic concentration in drinking water exceeded water standard level (0.05 mg/L). Even though China government has been working on building up water supply plants to ensure safe drinking water, the population at risk is still large: about 1.85 million according to the recent official data from Ministry of Health PRC et al. (2012) is drinking water with As level above 0.05 mg/L. Population exposed to drinking water with As level 0.01–0.05 mg/L was not included in that survey.

One goal of the present study is to draw a picture on arsenic sources of China and to know how the respective importance of each type of source is for elevating As concentration in drinking water. Secondly, we discuss some major documented hotspots that were well-known as As-affected areas. An additional goal is to summarize the features of environment where different types of natural high-As waters and the reasons why acute poisoning incidents happen.

2. Source of arsenic to surface and groundwater

2.1. Minerals

Arsenic is a metalloid which exists in earth crust and ranks 14th in element abundance order. The background concentration varies from 1.8 mg/kg to 2.1 mg/kg according to different reviews. Arsenic occurs as a major constituent in more than 200 minerals, including elemental As, arsenides, sulfides, oxides, arsenates and arsenites (Smedley and Kinniburgh, 2002). However, most of those arsenic containing minerals are rare in nature and generally found as sulfides associated with Au, Cu, Pb, Zn, Sn, Ni, and Co in ore zones. The most dominant minerals exist in environment are arsenopyrite (FeAsS), realgar (AsS) and orpiment (As_2S_3).

China has large amount of arsenic reserves: The known arsenic reserves were reported to be 3977 kt, and 2796 kt preserved reserves, of which 87.1% existed in paragenetic or associated ores up to the end of 2003 (Xiao et al., 2008). In 2011, China was the top producer of white arsenic with almost 50% world share, followed by Chile, Peru, and Morocco. (U.S. Geological Survey, 2012). Xiao et al. (2008) reported the distribution of arsenic deposits in China (Fig. 1). Documented arsenic ore reserves, as demonstrated in Fig. 1, are mostly located in southern and western regions of China. The sum of arsenic reserves locating in Guangxi, Yunnan and Hunan Province represents more than 60% of the total arsenic reserves explored in China. As reported, up to 87% of reserves are believed to be present in sulfide ores which are paragenetic or associated with transition metals ores.

2.2. Rocks, sediments, soils and air

2.2.1. Rocks

The arsenic abundance of China's continental lithosphere (CCL) has been reported to average 1.2 mg/kg (Li and Ni, 1997). Compositions of rocks in eastern China were with higher arsenic levels, particularly in clastic rock (5.0 mg/kg), pelite (7.8 mg/kg) and carbonatite (3.2 mg/kg) (Yan et al., 1997).

Concentrations in coals are variable depending on the area. Wang et al. (2005) sampled and analyzed 297 coal samples and found that 16% of the samples have arsenic concentrations higher than 8 mg/kg, while concentration ranged from 0.24 to 70.83 mg/kg. Other research reported much higher arsenic concentrations up to 32,000 mg/kg arsenic in coal sample (Ren et al., 1999). Guiz-

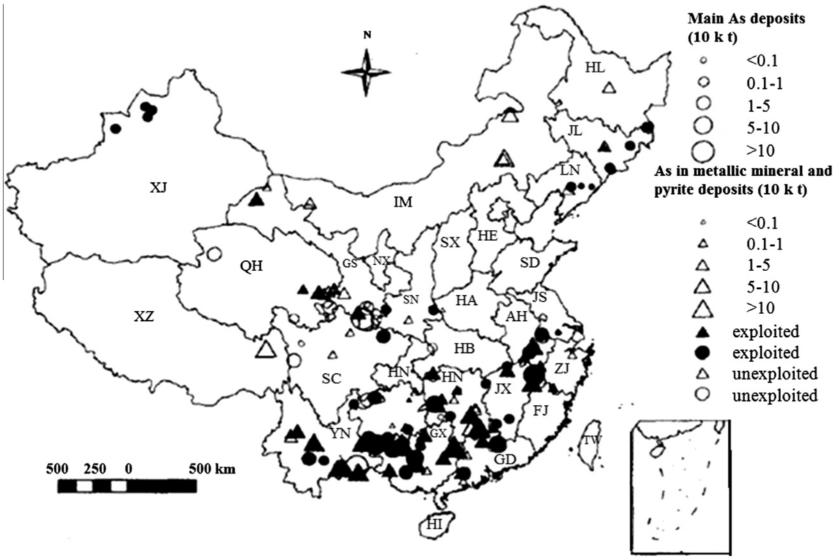

Fig. 1. Distribution of arsenic deposits in China (see abbreviations in Table 2) (from Xiao et al., 2008, revised).

hou province is well-known for its large amount of coal reserves, and these contain high arsenic concentration. In southwest of Guizhou, Emeishan basaltic rocks, which is believed to have close relationship with the forming of high-arsenic coal, contain 11–113 mg/kg of arsenic (Xie and Nie, 2007), although the basaltic rocks in earth crust are considered having low average arsenic concentration, around 2 mg/kg according to research of Turekian and Wedepohl (1961).

2.2.2. River and aquifer sediments

Average As concentrations for sediments of water system in China is 9.1 mg/kg (Luo et al., 2010). This value is notably higher than world average river sediments which is 5 mg/kg (Martin and Whitfield, 1983).Changjiang River (Yangzte River) is the 3rd longest river in the world. The Changjiang River Basin drains an area of 1.8 million square km, 18.8% of China's land area (CWRC, 2005-2013). Zhang et al. (1995) investigated 260 sediment samples from Changjiang River Basin and the average As concentration of raw sediments appears to be 7.6 mg/kg, while the fine grained parts (<63 μm) tends to be higher as 9.0 mg/kg. Huai River, another main river of China, has been reported to have an average As concentration in sediments of 12.6 mg/kg (9–21.8 mg/kg) (Luo et al., 2010). Studies carried out on the Yellow River have reported high arsenic abundance and notable cases of arsenic poisoning in various areas along the river. Fan et al. (2008) reported that As sediments concentrations in Shanyin (in Datong Basin area), Shanxi Province ranged from 3.09 to 26.25 mg/kg with the highest concentration found at depth of 28–34 m. Also within the Datong Basin, along Huangshui River and Sanggan River, Xie et al. (2008) collected aquifer sediment samples retrieved from 0 m to 50 m depths below ground level, and arsenic contents ranged from 4.9 to 118.2 mg/kg with mean value of 18.6 mg/kg. Hetao Plain located in the Great Bend of Yellow River in Inner Mongolia is one of the representative arseniasis-affected areas in China. In Hangjinhouqi County, northwest of Hetao Basin, sediments contained As concentrations ranging from 6.8 to 58.5 mg/kg, reported by Deng et al. (2009). The As concentrations of sediments from the Huhhot Basin which locates close to Hetao Plain lie in the range 3–29 mg/kg (Smedley et al., 2003).

2.2.3. Soils and atmospheric input

Baseline concentrations of As in soils are generally of the order of 5–10 mg/kg (Smedley and Kinniburgh, 2002). An average geochemical background level in world soils was set as 7.2 mg/kg by Boyle and Jonasson (1973). By analyzing 4095 soil samples from all over mainland China, Wei et al. (1991) investigated this geochemical background level for 61 elements. Table 1 shows As results in China compared with the As background levels for some other countries. Soil content for 95% of total samples was in the range of 2.5–33.5 mg/kg. Among different types of soil of China, regosol and mountain soils have relatively high As concentrations (around 16 mg/kg) and unsaturated siallitic soils have the lowest arsenic content 4 mg/kg. Based on the same dataset, Weng et al. (2000) studied the content distribution of As in Chinese topsoil (Fig. 2). Arsenic content in soils tended to associate with calcium content in the north and with iron content in the south of China. While As correlation with Fe is well recognized due to the high affinity of secondary iron oxyhydroxides for arsenic oxyanion (Charlet and Polya, 2006; Charlet et al., 2011), the correlation with calcium is less frequent and could be linked to the substitution of As(III) in calcite (Roman-Ross et al., 2006; Bardelli et al., 2011). Compare Fig. 2 with Fig. 1, we found the content of distribution to be positively correlated to ore deposits.

The deposition of atmosphereic arsenic may increase the arsenic content in aqueous system and soils slightly. Volcanic activities and eolian erosion of arsenic-containing minerals are natural processes which can release arsenic into the atmosphere. However, human activities are believed to play a more important role as source of arsenic air pollution. Fossil-fuel combustion for energy generation, mining and agricultural operations, such as biomass waste combustion, cause national wide problem both in urban and countryside areas. Combustion of coal is the principle source of China's outdoor air pollution (Sheldon et al., 1992). China relies on coal for 70–75% of its energy needs, consuming 1.9 billion tons of coal each year (Millman et al., 2008). Tian and Qu (2009) estimated atmospheric arsenic emission from coal combustion

Table 1
Comparison between background concentrations of arsenic in soils in Mainland China and some other countries (Wei et al., 1991).

Country	Range (mg/kg)	AM[a]	GM[b]
Mainland China	0.01–626	11.2	9.2
USA	<0.1–97	7.2	5.2
Japan	–	9.02	–
UK	–	11.3	–

[a] AM – Arithmetic mean.
[b] GM – geometric mean.

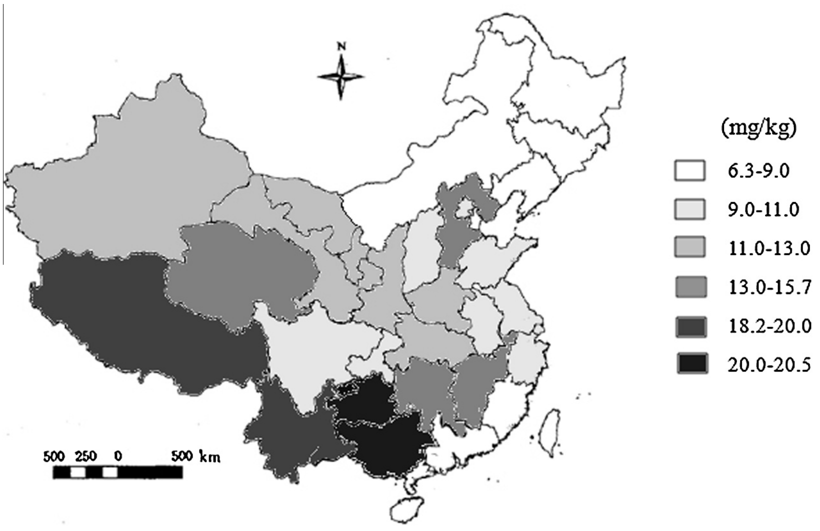

Fig. 2. Content distribution of arsenic in topsoil of Mainland China (from Weng et al., 2000, revised).

reached 1500 tons by China for year 2005. Atmospheric particles (PM2.5 and PM10) were analyzed for As concentration in some cities. As concentration in the air was 43.36 ng/m^3 (11.98–82.55 ng/m^3) in Taiyuan, 24.4 ng/m^3 in Guangzhou and 0.32 ± 0.17 mg/m^3 (0.07–0.79 mg/m^3) in Beijing (Xie et al., 2006; Huang et al., 2007; Yang et al., 2012).

3. Arsenic mobilization processes

3.1. Geogenic factors

Major processes responsible for observed concentrations of arsenic in surface and ground water include: mineral precipitation/dissolution, adsorption/desorption, chemical transformation, ion exchange, and biologic activity (Welch et al., 1988; Smedley and Kinniburgh, 2002). Ordinary solid phases containing arsenic around crustal abundance can give rise to high dissolved arsenic (0.05 mg/L) (Nordstrom, 2002).

3.1.1. Evaporation

In arid or semi-arid areas, use of groundwater is widespread due to lack of surface water. Extensive use of groundwater combined with high evaporation rate and low recharge rate leads to concentration of arsenic in groundwater, especially in low-lying places and closed basin where arsenic can hardly be flushed away by water flow. Studies on arsenic uptake by calcite and gypsum demonstrated that arsenic can be kept by accumulated salt contents (calcite and gypsum) in soil in saline area (Roman-Ross et al., 2006; Fernandez-Martinez et al., 2006). Rainfall or irrigation can release arsenic from those salts and then carry arsenic down into the subsoil and groundwater.

3.1.2. High pH

Adsorption and desorption reactions between arsenic and Fe, Mn and Al oxides and oxides and hydroxides surfaces are particularly important controlling reactions because those oxides are widespread in the hydrogeologic environment and arsenate adsorbs strongly to oxides and hydroxides surfaces in acidic and near-neutral-pH water (Dzombak and Morel, 1990; Waychunas et al., 1993). However, arsenate adsorption rapidly decreases in basic medium (Mamindy-Pajany et al., 2011). pH value plays an important role in the desorption of arsenic, because of its effects on the species distribution of anions, surface charge of the arsenic-bearing oxides and hydroxides, and subsequent electrostatic forces between arsenate and solids (Xu et al., 2012). The study of Bhattacharya et al. (2006) suggested the volcanic ash as the probable source of groundwater As. Locally, elevated pH values linked to carbonate dissolution, cation exchange, and dissolution of silicates promote release of adsorbed As. In high pH zones As remains dissolved in groundwater. Some studies also reported arsenic release from different iron-bearing minerals, soils and sediments by desorption process as pH values become alkaline (Appelo et al., 2002; Smedley and Kinniburgh, 2002; Breit and Guo,2012).

3.1.3. Geothermal

The release of arsenic from geothermal systems into surface and ground waters have been reported from several parts of the world. Arsenic concentrations in geothermal well fluids generally range from <0.1 mg/kg to 10 mg/kg. However, As concentrations of >20 mg/kg are not uncommon, at the other extreme. Most reservoir fluids are undersaturated with respect to arsenopyrite and other arsenic minerals (Ballantyne and Moore, 1988) and hence As leaching, rather than As precipitation, is predicted to occur in the reservoir. Hot springs, geysers and steam features with high As contents drain unimpeded into the nearest catchment system and contaminate shallow aquifer systems by natural upward movement of geothermal fluid and some other anthropogenic activities (Webster and Nordstrom, 2003; Aksoy et al., 2009). Dissolution of As oxide and orpiment, relevant to pH value, redox condition and fluid temperature, plays an important role in regulating arsenic mobility. Secondary minerals that form after the dissolution of Fe–As sulfides such as Fe-hydroxides exert a greater influence on the mobility of arsenic in the geothermal environment (Pascua et al., 2006). Study of Winkel et al. (2013) reveal that where Fe-(hydr)oxides are not sufficiently abundant to act as major scavengers for arsenic, arsenic can be closely associated with calcite matrix in a CO_2-enriched environment.

3.1.4. Sulfide oxidation

Arsenic-bearing minerals including arsenic-rich pyrite, arsenopyrite, orpiment, realgar and As-associated metal sulfides can release great amount of arsenic when the chemical environment has been changed. O_2-enriched environment leads to oxidation of these minerals followed by As mobilization. Insoluble As-bearing minerals are rapidly oxidized by exposure to atmosphere and then released soluble As(III) is carried by runoff and groundwater flow into surface and ground waters.

3.1.5. Reductive dissolution

Anaerobic microbial respiration, utilizing either sedimentary or suface-derived organic carbon, is one important process contributing to the mobilization of arsenic from host minerals, notably hydrous iron oxides (Charlet and Polya, 2006). Many studies have given evidences of arsenic release under reducing environments in presence of Fe-reducing bacteria. Burial of fresh organic matter, infiltration of fresh DOC and the slow diffusion of O_2 through the sediment lead to reducing conditions just below the sediment–water interface in lakes or in superficial groundwaters as abundantly described in SE Asia delta (Charlet and Polya, 2006; Fendorf et al., 2010). This encourages the reduction of As(V) and desorption from Fe and Mn oxides, as well as the reductive dissolution of As-rich Fe oxyhydroxides.

3.2. Anthropogenic factors

As a by-product of some human activities, arsenic can be added to natural waters by chemical industry, mining operations, and agriculture. Arsenical pesticide, industrial sewage/sludge and mining tailing have been reported as sources of arsenic and had contaminated groundwater and surface water via natural processes (e.g. oxidation of mine waste) or direct human manipulations, such as rainwater infiltration, irrigation, runoffs and sewage discharge. In addition to the chemical conditions of the As sources, the composition and states of the primary materials, treatment methods, storage design are influential as well (Breit and Guo, 2012).

4. High-As content in China waters – geogenic cases

4.1. Overview

One of the most pervasive problem afflicting people in rural area in China is inadequate access to clean water. By the end of 2010, the rural population with safe drinking water was 670 million, and only 54.7% of rural population had tap water (Ministry of Water Resources PRC, 2011). Before 1962, residents mainly relied on shallow well water and surface water (Sun, 2004). Deeper wells were drilled after shallow well water was discovered to induce fluorosis and after surface water was shown to be polluted. Since chronic endemic arsenicosis was found in Taiwan in 1968 and reported in Xinjiang Province in Mainland China in the 1970s, more areas have been identified to be affected by arsenicosis via drinking water. Up to year 2012, endemic arsenicosis distributed over 45 counties in nine provinces, while 19 provinces had the problem that arsenic concentration in drinking water exceeded water standard level (0.05 mg/L). In total, the recent official data from Ministry of Health PRC, et al. (2012) showed the population at risk reached to 1.85 million, less than 2.34 million which was reported by Xia and Liu (2004). Moreover, a systemic research was carried out on endemic arsenicosis affected and suspicious areas by China government during 2004 and 2010. There were 12,835 villages with a total population of around 1.25 billion under investigation. The result showed 844 villages with 697,000 people were exposed to high-arsenic drinking water (>0.05 mg/L) (Sun, 2011). Sun also gave the distribution of villages with high-arsenic drinking water (Table 2). Although in 2006, Ministry of Health PRC gave a revision of *Standards of drinking water quality* with permitted arsenic concentration as 0.01 mg/L instead of 0.05 mg/L as used in the past decades, it was considered as target rather than requirement (Ministry of Health of China and Standardization Administration of China, 2006). However, the new standards started to be subject to enforcement in July of 2012, which means more population would be taken into account if they have been exposed to drinking water with As concentration of 0.01–0.05 mg/L. Summary of documented cases of geogenic high-As groundwaters of China is shown in Table 3. Fig. 3 shows documented areas with groundwaters containing high-As content above 0.05 mg/L. As shown in Fig. 3, high-As groundwaters mainly appear in Zone I – i.e. in the north of China under arid or semi-arid climate.

While high-arsenic content in drinking water is widespread problem, it occurs only under special natural circumstances relating to geochemical environment and hydrological features. At least, two conditions are necessary: (i) Abundant source of arsenic and (ii) Arsenic transportation from the source to water and accumulation. Not all the areas which locate near the arsenic-contained minerals or rocks have arsenic contaminated problem. It is quite usual that one village is discovered to be with high-As content in water from wells at the same time the neighboring village has extreme safe water. Below, we will discuss some examples which exist in different areas of China and the main characteristics of them.

4.2. Different types of geogenic high-As waters

4.2.1. Oasis-like reducing/oxidizing water

Xinjiang Province is a very arid region and it is the place where arsenicosis was first reported in Mainland China. Kuitun, one of the arsenicosis affected cities, has annual rainfall of 160–185 mm while annual evaporation of 1800 mm (Wang et al., 1985). The affected areas in Xinjiang involve Dzungaria Basin on the north side of Tianshan Mountains in the west to Mamas River in the east, a stretch of ca. 250 km (Wang and Huang, 1994). According to a recent report (Sun, 2011) and to the map given by Shen et al. (2005), the south side of Southern Tianshan Mountains and the south side ofAltai Mountains were areas exposed to high-As drinking water. Wang (1984) found As concentrations up to 1.2 mg/L in groundwaters from this province. Most of arsenicosis areas in Xinjiang are low-lying lands with relatively low altitude. Although some researches believed in that the high-As groundwater was under reducing environment in Xinjiang, one research did analysis water samples which had As level above 0.6 mg/L and found only As(V) in these ground waters (Wang et al., 2004).

4.2.2. Geothermal waters

Hot springs with elevated As concentration have been reported in several parts of China, including geothermal zones of Guide County of Qinghai Province, Rehai and Ruidian of Yunnan Province, Tibet and Taiwan.

Guide County of Qinghai is one of the areas which are suffering serious endemic diseases. Both fluosis and arsenicosis exist in Guide County, affecting around 590,000 population. Geothermal water (18.5–34.6 °C) contains 0.32–4.57 mg/L fluoride and 0.112–0.318 mg/L arsenic. It's considered that the sources of arsenic are metamorphic as well as volcanic rocks in the north part of Guide Basin. The concentrations of arsenic were found to be positive associated with water depth and temperature of thermal water. The distributions of high-As and high-F groundwater had the same pattern as the abnormal geothermal regions (Shi et al., 2010).

Several water samples from hot springs in Rehe and Ruihai region of Yunnan were investigated. Almost all the hot springs water had pH value above 7.9 except one spring which had a pH of 3.5. The alkaline thermal waters contain arsenic ranging from 0.083 mg/L to 687 mg/L while the single acidic spring water had only 0.0436 mg/L of arsenic. Only inorganic arsenic had been found in those samples. The environment of those hot springs could be oxidizing and reducing as well. Four of eleven springs had As(V) as the dominant specie and other samples had considerable concentration of As(III) with the highest ratio up to 91% for As(III)/Total As (Liu et al., 2009).

Table 2
The distribution of villages with high-arsenic drinking water in rural areas of China (from Sun, 2011, revised).

As content	0.05–0.1 mg/L		0.1–0.15 mg/L		0.15–0.5 mg/L		>0.5 mg/L	
Provinces (abbreviation)	Village number	Population	Village number	Population	Village number	Population	Village number	Population
Xinjiang (XJ)	184	164,040	33	36,200	16	15,462	2	2114
Inner Mongolia (IM)	137	11,544	29	3165	36	2748	4	575
Bintuan, Xinjiang	61	62,723	7	4667	6	4741	2	12,125
Shanxi (SX)	48	73,994	22	22,139	4	8334	2	1675
Jilin (JL)	46	16,153	8	2604	17	5772	–	–
Shandong (SD)	26	30,642	5	7489	1	916	–	–
Anhui (AH)	24	32,390	9	16,952	18	32,686	4	7229
Henan (HA)	20	27,612	2	1893	4	5994	–	–
Shaanxi (SN)	10	12,826	1	1100	–	–	1	987
Yunnan (YN)	8	10,048	5	13,488	1	440	1	6201
Qinghai (QH)	6	4166	2	538	2	3470	6	1989
Gansu (GS)	6	4756	4	4501	1	146	2	3170
Hubei (HB)	4	5940	2	3202	1	983	1	1342
Jiangsu (JS)	1	1500	–	–	1	300	–	–
Heilongjiang (HL)	1	1713	–	–	–	–	–	–
Sichuan (SC)	–	–	–	–	–	–	–	–
Total	582	46,047	129	117,938	108	81,992	25	37,407

Table 3
Summary of documented cases of geogenic high-As groundwaters of China.

Province/Region	Concentration ranges ($\mu g/L^{-1}$)	Aquifer type	Groundwater conditions	References
Inner Mongolia (including Hetao Plain and Hubao Plain)	Up to 1740	Holocen alluvial and lacustrine sediments	Strongly reducing conditions, neutral pH, high alkalinity	Deng, 2008Deng et al., 2009
Xinjiang (Tianshan Plain)	40–750	Holocene alluvial plain	Reducing, deep wells (up to 660 m) are artesian	Wang and Huang, 1994
Shanxi (Datong Basin)	105–1932	Quaternary sedimentary basin	Reducing, high pH (8.09), high concentration of phosphate and organic matters	Guo et al., 2003, Guo and Wang, 2005
Jilin and Heilongjiang (Songnen Plain)	Up to 152.4	Quaternary sedimentary basin	Reducing, high pH(8.0–9.3), high concentration of organic matters	Zhang et al., 2010
Ningxia (Yinchuang Plain)	<10–177	Holocene alluvial and lacustrine sediments	Reducing and oxidizing, highest As level exists in reducing environment, pH (7.18–8.58)	Han et al., 2010
Kuitun, Xinjiang	Up to 880	Quaternary alluvial and lacustrine sediments	Reducing and oxidizing. Some tubewells water contain mainly As(V)	Wang et al., 2004Luo et al., 2007
Qinghai (Guide Basin)	<0.112–0.318	Artesian aquifer, metamorphic rocks and volcanic	Geothermal water (18.5–34.6 °C), high pH (>8)	Shi et al., 2010

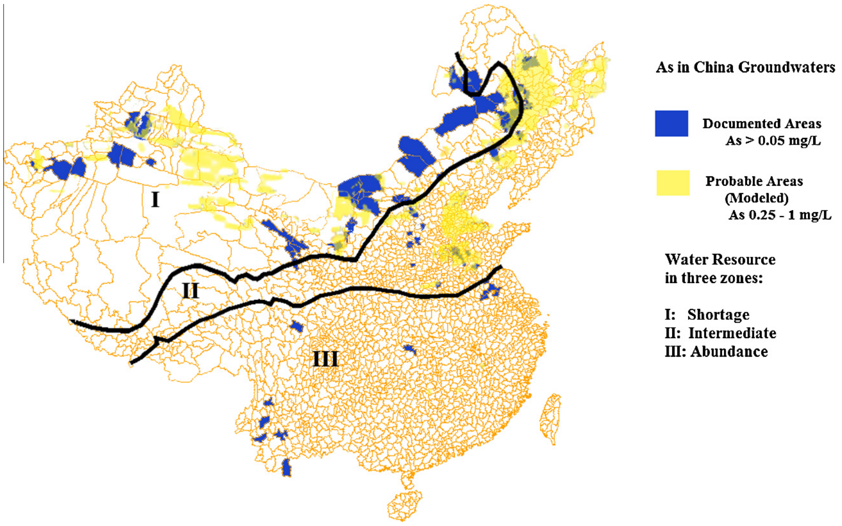

Fig. 3. Water resource distribution in China and comparison of map of documented high-As groundwaters and modeled map of probability of geogenic high-As groundwater (See abbreviations in Table 2.) (from Shen et al., 2005; Yu et al., 2007; Amini et al., 2008; Courrier International, 2009; Zhang et al., 2010).

4.2.3. Natural mineral waters

The arsenic contamination of mineral water in Hexigten Banner was first discovered in 1979, when more than 20 student campers who had done their intern exploration in the area ended up getting sick because of toxic spring water they had drunk in mountain area. After they were diagnosed as acute arsenic poisoning cases, local disease control and prevention centers carried out sampling and analysis in surrounding areas. Arsenic concentrations up to

2.43 mg/L were found in mountain spring water and arsenic content in groundwater ranged from 19.8 to 82.2 μg/L. Most of arsenicosis-endemic villages were located within this mountain area in valleys. Arsenic was derived from large outcrops of FeAsS, Zn and Pb ores distributed throughout the mountains where large mine tailings were frequently found (Ministry of Health of Inner Mongolia, 2009; Zhang et al., 2010).

4.2.4. Reducing environments

4.2.4.1. Irrigated Hetao Plain and Hubao Plain in Inner Mongolia

The Hetao Plain with an area of 13,000 km^2 is located in the western region of Inner Mongolia, bounded by the Yin Mountain in the north, by the Yellow River in the south, by Ulan Buh (Wulanbuhe) desert in the west, and by Ulansuhai Nur Lake (Wuliangsuhai) in the east. In the late Jurassic, a fault basin located in the northwest of the Hetao plain was formed. Lacustrine sediments with highly reducing conditions were formed in the closed basin. This region has groundwaters with high dissolved concentrations of arsenic (Yang et al., 2008). Concentrations up to 1.74 mg/L have been found in the groundwater in Hangjinhouqi, a county locating in the west part of Hetao Plain (Deng, 2008). Deng et al. (2009) reported results on arsenic concentrations and speciation in water samples collected in Hangjinhouqi, with an arsenic concentration ranging from 0.076 mg/L to 1.09 mg/L and speciation dominated by As(III): the As(III) to the total soluble As ratio ranging from 84% to 99%.

Hubao Plain lays in the east side of Hetao Plain and has similar geological structure. 74 groundwater samples were collected from chronic endemic arsenic poisoning areas around Hubao Plain in 1998 and 1999 respectively for water quality analysis. The results showed that high arsenic samples (>0.05 mg/L, As(III)/Total As = 82%) had low average levels of dissolved oxygen (0.0 mg/L) and low Eh value (39 mV) (Zhang et al., 2002). Lin and Tang (1999) also reported As-rich groundwater samples (As content: 0.42 mg/L), with 100% As(III) to total As ratio.

4.2.4.2. Datong Basin–Shanxi

Arsenicosis was revealed in early 1990s in Shanxi Province after local residents started to use water from deep wells (20–50 m). The geochemical features of Datong Basin are close to those found in Hetao Plain. The analysis of 66 groundwater samples collected from Shanyin area of Shanxi Province showed that 23 of them had high-As content (0.105–1.932 mg/L) and they have relative high As(III) to total As ratio ranging from 50% to 80.4% (Guo et al., 2003). Xie et al. (2011) identified the sources of arsenic in the shallow aquifers of Datong Basin through a thorough mineralogical, geochemical and zircon U–Pb dating study. Instead of pyrite which had arsenic content <1 mg/kg, Fe oxides (with up to 2000 mg/kg of arsenic) were shown to be the major mineral phases of arsenic enrichment in Datong bedrocks. The sedimentary rocks including coals were the most probable sources of arsenic in aquifer sediments according to zircon ages and geochemical data. The same group (Xie et al., 2008) reported that the aqueous arsenic levels were strongly depth-dependent in the Datong Basin and that the high arsenic concentrations were found at depths between 15 m and 60 m, with a maximum concentration equal to 1.82 mg/L. The hydrochemical characteristics of high arsenic groundwater from the study area indicated that the mobilization of arsenic was related to reductive dissolution of Fe oxides/oxyhydroxides and/or desorption from the Fe oxides/oxyhydroxides at high pH (above 8.0).

4.3. Prediction of geogenic potential As-affected groundwater

Statistical models based on the statistical relationship between As concentrations and relevant explanatory variables such as geology, climate, topography or soil organic matter content have recently been developed (Amini et al., 2008; Rodriguez-Lado et al., 2008; Winkel et al., 2008). Some researchers started to use logistic regression to assess the probability that As concentrations exceed a pre-defined threshold (Lee et al., 2009; Twarakavi and Kaluarachchi, 2006; Winkel et al., 2011). Amini et al. (2008) used a large databank of groundwater arsenic concentration from around the world as well as digital maps of physical characteristics such as soil, geology, climate, and elevation to model probability maps of global arsenic contamination. The relative significance of the variables in the arsenic model showed: The occurrence of arsenic under reducing aqueous conditions was most closely correlated to climatic, geological, and drainage parameters while under high-pH/oxidizing aqueous conditions it was most closely correlated to soil parameters (clay and silt), and drainage condition. Maps of modeled global probability of geogenic arsenic contamination in groundwater for reducing and high-pH/oxidizing conditions were delineated respectively. Nearly all the areas with high probability of arsenic occurrence in China groundwater were found in oxidizing conditions. The China part of their maps is compared with published and documented As-affected area in Fig. 3. Modeled map agreed well with the already established high As contaminated areas. However, documented high As regions such as Hetao Plain and Datong Basin were actually found to be under reducing conditions rather than predicted oxidizing conditions. Despite the moderate accuracy on the groundwater types, the map could still be used as a reference to investigate unknown areas, in order to prevent mass intoxication via groundwater with elevated As concentration.

Targeting the Shanxi Province, Zhang et al. (2012) applied stepwise logistic regression to analyze the statistical relationships of a dataset of As concentrations in groundwaters with some environmental explanatory parameters, where most of As investigations in this province focused on the Datong and Taiyuan Basins. They identified some environmental parameters are closely related to the distribution of high As concentrations, namely: (i) Holocene sediments; (ii) Topographic Wetness Index; (iii) Hydrological characteristics; (iv) Gravity and (v) Remote sensing information.

5. High-As content in China waters – anthropogenic cases

Table 4 gives a list of incidents of drinking waters polluted by industry in China in the last decade and three case studies were discussed below.

5.1. Mining and smelting activities

Arsenic is present in sulfide minerals associated with Au, Cu, Pb, Zn, Sn, Ni, and Co mineral ores and can be released in the water during the slow oxidation of As-rich pyrite waste tailings, or in the air during the smelting process. Up to 70% of total arsenic is abandoned as tailing by mine selections process. Only 10% of total arsenic is recovered while the remaining part is kept in intermediate material or found in solid waste residue and wastewaters (Wei and Zhou, 1992).

Acute poisoning has often occurred in China because of mine tailings. For example, in 1961, 308 people were poisoned and 6 were killed in Xinhua, Hunan Province. Arsenic-containing (5–13%) tailings from antimony mine were disposed close to drinking water tube well and contaminated the water (Yang, 1992). More recently, i.e. in the latest decade, this type of disaster happened

Table 4
Cases of drinking water source polluted by human activities in China.

Province/ Region	Polluted water type	Time	Contamination source	As level in polluted water (mg/L)	Notes	References
Dushan, Guizhou	Duliu River	2007	Sulfuric acid plant	Up to 14.2	65 poisoned	Chen et al., 2010
Chenxi, Hunan	Ground Water	2008	Sulfuric acid plant	Up to 19.5	17 poisoned	Chen et al., 2010
Hechi, Guangxi	Ground Water	2008	Smelter		15 poisoned	Chen et al., 2010
Yunan	Yangzonghai Lake, freshwater lake	2008	Chemical plants	>0.1	26,000 threatened	Liu, 2009; Environmental Protection Agency of Yunnan, 2008–2010; Wang et al., 2010
Henan	Dasha River, Minsheng River	2008	Sulfuric acid plant	Up to 2.56	Plant discharge with As level up to 445 mg/L	Peoples Court of Minquan County, 2010
Xichang, Sichuan	Well water	2006	Copper Smelter	–	17 poisoned	Chen et al., 2010
Yingde, Guangdong	Water channel	2005	Mining industry	–	34 poisoned	Chen et al., 2010
Jiangsu and Shangdong	Picang flood-diversion channel	2009	Chemical plants	Up to 1.987	500,000 threatened	Ling et al., 2009

quite often as a result of booming mining and smelting activities. In Hechi, Guangxi Province, arsenic acute poisoning happened at least three times between 2001 and 2008, with an increasing number of smelters being built in this region, famous as a non-ferrous metal-rich-area. The contamination was usually triggered by heavy rains or floods which flushed the waste water from smelter treatment tanks directly into surface waters. On the other hands, too many plants have been running without or under poor supervision from government. The lack of waste treatment system led to several problems as well.

5.2. Pesticide use

Inorganic arsenicals, such as lead, calcium, magnesium and zinc arsenate, zinc/sodium arsenite, Paris green (acetoarsenite) or organic arsenicals have been used extensively as agricultural chemicals. Those chemicals are used as herbicides, desiccants, antiseptics, toxic rat poisons, oncomelania hupensis poisons and so on. Because of their highly toxicity, many pesticides including arsenical products have been banned by China government. Although arsenical pesticides mainly draw some concerns related to soil pollution, it could also affect the quality of drinking water directly. It was reported that well water had been contaminated by pesticide and the concentration of As was around 1.15 mg/L in Yangshan, Anhui Province (Li et al., 2006).

Zhang and Xiao (1993) revealed that arsenic concentration measured in tube well waters was positively related to rainfall. The tube well was believed that have been contaminated by pesticide waste disposed around the well. Tube well water samples contained 0.008–0.044 mg/L arsenic in dry seasons and 0.03–2.00 mg/L of arsenic in rainy seasons. Arsenic concentration in soils and vegetables were studied as well. Soils contain 2.5–49.5 mg/kg at 40 cm depth and 7–97.5 mg/kg at 80 cm depth. The highest level of arsenic (5 mg/kg) was found in roots of vegetables.

5.3. Chemical industries

Sulfuric acid is produced by oxidation of sulfur rich ores. The use of As-bearing sulfide minerals as raw material in sulfuric acid manufactures is all-pervading in China chemical industries. Illegal use of low-class mineral ores that contain large amounts of arsenic, lack of safe waste treatment system and illegal discharge of waste waters into surface water made pollution incidents inevitable to happen. In the single year 2008, Heishui River of Guizhou, Groundwater of Chenxi, Hunan and Yangzonghai Lake of Yunnan were all polluted by sulfuric acid production plants. The Yangzonghai Lake is an exemplary case. Its waters had arsenic concentration less than 0.006 mg/L prior to September 2007. A contamination was noticed in April 2008 and resulted in arsenic concentration of lake water above 0.1 mg/L, according to analysis results of samples collected in July, September and October 2008 (Liu, 2009; Environmental Protection Agency of Yunnan, 2008–2010; Wang et al., 2010).

6. Conclusions and recommendations

This review has attempted to list the current available information on the occurrence and distribution of As contamination in China drinking water. The current facts related to drinking water contamination by arsenic in China appear alarming: (i) the problem is widespread throughout China; (ii) a large population has been exposed to drinking water with high level of geogenic As; and (iii) an increasing number of sudden and accidental As pollution events is taking place due to human activities.

Geogenic high As water are mostly found in arid or semi arid North of China. It results from: (i) use of groundwater as water supply is more common in the north, where As-rich groundwaters are more likely to be discovered, than in the south and (ii) climate, geochemical and hydrological conditions are favorable for formation of high-As water. Although natural arsenic resource is abundant and high-As in drinking water is widespread in China, the problem only occurs when certain geochemical and hydrological conditions are met. There are some common features among those cases. High-As spring or groundwater are found in closed basins where As is hard to be flushed away or be diluted. Affected areas are usually low-lying zones with high pH value (≈8.5), which is favorable for As being released and exchanged from minerals or rocks (Charlet and Polya, 2006). Reducing type As-rich groundwater bodies are mainly found in the north of China, under arid or semi-arid climate.

In contrast, in south of China, arsenic existence in drinking water is mainly due to human activities. Large amount of As ore reserves and As-containing mineral mines are located in the south where extensive mining activities and chemical industries increase the likelihood of As pollution accidents. Surface water in the south are contaminated by industrial discharge or runoff containing high As content from industries, triggered by heavy rains and floods in those subtropical or tropical zones.

Almost all arsenic-contamination cases were discovered thanks to poisoning accidents. It can be foreseen that under the current situation of water scarcity people will exploit more and more natural water for drinking or other uses. In order to ensure water safety as well as to prevent further environmental disasters, the government needs to conduct effective water quality monitoring and management. For those areas which have been identified as As-affected, continuous water quality test should be carried out on-site. Arsenic level as well as arsenic speciation should be examined.

Previous investigations of a number of affected sites in China have given people a better understanding of the As-contamination problem. However, research focused mainly on heavily contaminated water bodies with water As concentrations above 0.05 mg/L, and areas of groundwater with lower concentrations (0.01–0.05 mg/L) of arsenic will call out for government and researchers attention in the near future. Low concentration of arsenic could be dangerous if there's a high ratio of As(III) to total As. Knowledge on the speciation of arsenic is gaining increasing importance because toxicological effects of arsenic are connected toits chemical form and oxidation states. To have a better view of the danger of As-containing water and to give more comprehensive information to the exposed public, the analyses should not be restricted on the determination of total As. Arsenic speciation is also important and necessary to be measured.

Besides, more researches should focus on understanding the occurrence, origin and distribution of arsenic. Government should pay more attention to industrial and agricultural activities which lead to As pollution. More technical supports should be given to mining or chemical plants to deal with sewage and sludge storage and waste treatment. Supervision departments should increase the frequency of sampling and analyzing of the discharge from industrial plants.

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