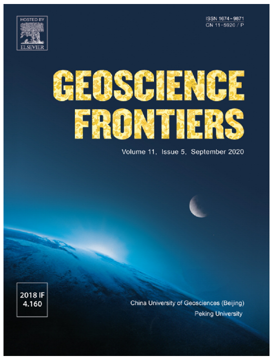

Arsenic contamination of groundwater: A global synopsis with focus on the Indian Peninsula

E. Shaji, M. Santosh, K.V. Sarath, Pranav Prakash, V. Deepchand, B.V. Divya

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Focus Paper

Arsenic contamination of groundwater: A global synopsis with focus on the Indian Peninsula

E. Shaji[a,*], M. Santosh[b,c], K.V. Sarath[a], Pranav Prakash[a], V. Deepchand[a], B.V. Divya[a]

a. Department of Geology, University of Kerala, Kariavattom Campus, Trivandrum 695 581, India

b. School of Earth Sciences and Resources, China University of Geosciences (Beijing), 29 Xueyuan Road, Beijing 100083, China

c. Department of Earth Science, University of Adelaide, Adelaide SA 5005, Australia

*corresponding author: shajigeology@gmail.com

Abstract

More than 2.5 billion people on the globe rely on groundwater for drinking and providing high-quality drinking water has become one of the major challenges of human society. Although groundwater is considered as safe, high concentrations of heavy metals like arsenic (As) can pose potential human health concerns and hazards. In this paper, we present an overview of the current scenario of arsenic contamination of groundwater in various countries across the globe with an emphasis on the Indian Peninsula. With several newly affected regions reported during the last decade, a significant increase has been observed in the global scenario of arsenic contamination. It is estimated that nearly 108 countries are affected by arsenic contamination in groundwater (with concentration beyond maximum permissible limit of 10 ppb recommended by the World Health Organization. The highest among these are from Asia (32) and Europe (31), followed by regions like Africa (20), North America (11), South America (9) and Australia (4). More than 230 million people worldwide, which include 180 million from Asia, are at risk of arsenic poisoning. Southeast Asian countries, Bangladesh, India, Pakistan, China, Nepal, Vietnam, Burma, Thailand and Cambodia, are the most affected. In India, 20 states and 4 Union Territories have so far been affected by arsenic contamination in groundwater. An attempt to evaluate the correlation between arsenic poisoning and aquifer type shows that the groundwater extracted from unconsolidated sedimentary aquifers, particularly those which are located within the younger orogenic belts of the world, are the worst affected. More than 90% of arsenic pollution is inferred to be geogenic. We infer that alluvial sediments are the major source for arsenic contamination in groundwater and we postulate a strong relation with plate tectonic processes, mountain building, erosion and sedimentation. Prolonged consumption of arsenic-contaminated groundwater results in severe health issues like skin, lung, kidney and bladder cancer; coronary heart disease; bronchiectasis; hyperkeratosis and arsenicosis. Since the major source of arsenic in groundwater is of geogenic origin, the extend of pollution is complexly linked with aquifer geometry and aquifer properties of a region. Therefore, remedial measures are to be designed based on the source mineral, climatological and hydrogeological scenario of the affected region. The corrective measures available include removing arsenic from groundwater using filters, exploring deeper or alternative aquifers, treatment of the aquifer itself, dilution method by artificial recharge to groundwater, conjunctive use, and installation of nano-filter, among other procedures. The vast majority of people affected by arsenic contamination in the Asian countries are the poor who live in rural areas and are not aware of the arsenic poisoning and treatment protocols. Therefore, creating awareness and providing proper medical care to these people remain as a great challenge. Very few policy actions have been taken at international level over the past decade to reduce arsenic contamination in drinking water, with the goal of preventing toxic impacts on human health. We recommend that that United Nations Environment Programme (UNEP) and WHO should take stock of the global arsenic poisoning situation and launch a global drive to create awareness among people/medical professionals/health workers/administrators on this global concern.

Key Words: Arsenic contamination, Groundwater, Indo-Gangetic alluvium, Orogenic belts, Global tectonics, Asia.

1. Introduction

Groundwater is one of the most precious natural resources in our planet. It is being exploited extensively in many parts of the world with a massive increase in extraction in the past few decades due to the availability of new and cheaper drilling and pumping technologies (Barbier, 2019). Hydrogeologists refer to this drastic change in groundwater utilization as 'the silent revolution', since it has occurred in many countries in an unplanned and totally uncontrolled manner (Stone et al., 2019 and references therein). The demand for good quality groundwater has increased with increasing population and developmental activities across the globe. Providing safe drinking water to the world's 7.8 billion people is one of the greatest challenges of the century. At the beginning of 20th century, the groundwater quality issues were minimal and total dissolved solids and pH were the only parameters of concern. However, during 21st century, there has been increased global attention on resolving groundwater quality issues. The chemical quality of ground water varies significantly depending on the type of aquifers, duration of rock-water interaction and the inputs from various natural and non-natural sources. During the last decade, groundwater contamination from various chemical constituents is being reported from aquifers throughout the world and often it becomes non-potable as the constituents exceed the limits prescribed by WHO. Geochemical processes during and after aquifer recharge can either improve or cause a deterioration of water quality (Maliva, 2020). In the recent years, pollution by arsenic (As) has become a serious issue of concern in view of its toxicity to humans (Polya et al., 2019 and references therein). Arsenic contaminants in groundwater can also affect the health of the aquifers.

This paper examines the current scenario of arsenic contamination of groundwater in countries across the globe with an emphasis on the Indian Peninsula. We review the global As contamination in groundwater, its ill effects on humans, sources characteristics, remediation, and also attempt to propose some recommendations for policy makers.

2. Geochemistry of arsenic

Arsenic, an element of the earth's crust with an abundance of 1.8 ppm by weight, combines with oxygen, chlorine and sulphur to form inorganic arsenic compounds. Arsenic and its compounds are widely used in agriculture, livestock feed, medicine, electronics, metallurgy, chemical warfare agents etc. Arsenic is of interest in terms of environmental issues and health impacts. Rock-water interactions in aquifer systems are the major cause of release of arsenic and causes deterioration in groundwater quality. Arsenic is the 12th most common element in nature, and it usually appears in three allotropic forms, including black, yellow, and grey. If heated, it rapidly oxidizes to arsenic trioxide (As_2O_3) and has a garlic odour (Fendorf et al., 2010). Arsenic is also known as 'king of poison' as it is a highly toxic element ranking number one in the 2001 priority list of hazardous substances and disease registry defined by WHO. Since 1993, the permissible value of the concentration of arsenic (As) in drinking water has been fixed as 10 μg/L. However, it was 50 μg/L prior to 1993. It is classified as carcinogen, mutagens, and teratogen. IARC (International Agency for Research on Cancer) has classified As is a class1 human carcinogen. In natural water bodies arsenic mostly found in two states trivalent arsenic (As^{3+}, Arsenite) and pentavalent arsenic (As^{5+}) both forms are highly toxic inorganic species (Fendorf et al., 2010). The toxicity of arsenite is much higher than that of arsenate. Generally, in groundwater, natural occurrences of high arsenic levels were reported in aquifers - especially unconsolidated sediment aquifers throughout the world and have been connected to several adverse health effects (Smedley and Kinniburgh, 2013; Mozumder, 2019). Arsenic contamination of groundwater is estimated to be affecting 500 million people around the globe. Continuous exposure to high arsenic water causes pigmentation, hyperkeratoses, ulceration, skin cancer and also affects liver, kidney, heart, and lungs (Sun et al., 2019 and references therein).

3. World scenario

The natural contamination of As in groundwater has been reported worldwide, and the majority of these belong to South Asian and South American regions (see Fig. 1; Ravenscroft et al., 2009; Bundschuh et al., 2012; Hashim et al., 2019). The severely affected countries include Bangladesh (Yang et al., 2014), India (Mukherjee et al., 2009; Bhowmick et al., 2018; Chakraborti et al., 2018; Bindal and Singh, 2019; Dhillon, 2020), China (Guo et al., 2014), Nepal (Pokhrel et al., 2009), Cambodia (Polya et al., 2010), Vietnam (Winkel et al., 2011; Stopelli, 2020), Myanmar (Van Geen et al., 2014), Laos (Cho et al., 2011), Indonesia (Winkel et al., 2008), the USA (Gong et al., 2014). In addition, countries like Argentina, Chile, Hungary, Canada, Pakistan, Mexico, and South Africa are also affected (Ravenscroft et al., 2009).

However, the South and Southeast Asian Belt is considered as the most arsenic polluted areas including India, Bangladesh, Nepal, Vietnam and China (Ravenscroft et al., 2009; McArthur, 2019). The developed countries, like USA and Canada, also experience widespread levels of arsenic contamination in groundwater although the concentrations are characteristically lower in comparison with the Asian countries (Sorg et al., 2014)

Our synthesis of the global data reveals that 107 countries are affected by arsenic contamination in groundwater (beyond WHO maximum permissible limit of 10 ppb) with highest reports from Asia (32) and Europe (31), followed by Africa (20), North America (11), south America (9) and Australia (4) (Fig. 1). Most of the arsenic pollution prone zones are located in the sedimentary basins close to the modern mountain belts and deltaic areas (Fig.

1). Regions with tropical climate are more vulnerable to arsenic contamination as this climate favours the release of As from arsenic compounds (Ranjan, 2019). The details of the 108 affected countries are shown in the World map (Fig. 1) and the list of countries are depicted in Tables 1 and 2.

In Latin America, the main sources of arsenic contamination in groundwater are geothermal fluids and volcanic activities (Simfors et al., 2020). In Mexico (North America), groundwater is the main source of drinking water (40%) and high As concentrations (>10 ppb) are reported in groundwater in different parts of Mexico (Bundschuh et al.,1997; Alarcón-Herrera et al.,2020). 1.5 million people in Mexico consume water with As above 25 µg/L, and about 150,000 people are exposed to As poisoning (Alfaro de la Torre et al., 2018). A new study by Alarcon-Herrera et al. (2020) explains that 8.81 million people are now exposed to high As groundwater. Similarly, the North American regions like Guatemala El Salvador also have high As content in water resources (Armienta et al., 2008; Libbey et al., 2015) and source is identified as volcanogenic.

In Bolivia, South America, high concentration of As (45.9 µg/L) in groundwater is recently reported (Alcaine et al., 2020) and the source is volcanic formations of the Neogene period. Similarly the groundwaters in the southern part of the Argentinian Chaco-Pampean plain are characterized by the elevated presence of arsenic (Smedley et al., 2002) and the Tertiary aeolian loess-type deposits in the Pampean plain and fluvial sediments of Tertiary and Quaternary age may be the source (Alcaine et al., 2020).

In Europe, many countries report (especially Greece, Hungary, Romania, Croatia, Serbia, Turkey, and Spain) elevated arsenic content in groundwater resources (Katsoyiannis et al., 2015). Similarly, the groundwater resources of the Pannonian Basin (Hungary, Romania, Croatia and Serbia) also show high values of naturally occurring arsenic in water (Rowland et al., 2011). A recent study by Zuzolo et al. (2020) reports As contamination of groundwater resources from central parts of Italy.

The worst affected African countries include Botswana, Burkina Faso, Ethiopia, Ghana, Morocco, Nigeria, South Africa, Tanzania, Togo, and Zimbabwe (Medunic et al., 2020). In all these African countries both surface and groundwater resources are affected by arsenic contamination, however, the severity varies from region to region.

4. Asian scenario

Thirty three Asian countries are significantly affected by groundwater arsenic contamination, which include Afghanistan, Armenia, Azerbaijan, Bangladesh, Cambodia, China, Georgia, India, Indonesia, Iran, Iraq, Japan, Jordan, Kazakhstan, Kyrgyzstan, Laos, Malaysia, Mongolia, Myanmar, Nepal, Pakistan, Philippines, Russia, Saudi Arabia, Sri Lanka, Thailand, Turkey, Turkmenistan, Uzbekistan, Vietnam, Tajikistan, and Korea (Fig. 1, Table 1). The most affected countries are China, Bangladesh, India, and Pakistan. This is followed by Vietnam. The problems related to arsenic pollution in selected countries are summarized below.

4.1 Bangladesh

The arsenic poisoning in groundwater was first reported by the Department of Public Health Engineering from the district of Chapai Nawabganj in 1993 (Huq et al., 2020 and references therein). Chakraborti et al. (2015) reported that the degree of the problem in the country was very critical and revealed that the centre part of the southeast Dhaka was the worst affected. Several studies confirm that the shallow aquifers of Bangladesh are badly affected by the high levels of As contamination (Yang et al., 2014; Edmunds et al., 2015). The As concentration in groundwater ranges from <0.5 to >4600 μg/L (Whaley-Martin et al., 2017 and references therein). Huq et al. (2020) reports that out of 64 districts, 61 districts contain

As exceeding the limit of WHO (10 µg/ L) standards for potable water and it affected more than 85 million people. Shallow groundwater of sedimentary lacustrine aquifers normally contains high As concentrations whereas deeper aquifers (> 300 m) is considered safe (Huq et al., 2020). In Bangladesh, the As contamination creates several social issues and economic problems (Rahman et al., 2018a, b). The manifestation of the arsenicosis is more intense among the poor, which is directly linked with the poverty circumstances (Rahman et al., 2018a).

4.2 Cambodia

In South and South East Asia, from Pakistan at the western periphery, to Cambodia and southern China at the eastern, groundwater used for domestic purposes is contaminated with arsenic (Richards et al., 2019). Arsenic contamination in Cambodian groundwater has a similar natural source as in Bangladesh and appears to descend from the Mekong River from natural processes in the Himalayan Mountains (Natasha et al., 2019). Agusa et al. (2002) found that groundwater wells in Kandal province in Cambodia contained an average of 178 µg/L of arsenic. In Cambodia, arsenic was also found in shallow tube wells in the low-lying Mekong delta at Prey Veng province. It is estimated that there are over 2.4 million people live in the As contaminated zone of Cambodia (Murphy et al., 2018).

4.3 China

China is one of the most affected countries facing health issues because of arsenic contamination in groundwater (Zhang et al., 2020). Although arsenic is present in several geographical regions in mainland China, Northern China has been identified as a high-risk area. It has been estimated that 19.6 million people are at risk of being exposed to arsenic-contaminated groundwater (Rodrigues-Lado et al., 2013). China faces groundwater quality issues from both industrial effluents and natural sources. The arsenic poisoning is noticed among people, who live in arid regions of the Northern provinces, probably because of scarcity of clean water for cooking and drinking from sources other than groundwater (Lado et al., 2013; Zhang et al., 2020). Arsenicosis was first reported in Kuitun region, located in the Xinjiang Uygur Autonomous Region of China in 1980.

In the Taiwan region, a good correlation was established between total arsenic and iron contents in groundwater samples and the marine sequences. The presence of clay layer within the aquifer may increase As contamination in groundwater (Liu and Wu, 2019). In Taiwan, concentration of arsenic in groundwater ranges from 10 to 1800 μg/L (Chen et al., 1995). Chronic arsenic problems were observed among a population of 40,421 in 37 villages in the country (World Bank, 2005; Liu and Wu, 2019). Rainfall significantly affects the arsenic concentration in the northern area. Black shale or pyrite material occurring in underlying geological strata are identified as the source of arsenic. Over-pumping groundwater introduces dissolved oxygen that oxidizes the immobile sulphide minerals, releases As by reductive dissolution of As-rich iron oxyhydroxides, and increases the mobile As in water (Liu et al., 2010; Maliva, 2020 and references therein).

The Chinese National Survey Program, conducted by the Chinese Ministry of Health between 2001 and 2005, tested groundwater samples from around 445,000 wells in 20,517 villages of 292 counties for arsenic contamination (Rodrigues-Lado et al., 2013 and references therein). In almost 5% of wells, arsenic levels were higher than the former Chinese standard of 50 ppb, and about 10,000 individuals were found to be affected by arsenicosis.

The Inner Mongolia is reported as another endemic area of severe arsenicosis in China. This area is mainly in the southern part of Mountain Yinshan, and the region connecting the plain north of Yellow River and the alluvial plain of Heihe River, where the groundwater is rich in natural arsenic. More than 600,000 people in 5 cities and 678 villages are identified as potential victims, and >3000 people were diagnosed with arsenicosis (Sun et al., 2004).

4.4 Pakistan

Rabbani et al. (2017) and Ali et al. (2019a) reported that 13 million people across 27 districts of Pakistan are prone to As contamination in their drinking water and people residing along the Indus River are facing higher threats. Total 9% of water resources in Pakistan were observed to carry elevated levels of arsenic above the allowable limit. Around 25%–36% population of two provinces in Pakistan, namely Sindh and Punjab, are exposed to drinking water As contamination over 10 ppb (Zudair et al., 2018 and references therein). About 11 districts of Punjab and Sindh provinces in Pakistan were found with As contamination in groundwater beyond the national defined permissible level (Ali et al., 2019b). The study by Ali et al. (2019) reports that about 656 villages and 6,173,680 people are at the risk of exposure to As in their water supply. In Sindh province, Tharparkar and Hyderabad along Indus River and in the Punjab province, Lahore and Kausar are well-known hotspot sites of natural geogenic As contamination in groundwater (Ali et al., 2019a). The most important aquifer of Pakistan is situated in Indus plain, and this area is characterized by Quaternary sedimentary deposits, mainly composed of alluvial and deltaic origin, and the sediment thickness ranges from few meters to several hundred meters in different parts. The aquifer which taps sedimentary deposit in this area has high arsenic contamination (Smedley, 2008; Ali et al., 2019b). In general, the sources are identified as both anthropogenic and natural (Smedley, 2008; Farooq et al., 2016). Coal mining activities and geothermal sources contribute in Jhelum and Chakwal Punjab provinces (Iqbal et al., 2001). In Tharparkar of Sindh province, arid environment and complex geology promote reductive dissolution of As minerals leading to contamination (100–2580 μg/L) in groundwater (Brahman et al., 2013). In Tharimirwah, Kotdigi, Sobo Dero, and Kingri, alluvial deposit of Indus river is the primary source of As contamination (Rabbani et al., 2017). In the northern region of Kohistan, mafic and ultramafic rocks are the main source of arsenic in groundwater.

4.5 Sri Lanka

Although arsenic-contaminated groundwater is not reported particularly in the metamorphic aquifers of Sri Lanka, elevated levels were found in sedimentary formations in certain parts (Mannar, Mulativu, Puttalam, and Jaffna) of the island (Chandrajith et al., 2020). In all these regions, groundwater is extracted from unconfined aquifers of the Holocene sand dunes that are underlain by Miocene limestones.

4.6 Thailand

In Thailand, arsenic in groundwater originates mostly from tin mining activities (Kim et al., 2011). Thailand is one of several countries in Southeast Asia, having problems with tin residue (Tiankao et al., 2018, and references therein). The waste piles, resulting from tin mining, contain high As (as arsenopyrite). The case studies from Ron Phibun sub-district, Nakhon Si Thammarat, a province in the southern part of Thailand (Williams et al., 1996; Smedley and Kinniburgh, 2002) recognized health problems. Approximately 1000 people have been diagnosed with As-related skin disorders, particularly in and close to the Ron Phibun town (Fordyce et al., 1995; Williams et al., 1996; Choprapawon and Rodcline, 1997). The affected area lies within the Southeast Asian Tin Belt (Schwartz et al., 1995). Arsenic concentrations are found at up to 5000 μg/L in shallow groundwater from the Quaternary alluvial sediment that has been extensively dredged during tin-mining operations (Tiankao et al., 2018).

4.7 Vietnam

Nguyen et al. (2019) reported that the people living in Nui Phao, Thai Nguyen in the northern Vietnam region have high risk of As poisoning from groundwater and vegetables. The study reveals that 75% of the groundwater samples had As exceeding the permissible limit of 10 µg/L by World Health Organization (WHO, 1999, 2004). The arsenic contamination of Vietnam was first reported in 2001 along the Red River alluvial tract (Bozack et al., 2019 and references therein). In Vietnam, a high concentration of pollutants such as As, Fe, and Mn were identified in groundwater pumped from tube wells which tap the Pleistocene aquifer (Smedley and Kinniburgh, 2002). The Red River delta is the primary source of As contamination and more than ten million people are at risk of adverse health effects (Bozack et al., 2019). The sediments in the Mekong River delta are rich in organic compounds that create an anoxic condition and that leads to the reduction of dissolved iron oxides, which in turn causes the release of arsenic (Le Luu, 2019). This is confirmed by another study that biological reduction of iron metal plays a vital role on the release of arsenic from Holocene sediments (Stopelli et al., 2020). In Vietnam, total arsenic concentration of 1–185 ug/L (average 39 µg/L) and 1–3050 ug/L (159 ug/L) were found in groundwater in the Mekong River delta and Red River delta, respectively (Berg et al., 2007).

Over all more than 230 million people worldwide, which include 180 million from Asia, are at risk of arsenic poisoning (See Table 2).

5. The scenario in Peninsular India

Groundwater plays a vital role in India to meet the water demands of various sectors, such as domestic, industrial and irrigational needs (Saha and Ray, 2019; Suhag, 2019). The alluvial tracts of Ganga and Brahmaputra rivers are the wealthiest groundwater province in the country. Most of the extraction occurs along the Indo-Gangetic basin in Northern and Northwestern India, which has resulted in significant drawdown and water table decline in many locations (Rodell et al., 2009; MacDonald et al., 2016). In India, it is reported that a population over 50 million is currently at risk from groundwater arsenic contamination. Several workers carried out extensive work on arsenic contamination in groundwaters in India, especially in Ganga basin (Chakraborti et al., 2018 and references therein). The Ganga River basin covers nearly 26% of India’s landmass and is home to a population of over 500 million (Chakraborti et al., 2018). The Ganga River basin is one of the most fertile and densely populated areas in the world (Khan et al., 2016). Presently, the Ganga is one of the world’s most polluted rivers, containing a number of toxins including chromium, arsenic, cadmium, lead, copper and mercury, as well as pesticides and pathogenic microbes nearly 3000 times higher than the safe limit prescribed by the World Health Organization (WHO, 2011; Tandon, 2018).

High arsenic (>10 ppb) groundwater has been reported in shallow aquifers from 10 states in India (CGWB, 2018), however, the deeper aquifers of India (>100 m) are free from arsenic. Arsenic contamination in groundwater was first reported from the Chandigarh region of north India (Datta and Kaul, 1976), and the second case was reported in the lower Gangetic plain of West Bengal (Garat et al., 1984). This was followed by reports from several states including West Bengal, Bihar, Uttar Pradesh, Jharkhand, Arunachal Pradesh, Assam, Manipur, Meghalaya, Mizoram, Nagaland, Sikkim, Tripura, Punjab, Himachal Pradesh, Chhattisgarh and Andhra Pradesh (World Bank, 2005; Mukherjee et al., 2006; Chakraborti et al., 2018). However, our synthesis reveals that in India, 20 states (West Bengal, Jharkhand, Bihar, Uttar Pradesh, Assam, Gujarat, Haryana, Madhya Pradesh, Panjab, Arunachal Pradesh, , Karnataka, Tamil Nadu, Himachal Pradesh, Telangana, Andhra Pradesh, Orrisa, Nagaland, Tripura, Manipur, Chhattisgarh) and 4 Union territories (Delhi, Daman and Diu, Puducherry, Jammu and Kashmir) are affected now (Fig. 2).

In India, high arsenic groundwater occurs in two categories: (1) alluvial terrane and (2) hard rock terrane. Alluvial aquifers are the main source (90%) of arsenic in India. Hard rock aquifers accounts only 10%, which includes states like Karnataka and Chattisgarh (Fig. 3). In Karnataka, arsenic is reported in association with sulfide mineralization, especially arsenopyrite. It is mainly restricted to the gold mineralized areas covering parts of Raichur and Yadgir districts. In Chattisgarh, it has been reported from the acid volcanic associated with Kotri lineament.

In the following sections, we present the state-wise details of As contamination in groundwater.

5.1 Assam

In Assam, arsenic in groundwater was first reported in 2004. Concentrations of As and other elements are higher at shallow depths, as reported for other regions of the Brahmaputra floodplain (Verma et al., 2015; Kumar et al., 2016a, b; Das et al., 2017, 2018; Patel et al., 2019).

5.2 Bihar

In Bihar, elevated levels of arsenic in groundwater was first reported in 2002, from two villages of Bhojpur district located in the middle Ganga plain (CGWB, 2018). The total population at risk is around 9 million (Thakur and Gupta, 2019). Most of the arsenic-contaminated districts tap groundwater from alluvial aquifer of the Ganga river basin in Bihar. Geological formations in the affected areas are Quaternary alluvium holding multi aquifer system, although As in groundwater is not reported from the hard rock aquifers of Bihar (Kumari et al., 2019).

5.3 Chhattisgarh

In Chhattisgarh, high concentration of As in groundwater was reported in the village of Kaudikasa in Rajnandgaon district (Shukla et al., 2010) and the high arsenic groundwater (up to 250 μg/L) wells are located in the granitic terrain with pegmatitic intrusions. Arsenic related skin cancer and keratosis were also identified from the area. High arsenic is also reported from tube-wells of Ambagarh-Chowki block (Acharyya et al., 2005), and is restricted to the N–S trending Dongargarh rift zone. The affected areas are underlain by acid volcanics, and granites. In the Chhattisgarh state, arsenic groundwater contamination is considered to be due to natural deposition of arsenic-rich pyrite, and its mobilization is due to microbial respiration of organic carbon (Ahmed, 2004).

5.4 Haryana

Arsenic concentration exceeding limit has been reported in groundwater in different parts of Haryana (Bhattacharya, 2017) Sediment containing arsenic-rich minerals which were dissolved under reducing conditions released As into the groundwater. The study of CGWB showed arsenic concentration in groundwater to be more than 50 ppb from different districts covered by alluvial aquifers (CGWB, 2018). The source of arsenic is geogenic and is occurring in alluvial sediments. The districts exposed to groundwater arsenic-contamination are drained by Yamuna river and its tributaries which originate from the Himalaya ranges (Bhattacharya, 2017).

5.5 Jharkhand

Groundwater with arsenic contamination above 50 ppb was first reported during 2004 from the Sahebganj district of Jharkhand located between the Middle and Lower Ganga Plains by CGWB. Later high arsenic groundwater (up to 133 μg/L) was reported from Sahibganj district along the river Ganga in Jharkhand (Alam et al., 2016 and references therein). Arsenic-contamination has been reported from the area close to the Ganga River and it is in those areas where the Ganga River has shifted course during the recent past. More than two hundred thousand people were affected by arsenic toxicity. Tirkey et al. (2016) studied the arsenic and other heavy metals in the groundwater samples of Ranchi city, Jharkhand.

5.6 Karnataka

In Karnataka, elevated As in groundwater is reported from hard rock aquifers and is reported mostly from the areas of gold mining and associated activities, greenstone belts of Yadgir district and southern region of Gulbarga district (Chakraborti et al., 2013 and references therein). The occurrence of arsenic is related to arsenopyrite present in the host rock. Examples are the Hutti Gold mining area in Lingasugur taluk of Raichur district and abandoned gold mining areas in Shorapur taluk of present Yadgir district. After extraction of the gold, the chemical waste was dumped on the ground surface in the adjoining areas of the mine. These dumped materials having arsenopyrite leached out arsenic during rainy season and joined the groundwater regime. Groundwater of Ingaldhal and surrounding villages of Chitradurga district also shows elevated levels of arsenic (Hebbar and Janardhan, 2016).

5.7 Manipur

Arsenic has been detected in ground water in the valley districts of Manipur namely, Kakching, Imphal East, Imphal West, and Bishnupur. These districts are located along the river courses which originate from the eastern Himalayas.

5.8 Punjab

CGWB (Central Ground Water Board) reports the presence of Arsenic in groundwater from Panjab during 2004 (CGWB, 2018). Arsenic concentration exceeding value of 10 ppb was found at 12 locations in districts of Amritsar, Gurdaspur, Hoshiarpur, Kapurthala and Ropar. All these arsenic-exposed districts are located along the river courses of the Ravi and Beas, which have also routes originating from the Himalayas.

5.9 Tamil Nadu

Sridharan and Nathan (2018) reported arsenic contamination in groundwater of Puducherry region. The Puducherry region is a sedimentary terrain composed of marine sediments, alluvium, laterites, thin seams of peat and lignite whose age ranges from Cretaceous to Recent. Over-pumping of groundwater, the rapid growth of industries, urbanizations, increase in population lead to ground water contamination.

5.10 Uttar Pradesh

Arsenic-contamination of groundwater was first reported in the Ballia district. The arsenic-affected villagers used to drink water generally from hand-pumps which tap groundwater from shallow aquifers of depth about 20–30 m. Twenty districts have been reported to have elevated arsenic in groundwater in scattered pockets. All the arsenic-affected districts in Uttar Pradesh and 12 districts in Bihar are aligned along the linear tract along the course of the river Ganga.

Shah (2015) summarised the status of arsenic contamination in groundwater resources of the north-eastern states of India. Arsenic in groundwater has been reported in the states of Assam, Arunachal Pradesh, Manipur, Nagaland and Tripura and the Brahmaputra Valley, Barak Valley and Manipur Valley are the most affected areas. Arsenic contaminated shallow tube-wells are located in the Holocene aquifer. The sources of arsenic are geogenic and the process of release of arsenic in groundwater is reductive dissolution of iron hydroxides.

5.11 West Bengal

Arsenic contamination in ground water in West Bengal was first detected in 1978, and around 16.66 million people (according to 2001 census) in 8 districts in West Bengal are at risk (Das, 2019). Das et al. (1996) and Chowdhury et al. (1999) reported that, in West Bengal, areas affected by arsenic contamination in groundwater are located in the upper delta plain, and are mostly reported from the abandoned meander belt. The source of the arsenic is geological. Bore-hole sample analyses show high arsenic concentrations only in soil layers rich in iron-pyrites (Das, 2019). It has been reported that more than 50 million people living along the Ganga–Brahmaputra basins are affected by the high levels of arsenic in drinking water. High arsenic groundwater has been observed mainly in the districts of North 24 Parganas, Burdwan, Howrah, Hooghly, Kolkata, and Nadia (Singh, 2006). The highest concentration of arsenic in groundwater was reported from Burdwan region, followed by Kolkata (Sen and Sarkar, 2019).

Here we present a new map to show relation between As contaminated areas of India and regional geology (Fig. 3). This map shows a good correlation between aquifer types and As contamination. The map shows that 90% of the affected areas belong to alluvial deposits of different ages. The hard rock aquifers of different ages spread across the country account only 10%. There are only limited reports from Decan traps and the regions with crystalline basement in the shield areas and 90% of the hard rock aquifers (including Cuddapah and Gondwana) are relatively safe zones (Fig. 3).

6. **Sources of arsenic contamination**

The main sources of arsenic contamination can be classified as natural and anthropogenic, and are evaluated below.

6.1 Natural sources

There are several natural sources, as well as anthropogenic actions that may introduce arsenic into groundwater and drinking water. The major natural sources include geologic formations (e.g., sedimentary deposits/rocks, volcanic rocks and soils), geothermal activity, coal and volcanic activities. Geothermal water can be a source of inorganic arsenic in surface water and groundwater (Welch et al., 2000). Although concentrations of arsenic in the earth's crust fluctuate, the average levels are commonly reported to range from 1.5 to 5 mg/kg. Arsenic is a significant component of many mineral species in magmatic, hydrothermal, and sedimentary rocks. It is widely present in sulfide ores of metals, including copper, lead, silver, and gold. There are over 100 arsenic-containing minerals, including arsenic pyrites (e.g., FeAsS), realgar (AsS), lollingite ($FeAs_2$, Fe_2As_3, Fe_2As_5), and orpiment (As_2S_3) (details of arsenic-bearing minerals with pictures are listed in Supplementary Table 1). Figure 4 shows arsenic concentration range (mg/kg) in common rocks, sediments and soils types. As seen in the table slate/phyllite has the maximum concentration followed by peaty soils and mudstones/marine shales. Therefore, high As groundwater is expected in sedimentary aquifers.

6.2 Anthropogenic sources

Anthropogenic related arsenic contamination in groundwater is reported in 54 countries and is largely created by human intervention, mining, coal and petroleum extraction.

The source characterisation, continent wise, is given in Fig. 5, where it is clear that the major source of As on all continents is sedimentary formations, particularly Holocene sediments. The details are also given in Table 2. In Asia, sedimentary formations contribute 45%, followed by mining (30%), coal (10%), petroleum (10%) and volcanic rocks (5%). In Europe, sedimentary formations and mining activities contribute equally followed by volcanic rocks, coal and petroleum. In America, sedimentary formations, mining activities and volcanic rocks contribute equally followed by coal and petroleum. In Africa, mining and sedimentary formations are the major contributors with little addition from coal, petroleum and volcanic rocks. In Australia all sectors contribute equally.

Anthropogenic related arsenic contamination may be categorised into different types such as mining-related, coal-related, or coal burning. Sulfides are frequently associated with gold ores, and are a potential source of arsenic. Mining and smelting of these minerals create environmental hazards of arsenic leaking into groundwater and surface water from slag pits, waste dumps, extraction basins, and mines. Mining-related (coal mining) arsenic contamination is being affected in 74 countries across the world. Petroleum-related arsenic has affected 17 countries in the world.

The main sources of As in the groundwater of India is alluvial sediments, and are mainly derived from Himalayan sediments due to erosion. Arsenic gets mobilized through the reductive dissolution of Fe^{3+} - oxyhydroxides in a reducing environment (Kumar et al., 2016a).

The main source of As in Bangladesh is largely from the Bengal Basin formed by the Ganga–Brahmaputra–Meghna (GBM) River system. This sedimentary basin has been formed by deposition of large volumes of arsenic-containing sediments that originated mainly from the Himalayas and was carried down by the mighty GBM rivers during the Pleistocene and Holocene periods. From these sediments, arsenic is leaching into the groundwater aquifers located in the fan deposit areas and Holocene alluvium.

7. Arsenic speciation in groundwater

The factors responsible for release of arsenic into the groundwater are pH, presence of organic matter in sediments (like peat, lignite and plant debris), water table fluctuation (Hinkle and Polette, 1999; Rodrıguez et al., 2004), water saturation of sediments, limited supply of sulphur and microbial activities (Matisoff et al., 1982; Chapelle, 1992; Lovely, 1997), groundwater flow direction, age of groundwater and topography (Fendorf et al., 2010), and marine transgression (Trafford et al., 1996; Berg et al., 2001). The mechanisms which cause the release of arsenic into groundwater include: (1) oxidation and dissolution of As and Fe bearing minerals (Welch et al., 2000; McArthur et al., 2001; Smedley and Kinniburgh, 2002); (2) weathering and reductive dissolution of arsenic-bearing primary and secondary minerals in the presence of natural organic matter (NOM) (Berg et al., 2001); (3) combination of both oxidative and reductive dissolution of arsenic-bearing iron oxides and oxyhydroxides (Nickson et al., 1998; Kinniburgh and Smedley, 2001; McArthur et al., 2001); (4) competitive exchange of As by other compatible ions such as nitrate, phosphate (Chowdhury et al., 1999) and bicarbonate (Nickson et al., 2000). As-rich minerals are linked with the Quaternary deposits of alluvial sediments belonging to the Holocene age (Mukherjee et al., 2009; Shah, 2010). As contamination occurs due to the reductive dissolution of As-bearing minerals (Postma et al., 2016), the As-rich sediments are transported by rivers originating from the Himalayas and are deposited into downstream basins and deltaic areas. The organic matter buried along with the sediment is utilized by microbes for metabolic activities. The microbial reduction of iron (Fe) from Fe^{3+} to Fe^{2+} due to the consumption of oxygen bound to As-bearing ferroxy hydroxides results in the subsequent release of As in water (Drahota et al., 2013; Verma et al., 2016). Arsenic contamination can take place in reducing aquifer environments, in oxidizing environments with high pH (Nickson et al., 2005), and with oxidative weathering of sulfide minerals and with geothermal activity. Soil texture also plays a significant role in providing the appropriate environment for As release into the groundwater (Hoque et al., 2009).

We use published chemical data on pH, Electrical Conductivity (EC) and As content to make scatter plots of As vs EC and As vs pH (Fig. 6a and b). It is observed that EC does not have any correlation with As content, however, pH has definite correlation with As content, with alkaline water favouring the release of As to the groundwater.

8. Arsenic contamination in the context of global plate tectonics

We show tectonic map of the world showing regions of high As in groundwater in order to evaluate the correlation between As contamination in the global plate tectonic context (Fig. 7). Arsenic contamination is markedly high in groundwaters of the circum-Himalayan region, and on the foot hills of the present-day mountain belts like Alpine-Himalayan-Tibet belt and Cordilleran-Andean belt (Fig. 7). The groundwaters hosted in Holocene aquifers, consisting of Himalayan sediments deposited by the great Asian rivers in deltaic environments, generally show high As content. The Ganga Brahmaputra river systems are the major contributors of the Bengal fan, which is considered as one of the largest modern deltas of the world. Guillot and Charlet (2007) proposed that the serpentinites enriched in arsenic and the arc-related rocks of Indus-Tsangpo suture zone could be one of the primary sources of arsenic. Further they argue that intense tectonic activity in frontal Himalayan belt associated with high rainfall conditions during the Holocene could be the possible reason for arsenic remobilization, transportation and disposition.

A close look at the global distribution of As-enriched areas in Fig. 7 reveals that the As enriched aquifers are associated with sedimentary basins adjoining major orogenic belts. Many of these sedimentary basins may be tectonically controlled and occur as foreland basins that developed by lithospheric flexure at the time of mountain building processes along convergent plate boundaries. High arsenic groundwater of these basins may thus be related to global plate tectonic framework of major orogenic belts. The transportation of As-enriched magmatic rocks from the depth to surficial deposits through tectonic processes such as exhumation, and subsequent release or mobilization of arsenic in groundwater occurs under conducive surficial biogeochemical processes. Mukherjee et al. (2014) attempted to correlate the widespread presence of As with orogens, as in areas as diverse as the Indus–Ganges–Brahmaputra basin (Himalayan orogen), the Chaco-Pampean basin (Andean orogen), Rocky mountain basin (Western Cordilleran orogen), and New England and north-eastern USA (the Appalachian orogen).

9. Prediction of spatial distribution of arsenic contamination

A few studies have used methods such as Thiessen polygon, inverse distance weighing (IDW) (Gong et al., 2014), global polynomial interpolation (Bhunia et al., 2016), and kriging (Gong et al., 2014; Sovann and Polya, 2014) to predict the spatial variation of contaminants in groundwater from different aquifers of the world. Although these methods are effective, non-availability of accurate spatial data points is the hurdle to produce meaningful outcomes. However, these methods do not account for spatial dependency of the data to predict the occurrence of the contaminants. There is always a paucity of reliable georeferenced data; thus, these models do not perform well. Therefore, predictive models that consider the factors responsible for contamination are used to overcome the limitations of the models that use interpolation for prediction. Logistic regression models (LRM) have been commonly employed to predict the spatial distributions of arsenic worldwide (Dummer et al., 2015). Several studies have used logistic regression (Zhang et al., 2013) to assess the likelihood of As contamination greater than the predefined limit of 10 mg/L by using limited As data points along with independent variables, such as geology, topography, and soil properties. Rodriguez et al. (2013) used proxies such as Holocene sediments, soil salinity, soil texture, topographic wetness index (TWI), drainage density, slope, distance to rivers, and gravity anomaly, out of which Holocene sediments, soil salinity, subsoil texture, and TWI were found to be most significant in predicting the occurrence of As in groundwater. A few studies used are linear regression (LR) (Zhang et al., 2013), principal component regression (PCR) (Luo et al., 2012), Bayesian modeling (Cha et al., 2016) and artificial neural network (ANN)-based regression (Cho et al., 2011; Bonelli et al., 2017) for As prediction models. Such models (Cho et al., 2011; Cha et al., 2016) have shown accuracies varying between 60% and 70%.

Of late, machine learning models have also been used for such studies. Machine learning models (e.g., random forests and neural networks) show higher prediction accuracy than LRM due to their strength in modelling complex relationships between response and predictor variables (Tesoriero et al., 2017). The machine learning algorithms develop sophisticated model subunits for capturing complex relationships to specify in parametric models. However, such machine learning model studies are rare in India, even though new regions have been added to existing high-As risk areas, affecting millions of people in India. A hybrid random forest model has been used by Bindal and Singh (2019) to predict the regions in Uttar Pradesh (UP) at risk due to As contamination. They predicted that 12% of the total population of Uttar Pradesh, which accounts for 23.48 million people are at risk in UP. The predictive abilities of the other models such as univariate, LRM, fuzzy, adaptive fuzzy regression (AFR) and adaptive neuro fuzzy inference system (ANFIS) were compared with that of a hybrid random forest model. Bretzler et al. (2017) used multivariate logistic regression method to create arsenic prediction models from the data sets collected from Burkina Faso, West Africa. They have predicted that aquifers of the Birimian formation with arsenic-bearing sulphide minerals, has the highest probability of yielding groundwater arsenic concentrations > 10 μg/L and further added that more than 560,000 people are potentially exposed to arsenic-contaminated groundwater in Burkina Faso. Winkel et al. (2008) predicted the groundwater arsenic contamination in Southeast Asia using logistic regression model by combining geological and surface soil parameters from the Bengal, Red River and Mekong deltas. Further, their study revealed that Holocene deltaic and organic-rich sediments are the major source of high arsenic groundwater in Southeast Asia.

This study predicts that As can be released to groundwater if the area is underlain by alluvial aquifers especially of Holocene age in foreland basins with high organic content. The mechanism of mobilisation could be reductive dissolution in humid / anoxic depositional environments under alkaline pH conditions.

To conclude, more As poisoning can be expected in areas underlain by Holocene aquifers consisting of Himalayan sediments, South and East Asia (SE China, Yangtze-Kiang basin Indonesia, Malaysia, Siberia), West Asia and Middle East (Arabian Peninsula, Turkey and Iran), South America (Western Amazonia, Pacific Plains) and in Europe (Danube delta, Baltic fringes). The other expected areas are (1) areas of intense sulphide mineralisation, (2) areas close to younger orogenic belts, and (3) areas of geothermal activity.

10. Health risk

It is believed that inorganic As^{3+} is more toxic than inorganic As^{5+} and the inorganic arsenic ingested is excreted through urine (Thomas and Bradham, 2016, and references therein). For many years, methylation and excretion were viewed as a detoxification system. It is now recognised that these methylated forms are probably the more damaging forms of As in respect of human metabolism. Long-term ingestion of water with high amount of As can cause a variety of cancers. The symptoms and the ill-effects are region specific and to some extent individual specific too. It is reported that well-nourished individuals suffer less damage than those of mal-nourished people. In India the first case of arsenic-induced skin lesions was identified at the Department of Dermatology, School of Tropical Medicine, Calcutta, India in 1983 (Saha, 2003). The illness that develops from chronic arsenic exposure is known as "arsenicosis". Skin lesions such as melanosis, keratosis, and leucomelanosis are the characteristic manifestations of arsenicosis. The WHO defines arsenicosis as a "chronic condition arising from a prolonged ingestion of arsenic above safe dose for at least 6 months, usually manifested by characteristic skin lesions of melanosis and/or keratosis with or without involvement of internal organs. Arsenicosis was first reported from Bangladesh in 1996, now it has become a severe worldwide problem (Smith et al., 2000 and references therein). The consumption of arsenic contaminated groundwater results in serious health issues like skin, lung, kidney and bladder cancer, coronary heart disease, bronchiectasis. hyperkeratosis, arsenicosis, hyperpigmentation of the palm and sole, hypertension, myocardial damage, liver damage, Bowens disease, and diabetes, among other diseases (Lalwani et al., 2004; Hopenhayn, 2006; Steinmaus, 2016; Chakraborti et al., 2018; Saha and Ray, 2019). Arsenic poisoning has also been linked to infant mortality, impaired intellect and motor dysfunction in children (Wasserman et al., 2004; Rahman et al., 2009; Parvez et al., 2011; Bhowmick et al., 2018; Saha and Ray, 2019). The details of the affected people are listed in the Table 2.

An overview chart showing the various impacts of As poisoning on humans health, collected from the above referred papers (Chakraborti et al., 2018; Saha and Ray, 2019 and references therein) is presented in Fig. 8.

11. Removal of arsenic from groundwater

Safe drinking water, nutritious food and adequate physical exercise are only the proven measures to fight chronic arsenic toxicity (Maeda, 1994). Proper watershed treatment and cost-effective conjunctive use of water along with creating mass awareness are effective the approaches to solve the arsenic crisis (Shakya and Ghosh, 2019). Inorganic arsenic can undergo microbially mediated biochemical transformation, i.e., the hydroxyl group of arsenic acid is replaced by the CH_3 group, thus getting transferred into relatively non-toxic form

(Frankenberger and Losi, 1995). The pathway of As^{5+} methylation initially involves the reduction of As^{5+} to As^{3+}, with the subsequent methylation of As^{3+} to dimethylarsine by coenzyme S-adenosylmethionine (Pierce and Moore, 1982). Methylation is often enhanced by sulfate-reducing bacteria. Several fungal species also have shown ability to reduce arsenic (United States Department of Health and Human Services, 2000).

Various technologies are being developed for the removal of arsenic contamination from groundwater in different parts of the world.

Electrocoagulation is considered an efficient technology to treat arsenic contaminated water and meet the drinking water quality standards (Mendoza-Chávez et al., 2020). A cement-based filter medium (CBFM) can also be used to remediate heavy metals from groundwater (Holmes et al., 2019).

Coagulation, chemical oxidation, advanced oxidation processes (AOPs), adsorption, ion-exchange, membrane filtration (nano-filter) and reverse osmosis are also common treatment technologies for the remediation of arsenic-contained water. The salient summary of each of these processes is given below.

Oxidation: Arsenite can be oxidized by oxygen, ozone, free chlorine, permanganate, hydrogen peroxide etc. Atmospheric oxygen, hypochloride and permanganate are commonly used for oxidation in developing countries. Air-oxidation of arsenic is very slow but chemicals like chlorine and permanganate can rapidly oxidize arsenite to arsenate under wide range of conditions (Wegelin et al., 2000).

Co-precipitation and adsorption processes: Water treatment with coagulants such as aluminium alum, activated alumina, ferric chloride and ferric sulfate are effective in removing arsenic from water. Ferric salts have been found to be more effective in removing arsenic than alum on a weight basis and effective over a wider range of pH. In both cases, pentavalent arsenic can be more effectively removed than trivalent arsenic (Pierce and Moore, 1982).

The Bucket Treatment Unit (BTU), designed for household need is based on the principles of coagulation, co-precipitation and adsorption processes. It consists of two buckets, each 20 L capacity, placed one above the other. Chemicals are mixed manually with arsenic contaminated water in one of the buckets by vigorous stirring and then flocculated by gentle stirring. The mixed water is then allowed to settle for 1–2 h. The water from the bucket is then allowed to flow into another bucket through a sand filter installed in the second bucket, very carefully avoiding the inflow of settled sludge in the first bucket. Now the second bucket practically contains treated water (Guha Mazumder, 2003).

Solar oxidation: It is a simple method of solar oxidation of arsenic in transparent bottles to reduce arsenic content of drinking water (Mukherjee et al., 2007). Ultraviolet radiation can catalyze the process of oxidation of arsenite in presence of other oxidants.

Large number of adsorbents have been utilized for removal of arsenic species (arsenite, As^{3+} and arsenate, As^{5+}) but the arsenite, As^{3+} removal requires pre-oxidation of As^{3+} to As^{5+} using oxidizing agents, which makes the process costly and sometimes produce unhealthy by-products (Zhang et al., 2007; Siddiqui and Chaudhry, 2017). Therefore, to avoid the pre-oxidation step using costly oxidizing agents, various solid materials with oxidative properties have been developed (Siddiqui and Chaudhry, 2017).

Graphene oxide (GO) and its composites have attracted widespread attentions as novel adsorbents for the adsorption of various water pollutants due to their unique physicochemical characteristics (Siddiqui et al., 2019).

Biological oxidation: Study of Pallier et al. (2010) reveals that some micro-organisms such as *Gallionellaferruginea* and *Leptothrixochracea* support and accelerate biotic-oxidation of iron, which makes a favourable environment for the adsorption of arsenic.

Arsenic can also be removed from groundwater by nano-filtration (NF) membrane configuration (Song et al., 2015).

Managed aquifer recharge also helps to reduce the As content in groundwater (e.g., Newman and Grey, 2019).

Laterite is used as a low-cost adsorbent in a sustainable filtration system to remove arsenic from groundwater in Vietnam (Nguyen et al., 2020). In India, clay rich laterite is easily available and can be tested as filtration system to remove arsenic from groundwater.

All the above methods described in this review have their own advantages and disadvantages. The filtration methods can be chosen based on the severity of the problem and affordability. The significant methods include awareness rising in the rural areas; providing nano-filters and As-removal appliances for the household as well as the community level.

12. Policy interventions

Safe drinking-water, as defined by the WHO (1999) guidelines, do not represent any significant risk to health over a lifetime of consumption. The health concerns associated with chemical constituents of drinking-water are more serious as these can cause adverse health effects after prolonged periods of usage. There are a few chemical constituents of water like As, that can lead to serious health problems.

In order to protect public health, a dual-role approach is essential, by defining the roles and responsibilities of service providers / authorities responsible for water supply and by prudent drinking-water supply quality surveillance. Organizational arrangements for the maintenance and improvement of drinking water supply services should be in place in all quality affected areas.

Surveillance of drinking-water quality can be defined as “the continuous and vigilant public health assessment and review of the safety and acceptability of drinking water supplies” (WHO, 2011). However, this recommended surveillance is not in force in many countries. Periodic quality assessment of various aquifers of globe is also not executed properly. The people exposed to As poisoning is reported to be more than 230 million across the globe, this may not be a correct figure, hence a global survey should be initiated by USGS or WHO to collect the real data.

Effective and sustainable programmes for the management of the aquifers require active support of hydrogeologists, hydrologists, water sector organisations, planners and all stake holders at large. These communities should be involved at all stages of groundwater development programmes, including initial surveys; decisions on siting of wells, siting of off-takes or establishing protection zones and monitoring and surveillance.

We urge that WHO and other relevant agencies formulate new policy recommendations to reduce the impact of As poisoning in groundwater resources and to improve safe water use in affected areas across the globe. The individual affected countries, especially Asian countries, should launch a public awareness campaign on safe water treatment or arsenic treatment equipment, providing the information on the status of As contamination and seeking additional safe water sources such as rainwater and treated groundwater. For the mitigation program, WHO or UNEP has to create a separate fund for the solution of the global As contamination of groundwater. It is essential to develop comprehensive management plans involving adequate medical, paramedical and infrastructural support within the umbrella of primary health care. The governmental agencies and NGOs need to reach out to the affected poor people, who still depend on the contaminated water for their domestic use. Erasing the threat of arsenicosis from the face of the Earth must be one of the prime mottos of policy makers. This paper recommends that a Global Policy for Arsenic Mitigation and Strategic Plan (GPAMSP) may specifically be formulated to solve the arsenic problem in drinking water.

13. Conclusions

Our study and synthesis lead to the following general conclusions.

(1) Arsenic contamination of groundwater in different parts of the world is an outcome of geologic and/or anthropogenic sources, leading to adverse effects in 230 million people.

(2) Globally, 107 countries are affected by arsenic contamination in groundwater.

(3) Southeast Asian countries, Bangladesh, India, Pakistan, China including Taiwan, Nepal, Vietnam, Burma, Thailand and Cambodia, are the most affected.

(4) In India, 20 states and 4 Union Territories have so far been affected by arsenic contamination in groundwater.

(5) While correlating the arsenic poisoning with aquifer types, it is observed that the groundwaters extracted from unconsolidated sedimentary aquifers, particularly along the younger orogenic belts, are the worst affected.

(6) Prolonged consumption of arsenic-contaminated groundwater has caused severe health issues like arsenicosis.

(7) We recommend that UNEP and WHO should view the world-wide arsenic poisoning with high priority, and launch a global drive for surveillance and to create awareness among people/medical professionals / health workers and work towards finding effective solutions.

(8) Global scale financial and logistic assistance may be essential to reduce arsenicosis. Besides, innovative interdisciplinary research should address the understanding of the occurrence, origin, distribution pattern and removal of arsenic in groundwater.

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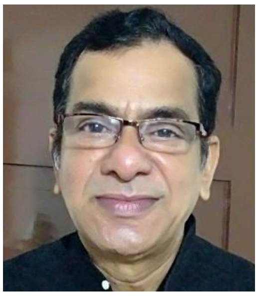
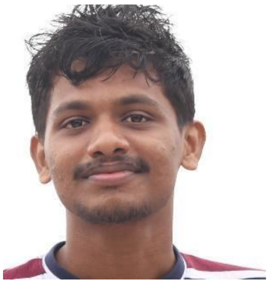
Dr. E. Shaji is an Associate Professor & Research guide in the Dept. of Geology, University of Kerala, India. Prior to the University service he worked as a professional hydrogeologist (Scientist D) in Central Ground Water Board, Ministry of Water Resources, Govt of India. He has 23 years of professional/ industry/teaching experience in Geosciences with focus on groundwater and petrology. Three scholars took PhD with him and guided several M.Phil. and M.Sc. students. He bagged Young Scientist Award from Kerala State Council for Science Technology and Environment, Govt. of Kerala. He is an Associate Editor of *Geoscience Frontiers*.



Professor M. Santosh is Foreign Expert at the China University of Geosciences, Beijing, China, Professor at the University of Adelaide, Australia and Emeritus Professor at the Kochi University, Japan. Founding Editor of *Gondwana Research* as well as the founding Secretary General of the International Association for Gondwana Research Editorial Advisor of *Geoscience Frontiers*. Receipt of National Mineral Award, Outstanding Geologist Award, Thomson Reuters Research Front Award and Global Talent award. He was recognized as Highly Cited Researcher continuously during last four years.

K.V. Sarath is a Research Fellow in the Department of Geology, University of Kerala, India. He completed his Master's degree in Geology from Central University of Kerala and Bachelor's degree in Geology from the Kannur University. His research interests include Hydrogeology, Petrology, Geochemistry and GIS/ Remote sensing.

Pranav Prakash is a Research Scholar in the Department of Geology, University of Kerala. He completed his Master's degree in Applied Geology from Pondicherry University and Bachelor's degree in Geology from the University of Kerala. His research interests include Hydrogeology, Sedimentology, Natural resources Management and GIS/ Remote sensing.



V. Deepchand is a Research Scholar in the Department of Geology, University of Kerala, India. He completed his M.Phil and Master's degree in Geology from University of Kerala and Bachelor's degree in Geology from Kannur University. His research interests include Petrology, Geochemistry, Hydrogeology and GIS.

B.V. Divya is a Research fellow in the Department of Geology, University of Kerala. She received her M.Phil and Master's degree in Geology from the University of Kerala and Central University of Kerala respectively and Bachelor's degree in Geology from Kannur University. Her research interests are in the field of Hydrogeology, Natural Resources Management and GIS/ Remote sensing.

Figure captions

Figure 1: Arsenic affected countries of the world with intensity shown by the size of the plots (see Tables 1 and 2). Note that South Asian and South American regions are the worst affected (source: Ali et al., 2019a and references therein and web sources (www.who.int/news-room/fact-sheets/detail/arsenic; www.sos-arsenic.net/)).

Figure 2: Arsenic affected states and Union Territories in India. 20 states and 4 Union territories are affected (source: www.mapsofindia.com; Chakraborti et al., 2018 and references therein; CGWB, 2018).

Figure 3: Arsenic affected areas superimposed on the general geology of India. Alluvial aquifers are the main source (90%) of arsenic in India and hard rock aquifer accounts only 10% (modified map of Aquifer systems of India; CGWB, 2017, 2018).

Figure 4: Average arsenic concentrations in rocks, sediments and soils. The main sources of As could be slate/phyllite/mudstone/marine shale/peaty soils (source: Smedley and Kinniburgh, 2002 and web resources (www.igrac.net/publications/143)).

Figure 5: Pie-chart showing continent-wise arsenic source characterisation. It is clear that the major source of As on all continents is sedimentary formations followed by mining (see Tables 1 and 2).

Figure 6: (a) Scatter plot of As vs. EC (Electrical Conductivity) in different countries. The published data show that EC does not have any correlation with As. (b) Scatter plot of As vs. pH As in different countries. The published data show that pH has correlation with As.

Figure 7: Arsenic contaminated regions superimposed on the tectonic map of the world. See that arsenic affected regions are mostly confined in the sedimentary basins close to the modern mountain belts and deltaic areas.

Figure 8: Flow chart showing various impacts of As poisoning on human health (source: Chakraborti et al., 2018; Saha and Ray, 2019 and references therein and web sources (www.sos-arsenic.net/)).

List of tables

Table 1. List of countries affected by As contamination in groundwater

Table 2. Country wise details of As-contaminated groundwater across the globe with details of population exposed and probable sources.

Supplementary Items

Supplementary Table 1: Details of As bearing minerals with pictures

Table 1. List of countries affected by As contamination in groundwater

Continent	**Number of countries affected**
North America	Canada, Costarica, Cuba, Dominica, Salvador, Guatemala, Honduras, Jamaica, Mexico, Nicaragua and USA (11)
South America	Argentina, Bolivia, Brazil, Chile, Colombia, Ecuador, Paraguay, Peru and Uruguay (9)
Asia	Afghanistan, Armenia, Azerbaijan, Kyrgyzstan, Bangladesh, Cambodia, China, Georgia, India, Indonesia, Iran, Japan, Jordan, Laos, Kazakhstan, Korea, Malaysia, Mongolia, Myanmar, Nepal, Pakistan, Philippines, Russia, Saudi Arabia, Sri Lanka, Tajikistan, Tibet, Thailand, Turkey, Turkmenistan, Uzbekistan and Vietnam (32)
Europe	Albania, Austria, Belgium, Croatia, Czech Republic, Bosnia-Herzegovina, Denmark, Estonia, Finland, France, Germany, Greece, hungry, Iceland, Ireland, Italy, Latvia, Lithuania, Macedonia, Netherlands, Norway, Poland, Portugal, Romania, Serbia, Slovakia, Spain, Sweden, Switzerland, Ukraine and United Kingdom (31)
Africa	Algeria, Botswana, Burkina Faso, Cameron, Egypt, Ethiopia, Ghana, Kenya, Libya, Malawi, Morocco, Namibia, Niger, Nigeria, South Africa, Tanzania, Togo, Uganda, Zambia, and Zimbabwe (20)
Australia and Oceania	Australia, Guam, New Zealand, Papua new guinea (4)

Table 2. Country wise details of As-contaminated groundwater across the globe with details of population exposed and probable sources.

Country	Location and maximum concentration (µg/L)	Population exposed	Environmental condition	References
Africa				
Burkina Faso	Mogtedo, Ouahigouy, Ganzourgou, Yatenga, Balé, Soum etc. (1630)	560,000	Sulphide minerals from volcanic rocks and schists. Gold mineralization in Birimian volcano-sedimentary rocks.	Bretzler et al., 2017
Cameroon	Ekondo Titi (2000)	4000		Mbotake, 2006; Ravenscroft et al., 2009
Ghana	Wassa West, Obuasi, Accra, Bolgatanga, Brong-Ahafo (4500)	Data not available		Bowell, 1994; Smedley, 1996; Buamah et al., 2008
Nigeria	Warri-Port Harcourt, Ogun State, Kaduna (750)	Data not available	Alluvial sediments, strongly reducing, slightly acidic	Oke, 2003; Edet and Offiong, 2004; Gbadebo and Mohammed, 2004
Asia				
Bangladesh	61 District (4730)	85 million	Holocene alluvial sediments (floodplain and deltaic sediments), organic matter, reducing, high alkalinity, arsenic-rich sediments	Rosenboom, 2004; Hossain, 2006; Chakraborti et al., 2015; Edmunds et al., 2015
Burma	Irrawady delta (630)	3.4 million	Holocene alluvial sediments strong reducing	Van Geen et al., 2014
Cambodia	Mekong delta, Kandal Province (1610)	2.4 million	Holocene alluvial sediments strong reducing	Berg et al., 2007; Phan et al., 2010, 2016; Kim et al., 2011; Ul Haque, 2015; UNICEF, 2016; Richards et al., 2019

China	Anhui, Beijing, Guangdng, Henan, Heilongjiang, Inner Mongolia, Jilin, Shanxi, Xinjiang etc. (2000)	19.6 million	Holocene alluvial sediments strong reducing, abundant hydrated ferric oxides, increased pH, iron phase in sediment aquifers	Guo et al., 2008; He et al., 2009; Rodriguez-Lado et al., 2013; Chen et al., 2017
India	20 District (3880)	50 million	Holocene alluvial and deltaic sediments, oxidation of arsenic rich pyrite or anoxic reduction of ferric iron hydroxides in the sediments to ferrous iron	Chakraborti et al., 2003, 2004, 2009, 2016, 2018; Mukherjee et al., 2006; Nickson et al., 2007; Bhowmik et al., 2018
Japan	Kyushu geothermal fields, Fukuoka; Kumamoto; Fukui; Takatsuki Torokuetc (25,700)	No data	Volcanic sediments, Holocene coastal sands, Quaternary alluvium aquifer	Tsuda et al., 1995; Mitsunobu et al., 2013; Even et al., 2017
Nepal	25 Districts (2620)	13 million	Geogenic and the dissolution of arsenic-bearing rocks, sediments and minerals; changes in reduction conditions, iron oxyhydroxides	Neku and Tandukar, 2003; Pokhrel et al., 2009; Thakur et al., 2010; Brikowski et al., 2014
Pakistan	27 Districts (2580)	13 million people out of total 40 million in 27 districts	Quaternary sediments (alluvial and deltaic origins; high percentage of fine to very fine sand and silt)	Fatmi et al., 2009; Brahman et al., 2013; Bibi et al., 2015; Rehman et al., 2016; Rasheed et al., 2017; Ali et al., 2019
Russia	Kamchatka (360,000)	Data not available	Hot springs	Karpov and Naboko, 1990
Australia and Oceania				
Vietnam	Red River Delta and Mekong Delta (3050)	10 million	Pleistocene and Holocene sediments; strongly reducing, high alkalinity, arsenic-rich sediments	Stanger et al., 2005; Berg et al., 2007; Nguyen and Itoi, 2009

Australia	Victoria region, New South Wales (300,000)	1976	Pyrite sediments, hydroxides and Fe oxyhydroxides, gold mining	Hinwood et al., 1999; Smith et al., 2003; Appleyard et al., 2006
Guam	Tumon Bay (1200)			Vuki et al., 2007
New Zealand	Waiotapu Valley, Rarangi, Marlborough (8500)	1939	Alluvial aquifers, reduced groundwater, geothermal water	Grimmett and Mclntosh, 1939; Webster and Nordstrom, 2003
Europe				
France	Tinee and Vesubie valleys, Vosges and the Pyrenees, Aquitaine basin (263)	17,000	Sedimentary basin, oxidation, ore deposits containing arsenopyrite	Saoudi et al., 2012; Barats et al., 2014; Drouhot e al., 2014
Germany	Bavaria, Saxony, Wiesbaden (550)	Data not available	Alluvium sediments, mineralized sandstone	Heinrichs and Udluft, 1999; Schwenzer et al., 2001
Italy	Limbardia, Emilia Romagna and Veneto (1300)		Shallow groundwater, hydrothermal, geothermal arsenic common around the volcanic canters	Tamasi and Cini, 2004; Giuliano et al., 2005; Vivona et al., 2005
Spain	Madrid basin, Duero Basin (613)	50,000	Alluvial sediments, strong reducing	Sanz et al., 2001; Garcia-Sanchez et al., 2005; Gomez et al., 2006
UK	Midlands, Cornwall, Liverpool, Northwest England (355)	Data not available	Limestone, sandstone, estuarine alluvium, mining, alluvial or glacial aquifers	Edmunds et al., 1989; Millward et al., 1997; Middleton et al., 2016
North America				
Canada	Nova Scotia, Saskatchewan, Ontario, British Columbia, Alberta (100,000)	Data not available	Thermal spring, Sulphide mineralization in volcanic rocks, high pH, sorption to ion oxides	Wyllie, 1937; Boyle et al., 1998; Kim et al., 2002; Kwong et al., 2007; Dummer et al., 2015; Bondu et al., 2017
Mexico	Region Lagunera, Valle del Guadiana, valle da Zimapan	2 million	Volcanic sediments oxidation of sulphide and arsenopyrite, dissolution of	Wyllie, 1937; Boyle et al., 1998; Kwong et al., 2007; Dummer et al., 2015; Boochs et al.,

	(2400)		scorodite	2017
USA	Alaska, Arizona, California, Hawaii, Idaho, Maryland Massachusetts, Nevada, Oregon, Utah, Washington etc. (24,300)	Data not available	Holocene and older basin-fill sediments, high pH, up-flow of geothermal water; dissolution of or desorption from iron oxide; dissolution of sulphide minerals	Southwick et al., 1983; Stauffer and Thomson, 1984; Welch, 2000; Flanagan et al., 2014; Luczaj and Masrik, 2015
South America				
Argentina	Chaco-Pampean Plain, Cordoba, Salta, Jujuy, La Pampa (14,969)	2 million	Tertiary- Quaternary volcanic deposits, post volcanic geysers and thermal springs, excessive irrigation strongly affects local geochemical and hydrochemical conditions	Nicolli et al., 1989, 2012; Auge, 2014; Panigatti et al., 2014; Robles et al., 2016
Brazil	Ribera Valley, Amapa State, Rio das Valihas, Minas, Gerais, Rondonia State, Amazon (100,000)	Data not available	Sulphide-rich gold-bearing rocks that constitute the aquifers	Matschullat et al., 2000; Figueiredo et al., 2007; Bidone et al., 2016; Ciminelli et al., 2017; Tea et al., 2019
Chile	Northern and Central Chile (27,000)	500,000	Quaternary volcanogenic sediment, oxidizing, arid conditions, high salinity	Borgono et al., 1977; Romo et al., 2003; Herera et al., 2014; Corradini et al., 2017

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Highlights

- Nearly 108 countries of the globe affected by arsenic contamination in groundwater.
- More than 230 million people, including 180 million from Asia are at risk.
- More than 90% of arsenic pollution is inferred to be geogenic.
- Prolonged consumption of arsenic-contaminated groundwater results in severe health issues.