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ARSENIC POLLUTION IN THE GLOBAL ENVIRONMENT

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ABSTRACT

Arsenic pollution is a public health concern globally because it's a toxic metalloid with severe health effects such as cancer, cardiovascular diseases, reproductive health problems and impaired cognitive development in children. Arsenic concentrations up to 2000 µg L^{-1} in groundwater and 46,049 µg L^{-1} in surface water have been reported in North and South America respectively. In Africa, As concentrations up to 2300 µg L^{-1} in groundwater and up to 19,000 µg L^{-1} in surface water have been reported. High As concentrations in groundwater have been associated with natural processes such as reductive dissolution of Fe/Mn oxyhydroxides, microbial oxidation of organic matter, reduction of sulfide minerals, and hydrothermal processes in volcanic regions. In surface waters, anthropogenic activities such as ore roasting, mining and smelting have contributed immensely to high As concentrations. The highest As concentrations in groundwater (7754 µg l^{-1}) was reported in the west region of Turkey in Europe due to geothermal waters. However, the highest population exposed to high As concentrations was reported in Bangladesh (77 million) and India (50 million) with up to 2000 µg L^{-1} As and 3200 µg L^{-1} As in groundwater of Bangladesh and India respectively. Although, less people are now drinking the As contaminated water in Bangladesh, they were still irrigating and cooking with it leading to exposure via dietary sources such as rice. Dietary sources such as rice also need to be regulated just like drinking water.

Keywords: Arsenic; water pollution; global environment; public health

1. INTRODUCTION

Arsenic (As) is ubiquitous in the environment and can be detected in rocks, minerals, soils, sediments, and biota in aquatic and terrestrial environments. Sources of As may be from natural processes such as rock erosion or weathering, forest fires, volcanic activity or may be due to human activities such as smelting, mining, use of agrochemicals such as fertilizers, pesticides, industrial emissions, wood preservation, and poor waste management practices (Chen et al., 2015; Gallego et al., 2015; Liu et al., 2015). The mobilization of As in underlying rocks and minerals by chemical and biological processes could lead to contamination of groundwater while

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mining and industrial drainage into surface waters could potentially increase As concentrations in the water.

Arsenic pollution of groundwater, surface water, and sediments has been reported in virtually every continent in the world for decades making it a global environmental issue (Das and Sarkar, 2018). It has been estimated that about 150 million people worldwide are affected by As contamination of drinking water (Ravenscroft et al., 2009). Groundwater As pollution has been reported in Asia, North America, South America, Europe and Africa. In particular, the continent Asia, in countries such as Bangladesh and India have been exposed to extreme As pollution which has led to severe health effects in majority of the populace (Rahman et al., 2014; Chakraborti et al., 2015).

Exposure to As in the environment over a long period of time is deadly because As is toxic and has been classified as a human carcinogen. (IARC, 2012). Exposure to As on the long term could lead to an accumulation of As in tissues such as skin, hair and nails, resulting in symptoms such as hyperpigmentation and keratosis which could lead to an increased risk of skin, internal organ, and lung cancers (Kapaj et al., 2006). Arsenic poisoning has also been linked with respiratory diseases, gastrointestinal disorder, liver malfunction, hearing loss, reproductive health problems, impaired cognitive development in children, cardiovascular disease and neuropathy (Guo et al., 2007; Ravenscroft et al., 2011; McClintock et al., 2012; Santra et al., 2013).

Regulatory bodies like the United States Environmental Protection Agency (USEPA) in the USA and similar bodies in other nations have been tasked with the job of controlling the release of toxic contaminants like As into the environment to protect public and ecological health. Regulatory standards (maximum contaminant level-MCL) for As concentrations in drinking water have been reduced from 50 µg L^{-1} to 10 µg L^{-1} in Canada and the United States about seventeen years ago by regulatory bodies in these countries (USEPA, 2001; Health Canada, 2006). This is intended to further reduce As release into the environment and thereby reduce exposure to this toxic contaminant. Most developing nations have MCL > 10 µg l^{-1} the regulatory limit for the United States and WHO. For example, the current MCL for arsenic in drinking water (25 µg l^{-1}) in Mexico is two and a half times higher than that of the United States and World Health Organization (WHO). In Bangladesh and India with notable episodes of As contamination, the MCL for As is 50 µg l^{-1}, five times that of the US and WHO.

The deadly health effects of As can be prevented by increasing our understanding of the sources and levels of As contamination in the environment. The last review on status of As contamination worldwide was written about 16 years ago in 2002 and so there is need to reassess the situation on a global level. Besides, As is on the list of priority pollutants written by USEPA which means we need to monitor and regulate its release into the environment. Although there

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are numerous reviews on As, they have been limited to countries or regions. This paper attempts to provide information on the current status of arsenic pollution and provide feedback on levels of As contamination in the global environment. Hence, the objectives of these review are to 1) review arsenic contamination in the past two decades and 2) assess the current status of arsenic levels in the global environment in order to provide direction for protecting public health.

2. ARSENIC CONTAMINATION IN THE GLOBAL ENVIRONMENT

Even though As contamination has been reported in virtually every continent, there has been a lot of havoc done in South East Asia more than other locations. Hence, we will discuss more extensively about As contamination in Asia and also describe episodes of As contamination in other nations and continents across the world. In particular, Bangladesh and India have reported As contamination on a large scale along with the most devastating health effects in the world history.

2.1 Arsenic Contamination in Asia

2.1.1. Bangladesh

The arsenic contamination of groundwater sources in Bangladesh was reported in 1992 and thereafter presented at an International Conference on As in Groundwater in Calcutta (now Kolkata, India) in 1995 (Das et al., 1995; Dhar et al., 1997). The arsenic contamination in Bangladesh is the most tragic and extensive arsenic pollution in history. Groundwater As concentrations up to 2000 µg L^{-1} about 200 times higher than the MCL (10 µg L^{-1}) for As were detected in some areas of Bangladesh (Tondel et al., 1999).

Arsenic concentrations above regulatory standards (then 50 µg L^{-1}) were found in 41 districts out of 60 districts. Arsenic concentrations in 97% of the hair samples from people living in As-affected villages were above the toxic level and 95% of the nail samples contained As above the normal level (Table 1). People suffering from As-induced skin lesions were identified in 21 districts out of the 22 districts surveyed (Chowdhury et al., 1999). A research team identified eight thousand and five hundred arsenic patients suffering from various skin lesions, gangrene in leg, skin, lung, bladder, liver, and renal cancer (Anawar et al., 2002).

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Table 1: Highlights of Arsenic Contamination in Bangladesh.

		References
Discovered	1992	Das et al., 1995; Dhar et al., 1997
Presented at conference in India	1995	Das et al., 1995; Dhar et al., 1997
% of districts affected	68.3 %	Das et al., 1995; Dhar et al., 1997
Arsenic in nail	95% samples above normal level	Chowdhury et al., 1999
Arsenic in hair	97% samples above normal level	Chowdhury et al., 1999
As-induced skin lesions	95.5 % of districts	Chowdhury et al., 1999
Population suffering from lesions and gangrenes	8,500 people	Anawar et al., 2002

Das et al. (2004) analyzed rice, water and soil samples and reported that the water and soil samples exceeded the MCL set for As by WHO and Bangladesh while samples of rice grains did not exceed the regulatory limit of 1.0 mg/kg DW. The results of the study by Das et al. (2004) showed that the As concentrations were higher in the roots of rice with the highest concentrations of plant As reported in samples collected from fields irrigated with contaminated As water. In addition, higher As concentrations in the roots of rice relative to the rice shoots indicates that the tolerance mechanism of rice was based on reduction of As uptake from roots to shoots. This indicates that rice is not an As accumulator or hyperaccumulator since the plant could not translocate As from roots to the shoots.

By the year 2007, Ohno et al. (2007) reported that As intake via drinking water was not high anymore despite the highly contaminated groundwater in the area because many families had changed their drinking water sources to less contaminated ones. They determined the average

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contributions to the total arsenic intake (Fig. 1) to be 13% from drinking water, 4.4% from liquid food, 56% from cooked rice, 11% from solid food, and 16% from cereals (Ohno et al., 2007). Even though they were not drinking the contaminated water anymore, they were still cooking with it. Suave (2014) concluded that arsenic exposure from rice is equivalent to or even greater than exposure from drinking water and suggested As in rice should be regulated as well.

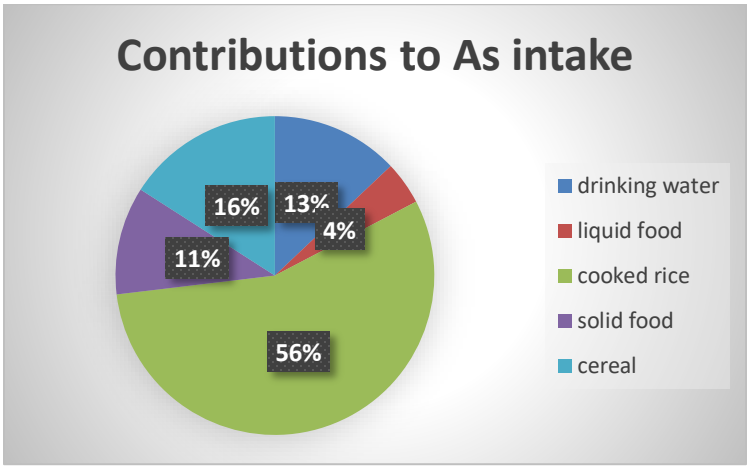

Figure 1: Average contributions to Arsenic Intake. Data from Ohno et al. (2007)

Several studies have indicated that the source of As in groundwater in Bangladesh is due to natural occurrence of As rich parent rock or minerals. Geochemical studies suggest that arsenic may be released from both reductive dissolution of Fe and Mn (oxy) hydroxide and microbial oxidation of organic matter (Anawar et al., 2002; Tareq et al., 2003; Zheng et al., 2004). Leaching experiments have shown that As is mobilized under acidic conditions when in association with Fe/Mn oxides while alkaline conditions favor As release from organic matter (Bibi et al., 2008). In addition to a strong correlation between As and both dissolved organic carbon (DOC) and Fe. Halim et al. (2009) reported a negative correlation between As and sulfate (SO_4^{2-}) suggesting that As may not be directly mobilized from sulfide minerals such as arsenopyrite in the Ganges Delta Plain, southwestern Bangladesh.

2.1.2 India

The most severe arsenic contamination in India was reported in West Bengal where As was detected above the MC L (50 µg L^{-1} for India) in 9 out of 19 districts with a population of 50 million people (Chakraborti et al., 2009). Arsenic concentrations up to 3192 µg L^{-1} were detected in tube well water samples in the upper and middle Ganga plain, India. About 46.5% tube well water samples exceeded 10 µg L^{-1}, 26.7% exceeded 50 µg L^{-1} and 10% exceeded 300 µg L^{-1} limits (Ahamed et al., 2006). Arsenic mobilization was attributed to arsenopyrite oxidation and

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reduction of Fe hydroxides underground in favorable Eh conditions (Ghatak, 2001; Pandey et al., 2002).

Pandey et al. (2002) reported that 30, 000 people residing in 30 villages and towns of central east India were exposed to groundwater with As concentrations above standard limits (>10 μg L^{-1}). The study conducted for three years showed a continuous increase in the arsenic levels in both the dug wells and hand pumps with a maximal mean As concentration of 3050 μg L^{-1} at one hand pump. However, the As concentrations in dug wells were much lower than that of hand pumps.

Rahman et al. (2014) conducted a study to determine the extent and effect of As contamination in Nadia, one of the nine As affected districts in West Bengal, India. They discovered that 51.4% and 17.3% of the tube-wells and groundwater of all 17 blocks had As above regulatory limits. They estimated that about 2.1 million people could be drinking As contaminated water above 10 μg L^{-1}. As concentrations at 300 μg L^{-1} was associated with the skin lesions diagnosed in regions with high As concentrations in drinking water. About 0.048 million in West Bengal, India was exposed to As contaminated water above 300 μg L^{-1} and at risk of developing arsenical skin lesions (Rahman et al., 2014). Analysis of biological samples such as hair, nail and blood revealed that 95% had As concentrations above the normal level. They also found evidence for dermal and neurologic effects of As and arsenical toxicity in children (Rahman et al., 2014).

Chakraborti et al. (2016) conducted a similar study in five blocks in Patna district, Bihar, India. They reported that 61% and 44% of the tube-wells had As above 10 and 50 μg l^{-1}, respectively, with maximum concentration of 1466 μg l^{-1}. Even though all the biological samples tested had As concentrations above the normal level, about 9.7% of the villagers had arsenical skin lesions. Arsenical neuropathy was also observed in 40.5% of 37 arsenicosis patients with 73.3% prevalence for predominant sensory neuropathy and 26.7% for sensor-motor.

2.1.3 China and Vietnam

China has been categorized as a region with high As concentrations because 20 out of 34 provinces have groundwaters with high As concentrations in arid/semi-arid inland basins and river deltas (Guo et al., 2014). Guo et al (2014) reported that these inland basins and river deltas are characterized by reducing conditions, high Fe and Mn concentrations, high pH and hydrogen carbonate (HCO_3^-) concentration, relatively low nitrate (NO_3^-) and sulfate (SO_4^{2-}) concentrations. They added that high pH and high HCO_3^- lead to As desorption from adsorption sites in the aquifer systems while both reductive dissolution of Fe oxides and reductive desorption of As could also result in As mobilization under reducing conditions (Guo et al., 2014).

High As concentrations due to geothermal waters have also been reported in China. Even though hydrothermal waters are found in all provinces of China, the high temperature hydrothermal

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systems with high As geothermal waters are mainly found in the Tibet autonomous region, western Yunnan Province and western Sichuan Province commonly called the Yunnan-Sichuan-Tibet Geothermal Province (YST) (Guo et al., 2017). Arsenic concentrations up to 1.1 mg L^{-1} was reported in the geothermal waters of the Rehai hydrothermal area, located in the Tengchong volcanic region of Yunnan Province (Guo et al., 2017). Both neutral and acid hot springs occur in Rehai with higher As concentrations in the neutral hot spring than the acid hot springs (Guo et al., 2017).

In addition to natural sources, agricultural and industrial discharge into surface waters have also been identified as a potential source of As pollution in China. Elevated As concentrations (128 μg L^{-1}) was reported in Yangzonghai Lake, a large plateau lake in China due to illegal discharge of As into the lake by industrial enterprises (Chen et al., 2015). The use of animal manure from concentrated animal feeding operations as fertilizers has led to elevated As concentrations in agricultural soils of the Pearl River Delta, south China (Liu et al., 2015). This is due to the use of arsenicals such as phenylarsonic acid compounds as additives to animal feeds to control parasites, promote growth, and increase feed utilization efficiency over the past two decades (Jackson et al., 2006; Liu et al., 2015).

There were three occurrences of arsenic pollution in three locations in Cambodia and Vietnam, both in Southeast Asia; city of Hanoi, Mekong delta and Red River delta. Groundwater arsenic pollution reported in the Mekong delta had As concentrations ranging from 1–1610 μg L^{-1} in Cambodia (mean 217 μg L^{-1}) and 1–845 μg L^{-1} in southern Vietnam (mean 39 μg L^{-1}) (Berg et al., 2007). Ten million people were exposed to groundwater As concentrations varying from 1–3050 μg L^{-1} (mean 159 μg L^{-1}) in Red River delta in northern Vietnam (Berg et al., 2007).

2.2 Arsenic Contamination in the Americas

2.2.1. North America

Only 10% of 30,000 groundwater samples in the USA exceeded the 10 μg L^{-1} regulatory limit while slightly less than half was < 1 μg L^{-1} (Welsh et al., 2000). Arsenic concentrations higher than regulatory limit of 10 μg L^{-1} were observed more in the Western half of the United States than the Eastern half (Welsh et al., 2000). Arsenic concentrations in ground water of the Appalachian highlands and Atlantic Plain of the Eastern United States were < 1 μg L^{-1} which was much lower than the Interior Plains and the Rocky Mountain Systems in the West (Welsh et al., 2000).

Eight years later, high As levels in drinking water wells were reported in the Northern Appalachian Mountain belt stretching approximately from Eastern Pennsylvania northward through Northeastern Maine, in the United States by Peters (2008). The source of arsenic in

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drinking water was identified as naturally occurring geologic materials, typically arsenopyrite, substituted sulfides such as arsenian pyrite, and nanoscale minerals such as westerveldite. In addition, the reductive dissolution of iron minerals may also lead to elevated levels of As in groundwater especially in landfills (Peters, 2008). Between 6% and 22% of households using private drinking water wells in this high arsenic region are exposed to arsenic concentrations higher than USEPA's MCL of 10 μg L^{-1} (Peters, 2008).

Stone et al. (2007) also reported As concentrations above the USEPA MCL of 10 μg L^{-1} in ten community water systems in Oregon, USA. High As concentrations within groundwater of the Madison, Upper Missouri River Valley, South Western Idaho and North Central Nevada in the United States were associated with geothermal waters (Welsh et al., 2000). High As concentrations associated with alkaline groundwater with pH > 8 was found in South Dakota, Oregon, Central Oklahoma, Northwestern Washington, and Arizona while acidic groundwater was observed in some mined areas (Welsh et al., 2000).

The long-term impact of a metal smelter in Ruston, Washington, USA, closed 20 years ago, on As mobility in sediments of nearby lakes was investigated by Gawel et al. (2014). This study reported that arsenic was highly mobile in the urban lakes, with maximum dissolved arsenic concentrations proportional to surface sediment levels and reaching almost 90 μg L^{-1} As. About 83% of the lakes in the deposition zone had surface sediments that exceeded probable effects concentration (PEC) for arsenic (Gawel et al., 2014).

The arid region of northern Mexico (states of Chihuahua and Coahuila) and bordering states of the southwestern US (New Mexico, Arizona, and Texas), are known for high As concentrations in groundwater (Camacho et al., 2011). Natural sources include volcanic processes and anthropogenic sources include mining and smelting of ores containing arsenic. The surface waters in these regions are not polluted due to the affinity of As for solid phases in alkaline conditions common to arid areas (Camacho et al., 2011).

Sediments from the cold Lake area in Alberta, Canada were also characterized and results show that high As, pyrites and arsenopyrites were detected in sediments from shale (Javed et al., 2014). This suggests that sulfide minerals could be the potential source of As in the groundwater. Arsenic concentrations in groundwater up to 580 μg L^{-1} were reported in an area of sulfide mineralization on Bowen Island, British Columbia, Canada (Boyle et al., 1998) while As concentrations of surface waters in Canada range from 0.28 μg L^{-1} to 556 μg L^{-1} (Wang & Mulligan, 2006).

2.2.2. Latin America

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It has been estimated that about four and a half million people in Latin American countries are chronically exposed to high levels of As in drinking water, based on the 50 μg L^{-1} regulatory limit in many of the Latin American countries (Castro de Esparza, 2009). Elevated concentrations of As in drinking water sources have been reported in Argentina, Bolivia, Brazil, Chile, Colombia, Cuba, Ecuador, El Salvador, Guatemala, Honduras, Mexico, Nicaragua, Peru, and Uruguay (McClintock et al., 2012). The largest known As exposed population in Latin America is in Argentina with about 4 million people exposed to high levels of As from drinking water based on the 10 μg L^{-1} limit (McClintock et al., 2012).

High As levels in Latin America has been attributed to the release of As from volcanic rocks and their sediments into drinking and irrigation water, mining activities, and use of As-based pesticides and wood preservation agents (Bhattacharya et al., 2006; Bundschuh et al., 2004). The mixing of geothermal waters (usually associated with volcanism) with cold aquifers are also a primary source of As contamination in the Pacific zone of Latin America (Lopez et al., 2012).

In Ecuador, high As concentrations have been reported in thermal waters (39–10,560 μg l^{-1}) due to hydrothermal processes in volcanic regions, in drinking water wells (9–125 μg l^{-1}) of three provinces and in rivers affected by gold mining activities (2.0–46,049 μg l^{-1}) (Cumbal et al. 2010; Bundschuh et al., 2012; Otero et al., 2016). High As concentrations has also been reported in groundwater (up to 740 μg L^{-1}) and in river water samples (up to 503 μg L^{-1}) from Mexico (Rosas et al., 1999; Gutierrez et al., 2008; Espino-Valdes et al., 2009; Camacho et al., 2011).

In Colombia, As concentrations in surface water and groundwater range from not detected to 255 μg l^{-1} with maximal concentrations of about 20 times the regulatory limit of 10 μg L^{-1}. (Alonso et al., 2014). High As concentrations in Colombia (Table 2) were detected in mining regions with soil As concentrations up to 148 mg kg^{-1} and 1400 mg kg^{-1} in sediments and in agricultural areas with vegetables containing up to 5.4 mg kg^{-1} As (Alonso et al., 2014). A recent study in Peru revealed widespread As contaminated groundwater with As concentrations above WHO permissible limit (George et al., 2014).

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Table 2: Arsenic contamination in North and South America

Country	Ground water ($\mu g\ L^{-1}$)	Surface water ($\mu g\ L^{-1}$)	Sediments ($mg\ kg^{-1}$)	References
USA	220-2000	90	5.0 -1370	Gawel et al., 2014; Serfes, 2005
Canada	580	0.28 - 556	6.3 – 10000	Galloway et al., 2017; Boyle et al., 1998; Wang & Mulligan, 2006
Mexico	740	503	NA	Camacho et al., 2011
Colombia	255	NA	1400	Alonso et al., 2014
Bolivia	964	11140	NA	Quintanilla et al., 2009; Van Den Bergh et al., 2010
Ecuador	9-125	2.0–46,049	NA	Otero et al., 2016

NA-not available

In Bolivia, As concentrations of groundwater within the Lake Poopo Basin were up to 245 $\mu g\ l^{-1}$ while in the stream affected by acid mine drainage and thermal waters, As concentrations up to 11140 $\mu g\ l^{-1}$ were reported (Quintanilla et al., 2009). Drinking water wells in the Southern and Western part of Lake Poopo had As concentrations up to 299 $\mu g\ l^{-1}$ while in the North and Northeast, drinking-water wells contained As up to 964 $\mu g\ l^{-1}$ (Van Den Bergh et al., 2010). A recent study has also reported that 90 % of drinking water wells within the Lake Poopo region had As concentrations exceeding the regulatory limit of 10 $\mu g\ l^{-1}$ (Munoz et al., 2016).

2.3 Arsenic contamination in Europe

Arsenic contamination of groundwater has been reported in some countries in Europe including United Kingdom, Hungary, Norway, Denmark, Ireland, Croatia, Italy, Spain, Greece, Sweden, Romania and Slovakia (Van Halem et al., 2009; Selinus et al., 2010; Katsoyiannis et al., 2014; McGrory et al., 2017). Maximal As concentrations (Fig. 2) reported in Norway, Denmark, Ireland, Hungary, Italy, Croatia and Greece were 19, 30, 234, 240, 431, 612, 1840 $\mu g\ l^{-1}$

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respectively (Frengstad et al., 2000; Jessen et al., 2005; Ćaver et al., 2005; Kouras et al., 2007; Rowland et al., 2011; Carraro et al., 2013; McGrory et al., 2017). The maximal As concentrations reported for Cyprus is slightly higher than values for Norway and Denmark with As concentrations in drinking water wells in Cyprus varying from 0.3 to 41 µg l^{-1} in 2007-2009 while it was 0.3-64.2 µg l^{-1} in 2010 (Christodoulidou et al. 2012). Sources of the As contamination in Cyprus were not identified and no explanations were given for the increase in As concentrations.

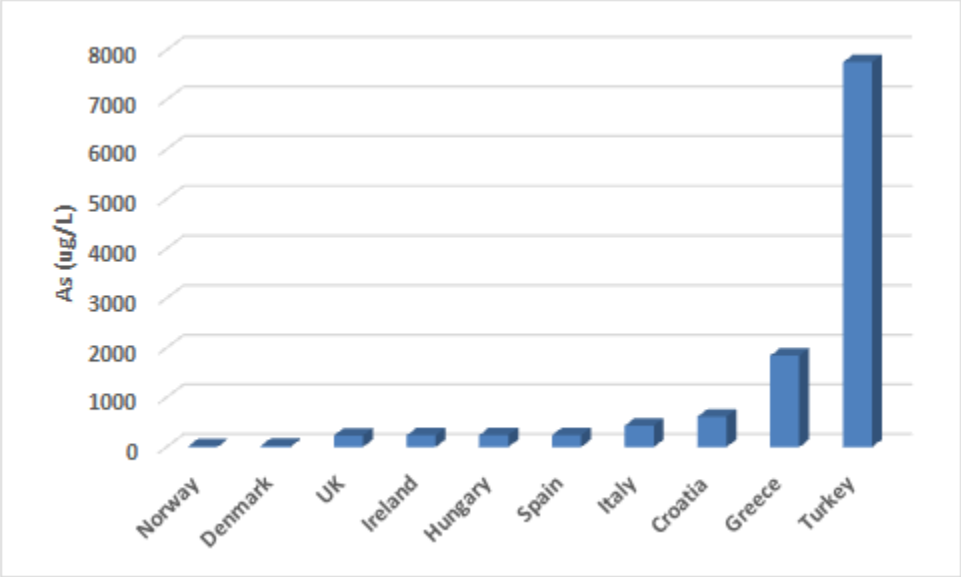

Figure 2: Maximal As concentrations (µg l^{-1}) in groundwater of some European countries

In a study covering nearly 60% of the population in Spain where average municipal drinking water arsenic concentrations ranged from 1 to118 µg l^{-1}, cardiovascular mortality rates in municipalities with arsenic concentrations in drinking water >10 µg l^{-1} were 3.2% increased in women and 2.4% increased in men compared to municipalities with concentrations <1 µg l^{-1} (Medrano et al., 2010). Arsenic concentrations from public water supply systems in Serbia (5-420 µg l^{-1}) were much higher than that of Spain with almost two-thirds of water samples in ten municipalities in Vojvodina, Serbia exceeding both Serbian and European drinking water standards (Jovanovic et al., 2011).

Arsenic concentrations of water from 10 private wells, 29 public supplies, 9 springs, 11 dams, including 9 small dams, 2 rivers, and 1 distribution network in Aksaray province, Turkey were determined. There were 27 water sources with concentrations above the regulatory limit of 10 µg l^{-1} (Altas et al., 2011). Another study in Turkey reported As concentrations up to 7754 µg l^{-1} in water resources of the west region of Turkey (Baskan and Pala, 2009). Abundant geothermal

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activity may be responsible for the high As concentrations reported in western Turkey (Baba & Sozbilir, 2012).

Groundwater with extremely high arsenic concentrations (1.6–1.9 mg L^{-1}) and high temperature were also associated with geothermal wells in Greece. Almost 65% of groundwater samples from public water supply wells and private wells in the area of Chalkidiki, Northern Greece had As concentrations higher than the maximum contaminant limit of 10 μg l^{-1} with concentrations ranging from 1-1840 μg l^{-1} (Kouras et al., 2007). In Poland, total As concentrations of drinking water, surface water, waste water and snow were determined in the provinces of Wielkopolska, Kujawy-Pomerania, and Lower Silesia (Komorowicz and Barałkiewicz, 2016). Results showed that up to 1.01 μg L^{-1} As was present in drinking water and waste water samples collected from the provinces (Komorowicz and Barałkiewicz, 2016). The maximum As concentration of 3778 μg L^{-1} in surface water (Trująca Stream) was detected in the area affected by geogenic arsenic contamination (Komorowicz and Barałkiewicz, 2016). On the contrary, high As concentrations (up to 1770 μg l^{-1}) detected in surface water of Glendinning, Scotland was not due to geogenic sources but because of its close proximity to a former Sb mine (Macgregor et al., 2015).

A previous study has reported that As distribution in European soils of agricultural and grazing land are directly related to the underlying geology (Tarvainen et al., 2013). Average As concentrations were three times higher in southern European soils than in northern European soils. More than 95% of all values determined in agricultural soils and 93% of all values in grazing land soils were below the action levels for As in agricultural European soils. Most of the high As areas have sulfide mineralization and have undergone mining and smelting operations (Tarvainen et al., 2013).

Mining is a major source of As pollution with extremely high concentrations of As reported in mine waste and nearby surface waters. The concentrations and mineralogy of arsenic in mine wastes and stream sediments in a former metal mining area of the UK was determined by Rieuwerts et al. (2014). They reported that concentration of As in mine wastes (1.8×10^5 mg kg^{-1} As) and stream sediments (2.5×10^4 mg kg^{-1} As) exceeded background and guideline values for the UK. The scorodite (As mineral) discovered in the mine waste was inversely correlated with potentially mobile As (Rieuwerts et al., 2014). Scorodite was also detected in Jedova jama, one of two 50-year-old mining waste dumps in Czech Republic in central Europe by Drahota et al. (2016). They reported that high concentrations of As (4600 μg l^{-1}) was present in pore water after rainfall events in Jedova jama while it was negligible at Dlouha Ves (up to 1.5 μg l^{-1}). Arsenic mobility at Jedova jama was attributed to the dissolution of amorphous ferric arsenate and simultaneous precipitation of Fe hydroxides under mildly acidic conditions (pH ~

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4.4) while immobile As at Dlouhá Ves is due to the efficient adsorption on Fe hydroxides and hydroxosulfates under acidic pH of ~2.8 (Drahota et al., 2016).

2.4 Arsenic contamination in Africa

It has been estimated that about 413 t yr^{-1} As entered the Egyptian northern delta lakes via agricultural drains and waste water discharge (Abdel-Moati, 1990). According to Abdel-Moati (1990), As concentrations of the lakes varied from 1.2 to 18.2 μg L^{-1} and between 1.2 and 8.7 μg g^{-1} for dissolved and particulate species respectively. Abdel-Moati (1990) explained that main As sources to the drainage system are phosphate fertilizers, detergents, herbicides, and loamy Nile deposits while about 52% of the total As derived to the Nile delta lakes is transported to the Mediterranean coastal seawater through the lakes' inlets.

On the contrary, As contamination of ground water and surface waters in Botswana was attributed to natural sources. Elevated As concentrations up to 3.2 mg L^{-1} was reported in shallow water underlying Camp island in the Okavango Delta of Botswana (Huntsman-Mapila et al., 2011). The Okavango Delta of Botswana is a 20, 000 km^2 wetland system located in an arid zone that includes over 100,000 islands, ranging in size from a few meters to over 50,000 ha (Gumbricht et al., 2004). Arsenic concentrations increased from below detection limits in surface water to 180 μg l^{-1} in the groundwater in New Island probably due to high evapotranspiration rates, alkaline pH which leads to desorption of arsenic or dissolution of arsenic sulfides, and formation of thio-arsenic complexes which keeps arsenic in solution (Mladenov et al., 2014).

In Nigeria, As concentrations in wells and boreholes varied from 2 – 510 μg l^{-1} with 75% of samples above the WHO limit of 10 μg l^{-1} in Zaria (Musa et al., 2008). As concentrations in shallow well water was higher than the bore hole water samples. Several studies have reported As concentrations higher than 10 μg l^{-1} in reservoirs and freshwater ecosystems in Nigeria (Benzer, 2016; Egbinola and Amanambu, 2014; Garba et al., 2012) but only one has reported symptoms of As poisoning in the Biu Volcanic Province of North East Nigeria where As concentrations higher than 10 μg l^{-1} was detected in surface water used for drinking purposes (Usman and Lar, 2013). Geothermal waters associated with volcanism may be responsible for high As concentrations in the Biu Volcanic Province of North east Nigeria.

In the Ethiopian Rift valley, 35% of groundwater wells and 70% of the hot springs (and geothermal wells) have arsenic concentrations above the WHO limit of 10 μg l^{-1} (Rango et al., 2010). The highest As concentrations in groundwater and surface water (Table 3) were reported in African countries such as Ghana and Cote d'Ivoire where gold mining was in close proximity to the sample locations.

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Table 3: Arsenic contamination in Africa

Country	Ground water (µg L^{-1})	Surface water (µg L^{-1})	Sediments (mg kg^{-1})	References
Botswana	3.2-116.6	2.3	0.2-7.0	Huntsman-Mapila et al., 2006
Morocco	NA	38.9-199.6	110 – 390	El Azhari et al., 2016; El Hachimi et al. (2007)
Cote d'Ivoire	0.2-2300	2-19000	1.6 – 294	Kinimo et al., 2018; Sako et al., 2018.
Egypt	1.0	1.2-18.2	10 – 44	Abdel-Moati, 1990; El-Badry & Khalifa, 2017 Saad &Hassanien, 2001
Nigeria	20-810	1.3	2.44	Usese et al., 2017, b; Musa et al., 2008
Ghana	1-141	0.2-2.2	942-10, 200	Smedley, 1996; Gbogbo et al., 2017; Serfor-Armah et al., 2006.
Ethiopia	21.4	0.9-77	2.5-6.6	Rango et al., 2010
MCL/PEC	10	10	33*	MacDonald et al., 2000

NA-not available *MCL-maximum contaminant limit **PEC-probable effect concentration

In Ghana, As concentration in water samples varied from 900 – 8250 µg L^{-1} while sediment samples had 942-10200 mg kg^{-1} As in a gold mining town (Serfor-Armah et al., 2006). The As rich mineral known as arsenopyrites ($FeAsS_2$) is commonly associated with gold mining because the weathering of As bearing minerals and deposits can lead to lowering of pH of nearby waters which mobilizes the As increasing the As concentrations of such waters. In aqueous environments, As can also be mobilized when cyanide is used to leach gold ores leading to high water pH (Straskraba and Moran, 1990). Developed nations such as the United States have been able to regulate mining and control release of contaminants into the environment.

In Morocco, As concentrations in sediment samples from the Moulouya River, the Ansegmir tributary and the Hassan II Dam located downstream of an abandoned mine range from 110 to

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390 mg kg^{-1} (El Azhari et al., 2016). The geoaccumulation index (Igeo) showed a very high As contamination of all sediment samples while the potential ecological risk index indicated that the sediment As concentrations are expected to be highly toxic to living organisms in the sediments (El Azhari et al., 2016). The sediment concentrations reported are higher than the probable effects concentration, (PEC) (concentration above which there might be harmful effects). Sediments in Morocco, Cote d'Ivoire and Ghana have As concentrations (Table 3) higher than the PEC which indicates they are polluted and harmful to the aquatic organisms in the water.

In a study conducted by Nyanza et al. (2014), total As concentrations in soils, water and cassava plants were determined at Rwamagasa village in northwestern Tanzania where daily living activities occur in close proximity to extensive artisanal and small-scale gold mining. Chemical analyses showed that 12.5% of water sampled had total As concentrations above WHOs regulatory limit for 10 µg L^{-1}. Total As in soil samples ranged between 183-20, 298 µg kg^{-1} while total As in cassava leaves ranged from 60 to 1,120 µg kg^{-1} and from 25-310 µg kg^{-1} in cassava roots (Nyanza et al., 2014).

3. CURRENT STATUS OF ARSENIC CONTAMINATION

After about two decades since the initial report of severe arsenic pollution, Chakraborti et al. (2010) reviewed the status of As contamination of groundwater in Bangladesh. Their analysis of 52,202 water samples from hand tube-wells in all 64 districts of Bangladesh showed that 27.2% and 42.1% of the tube-wells had As above 50 and 10 µg/l, respectively while 7.5% contained As above 300 µg/l. They explained that water analyses from the four principal geomorphological regions of Bangladesh showed that hand tube-wells of the Tableland and Hill tract regions are primarily free from As contamination, while the Flood plain and Deltaic region, including the Coastal region, are highly As-contaminated (Chakraborti et al., 2010). They added that due to mitigation efforts by the government, non-governmental organizations and international aid agencies, most of the people living in arsenic contaminated areas have been drinking As safe water for the past five years (Chakraborti et al., 2010).

There is growing global concern about As contamination of drinking water which has led to numerous studies and publications. We present some studies published within the past one year that has investigated As concentrations in groundwater, surface water and sediments. There is a public health concern in Northern Italy because of increasing As concentrations in some wells in aquifers 160 to 260m deep used for public water-supply in the Po Plain of Northern Italy due to drawdown of As-polluted groundwater (As ≤144 µg L^{-1}) from overlying aquifers (Rotiroti et al., 2017). Arsenic concentrations greater than the permissible limit of 10 µg L^{-1} was reported in the entire Po Plain due to natural sources (Rotiroti et al., 2017).

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A study by Gimenez-Forcada et al. (2017) in the southern part of the Duero Basin in Spain reported As concentrations up to 241 μg l^{-1} in groundwater. About 47% of groundwater samples collected contained arsenic above 10 μg/L from sources identified as geothermal waters and an oxidizing alkaline environment (Gimenez-Forcada et al., 2017). A recent study by Shahab et al. (2018) in Pakistan reported As concentrations in surface and groundwater in the Sindh Province of Pakistan. Arsenic concentrations up to 125 μg L^{-1} in groundwater and up to 35 μg L^{-1} in surface water was detected. Majority of the water samples exceeded WHO permissible limit of 10 μg L^{-1} with a mean concentration of 46.8 μg L^{-1} in groundwater samples and 15.4 μg L^{-1} in surface water (Shahab et al., 2018).

In North America, As concentrations higher than the 10 μg L^{-1} limit were detected in more than half of 59 private wells in fractured crystalline bedrock aquifer in western Quebec, Canada while shallow wells in unconsolidated surficial deposits were uncontaminated (Bondu et al., 2017a). Further study showed that the local weathering of sulfide minerals is not the main mechanism of arsenic mobilization in borehole waters but the mobilization of arsenic is related to the reductive dissolution of Fe and Mn oxyhydroxides in the downgradient part of the aquifer (Bondu et al., 2017b).

Elevated As concentrations (6.3 – 10000 mg kg^{-1}) were reported in the sediments of 95 lakes in the Yellowknife region, Northwest Territories, Canada which could be a result of geogenic input from the weathering of mineralized bedrock and/or as a result of anthropogenic input from ore roasting at the Giant Mine (Galloway et al., 2017). Schuh et al. (2018) showed that the elevated As concentrations in sediments were largely derived from ore roasting in the region. The maximal concentration of 10,000 mg kg^{-1} is about 2000 times higher than Canada's Interim Freshwater Sediment Quality Guideline of 5.9 mg kg^{-1} (Canadian Council of the Ministers of the Environment, 2002).

Characterization of arsenic solid and aqueous phases in lake waters and sediment within 5 km of the giant mine in Canada shows that arsenic trioxide accounts for up to 6 % of the total arsenic in sediments while the bulk (>80 %) of arsenic is contained in the form of secondary sulfide precipitates, with iron oxy-hydroxides hosting a minimal amount of arsenic (<1 %) (Van Den Bergh et al., 2018). Soluble arsenic trioxide acts as primary source of As to sediment pore-waters while geogenic As is present in low concentrations in sediments and are not significant sources of As to lake waters (Van Den Bergh et al., 2018).

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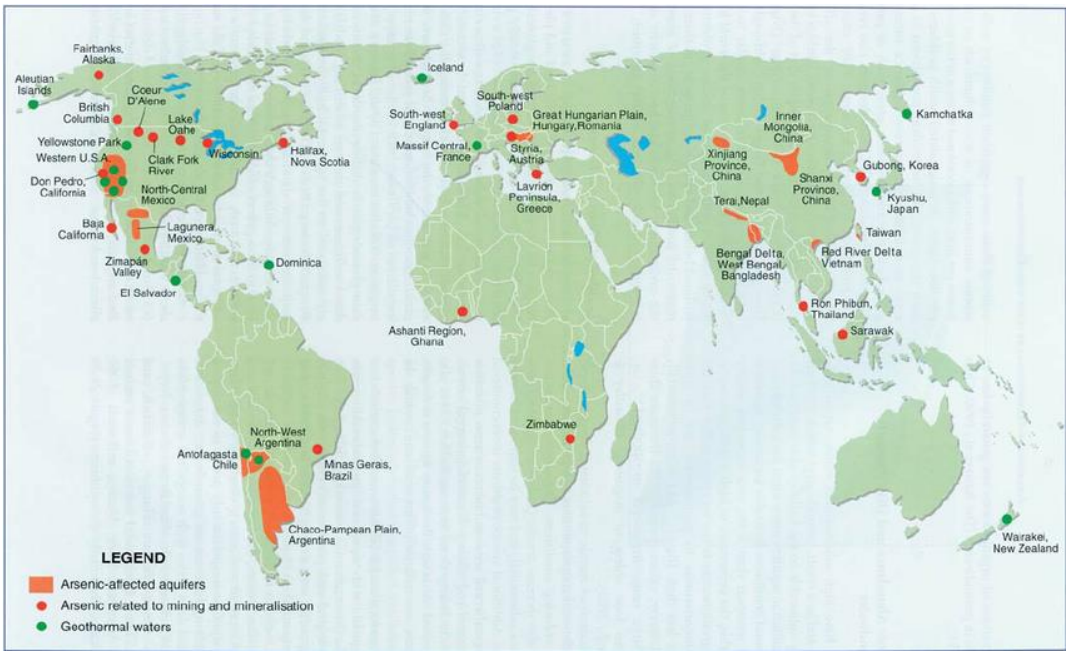

Figure 3: World Map showing locations with As concentrations in groundwater > 50 µg L^{-1}. Culled from Centeno et al., 2007.

A global map (Fig. 3) showing locations with As contamination published about a decade ago by Centeno et al. (2007) showed As contamination in Ghana and Zimbabwe due to mining and mineralization but recent studies have reported As contamination in some other African countries such as Cote d'Ivoire. The impact of industrial and artisanal gold mining on wetland sediment quality was evaluated in Cote d'Ivoire, a country in West Africa by Kinimo et al. (2018). In industrial mining areas, As concentrations varied from 7.38 µg g^{-1} at Bonikro to 294.38 µg g^{-1} at Afema while in artisanal mining areas, it ranged from < 1.6 µg g^{-1} at Agbaou and Bonikro to 5.1 µg g^{-1} at Afema (Kinimo et al., 2018). When all the sites are taken into consideration, the industrial mining areas were significantly contaminated than artisanal and non-mining areas with the average As concentration in sediments being the highest in Afema (abandoned mine) and the lowest in Agbaou site (new active mine) (Kinimo et al., 2018). Geoaccumulation index showed extreme sediment contamination in As in industrial mining areas at Afema (Kinimo et al., 2018). The contamination factor and pollution load index indicated that the concentration of As in Kafue river sediment in Zambia was very high at sites in the Copperbelt mining area (M'kandawire et al., 2017). Potential ecological risk showed that As is likely to cause adverse biological effects to aquatic organisms in the Copperbelt mining region of the Kafue River (M'kandawire et al., 2017).

Agricultural and industrial drainage was identified as a major source of pollution for surface waters in Egypt because the highest As concentrations were recorded adjacent to the agricultural

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and industrial drains (El Badry & Khalifa, 2017). Arsenic concentrations in sediment samples from Burullus lagoon, one of four natural body waters in northern Egyptian delta lakes varied from 10 to 44 mg kg^{-1}. The maximum As concentration of the sediments is higher that the probable effects concentration (PEC) for As in sediments which is 33 mg kg^{-1}. (MacDonald et al., 2000). In Nigeria, As concentrations in the Lagos lagoon decreased from 100 μg l^{-1} in 2009 to below 10 μg l^{-1} (1.29 μg l^{-1}) in 2017 (Aderinola et al., 2009; Usese et al., 2017). It is not clear what is responsible for the reduction in the surface water As concentrations.

4. REGULATION OF ARSENIC

There is growing concern over the effectiveness of the current regulation of As in drinking water. A recent study in Iowa state, USA, has shown a significant dose-dependent association between low-level arsenic exposure and prostate cancer (Roh et al., 2017). They explained that the replication of this result in future individual-level studies may suggest that 10 μg l^{-1} is not protective for human health (Roh et al., 2017). On the contrary, Welch et al. (2018) examined trends in As exposure in US general populations between 2003 and 2014 by determining urinary As concentrations by drinking water source. They reported that As exposure declined by about 35% between 2003 and 2014 among non-smoking individuals using public community water systems. Their results suggest regulation and prevention strategies to reduce arsenic exposures in the U.S. may be succeeding (Welch et al. 2018).

Sixteen years after the first arsenic contaminated groundwater with As concentrations up to 850 μg L^{-1} was reported in the region of Kuitun and Chepaizi, Dzungaria district, Xinjiang, People Republic of China, the health status of 178 villagers from endemic and 179 villagers from control sites were evaluated (Liu et al., 2013). The study revealed that 51 out of 178 people from the arsenic endemic area showed skin lesions related to arsenicosis which were absent among villagers from the control site. Furthermore, skin lesions related to arsenicosis were observed in 4 out of 9 subjects that were16 years of age or younger from different villages and born after the completion of water intervention. They concluded that there may be sporadic exposure or voluntary drinking of contaminated water or there may be other sources of dietary exposure to arsenic. (Liu et al., 2013).

Rice is a major source of As because it accumulates As more efficiently than any other food crop probably because it is grown under water logged conditions. An exposure study comparing drinking water and rice consumption as sources of As used data from a survey of 49,473 U.S. public water utilities serving approximately 230 million people and dietary data from What We Eat in America survey to estimate the exposure to As in the United States (Barrett, 2017). Barrett (2017) used the Stochastic Human Exposure and Dose Simulation (SHEDS) model to estimate that inorganic arsenic exposures attributable to drinking water and rice consumption averaged

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4.2 and 1.4 μg/day, respectively. Since about two-thirds of the drinking water samples had concentrations below the detection limit, most people will have greater arsenic exposure from rice consumption compared with drinking water (Barrett, 2017). Previous studies have shown an association between rice consumption and urinary As in women of Korean descent in Washington State, USA and pregnant women in New Hampshire, USA (Cleland et al., 2009; Orloff et al., 2009). Rice is considered a major source of inorganic arsenic exposure for the women population in Washington state where the average intake of 16.3 μg of inorganic arsenic from rice per day was estimated (Lai et al., 2015).

5. CONCLUSION

High As concentrations has been reported in virtually every continent because As is ubiquitous in the environment. A lot of emphasis has been placed on As concentrations in drinking water sources because drinking water is a major exposure route for the toxic metalloid. Hence, most of the existing literature have reported As concentrations in drinking water and population exposed to it. However, there are also some studies reporting As concentrations in surface water and sediments. Natural sources such as reductive dissolution of minerals and parent rock are mainly responsible for As contamination of drinking water in Asia while in the Americas and Europe, studies reported geothermal sources as well. Most of the studies in Africa worked on surface waters and sediments in places impacted by mining and smelting. The population that was exposed to As contamination in drinking water in Bangladesh and India over two decades ago are no longer drinking contaminated water. But they are still exposed to As via dietary sources such as rice which are still being irrigated and cooked with contaminated water. Dietary sources of As such as rice becomes the major exposure route in places where the As in drinking water is below MCL for As.

There should be a committee on As contamination worldwide to control and manage issues relating to As in all nations. This committee will look at reducing exposure to As in drinking water or through dietary exposures in food and drink such as rice and fruit juices. They will also look at ensuring that the maximum contaminant limit set by each nation is able to protect the citizens from the health effects of As exposure in their region. Most developing nations have MCL > 10 μg l^{-1} the regulatory limit for the United States and WHO. For example, the current maximum contaminant level (MCL) for arsenic in drinking water (25 μg l^{-1}) in Mexico is two and a half times higher than that of the United States and World Health Organization (WHO). In Bangladesh and India with notable episodes of As contamination, the MCL for As is 50 μg l^{-1}, five times that of the US and WHO. All nations need to provide high quality drinking water for the general public and maintain a record of As concentrations in drinking water available to communities to protect public health. However, in some places, drinking water is not the main exposure route for As but rather dietary sources. Hence. dietary sources of As such as rice,

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vegetable and fruits need to be more closely monitored and regulated like drinking water sources.

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