Advances in Water Security

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Sushant K. Singh *Editors*

Arsenic Water Resources Contamination

Challenges and Solutions

Springer

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Preface

Ensuring potable water to billions of lives across the globe is a serious challenge facing humanity. Arsenic water contamination through natural or anthropogenic sources has been causing global mass poisoning. Nearly 300 million individuals are potentially at risk due to their exposure to water resources with unsafe levels of arsenic in more than 100 countries. The impact of arsenic contamination is severe in low-income communities with children being the most vulnerable. In this book, we synthesize research conducted on various aspects of arsenic water contamination across the globe.

Topics covered in this book are grouped into four main themes: (1) arsenic contamination, (2) routes of arsenic exposure and potential health risks, (3) sustainable arsenic mitigation technologies and management policies, and 4) cases studies. Chapters 1 through 3 cover the arsenic contamination and its associated geochemical and hydrogeochemical aspects. Chapters 4 and 5 have detailed information of the routes of arsenic exposure to human beings and associated health risks. Various aspects of arsenic mitigation policies and sustainability are discussed in Chaps. 6 through 10. Finally, two case studies are covered in Chaps. 11 and 12.

This is the second book in the Advances in Water Security Book Series. It is a unique book as it uses a holistic approach in investigating the risks related to groundwater resources. Overall, the book contributes to a better understanding of groundwater resources contamination and human health topics of interest to different stakeholders who are interested in a better understanding of these issues and need reliable information to help them make well-informed decisions. This book sheds the light on this global environmental menace and will draw the attention of a diverse group of researchers and decision-makers interested in environmental sciences, management, and sustainability to public health and policy. In addition, this book can serve as a reference for many groundwater hydrologists, students, and professionals.

Prairie View, TX, USA — Ali Fares
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Contents

Chapter 1
Groundwater Arsenic Contamination and Availability of Safe Water for Drinking in Middle Ganga Plain in India

Sudarsan Sahu and Dipankar Saha

Abstract The Middle Ganga Plain (MGP) is the second largest arsenic (*As*) contaminated groundwater zone in south-east Asia after the Bengal Delta Plain. About 90% of the total population in the MGP depends on shallow aquifer based groundwater supply for their drinking and irrigation need. More than ten million people in the alluvial belt of MGP are exposed to elevated levels of *As* (more than the limit set by Bureau of Indian Standards (BIS) 2012 drinking limit of 50 μgL^{-1}) in their drinking water. In the light of synthesis of groundwater *As* distribution in MGP, the availability of safe sources of water for drinking have been analyzed. The current study delineates two aquifer systems in MGP through the study of borehole lithologs ($n = 27$). The first aquifer system persists from below the top aquitard to up to ~87–126 m below ground level (bgl), while the second aquifer system starts at various depths ranging from ~116 to 139 m bgl and continues up to the depth of ~250 m bgl. Suitability of the deeper (second) aquifer system for *As* free drinking water has been assessed through analysis of pumping test data. The Newer Alluvium comprising the top aquitard and the upper 15–20 m slice of the first aquifer system hosts the contaminated aquifers for the depth of ~50 m below ground surface. The Pleistocene aquifers (the bottom three-fourth part of the first aquifer system and the entire second aquifer system) are low in groundwater *As*. The second aquifer system, existing in confined condition, can be used for community water supply through the installation of deep tube wells with a regulated draft. Community hand pumps in the depth range of 5–15 m below ground can be constructed in the sandy areas in the floodplain for drinking need of the local people. Large diameter dug wells can also be constructed to tap the replenished dynamic groundwater, which normally remains oxic and low in *As*.

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1 Introduction

The United Nations Security Council (UNSC) has adopted universally applicable Sustainable Development Goals (SDGs) for transforming the world and to end all forms of poverty by the year 2030 (UNSC 2017). Among those, sustainable development goal 6 (SDG 6), which reads as ‘Ensure access to water and sanitation for all,’ seems to bear the greatest significance in the contemporary world where around 40% of the people are affected by the scarcity of water. It is a major challenge for the governments and other authorities responsible for providing safe and affordable drinking water to all in a sustainable way by 2030. During the last few decades, besides the physical and/or economic water scarcity, deterioration in the quality of groundwater owing to both geogenic and anthropogenic reasons has reduced fresh water available for drinking supplies. It has been causing a kind of water scarcity in a scenario where the resource is plentifully available (Bigas 2012; Sharma et al. 2017).

Groundwater arsenic (*As*) of geogenic origin has been the major and serious health concern worldwide for the last several decades (Polya et al. 2005; Acharyya and Shah 2007). More than 296 million people are exposed to unsafe levels of *As*, beyond the World Health Organization (WHO 1996) set limit of 10 μgL^{-1}, in their drinking water and food supplies (Chakraborti et al. 2017). The contamination is widely pervasive in low-lying alluvial settings such as floodplains of river systems and fluvio-deltaic regions of southern and south-eastern Asia. The deltaic region of Ganga-Brahmaputra-Meghna River system, also referred as Bengal Delta Plain (BDP), is the most severely affected region of the world (Nickson et al. 1998; Burgess et al. 2010) where more than 50 million people were affected by the elevated levels of *As* in drinking water (Nickson et al. 1998; Burgess et al. 2010; Chakraborti et al. 2017).

Groundwater *As* contamination (greater than 50 μgL^{-1}) in the middle reaches of Ganga Plain was first detected in 2002 (Chakraborti et al. 2003). In the subsequent decade, a number of studies and researches focused on the groundwater *As* contamination in the Middle Ganga Plain (MGP). The eastern parts of Uttar Pradesh (U.P.), the state of Bihar and the north-eastern parts of Jharkhand state falling in the stretch of MGP (Fig. 1.1) has been identified as the second largest groundwater *As* contamination zone in south-east Asia after the BDP (Acharyya and Shah 2004; CGWB and PHED 2005; Shah 2008; Saha 2009; CGWB 2010; Saha et al. 2011a; CGWB and BIT 2013; Saha and Shukla 2013; Sahu and Saha 2015b; Saha and Sahu 2016). Wide alluvial tracts in the floodplains (of Holocene and Recent) along both the banks of the axial drainage Ganga and a few other Himalayan rivers exhibit elevated concentrations (greater than 50 μgL^{-1}) of groundwater *As* (limit set by Bureau of Indian Standards (BIS)) (BIS 2012) in the shallow aquifers largely within ~50 m below ground level (Pandey et al. 2009). In the state of Bihar only, an estimate indicates ~ten million people reside in the risk zone being exposed to the threat of *As* contamination (Sahu and Saha 2015b). If 10 μgL^{-1} limit of WHO is considered, this number can be in many folds (Sahu and Dwivedi 2012).

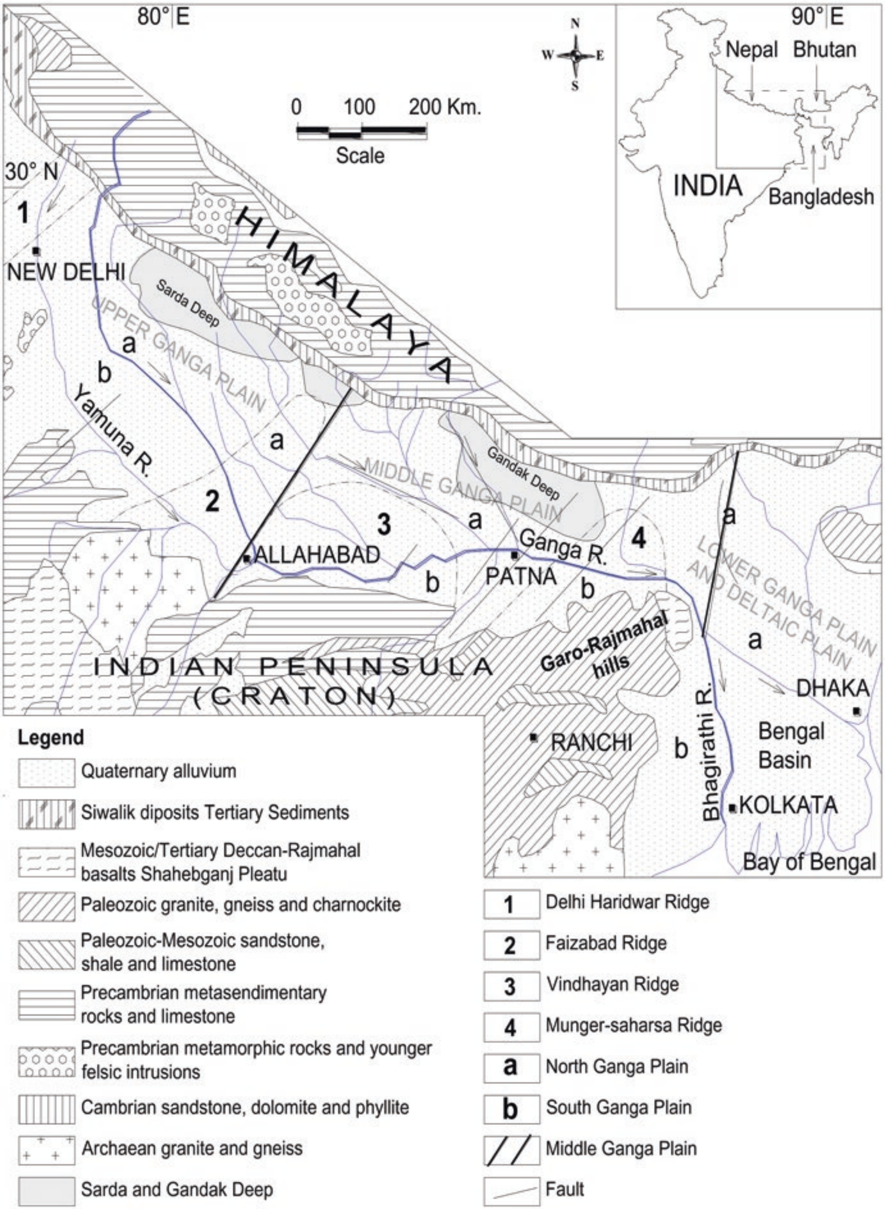

Fig. 1.1 Location map of the study area. The MGP lies between the Faizabad Ridge at the west and the Garo-Rajmahal hills at the east: the figure was cited from (Saha and Sahu 2016)

Several very significant works of research related to groundwater *As* in MGP describe its occurrence and probable mobilization mechanisms (Acharyya and Shah 2004; CGWB and PHED 2005; Mukherjee et al. 2007; Kumar et al. 2016; Nayak et al. 2008; Shah 2008; Saha 2009; MDWS 2011; Singh et al. 2016; Chakraborti et al. 2013). Limited studies are available which focus on the role of geology in the occurrence of groundwater *As* and the spatial distribution of the contaminant in different aquifer systems in MGP (Pandey et al. 2009; Saha et al. 2011a; Saha and Shukla 2013; CGWB and BIT 2013; Sahu and Saha 2015b; Saha and Sahu 2016). The present work delineates alternate safe deeper aquifer in MGP (Fig. 1.1), which can be tapped for drinking water supply. In order to identify the availability of such aquifers, the current study has analyzed the mobilization path of groundwater *As* in relation to the Holocene geomorphology and the stratigraphic architecture. It encompasses the study of aquifers geometry and their hydraulic characteristics to ascertain their aloofness from future contamination. The study also aims at delineating the probable alluvial patches in a part of the affected corridor where the shallow aquifers can host potable water with a lower concentration of *As*. In doing so, the study detects the vulnerable environments in the floodplains which are susceptible to groundwater *As* contamination.

2 Study Area Set Up

The Ganga Plain in the Himalayan foreland basin is a shallow asymmetric depression that slopes easterly (Singh 1996). The deepest part lying close to the southern margin is occupied by the axial drainages, the Yamuna in the west and the Ganga in the east. The southern part slopes north/northeastward, whereas the northern part slopes south/southeastward, converging along the axial drainage line (Singh 1996). The plain has remained an active fluvial depositional basin, and has been configured in relation to the active response of the basin to the extra- and intra-basinal tectonics, climate-driven sediment and water regime (Singh 2004; Sinha et al. 2005a; Sahu et al. 2015). The plain has been formed out of the Quaternary alluvial deposits laid down by the rivers emerging from north (the Himalaya) as well as the south (the Indian peninsula) (Sahu and Saha 2015b; Singh 1996).

The basement of the Ganga Plain is criss-crossed with several ridges, depressions, and faults (Fig. 1.1). The Delhi-Hardwar Ridge at the west, the Faizabad Ridge at the middle and the Munger-Saharsa Ridge at the east (Rao 1973; Singh 1996; Narula et al. 2000; Sinha et al. 2005b) divide the plain into three major physiographic units from west to east; (i) the Upper Ganga Plain (UGP), in the west of Faizabad ridge, (ii) the MGP, between Faizabad and Munger-Saharsa ridge/Garo-Rajmahal hills, and (iii) the Lower Ganga Plain (LGP) in the east of Munger-Saharsa ridge. The LGP merges with the deltaic region of GBM (Singh 1991; Thomas et al. 2002). In the current paper, the alluvial plain bounded between the Ganga-Yamuna confluence (on the eastern fringe of the Faizabad Ridge) at the west and Rajmahal

Hills (on the eastern fringe of Munger-Saharsa Ridge) at the east has been considered as the MGP (Saha and Sahu 2016).

The MGP is divided into two major geomorphic units by the axial river Ganga; the North Ganga Plain (NGP) and the South Ganga Plain (SGP) (Fig. 1.1). In SGP, the northerly thickening Quaternary deposits are unconformable laid directly over the Precambrian basement (Prakash et al. 1990; Agarwal et al. 2002) in the western and the central parts, while the Gondwanas and the Rajmahal Trap of Mio-Pliocene age lie between the Quaternary sediment and the Precambrian basement in the eastern part. In the NGP, the Quaternary sediment overlies the Siwaliks deposits (Agarwal et al. 2002; Singh 2004). The southern extent of the Siwaliks is yet to be ascertained (Sastri et al. 1971; Singh 1996). The basement faults, ridges and depressions have affected the river channel behaviour on the surface (Singh 2001), consequently affecting the nature and thickness of sediment fill at different parts of the basin (Singh and Bajpai 1989; Singh et al. 1999).

The major northern tributaries of the Ganga, such as the Ghaghra, Gandak, Kosi, and Mahananda, originating either from Himalayan mountains or its foot-hills are located in MGP (Singh 1991, 1996). Among the southern tributaries, the Sone is the mightiest one (Fig. 1.2). The Himalayan rivers get ample contribution to drainage from the glacial melt (Owen et al. 2002). They are generally multi-channel and braided with higher annual average discharge and sediment load. The southern tributaries, in contrast, are ephemeral, and most of them remain dry during the summer months. The MGP with an average slope of ~2% is characterized by both the degra-

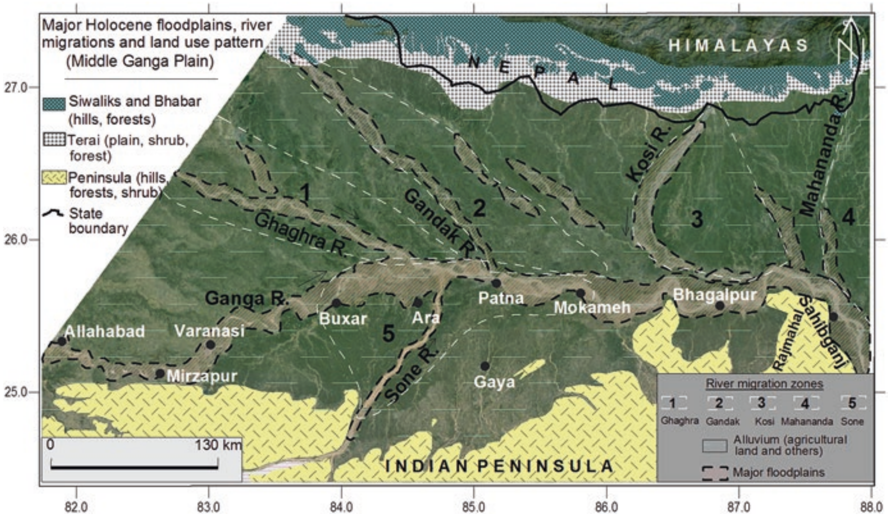

Fig. 1.2 Major Holocene floodplains and broad categories of land use in MGP (after Saha and Sahu 2016). The Himalayan rivers such as Ghaghra, Gandak, Kosi, and Mahananda form floodplains by shifting within the span of their megafans through channel migrations and avulsions. The floodplains along both the banks of the Ganga River are formed due to the oscillation and migration of the river. The Bhabar and Terai sub-zones together form the piedmont zone (Singh 1996)

dational and aggradational regimes. While the western parts adjoining to the UGP show erosional behavior, the rest of the eastern part of the unit shows predominantly a depositional setting (Sinha 2005). Sinha et al. (1996) have established a rapid rate of sediment accumulation during the late Holocene (0.7–1.5 mmyr^{-1} in last ~2400 years) in part of the MGP.

Channel migrations and avulsions are more frequently observed in MGP, resulting in frequent occurrences of backs-swamps, meander scars, channel cut-off lakes, abandoned channels/crossbar channels, short length natural levee, etc. For instance, the Kosi has shifted ~150 km westward in the last 200 years by a series of avulsions (Wells and Dorr Jr 1987; Agarwal and Bhoj 1992). Similarly, the Gandak, the Sone and even the smaller rivers in the eastern half of MGP like Bagmati and Burhi Gandak also showed dynamic characters (Mohindra et al. 1992; Jain and Sinha 2005; Sahu et al. 2010; Sahu and Saha 2014). In the case of the Ganga, the north-south oscillation at different segments (Swamee et al. 2003; Shah 2008; Sahu and Saha 2014) through channel migration and meander cut-offs are quite visible in the satellite images (Fig. 1.2).

Morphologically, the Gangetic Plains is sub-divided into three distinct zones: (i) the piedmont zone, located close to the Siwalik Hills (including the Bhabar and Terai sub-zones) (Fig. 1.2), (ii) the central alluvial plain, occupying the area between the piedmont zone and the axial rivers (the Yamuna at the west and the Ganga at the east), incorporating the floodplain of the Ganga, and (iii) the marginal alluvial plain between the axial drainage at north and the Indian peninsula (the craton) at south (Karunakaran 1976; Agarwal et al. 2002). The Bhabar[1] possesses graveliferous sediments with steep slopes, whereas the Terai[2] is characterized by low-lying areas with the development of swamps, ponds and small sandy layers (Pathak 1982; Singh 1996). The Bhabar and Terai together constitute the piedmont zone at the base of the Siwalik foothills (Singh 1996). The central alluvial plain, which occupies the major part, consists of (i) interfluve upland terrace (T_2) representing the Older Alluvium (OA), and (ii) river-valley terraced formations, referred as Newer Alluvium (NA) (Fig. 1.3). The OA is made up of yellow to brownish yellow, medium to coarse sand with profuse calcareous nodules and iron concretions, inter-layered with clay and sandy clay (Shah 2010). The NA of Holocene age characteristically consists of unoxidized fine to medium sand, silt and clay (gray to dark gray in colour) deposited in the fluvial/fluvio-lacustrine setting. Within the NA the floodplain of the Ganga and its tributaries are incised on T_2 surface. The entrenched river valley terrace (T_1) upon the T_2 surface, also referred as Older Floodplain (OFP), normally occurs above the regular flood level of the rivers. The Active Floodplain (AFP), also mentioned as T_0 surface, are aligned as narrow zones along the rivers, entrenched

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[1] Bhabar is the term used for the proximal part of the piedmont zone which comprises coalescence of several gravelly alluvial fans with sediment derived from the Siwaliks.

[2] The Terai belt to further south is the distal part comprising fine sandy layers within the silty-clayey framework. The entire piedmont zone is 10–50 km wide (Pathak 1982; Goswami and Yhokha 2010).

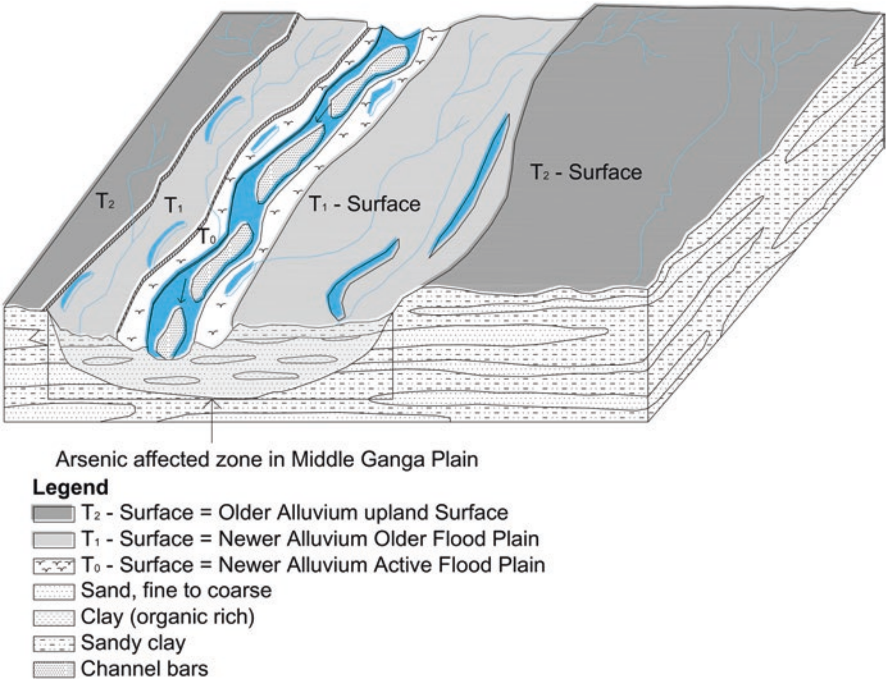


Fig. 1.3 Schematic model of MGP showing the disposition of NA and OA. The OA upland surface comprising of Pleistocene sediments is low in *As* content (Saha and Sahu 2016; Shah 2008)

upon OFP and are marked with Recent sediments. The T_0 surface is subjected to annual flooding (Acharyya 2005; Sahu and Saha 2015b).

2.1 Floodplain Geomorphology

The floodplain of the axial drainage Ganga is dotted with a series of abandoned channels and meander scars, specifically on its right bank, which are left behind in the course of migration of the river (Singh 1996; Sinha et al. 2005a; Sahu et al. 2010). The geomorphic sub-features include low-lying mudflats, channel cut-off lakes, levees, and point bars with the typical ridge (scroll bar) and swale topography. The active cut-off lakes are observed particularly in the AFP (Sahu and Saha 2015a, b).

The cut-offs as well as small abandoned cross-bar channels associated with the meander scars, mainly in the OFP, have been leveled with finer argillaceous clayey sediments. However, those still form depressions and remain under seasonal flood water. When flood water recedes, those behave as mudflats. The point bar surfaces show alternating ridges and swales or depressions (meander scroll topography), formed as a result of channel shifting and formation of small and large cross-bar channels. The ridges are sandy with only a minor mud drape (0.5–1.0 m thick) at the surface. The depressions (swales), however, possess considerable clay/mud of ~2.0–7.0 m thickness. Levees in the study area are wide and subdued in comparison to those observed in the downstream parts particularly around Patna urban area. They dip gently towards the south and merge with the floodplain. The levees along the cut-bank of the old meander scars are composed of silt and mud with infrequent lenses of very fine sand. However, the levees on the inner side of the meanders are relatively rich in the sand with minor lenses of mud (Sahu and Saha 2015a, b).

3 Methodology

The study takes the help of published works on groundwater *As* contamination and its spatial distribution in MGP (Chakraborti et al. 2003; CGWB and PHED 2005; Ahamed et al. 2006; Ramanathan et al. 2006; Mukherjee et al. 2007; Nayak et al. 2008; Shah 2008, 2010, 2014, 2015; Saha 2009; Saha et al. 2011a; PHED 2009; CGWB 2010; MDWS 2011; CGWB and BIT 2013; Chakraborti et al. 2017; Saha and Shukla 2013; Sahu and Saha 2015b; Chakraborti et al. 2013). The heterogeneity in the distribution patterns of *As* and their relation with geology helps to define specific environments in MGP and their groundwater *As* character. Based on the understanding, and the sedimentary facies and sequence models as proposed by Sahu and Saha (2015b), a geomorphic model has been proposed to describe the spatial variability of groundwater *As* in MGP. The available borehole lithologs (CGWB 2013) of shallow depth (~30 m bgl) have been studied in sub-meter scale to differentiate the Himalayan derived gray sediment from the brownish yellow sediment derived from the southern Indian peninsula (the craton). Aquifer systems in MGP have been delineated assessing the available borehole lithologs ($n = 27$) up to 300 m below the ground surface (CGWB 2013, 2015). Sahu and Saha (2016) and Sahu et al. (2018) have broadly assessed hydraulic conductivities and groundwater potential in various granular zones in the stratigraphic framework and the aquifer systems thereof based on the borehole sediment grain size analysis and the use of empirical methods. Sahu et al. (2018) have also analyzed the pumping test data of several deep tube wells ($n = 19$). The present work re-interprets the pumping test data of seven observation wells and one piezometer, which were constructed near the pumping wells tapping the deeper aquifer system in the *As* affected parts of MGP (CGWB 2013). Suitability of the aquifer system has been assessed for the supply of safe water for drinking.

4 Occurrence of Groundwater Arsenic

Various government agencies and other researchers have carried out several analysis campaigns and studies pertaining to the groundwater *As* contamination in various parts of MGP since 2003 (Kumar et al. 2010, 2012, 2016; Acharyya and Shah 2004; CGWB and PHED 2005; Nickson et al. 2007; Ahamed et al. 2006; Ramanathan et al.

2006; Mukherjee et al. 2007; Nayak et al. 2008; Shah 2008, 2010, 2014, 2015; Saha 2009; Kunar et al. 2009; Pandey et al. 2009; Saha et al. 2009, 2011a; Chauhan et al. 2009; CGWB 2010; MDWS 2011; PHED 2009; Saha and Dhar 2012; Yano et al. 2012; CGWB and BIT 2013; Chakraborti et al. 2013, 2016a, b, 2018; Singh and Ghosh 2011, 2012; Singh et al. 2014; Saha and Shukla 2013; Sahu and Saha 2015b).

Arsenic contamination has been observed largely in alluvial corridors in the valleys of particularly the Himalayan rivers with severe incidence along the axial drainage Ganga (Fig. 1.4). The corridors mark the floodplains of the rivers where large number of shallow hand pumps (depth range: less than 50 m bgl) have been found to be affected by elevated concentration of groundwater *As* (CGWB and BIT 2013; Kumar et al. 2016; Saha et al. 2011a; Saha and Shukla 2013; Sahu and Saha 2015a, b; Shah 2008, 2010, 2014, 2015; Singh et al. 2014, 2016; Singh and Ghosh 2011, 2012; Chakraborti et al. 2013). In the state of Bihar, the contamination zone covers a geographical area of 9104 km^2 falling mostly in the rural parts of 15 districts (out of total 38) (Fig. 1.4) encompassing 57 blocks including more than 1600 habitations. *As* per the estimate of Sahu and Saha (2015a), considering the BIS limit of 50 µgL^{-1}, more than ten million people (~10% of state populace) who depend on the shallow contaminated aquifer for their entire drinking need reside in the risk zone. If 10 µgL^{-1} limit (WHO 1996) is considered, this number can increase in many folds (Sahu and Dwivedi 2012). Figure 1.4 shows the districts in MGP in the states of U.P., Bihar and Jharkhand where groundwater *As* contamination is reported.

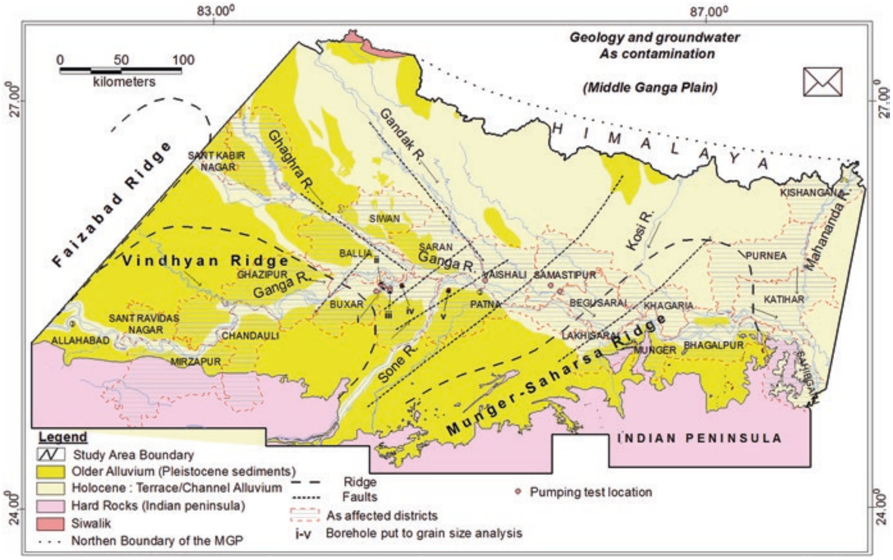
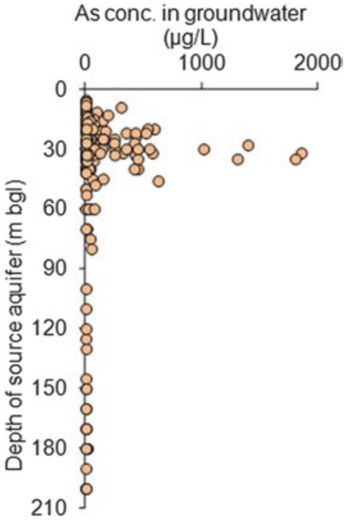


Fig. 1.4 A generalized geological map of MGP. It also depicts the districts with elevated concentration of *As* in groundwater in the shallow aquifers (the figure has been cited from Saha and Sahu 2016). The locations of tube wells where pumping tests have been carried out and the boreholes used for grain size analysis of drill-cut samples have been indicated in the map (see later sections)

In the similar line of West Bengal and Bangladesh in BDP, considerable spatial heterogeneity in groundwater *As* concentration is also observed in MGP. The contamination occurs in patches and not the entire alluvial stretch of any locality shows the contamination. It is associated with the local geomorphic and sedimentary units (Sahu and Saha 2015a, b). Spatial heterogeneity in the distribution of various sedimentary units associated with different geomorphic setups causes multi-fold differences in *As* concentrations in the hand pumps (Saha and Sahu 2016; Sahu and Saha 2016). Even in the same village, more than a hundred-fold difference in the *As* content in the hand pump water has been noticed (Chakraborti et al. 2003; Singh et al. 2014). The spatial variation in concentration of *As* along the northern bank, as wide as in the areas from the southern bank of the Ganga, has not been reported. Higher *As* concentration in the range of 500–1861 μgL^{-1} have been recorded from various parts of MGP (Ahamed et al. 2006; CGWB and BIT 2013; Chakraborti et al. 2003; Chauhan et al. 2009; Kumar et al. 2010; Kunar et al. 2009; MDWS 2011; Mukherjee et al. 2007; Pandey et al. 2009; Saha et al. 2011a; Sahu and Saha 2015b; Shah 2014; Chakraborti et al. 2013). Various workers in specific research areas from major parts of MGP report ~40–80% of the hand pumps showing groundwater *As* beyond 10 μgL^{-1}, whereas, in ~20–60% of the hand pumps, *As* exceeds 50 μgL^{-1} (e.g., Chakraborti et al. 2003, number of hand pumps (n) = 206; Shah 2008, 2010; Kumar et al. 2010, $n = 36$; Saha et al. 2011a, $n = 60$; CGWB and BIT 2013, $n = 6364$; Saha and Shukla 2013, $n = 57$; Sahu and Saha 2015a, $n = 285$). In Sahibganj district of Jharkhand State, however, comparatively lower proportion of hand pumps of ~36% and ~15% are found to exhibit *As* exceeding 10 μgL^{-1} and 50 μgL^{-1}, respectively, in ($n = 3354+$) (Chakraborti et al. 2013; Nayak et al. 2008). In an extensive sampling and analysis from 36 blocks in Bihar, all bordering the Ganga, Chakraborti et al. (2013), however, has reported 33% of the samples exceeding 10 μgL^{-1} ($n = 19{,}961$), while only 18% ($n = 5558$) showed *As* content beyond 50 μgL^{-1}. In the same study, the authors report 44% and 26% of the samples ($n = 4780$) from the western parts of MGP in U.P. with *As* concentration beyond 10 μgL^{-1} and 50 μgL^{-1}, respectively. The density of *As*-contaminated tube wells from Patna-Ballia alluvial stretch is higher compared to that of Buxar-Mirzapur stretch in the western parts of MGP (Saha and Sahu 2016).

A depth vs. *As* concentration cross-plot of samples from MGP ($n = 183$), reveals a rapid decrease in *As* concentration beyond ~40 m bgl. Figure 1.5 depicts the depthwise variation in groundwater *As* gathered from various studies undertaken in MGP, indicating the maximum concentration in the depth range of 15–35 m below ground (CGWB and PHED 2005; Saha et al. 2011a; Saha and Shukla 2013; Sahu and Saha 2015b; Shah 2008, 2010). The groundwater samples collected by various researchers were generally from hand pumps with depth <40 m. A few of them (specifically irrigation wells) reaches up to 75 m bgl. Because of potential sand zones at shallow depth, the depths of wells (including hand pumps) are shallower (10–45 m bgl) in the middle (including Patna-Ballia stretch) and eastern parts of MGP, whereas, at the west, beyond Buxar, the wells often go deeper, up to 75 m bgl. Pandey et al. (2009) reported up to 345 μgL^{-1} in the moderately deep (25–64 m bgl) hand pumps. However, Chauhan et al. (2009) analyzed 9 water samples collected from deeper hand pumps (depth range: 66–75 m bgl) and documented *As* in the safe

Fig. 1.5 Depth vs. groundwater *As* concentration in MGP (CGWB and PHED 2005; Chauhan et al. 2009; Pandey et al. 2009; Sahu and Saha 2015b; Shah 2008, 2010, 2013)



range of 12–20 μgL^{-1}. As shown in Table 1.1, the *As* concentration remained below even the WHO standard of 10 μgL^{-1} in tube wells tapping deep aquifers at depths beyond 50 m bgl (CGWB 2013).

The studies pertaining to the seasonal variation of groundwater *As* in the shallow phreatic aquifer in MGP is very limited. CGWB (2007), however, has reported a decline in *As* concentration during the monsoon months. It is to be noted that monsoon rainfall is the major source of recharge in the Gangetic Plains. Researchers have also reported an improvement in the chemical quality of groundwater in terms of major constituents during the monsoon months (Saha et al. 2008). The timescales over which temporal variation in groundwater *As* may take place vary from hours through seasons to years or decades. Though, in Bangladesh, the studies pertaining to temporal variations of *As* are inconclusive (BGS and DPHE 2001; Tareq et al. 2003), workers from West Bengal in the lower Ganga Plains (Chadha and Ray 1999; Chatterjee et al. 1995) indicate seasonal variations with minima during the post-monsoon period (November). Other than rainfall recharge, various rates of groundwater pumping and climate can be potential factors inducing such changes. Investigation on temporal variations in groundwater *As* in MGP may help in finding mitigation measures.

5 Holocene Floodplain of Newer Alluvium (NA) Hosting Groundwater Arsenic

The Holocene floodplain comprising NA has been found to be contaminated with *As*. The *As* contaminated aquifers are phreatic type and mainly confined within the shallow depth of ~50 m below ground which is predominated by NA (Pandey et al. 2009; Saha et al. 2011a). Thin to moderately thick (~2–15 m)

Table 1.1 Arsenic concentration reported from tube wells tapping deep aquifers in MGP

Sl No	Location	District	Long. (E)	Lat. (N)	Slot position (m bgl)	Max. *As* conc. (μgL^{-1})	Reference
Bihar							
1	Nargada Narayanpur	Bhojpur	84.422	25.607	136–142, 156–162, 179–188, 191–194, 205–208, 214–217, 222–228	*3.0*	CGWB (2013) Saha et al. (2011a) Sahu (2013) Sahu and Saha (2016)
2	Karnamepur	Bhojpur	84.358	25.654	136–142, 147–153, 161–167, 173–179, 187–199	*BDL*	
3	Shahpur	Bhojpur	84.399	25.596	127–133, 139–145,150–156, 169–175, 206–212	*BDL*	
4		Bhojpur	–	–	127–133, 139–145, 169–175, 206–212	*BDL*	
5	Bariswan	Bhojpur	84.436	25.626	170–182, 191–197	*BDL*	
6		Bhojpur	–	–	94–106, 109–115, 128–140, 156–162, 169–181, 187–199	*BDL*	
7	Paharpur	Bhojpur	84.471	25.649	140–146, 185–191, 199–205, 244–250	*BDL*	
8	Bharauli	Bhojpur	84.380	25.631	162–171, 174–180, 183–186, 189–196, 203–215	*BDL*	
9	Amrahi Nawada	Bhojpur	84.553	25.544	124–129, 135–141,166–170, 173–177, 181–187,193–196	*5.0*	
10	Neazipur	Buxar	84.158	25.667	150–156, 164–176, 182–188, 194–200	*BDL*	
11	Brahmpur	Buxar	84.317	25.605	120–132,156-162,156-162,176-182,190–202	*3.5*	
12	Churamanpur	Buxar	84.031	25.581	158–164, 176–182, 208–220	*BDL*	

13	Madudabad	Samastipur	85.728	25.586	90–96,116-128,132-138,222–228	*BDL*	Saha and Shukla (2013)
14		Samastipur	–	–	126–132	*8*	
15	Shahpur Patori	Samastipur	85.560	25.626	80–86, 99–105	*9*	
16	Vidyapatinagar	Samastipur	85.798	25.601	210–222	*BDL*	
17	Gangajal	Saran	85.161	25.710	56–62, 66–72, 88–100, 115–127, 142–154, 165–171	*6.0*	
18	Sitabdiara	Saran	84.641	25.756	105–117, 123–135,175–187, 215–224	*5*	
19	Narepur	Begusarai	85.916	25.595	55–61, 89–95, 122–128	*4.5*	
20		Begusarai	85.902	25.599	122–125	*9.4*	
21	Bihat	Begusarai	86.046	25.432	140–146, 160–166, 175–187	*BDL*	
22	Barauni	Begusarai	85.965	5.4643	154–160, 194–200, 221–223	*BDL*	
23	Barh	Patna	85.692	25.473	122–128, 134–140, 170–176, 179–182, 205–211, 220–226	*BDL*	
24	Gyaspur	Patna	85.469	25.474	92–100, 116–122, 140–146, 150–156, 168–174, 180–188	*BDL*	Saha et al. (2011a)
25	Maner	Patna	84.888	25.642	125–131, 149–155	*BDL*	
Uttar Pradesh							
26	Ramgarh	Ballia	84.399	25.786	196–208, 244–252, 285–289, 322–334	*BDL*	MDWS (2011), Chauhan et al. (2009), Pandey et al. (2009)
27	Dalan Chapra	Ballia	84.518	25.725	186–194, 203–215	*BDL*	

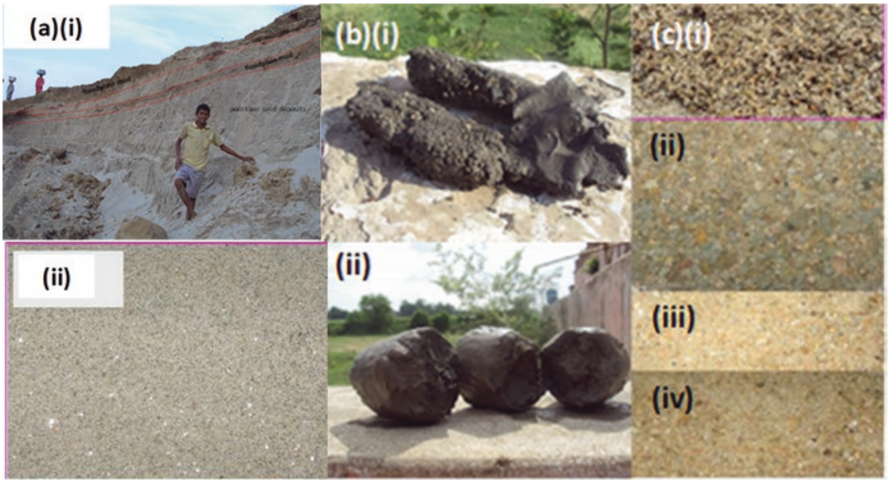


Fig. 1.6 (**a**.i) Dry sand (lying above the water table) of a point bar of Ganga in its floodplain (OFP) exposed in a sand mine pit. (**a**.ii) Fine to very fine micaceous Ganga sand. (**b**.i–ii) Black clay cores recovered from a borehole. They are rich in organic carbon (often impressions/remains of aquatic plants and shelly matter of aquatic organisms are observed) deposited in a lacustrine environment. Sometimes gravels of quartz, feldspar, and kankars (carbonate concretions) are found mixed with the clay, which might have been brought to the channel cut-off lake by floods. (**c**.i–iv) Typical craton sand of brownish yellow colour and fine to coarse in size (Sahu 2013, Sahu and Saha 2015b)

saturated fine sand lenses constitute these aquifers, which are widely tapped locally by dug wells and shallow tube wells for both drinking and irrigation purposes. However, differences of opinions exist on contamination level between the two geomorphic sub-units of NA; T_0 (AFP) and T_1 (OFP) surfaces. Few researchers advocated that T_0 surface or the AFP is more contaminated (Acharyya 2005; Sahu and Saha 2015b). In Bhojpur district, however, Sahu and Saha (2015b) have observed higher contamination in T_1 than T_0 surface. In this part, 32% (n = 34) samples from T_0 and 45% (n = 249) samples from T_1 surface exceeded the 50 μgL^{-1}. The Himalaya has been indicated as the source of *As* rich sediment in NA (Acharyya 2005; Mukherjee et al. 2007; Sahu and Saha 2015a, b). The sediment comprises fine to very fine micaceous gray to dark gray sand and mud/clay, which is in an un-oxidized state (Mukherjee et al. 2007; Sahu 2013) (Fig. 1.6a, b). In contrast, the OA sediment, derived from peninsular highlands is oxidized, yellow to brownish-yellow in colour, coarse-grained and mixed with carbonate concretions, widely known as *kankars* (Saha et al. 2014; Sahu et al. 2015) (Fig. 1.6c.i–iv). In a similar trend as reported from BDP (Ravenscroft et al. 2005), the OA in MGP also shows lower *As* concentrations in groundwater (Pandey et al. 2009; Saha et al. 2011a; Shah 2008).

5.1 Sedimentary Facies and Sequence Models in Newer Alluvium

Sahu and Saha (2015a) have identified ten sedimentary facies (Table 1.2) types in the NA in the floodplain in the depth range of 0–30 m below ground. These facies include: (i & ii (Fig. 1.6)) channel lag deposits (*l*1*, l*2 (Table 1.2))- comprising reworked carbonate nodules and sand mostly in 1000–3000 μm size range, (iii & iv) a well-sorted fine sand (*f*1*, f*2)- where ~75–85% of the particles are in 130–240 μm size containing proportions of mica flakes (Fig. 1.6a), (v) a moderately well-sorted very fine sand with *(f*3) in which 70–85% of the grains fall in 92–110 μm size range with varying proportions of silt, (vi) a clean, fine to coarse silt with a proportion of very fine sand (*f*4); 69–82% of the particles remain in the size range of 22–45 μm (vii & viii) mud with varying proportions of silt and clay (*m*1*, m*2), and (ix & x) black clay/mud and organic matter with varying minor silt (*c*1*, c*2) (Fig. 1.6b.i-ii).

Various patterns of spatial distribution of the above sedimentary facies in different geomorphic environments in the floodplain have given rise to eight sedimentary sequences (*Sed. Seq.*) models (Fig. 1.7) in MGP (Sahu 2013; Sahu and Saha 2015b). The authors have studied the level of groundwater *As* in each model and also the vulnerability of the models to contamination. About 94% of the samples ($n = 85$) from ridges on the point bars exhibited concentration less than 50 μgL^{-1} (*Sed. seq. S1*). The wells located on palaeo-levee were also less contaminated, where 87% of the samples ($n = 71$) remained <50 μgL^{-1} (*Sed. seq. S2 and S8*). On the other hand, the wells located either on or near the filled-cut off channels (generally with fine-grained material) were found to be significantly contaminated, with 51% of them ($n = 73$) exceeding 50 μgL^{-1} and 30% even beyond 100 μgL^{-1} (*Sed. seq. S3, S4, S6, and S7*). Those also include the abandoned crossbar channel areas forming swales on point bars.

The most critically contaminated villages are located on earth-filled palaeochannels (may be the locations of channel bar islands), which are raised by 2–3 m above the local flood level (*Sed. seq. S7*). A typical example of such villages is Semaria-Ojhapatti, where 80% of the wells with *As* concentration greater than 50 μgL^{-1} (Sahu and Saha 2015b). Due to the non-availability of data, groundwater *As* concentrations related to *Sed. seq. S5* could not be produced here. The relation of the *As* concentration with different geomorphic features and the shallow alluvial stratigraphy has been depicted schematically in Fig. 1.8. Most of the hand pumps (range: 87–94%) in the scroll bar ridges, point bar platforms and palaeo-levees, which cover ~70% of the area under meander scars, recorded lower *As* concentrations (<50 μgL^{-1}). Exceptions do occur in the palaeo-levee at the channel cut-offs forming active lakes, where *As* concentration beyond 50 μgL^{-1} are also obtained. The channel-lakes of Recent meander cut-offs are areas of active biomass accumulation (Fig. 1.8).

The research findings of Sahu and Saha (2015a, b) indicated that groundwater *As* contamination has got a specific relation with the Holocene geomorphic sub-features and the shallow sedimentary sequences in the area. The contaminated areas coin-

Table 1.2 Sedimentary facies types identified in MGP (Sahu 2013; Sahu and Saha 2015b)

Facies	Lithology	Colour	Sand: silt: clay (% ratio)	Average mean grain size (μm)	Organic Carbon content in g/kg	Interpreted depositional setting
*l*1	Coarse to gravel size reworked carbonate nodules	Gray	–	–	–	Channel lag deposits of Ganga/channel deposit of Sone River
*l*2	Medium to coarse sand with carbonate nodules and infrequent iron flakes and nodules.	Light yellow to yellowish brown	100:0:0	595	–	
*f*1	Fine sand with mica flakes. Basically transitory sand bodies on channels.	Gray	93–98:7–2:0	210	–	Channel sand as bar deposits (braid bar and lower reaches of point bars) and other transitory sand on the channel bed
*f*2	Fine sand with a dominating proportion (~ 70–80%) of mica flakes.	Gray, shining dark gray (when biotite predominates)		107	0.1–0.8	Channel sand as bar deposits
*f*3	Very fine sand with a proportion of silt/ mud.	Light gray to Brown	92:6:2	91	0.1–0.6	Low energy channel deposits (crossbar channels)/over bank deposits (upper reaches of point bars)
*f*4	Silt/mud with a good proportion of very fine sand.	Brown	14:69:14	52	0.6–1.2	
*m*1	Silt and clay. Basically flood plain mud deposits, with higher content of silt.	Brown to light yellow	5:60:35	31	0.8–1.6	Over bank flood plain deposits
*m*2	Silt/ clay. Basically flood plain mud deposits, with higher content of transgenic clay.	Light yellow to gray	5:25:70	15	3.2–6.1	
*c*1	Clay-rich with organic matter, with minor silt.	Light black (locally termed as *kalli mitti*).	3:25:72	12	5.9–6.6	Lacustrine deposits
*c*2	Organic black clay with minor or no silt.	Grayish black to black	1:10:89	4		

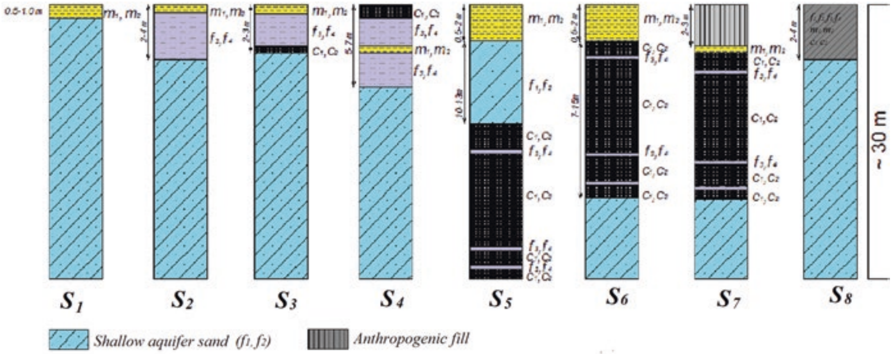


Fig. 1.7 Eight sedimentary sequences (*S1-S8)* identified in the area within 30 m below ground. *f*1-*f*4, *m*1-*m*2, and *c*1-*c*2 represent the sedimentary facies (Sahu 2013; Sahu and Saha 2015a, b)

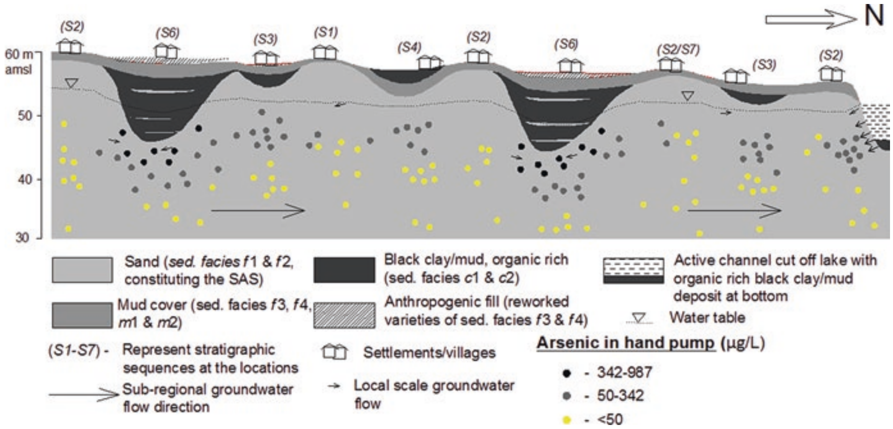


Fig. 1.8 A schematic section in the Ganga floodplain (south to north) showing generalized topography, floodplain morphology and shallow alluvial stratigraphy. It depicts the concentration of *As* in hand pumps (within ~30 m bgl) and their distribution in the settlement areas distributed in different geomorphic environments in the floodplain. High *As* (50–342 μg/L) has got an association with the hand pumps (depth: ~10–30 m bgl) in the villages settled on anthropogenic fills over the channel fill sequence. The sandier areas in the scroll bar ridges, levees (particularly in OFP) and proximal floodplain sequences often exhibit lower concentration of *As* (less than 50 μg/L). It is noteworthy that these environments together make ~70% of the area of a typical meander scar of the Ganga (Sahu and Saha 2015a, b)

cide with the palaeochannel cut-offs of the Ganga and the narrower belt on the inner sides of the cut-offs, which were earlier occupied by cross-bar channels of various dimensions. The organic carbon (OC) rich black clay/mud deposits reveal variable thickness from <1 m in cross-bar channels to 10–15 m in palaeochannel cut-offs as clay-plugs. The OC rich soil in the paddy fields in the floodplain might also be helping in the leaching OC to the water table (Sahu and Saha 2015a, b). The presence of organic material increases the structural stability of soil and its resistance to rainfall impact, which otherwise clogs the pores by particle breaking. The retaining of pores in the soil enhances the infiltration rate through it (Roose and Barthes 2001). Analyses show 8.5–12.2 Tritium Units in groundwater in the shallow aquifer, suggesting recent recharge (<40 years) (Saha et al. 2011b). A significant part of the recharge is possibly contributed from impounded surface water bodies like channel cut-off lakes and other floodplain depressions (buried cut-offs) as indicated by δ^{18}O distribution (Saha et al. 2011b). The surface water while percolating downward collects the OC through dissolution from the clay bodies. The OC might also be coming to the groundwater continuously at the interface between the aquifer and clay bodies in the palaeochannels. On the banks of the active cut-off lakes, the modified hydraulic gradient owing to groundwater withdrawal in the settlements brings OC of lake water and underlying sediment to groundwater, which subsequently percolates downward and moves village-ward along with the groundwater.

5.2 Release of Arsenic in Groundwater in MGP

A clay-plug in the palaeochannel of Ganga measures 12–15 km in its arc-length with an average width of ~1.20 km. On average, the clay continues up to ~13 m depth (Ave. volume: 0.21 km^3). The bottom surface of the lower part of this huge clay plug, which remains below the local water table, continuously remain in contact with the aquifer and releases OC to the groundwater. The OC is consumed by the microbes present in the aquifer for their survival, which in turn consumes the available oxygen in groundwater. This produces a reductive environment and causes dissolution of hydrated iron oxide (HFO), releasing both the *As* and iron into groundwater. The reductive dissolution of HFO in *As* affected areas of MGP is also evident by a high concentration of dissolved iron in groundwater (Shah 2008, 2013). Reductive dissolution of adsorbed *As* from HFO has been reported in Bangladesh (Nickson et al. 1998; BGS and DPHE 2001). Arsenic is inherently present in the aquifer sediment, and Kumar et al. (2012) have detected ~15–40.81 mg/kg of total *As* content in sediments in the Semaria Ojhapatti village within 32 m depth. The OC only creates a suitable environment for *As* release to groundwater. In general, pyrite or any other *As* rich mineral that can release *As* to groundwater is not present in the sediment in MGP (Shah 2008).

The spread of the reducing environment from the OC bearing clay-plug depends upon the hydraulic conductivity, groundwater flow direction, groundwater development and the volume of fresh oxic water recharge that reaches the spreading anoxic front (Sahu and Saha 2015a, b). The sandier areas along the scroll bar ridges and levees yield a low concentration of *As* as the oxic groundwater in such areas favours stability of hydrated iron oxide. Good hydraulic conductivity in the aquifer also helps in flushing out whatever contamination is there. There may be a source-distance relation between the *As* concentration and the OC rich clay plugs in the sedimentary sequence. Ideally, it can be said that in case of a clay-plug in an aerially extensive sand body, the concentration of groundwater *As* may decrease with an increase in distance from the plug. In the downgradient of groundwater flow, away from the plug, the anoxic front gets weaker due to its confrontation with more oxic groundwater, and thus, the groundwater *As* concentration may decrease (Saha et al. 2009; Sahu and Saha 2015a, b).

5.3 Spatial Variability of Groundwater Arsenic in MGP-A Geomorphic Model

The OA upland sediment in the MGP is low (<10 µgL^{-1}) in groundwater *As* concentrations (CGWB and PHED 2005; Shah 2008). The OA in MGP form the deeper aquifer system, as well as the lower parts of the shallow aquifer system, are also low in *As* (Saha et al. 2011a). Groundwater *As* contamination derived mostly from the Himalayan sources is confined within the NA sediment in the floodplains of Ganga. However, there exists a spatial variability in the incidence of groundwater *As* within the affected areas itself. Considering the regulating limit of 50 µgL^{-1}, Sahu and Saha (2015b) identified that only 34% of the tested sources as *As* contaminated, which could reach up to ~78% if the WHO (1996) standard of 10 µgL^{-1} is considered. The frequency of affected hand pumps is more in OFP than in AFP.

The schematic model depicted in Fig. 1.9 shows the role of organic-rich argillaceous sediment bodies in groundwater *As* concentration in various morphostratigraphic units in the entire floodplain. The hand pumps of different depths within the contaminated zone of the shallow aquifer itself yield different

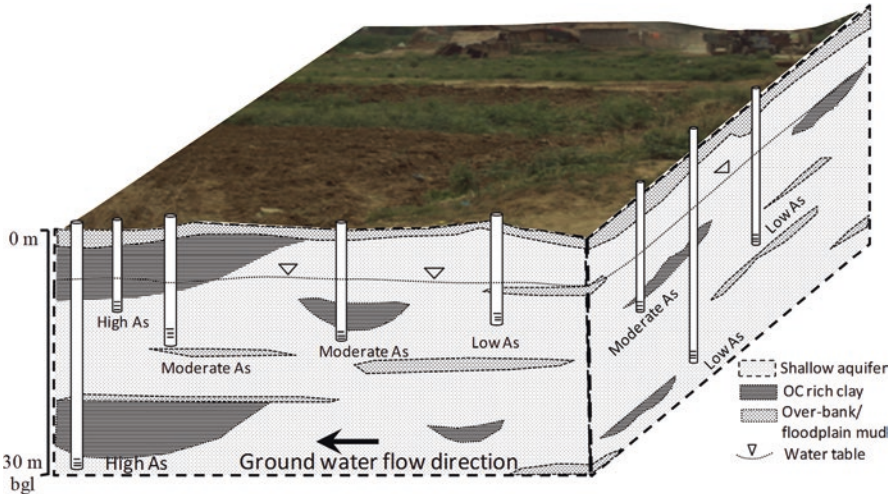


Fig. 1.9 A schematic model of the distribution of clay-plugs of various dimensions in the floodplains and associated variability in the concentration level of groundwater *As*

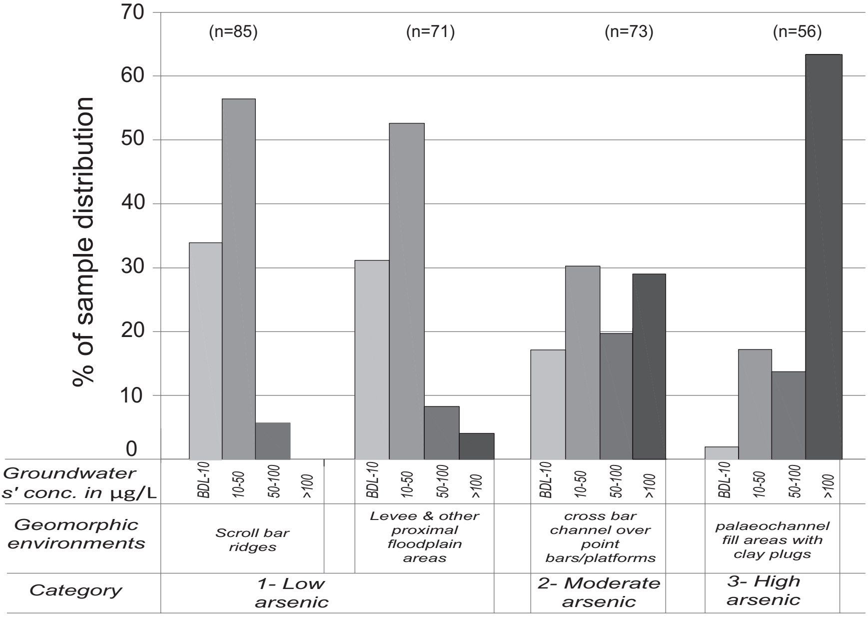


Fig. 1.10 Relations between the frequency of water samples and abundance of *As* in different geomorphic environments in the floodplain with different sub-surface stratigraphy. 'n' shows the number of groundwater samples tested (Sahu and Saha 2015b)

concentration levels. The possibility of getting low *As* in hand pumps of shallower depth (10–20 m bgl) is more owing to their location in sandier areas of scroll bar ridges, levees, and point bar platforms, whereas the chances of high *As* is more in moderately deep hand pumps (30–40 m bgl) in the clayey areas (clay-plugged palaeochannels). Sahu and Saha (2015b) have also observed similar pattern of *As* contamination in hand pumps in different geomorphic environments in the floodplain of NA in MGP (Fig. 1.10).

The sedimentary units in NA were deposited under fluvio-lacustrine environments. The MGP lies sufficiently upstream of the coastline and has been argued to remain out of the reach of marine influence (Tandon et al. 2008). As such the *As* contamination levels correlated with the low groundwater flushing from the Newer Alluvial deposits during the high-stand setting of sea level during Holocene, as suggested earlier (Shah 2008), seem to bear a little scope. The channel entrenchment and its migration in MGP were driven by the forcing factors like climate and tectonics (Sahu et al. 2010; Sahu and Saha 2014). The nature of the low-lying fluvial and fluvio-lacustrine features gave rise to various depositional environments, which evolved into various sedimentary sequences in the area. The thickness of the mud overlying the shallow aquifer and the dimension of the organic-rich argillaceous deposits control the level of *As* concentration in groundwater. The spatial distribution of such argillaceous bodies is reflected in the spatial variation in the level of groundwater *As*. A spatial lithology dominated by clay, no doubt, obstructs the groundwater flow, thereby increasing the resident time of water, which reacts with clay to release more OC and in turn increasing the *As* content in groundwater. Of course, the groundwater recharge, discharge, and flow regime play significant roles as modifying factors in this model. Groundwater development augments the concentration of *As* by modifying the local groundwater flow regime and activating the release of enhanced dissolved OC and biomass to groundwater.

6 Aquifer Systems in the Middle Ganga Plain

The floodplain sediment, comprising NA in the top part of the stratigraphic column (Fig. 1.11), largely acts as an aquitard (Saha et al. 2014; Sahu et al. 2018). With a mud cover at the surface, it is inter-layered by fine sand lenses of various dimensions. The sediment in the majority of the floodplain is purely Himalayan derived, deposited by the Ganga and other Himalayan rivers. The bottom part (~2–5 m) of the aquitard displays a mixed zone with alternate layers of the craton and Himalayan sediment (Fig. 1.11). The transitory zone is followed downward by thicker columns of coarser brownish yellow sediment of purely craton origin. The granular zones therein form moderate to high potential aquifer systems that spread regionally in MGP. The clay intercalations are often thin and less in number that gives rise to higher sand to clay ratio varying between ~70–95% in the depth range of ~50–200 m below ground in the axial part of the MGP where there is more incidence of groundwater *As* contamination. The craton sediment comprises two principal assemblages of litho-types: (1) assemblage-1, dominated by coarse to very coarse sand and gravels (often highly loaded with *kankars*) of craton origin, with interlayers of fine sand and minor scale clay lenses at places, form moderate to high potential aquifer (30–60 m depth range), and (2) assemblage-2, consisting of mainly medium to very coarse sand (and gravels) and at places with lenses of fine to medium sand of craton origin.

Studies indicate that a few clay zones appear at different depth ranges, though in major cases they remain local in extent. However, the clayey zone forming an aquitard within the depth range of 87 and 139 m bgl is widely pervasive in a larger part of MGP (Fig. 1.12). The thickness of the aquitard hereafter referred as ‘middle clay,’ varies within 8–24 m. It broadly divides the vertical sand sequence into two aquifer systems, particularly in the central and eastern parts of MGP. The first aquifer system ranges from the bottom of the top aquitard down to the depth of ~87–126 m bgl. The upper half of the first aquifer system is predominated by the sediment assemblage-1. A mixed zone of craton and Himalayan sediment follows at the top of the assemblage. In this transition zone, it appears that there are several clay zones of the centimeter scale in thickness. The mixed zone is followed upward by the pure fine sand of Himalayan origin which hosts groundwater *As* at several locations. The assemblage-2 at the bottom half forms the most potential part of the aquifer system in the entire area.

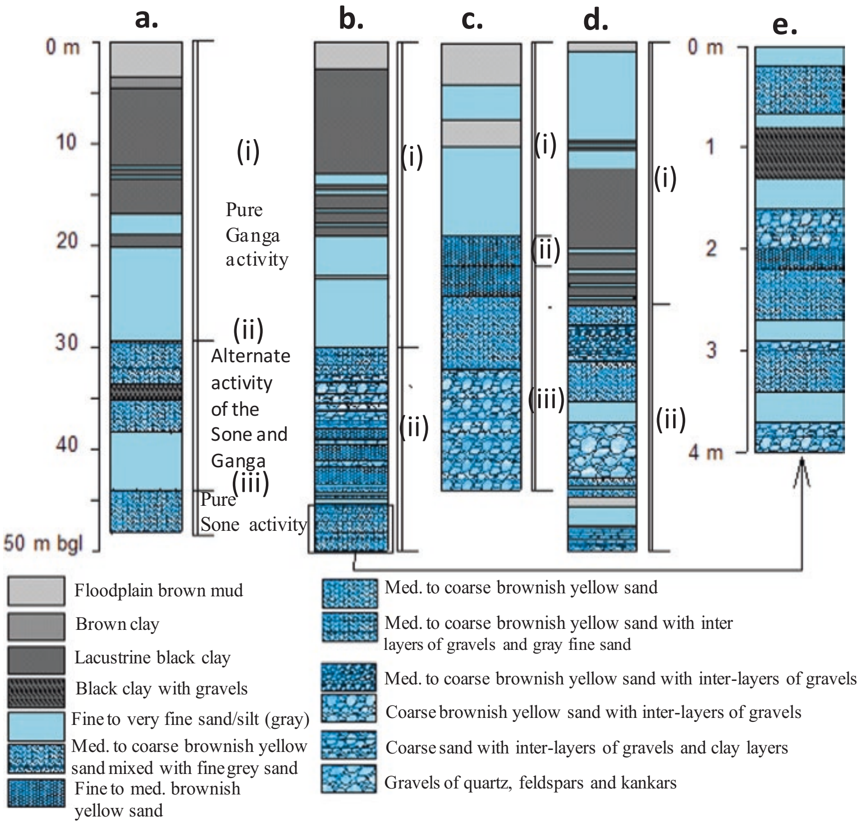


Fig. 1.11 Few representative shallow lithologs from different geomorphic environments in the *As*-contaminated floodplain of Ganga in Bihar in MGP (source: CGWB 2013). (**a**, **b**) Channel fill sequence, (**c**) Point bar platform sequence with cross bar channel activity. (**d**) Point bar sequence in the scroll bar ridge area with buried channel fill clay plug. (**e**) depicting the detail lithology in the lower part of 'b' litholog. (i) Represents the upper parts of the boreholes, dominated by Ganga sediment. (ii) Represents alternate sequence of coarser Sone sand and the finer Ganga sand. (iii) Represents coarser Sone sediment that prevails beyond the alternate sequence of the Sone and Ganga sand

Most of the tube wells and some hand pumps in the area, however, remain confined within the NA at the top and the assemblages-1 in the first aquifer system.

The second aquifer system starts at various depths ranging from ~116 to 139 m bgl and continues up to the depth of ~250 m below ground (Fig. 1.12). Lying below the middle clay, it is laterally extensive and continues southward in plain forming the deeper aquifer system in OA. Groundwater occurs under semi-confined to the confined condition. In comparison to the first aquifer system, its clay content is significantly low. At the eastern and central parts of the area, bottom clay is noticed at the base of the second aquifer system. The aquifer system is broadly uniform in the area, except the variation in the size of the sand at vari-

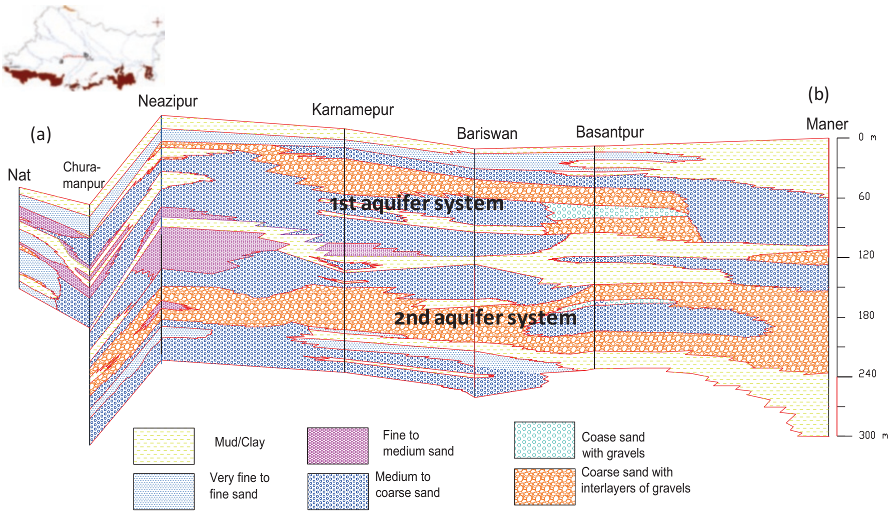


Fig. 1.12 A hydrogeological transect in the *As* affected area in MGP in the western parts of Bihar. The map of MGP (inset) shows the location of the section

ous depths. The thickness of the aquifer system increases westward because of the absence of the bottom clay.

At certain locations in the eastern parts of MGP (e.g., around Patna Urban Area), the first and second aquifer systems merge and seem to behave as a single aquifer system. However, the aquitard at the top of the sequence at surface remains quite thick (range: ~18–80 m) which gives a confined character to the aquifer system. In the northern parts of MGP, along the left bank of the Ganga, finer gray sand appears at deeper levels and also in the stratigraphic columns. However, the black clay (rich in organic matter) as found in NA at the top levels is generally absent at those deeper levels. Two similar aquifers systems are also observed in sufficiently large areas in the northern parts owing to the existence of middle clay layers. In certain parts of MGP, a third aquifer system is also noticed beyond the base of the second aquifer system. It probably exists within ~250–300 m below ground.

7 Depth to Water Levels in the Shallow Phreatic Aquifer

The depths to water levels (DTWs) in the phreatic aquifer in MGP reflect strong relation with the monsoonal rainfall. The deepest and shallowest water levels are observed at some point in time during the months of April–June and Aug-Sept. Respectively, depending on the onset and retreat of monsoonal rainfall. In parts of MGP, about 65% of the total annual groundwater recharges being contributed from monsoon rainfall. In the year 2015, the contaminated parts in

Bihar showed the pre-monsoon DTWs (CGWB 2016) varying between 1.27 m and 12.00 m bgl, whereas, during the post-monsoon period the DTWs remained in the range of 0.58–10.10 m bgl. The fluctuations in the water levels during the pre- and post-monsoon period of the year varied between −2.02 m and 8.47 m. Some of the wells recorded a fall in water level during the post-monsoon period owing to a deficit (~30%) in rainfall in the year. The annual mean water levels for the year (May 2015-Jan 2016) varied between 1.0 and 11.50 m bgl. The groundwater flows north-easterly to easterly coinciding with the direction of flow of the Ganga River. The Ganga remains a gaining river, receiving water from the aquifer/aquitard. The hydraulic gradient in NA (1:1700–1:5750) indicated sluggish groundwater movement towards the Ganga (Saha et al. 2009; Saha and Shukla 2013). The historical water level data (1985–2007) of four wells located in the contaminated areas of Bihar state in MGP exhibited stable or only marginal decline (<0.5 myr-1) in water levels (Saha et al. 2009; Saha and Shukla 2013).

8 Arsenic Safe Deeper Pleistocene (OA) Aquifers

The studies in MGP (Acharyya 2005; CGWB and PHED 2005; Saha et al. 2011b; Shah 2008; Sahu and Shukla 2010) as well as in BDP (Acharyya et al. 2000; Ravenscroft et al. 2005) indicate that the groundwater *As* concentrations in the Pleistocene aquifers in OA remain low. Within the permissible limits of drinking (10 μgL^{-1}). The thick aquifer systems underlying the thin veneer of the Holocene deposits comprise fine to coarse sand and gravel of brownish yellow colour which were deposited in an oxidizing environment (Saha et al. 2011a; Sahu 2013). The Pleistocene sediment that underlies the major part of the affected area in MGP is craton derived. Invariably containing calcium carbonate concretion (*kankars*) of various size, the sediment was laid down by the braided Sone River in its megafan over a large area (Sahu et al. 2015).

9 How Safe the Pleistocene Aquifers Are?

Along with the top aquitard, the top 15–20 m slice of the first aquifer system is reported to host elevated levels of groundwater *As*. Though locally the bottom part of the first aquifer system is separated from the contaminated upper part by clayey layers, in most areas, it remains vulnerable to the downward spreading of groundwater *As* (Fig. 1.13). Increase in agitations in aquifer owing to groundwater development may help in the downward spreading of the contamination front. However, the second aquifer system in major part of the affected area possesses a confining layer (the middle clay) at its top. The overlying first aquifer system and the middle

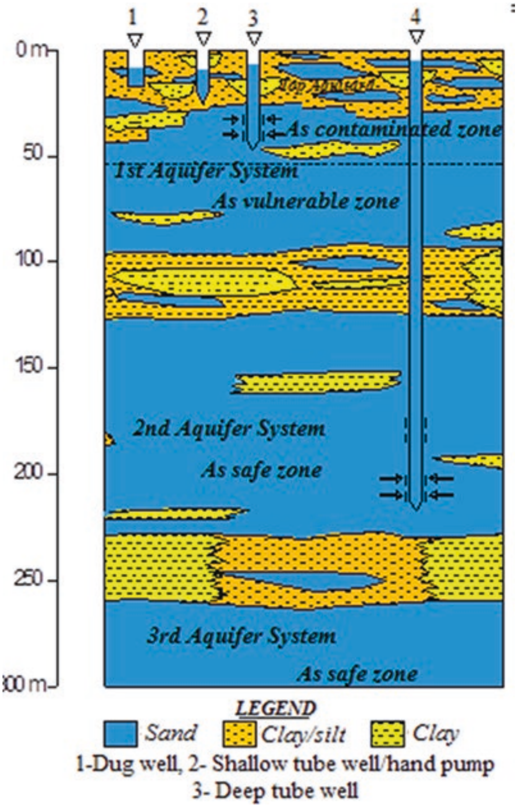


Fig. 1.13 Schematic diagram showing aquifer-aquitard configuration and distribution of *As* in the multi-layered alluvial aquifer in the *As* affected area in MGP (Saha et al. 2009)

clay act as the buffer zone for the second aquifer system. It will remain safe as long as the first aquifer system remains safe. Leaching of *As* water through the middle clay, which may act as an aquitard, may only contaminate the deeper (2nd) aquifer system. It may happen only when the piezometric head of the second aquifer system drops below that of the first aquifer system. The piezometric levels belonging to different times collected from the tube wells (CGWB 2013) tapping the second aquifer system varies between 0.8 and 12.98 m bgl (with a mean of 6.8 m bgl). Around Patna urban area in the eastern parts of MGP, the pre-monsoon water levels in the aquifer system were observed in the range of 4.1–12.7 m bgl, whereas those during the post-monsoon period varied between 1.49 and 10.08 m bgl (CGWB 2015). Resting of the water levels much above the bottom of their confining layer of the aquifer system at its top indicates the piezometric nature of the hydraulic heads and the confined condition of the aquifer systems. It is observed that the piezometric head in the second aquifer system rests above that of the first aquifer system (Saha et al. 2011a; Sahu 2013), which may get reversed owing to increase in groundwater draft from the former one as found around Patna urban area (CGWB 2015).

9.1 Yield Potential and Drawdown in the Second Aquifer System

The existing tube wells in MGP, which have been constructed tapping the second aquifer system (CGWB 2013), record discharges in the range of ~46–535 m^3/h with the majority values ranging between ~100 and 300 m^3/h. The maximum observed drawdown values varied between 1.1 and 10.8 m. It indicates that the aquifer system, comprising Pleistocene sediment of craton origin, is highly productive. The aquifer system with coarser granular material was laid down by the cratonic major Sone River in its megafan. Lower drawdown values in spite of higher discharges (Fig. 1.14) indicated a high coefficient of storage and transmissivity of the aquifer system (Driscoll 1986; Kruseman et al. 2000). Similarly, the binary plot between the drawdown and transmissivity (m^3/d) of the aquifer system shows lower drawdown values associated with higher values of transmissivity (Fig. 1.14).

9.2 Pumping Tests to Ascertain the As *the Safety of the Second Aquifer System*

In the present section, it has been attempted to assess hydraulic parameters of transmissivity (*T*), storativity (*S*) and hydraulic conductivity (*K*) of the second aquifer system and the vertical conductivity (*K*′) in the middle clay overlying it. The pumping test data of 7 observation wells and 1 piezometer have been re-analyzed using the Walton (1962) curve fitting method considering the aquifer as semi-confined

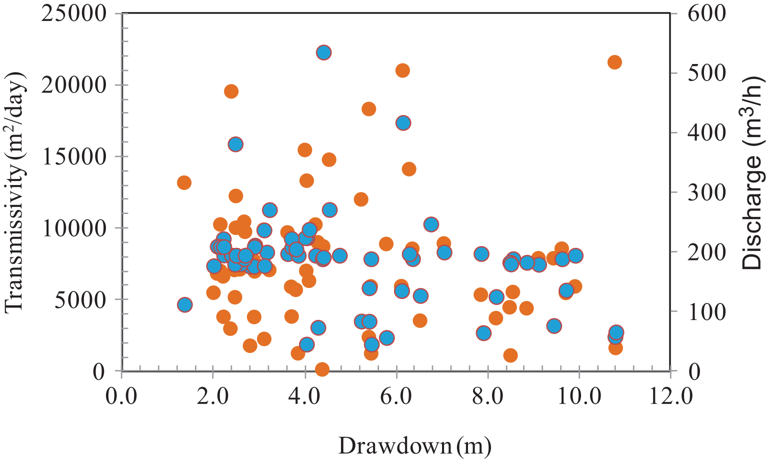


Fig. 1.14 Drawdown vs. discharge and aquifer transmissivity plot for the tube wells tapping second aquifer system in MGP

Table 1.3 Tube well construction depth and various attributes of pumping tests in tube wells in the *As* affected parts of MGP. The 'r' denotes the distance between the PW and the OW/PZ

Sl. No	Location	Distance ('r' in m)		Zones tapped (range in m)	Static water level (m bgl)			pumping duration (min)	Pumping Discharge (m^3/h)	Drawdown (m)		
		OW	PZ		PW	OW	PZ			PW	OW	PZ
1	Kancha	5	–	98–128	–	3.6	–	985	74	–	0.3	–
2	Vidyapati nagar	5.45	–	210–222	2.8	3.1	–	1000	57	5.8	1.3	–
3	Brahmpur	–	4.5	120–202	3.0	–	3.8	500	200	11.0	–	0.7
4	Konharaghat	5	–	86–248	13.0	12.1	–	360	176	2.8	1.2	–
5	Bariswan	19.56	–	94–199	4.6	4.6	–	480	189	6.4	0.3	–
6	Karnamepur	6	–	136–199	6.6	6.6	–	700	195	9.9	0.8	–
7	Nargada Narayanpur	6.9	–	136–228	4.2	4.6	–	760	189	9.5	0.5	–
8	Bharauli	12.31	–	162–215	4.4	3.7	–	800	183	8.5	0.4	–

with the assumptions of non-steady flow of water in the aquifer and insignificant storage in the overlying aquitard. The pumping wells (PWs), as well as the observation wells (OWs) and piezometers (PZs), tap the granular zones in the second aquifer system (Table 1.3). The effect of pumping on the top aquitard and the first aquifer system were studied by measuring water levels from shallow tube wells available at Bharauli (zones tapped 27.5–30.5 m) and Karnamepur (zones tapped 16.0–18.0 m) within a radius of 7.0 m from the pumping wells.

The duration of pumping were kept between 360 and 1000 min (Table 1.3). Pre-pumping water levels in observation wells varied between 3.1 and 12.1 m bgl. Pumping discharges varied considerably between 57 and 200 m^3 per hour. While the drawdown in the PWs remained within 2.8–11.0 m, those in the OWs/PZ varied in the range of 0.3–1.3 m. The water levels in shallow piezometers at Bharauli and Karnamepur were not affected until the end of pumping.

Analyses of the log-normal plot of time-drawdown data at Bharauli reveal minor increment in drawdown after 500 min of pumping (Fig. 1.15a), indicating a semi-confined nature of the aquifer. Similarly, incremental drawdowns almost cease after 300–400 min of pumping at Bariswan, Karnamepur, Kancha, and Nargada Narayanpur. At Konharaghat and Vidyapati Nagar, further drawdowns are observed even after 300 min of pumping. In the latter case, the drawdown ceases after only 600 min. During pumping, the flow in the aquifer is considered to be horizontal but the flow induced in the aquitard remains vertical (Kruseman et al. 2000). The error introduced by this assumption is less than 5% if the K of the aquifer is two or more orders of magnitude greater than that of the aquitard (Neuman and Witherspoon 1969).

The Walton match points in the log-log plot (Fig. 1.15b) of the time-drawdown data are produced in Table 1.4. The *T* values determined by the method range the

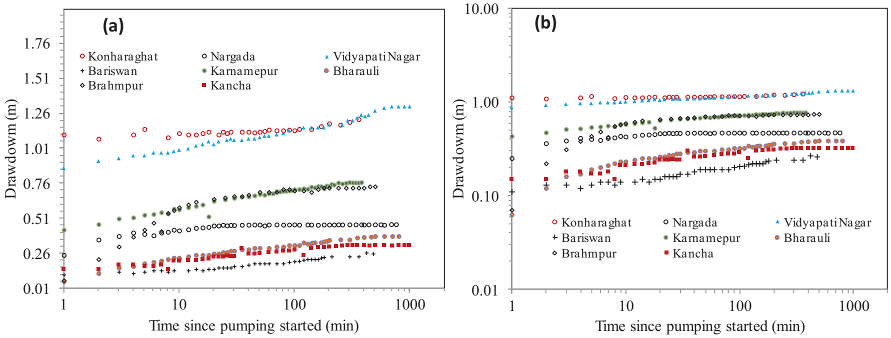


Fig. 1.15 (**a**) Log-normal and, (**b**) the log-log plots of the time-drawdown data of the pumping tests in the *As* affected areas in MGP

Table 1.4 The parameters in the Walton method and the estimated transmissivity (T) and storativity (S) values for the second aquifer system. The last two columns show the saturated thickness of aquifer and the estimated average K values

		Walton (1962) match points[a]							
Sl. No	Location	W(u, r/L)	1/u	t	s	'T' (m^2/d)	S	'D' (m)	Ave. 'K' (m/d)
1	Kancha	6.00	40,000	400	0.20	4243	4.7×10^{-3}	30	141
2	Vidyapati nagar	6.00	10,000	30	0.60	1085	3.0×10^{-4}	12	90
3	Brahmpur[b]	3.55	9500	40	0.30	4522	2.6×10^{-3}	82	55
4	Konharaghat	4.00	15,000	50	0.40	3367	1.25×10^{-3}	107	31
5	Bariswan	3.00	1000	32	0.11	9834	2.28×10^{-3}	97	101
6	Karnamepur	4.00	34,298	89	0.30	6197	9.4×10^{-4}	63	98
7	Nargada Narayanpur	4.00	10,000	65	0.23	6271	2.38×10^{-3}	92	68
8	Bharauli	4.00	6159	49	0.20	6986	6.4×10^{-4}	53	132

[a] Walton (1962) match point: An arbitrary point below the Walton type curve, matched in maximum part by the time-drawdown observed data curve. *W(u, r/L)* and *1/u* are the coordinates of the match point on the Walton map, while *t* and *s* are the coordinates of the match point on the time-drawdown log-log plot of pumping test data (Walton 1962)

[b] Tube well is a piezometer, while all others are observation wells

minimum of 1085 m^2/d to the maximum of 9834 m^2/d at Bariswan (Table 1.4) indicating moderate to good potentiality of the second aquifer system. The storage coefficient values are estimated below 0.005 (range: $4.7 \times 10^{-3} - 3.0 \times 10^{-4}$), indicating the confined condition of the aquifer (Kruseman et al. 2000).

The *K* values for the second aquifer system were determined from the pumping test data based on the following relation:

$$K = \frac{T}{D} = \frac{T}{d - d'}$$

Where,

D = Saturated thickness of the aquifer system.
d = Sedimentary thickness (in m) between the lower level of the deepest slot and the upper level of the shallowest slot in the tube well.
d' = Cumulative thickness of clay and sandy clay in between in m.

The saturated thickness of the aquifer system at the pumping test locations vary between 30 and 107 m. The estimated *K* values fall in the range of 31–141 m/d, indicating good potential of the aquifer system (Table 1.4). The vertical hydraulic conductivity (K') of the middle clay (the aquitard) confining the second aquifer system has been worked out from the Walton method, based on the following relation;

$$c = \frac{L^2}{T} = \frac{D'}{K'}$$

Where,

L = leakage factor in m.
c = Hydraulic resistance of the aquitard in day
D' = saturated thickness of the aquitard (confining bed) in m.
K' = hydraulic conductivity of the aquitard for the vertical flow in m/day.

The values of *L* are derived from *r/L* values of Walton Type Curve with which the time-drawdown curve has been matched during interpretation. The D' varies from 10 m at Vidyapati Nagar to 32 m at Kancha (Table 1.5). The values of *L* range between the minimum of 450 m at Brahmpur to the maximum of 4103 m at Bharauli. Large values of *L* indicate higher hydraulic resistance in the aquitard, and consequently smaller leakage rate through the aquitard whereas-small values of *L* mean a high leakage. The K' values of the clay have been estimated in the range of 0.01 at Bharauli and Bariswan to 0.33 m/d at Brahmpur. The low conductivity values show the impermeable character of its predominantly sandy clay to the clayey character (Bouwer and Bouwer 1978).

Table 1.5 Thickness of the middle clay between the first and second aquifer systems at pumping test locations and the estimated leakage factors (L), hydraulic resistances (c) and the vertical conductivity (K′) values

Sl. No	Location	Thickness of mid- clay (*D'*) (m)	*r/L*	Leakage factor ‘*L*’ (m)	Ave. *T* (m^2/d)	Hydraulic resistance ‘*c*’ (L^2/T)	*K′* (m/d)
1	Kancha	32	0.005	1000	4243	235.7	0.14
2	Vidyapati nagar	10	0.01	545	1085	273.8	0.04
3	Brahmpur	15	0.01	450	4522	44.8	0.33
4	Konharaghat	29	0.005	1000	3367	297.0	0.10
5	Bariswan	15	0.005	3912	9834	1556.2	0.010
6	Karnamepur	18	0.003	2000	6197	645.5	0.028
7	Nargada Narayanpur	22	0.005	1380	6271	303.7	0.072
8	Bharauli	25	0.003	4103	6986	2410.2	0.010

9.3 Groundwater Potential of Second Aquifer System Based on Sediment Grain Size Analysis

The horizontal *K* values of granular layers in the second aquifer system at five locations, namelyKarnamepur, Bharauli, Nargada Narayanpur, Paharpur and Simri (see Fig. 1.4 for locations), have been estimated using empirical relation based on the sediment grain size parameters of every layer (CGWB 2015; Sahu 2013; Sahu and Saha 2016; Sahu et al. 2018). The average effective conductivity values (K_e) of multilayered aquifer systems have been worked out based on the following relation:

$$K_e = \frac{1}{D}\left(K_1 d_1 + K_2 d_2 + K_3 d_3 \ldots\right)$$

Where,

K_e = average effective horizontal *K* of the aquifer system.
D = total saturated thickness of the aquifer system (m)
$= d_1 + d_2 + d_3 \ldots\ldots$
d_1, d_2, d_3 = thickness of each layer (m)
$K_1, K_2, K_3 \ldots.$ = horizontal hydraulic conductivity (m/d) of each layer

The K_e value coupled with the cumulative thickness of the granular zones has been used to estimate the T_e (=K_eD) approximations of the aquifer system, which can be expressed by the following relation:

$$T_e = T_1 + T_2 + T_3 \ldots$$

The grain size based empirical equation of Breyer (1964) has been used to estimate the hydraulic conductivity of the aquifer systems (Breyer 1964). The equation is shown below:

$$K = \frac{g}{v} C_b d_{10}^2$$

Where C_b is Beyer empirical coefficient $= 6 \times 10^{-4} \log \frac{500}{U_c}$.
g = acceleration due to gravity (m/s^2).
v = kinematic viscosity of water (m^2/s).

The method is useful for analyzing heterogeneous porous media with the poorly sorted grains characterized by $1 < U_c < 20$ and $0.06 \text{ mm} < d_{10} < 0.6 \text{ mm}$. The method is considered as the most suitable for the assessment of the *K* approximations of the aquifers consisting of unconsolidated sand in the Ganga Plain (Sahu and Saha 2016).

The depth ranges of the second aquifer system vary between 130 and 250 m bgl for the Karnamepur and Bharauli sites, and 134–247, 131–250 and 111–242 m bgl for the sites of Nargada Narayanpur, Paharpur, and Simri, respectively, (Table 1.6). The aquifer system is continuous within the depth ranges at the sites of Bharauli and Nargada Narayanpur. However, intervening clays appear at the other sites; in the depth range of 203–222 m bgl at Karnamepur, 206–213 m bgl at Paharpur, and 211–214 m and 224–227 m bgl at Simri. Nevertheless, these clays are lensoid type and local in extent which pinch out quickly. Hence, the aquifer system at these locations also possesses hydraulic continuity within the depth ranges as mentioned above. Thus, the estimated thicknesses of granular zones vary between the minimum of 101 m at Karnamepur to the maximum of 125 m at Simri. The estimated range of *K* values stands high at 43–196 m/d (*average effective K:* 80 m/d) for the Paharpur site, whereas the minimum range of 31–48 m/d (*average effective K:* 40 m/d) is assessed for the Bharauli. For Karnamepur, Nargada Narayanpur and Simri, the *K*-estimates fall in the ranges of 32–108 m/d (*average effective K*: 67 m/d), 36–165 m/d (*average effective K:* 53 m/d) and 22–118 m/d (*average effective K:* 88 m/d) respectively. The estimated *T* values vary between the maximum of 11,000 m^2/d at Simri and the minimum of 4800 m^2/d at Bharauli (average: 7503 m^2/d) (Table 1.6). The estimated values of *K* and *T* suggest again the highly prospective character of the second aquifer system from groundwater development point of view.

Table 1.6 Cumulative thicknesses of granular zones and grain size based estimation of the aquifer parameters K (average effective conductivity) and *T* (transmissivity) for the second aquifer system in the *As* affected areas in MGP

	Parameters of the second aquifer system				
Location	Depth range (m)	Granular. zones, D (m)	Range of K (m/d)	average effective K (m/d)	T (m^2/d)
Karnamepur	130–250	101	32–108	67	6767
Bharauli	130–250	120	31–48	40	4800
Nargada Narayanpur	134–247	113	36–165	53	5989
Paharpur	131–250	112	43–196	80	8960
Simri	111–242	125	22–118	88	11,000

9.4 Groundwater As Safety in the Second Aquifer System

It has been increasingly evident from the research in BDP (Bangladesh, and West Bengal in India) and MGP that groundwater from yellow to brownish-yellow Pleistocene sediments exhibit relatively low *As* concentration, whereas the gray to dark gray Holocene sediments rich in silt/clay bear moderate to high *As* in groundwater (Acharyya et al. 2000; Ravenscroft et al. 2005; Saha and Sahu 2016). The aquifer systems in OA in MGP comprising oxidized and yellow-brownish yellow Pleistocene sand of craton-origin also remain low in groundwater *As* (Pandey et al. 2009; Saha 2009; Sahu and Saha 2015b). But, arises the question of whether the second aquifer system can sustain the leaching of *As* bearing groundwater from the overlying aquifers? The answer to this question is that it cannot happen unless the entire first aquifer system of up to ~100 m below ground gets contaminated. We know that the bottom part of this aquifer system comprising brownish-yellow Pleistocene sediment is yet safe from *As*. It might be acting as a physical buffer zone for the second aquifer system and protecting it from *As* contamination. Michael and Voss (2008, 2009) suggested about the *flow-pattern defense* while working on models related to groundwater heads and ages in BDP. The defense pattern protects the groundwater at depths greater than 150 m (modeled as more than 1000 years) if deep groundwater abstraction was limited to domestic supply and distributed among hand-pumped wells. However, large scale irrigation pumping from the deeper aquifer, on the other hand, will amplify the downward flow of contaminated water and considerably shorten the travel time of *As* groundwater from the shallow aquifer to reach the deeper aquifer (Michael and Voss 2008, 2009). The geological formations characterized by more permeable materials below can augment the rate of downward leaching of *As* water (Ravenscroft et al. 2009; Burgess et al. 2010). In contrast, shallow irrigation pumping does not impede the flow-pattern defense of deep groundwater. Instead, it provides an extra protection by creating a hydraulic barrier against downward *As* migration (Burgess et al. 2010).

Geochemical defense is another mechanism which might be helping to keep the groundwater safe from *As* contamination in the brownish yellow Pleistocene sediment (Burgess et al. 2010). The concept relies on the reactivity of the sediment to the *As* coming to groundwater. The oxidized Pleistocene sediment in the deeper aquifer contains ferric oxyhydroxides which have a large capacity to adsorb dissolved *As* (Dixit and Hering 2003). The oxic condition in the aquifer is expected to induce higher adsorption of *As* onto the sediment surfaces.

10 Other Options of Safe Source of Water for Drinking

Other than the low *As* bearing deeper second aquifer system, the following groundwater based options can be chosen to meet the drinking need in the contaminated parts of the Holocene floodplain in MGP.

10.1 Dug Wells

Dug well (DW) is the oldest kind of structure for groundwater abstraction. Various studies from the contaminated areas in BDP (Chakraborti 2001; BGS and DPHE 2001; Ahmed and Rahman 2000; Warner et al. 2008; Bennett et al. 2010) and MGP (CGWB and PHED 2005; Saha and Chandra 2010; Saha and Sahu 2016) indicated low *As* concentrations in the open dug wells, complying with the WHO guideline value of 10 μgL^{-1}. Most groundwater samples analyzed from dug wells in Bangladesh, West Bengal, Myanmar, and Nepal have been found to contain arsenic concentrations less than 50 μgL^{-1} (the national standard for most countries in Asia) (World-Bank 2012). The National Policy for *As* mitigation in Bangladesh (DPHE 2004; Chakraborti 2001; Edmunds et al. 2015; World-Bank 2012) has suggested DWs as the preferable alternatives of safe drinking water in areas with groundwater *As* beyond the drinking limit. The exposure of water in the dug well to the open air and agitation during water withdrawal causes oxidation which causes precipitation of the dissolved *As* and even iron in water. The dug well collects water from the top layers in the shallow aquifer which gets fresh recharge in every year from rainfall and other surface water bodies. The recharge helps to contain the *As* concentration in the dug well zone in three ways; (1) flushing out of the dissolved *As* in the down-slope of the water table and getting discharged in stream/rivers, (2) diluting the *As* level in groundwater, and (3) the percolating water collects air from the aerated zone and helps in precipitating the *As* through oxidation. The air and aerated water in well may also build an oxidized zone of soil around dug well, which can significantly reduce the *As* level in water that infiltrates to the well through that zone. However, the *As* level in the dug well water needs to be checked regularly. The low residence time of water in the well owing to the high rate of withdrawal may not provide ample time for settling down of *As*.

Moderate to large diameter dug wells can no doubt be the good source of *As* safe water for drinking and other domestic uses. However, the contamination owing to bacteria and other surface derived pollutants remain as prime concerns that should be addressed properly before suggesting its use. Pollution in surface water can easily reach the dug well water. Thus, dug wells for the purpose should be constructed with requisite sanitary seals. The construction of an apron around the well can prevent entry of contaminated used water at the well site by seepage into the well. Water may be withdrawn by the installation of a manually operated hand pump.

10.2 Low As Zones in Shallow Aquifers

The entire geographic area of any block/taluka falling in the floodplain may not be affected by groundwater *As*. Around ~20–50% of hand pumps in the affected areas in MGP yield safe water with *As* less than 10 μgL^{-1}. Proper understanding of the relation between elevated *As* content and aquifer hydraulics, groundwater flow regime and local lithologic variations of the shallow aquifer can be used for mapping the *As*-safe areas in otherwise the affected block. The prominent ridges on point bars, levees, and point bar platforms, predominated by sand facies, are by and large low in *As* (Weinman et al. 2008; Sahu and Saha 2015a, b). The *As* contamination spreads along with spreading of the reducing front from the OC rich sediment unit. The rate of spreading depends upon the volume of OC release, hydraulic conductivity in the aquifer, groundwater flow direction, the rate of groundwater development and the volume of fresh oxic water recharge that reaches the spreading anoxic front. The sandier areas along the scroll bar ridges and levees exhibit low concentration of *As* as the oxic groundwater in such areas favors stability of hydrated iron oxide. There may be a source-distance relation between the *As* concentration and the OC rich clay plugs in the stratigraphic sequence (Sahu and Saha 2015a).

11 Conclusion

The physical dynamics in the Ganga and other Himalayan rivers in MGP, such as their meandering and migration, and at places channel avulsion during the Holocene period have formed narrow to wide floodplains characterized by fluvio-lacustrine/lacustrine environments of sediment deposition. Other than lithology, climate and tectonics have also played their roles in the shifting of the river channels in MGP. The geomorphology and the distinctive sedimentary facies associated with different sub-environments display characteristic groundwater *As* levels. Typical sedimentary sequences with rich OC content in the lithology display higher levels of *As* in groundwater. The distribution pattern of groundwater *As* bears significant correlation with the floodplain geomorphologic elements. Groundwater *As* contamination is observed in the NA affecting both the AFP and OFP. Vertically the contamination is confined largely within the depth of the top ~50 m of the sedimentary sequence in MGP.

A two-tier aquifer system is noticed in MGP within 300 m bgl, overlain by an aquitard at the top. The Holocene deposits comprising the top aquitard and the upper slice (15–20 m) of the first aquifer system host the contaminated aquifers in MGP largely within the depth of ~50 m below ground. The sediment in the top aquitard is predominantly Himalayan derived, un-oxidized and gray in colour. The first aquifer system and the top aquitard are hydraulically connected and possess shallow water levels (<8 m bgl). This unit gets ample recharge from monsoon rainfall.

The Pleistocene aquifers (the bottom three-fourth of the first aquifer system and the entire second aquifer system) in the OA of MGP are low in groundwater *As*. The lower (second) aquifer system (beyond 100 m depth below ground) is separated from the overlying first aquifer system by middle clay. The storage coefficient values of the second aquifer reveal its confined character and hydraulic separation from the upper aquifer system. The middle clay is characterized by large values of hydraulic resistance (range of L: 450–4103 m) which indicate smaller leakage rate through the aquitard. The low vertical conductivity values (range of K': 0.01–0.33 m/d) show the impermeable character of the aquitard. The aquifer system has got its recharge areas located at further south in the basin in the exposed Pleistocene sediments. The transmissivity values of the aquifer system indicate that it can be used for community water supply with a regulated draft so that its hydraulic head remains well above the head of the first aquifer. The water supply wells should adopt proper designs like cement sealing against the *As*-contaminated zones and the middle clay to prohibit downward leakage of *As*-contaminated groundwater from the top aquitard.

The entire area of the floodplain is not contaminated, and there are prominent ridges on point bars, levees and point bar platforms, predominated by sandy facies, which are by and large low in *As*. Most of the hand pumps (89–94%) from these units exhibit low *As* concentrations, often within the drinking limit prescribed by WHO. Community hand pumps in the depth range of 5–15 m below ground can be constructed in those areas for drinking need of the local people. However, regular quality check of such tube wells is a must to ensure the supply of low *As* groundwater.

Large diameter dug wells in the floodplain can also be constructed to tap the replenished dynamic groundwater, which is usually oxic and low in *As*. The air-contact and the surging in dug wells during groundwater withdrawal help in precipitating the *As* through oxidation. Other than diluting the *As* level in groundwater, the recharge in the dynamic zone of the shallow aquifer also removes *As* through flushing. However, the dug wells need to be constructed carefully with proper sanitary measures to ensure the safety of well water from bacteriological and other pollutions coming from the surface.

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Chapter 2
Geomorphic Controls on Spatial Arsenic Distribution in Aquifers of the Brahmaputra River Floodplains

Runti Choudhury, Chandan Mahanta, Pallavi Sharma,
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Abstract The present study was conducted to explore the controls of geomorphic features on arsenic (*As*) mobilization in groundwater aquifers of the Brahmaputra River floodplains. Linear Imaging Self-Scanning Sensor-4 (LISS IV) imagery was used to demarcate different geomorphic units based on spectral signatures and field investigations using ArcGIS 9.3 software. A total of 132 groundwater samples covering four different geomorphic units were tested in the laboratory using Atomic Absorption Spectrophotometer (AAS). Overlay of test results of groundwater samples on classified geomorphic map revealed that out of a total 132 samples 77% sources in younger alluvium and 89% sources tested in older alluvium were found to be affected with *As* beyond permissible limit of 10 μg/L set by the World Health Organization. *As* enrichment along the different geomorphic units followed the trend as Younger Alluvium> Older Alluvium> Natural Levees> Floodplain Deep.

1 Introduction

Arsenic contamination of groundwater is a major drinking water concern, and several million people worldwide are affected by these crises (Smedley and Kinniburgh 2002; Ravenscroft et al. 2009; Chakraborti et al. 2017; Mukherjee et al. 2018; Biswas et al. 2012). The reductive dissolution of Fe-oxyhydroxides is the primary mechanism of arsenic release into groundwater, yet factors controlling the spatial

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variation of arsenic from meters to tens of meters remains elusive. Mode of arsenic occurrence and mobility in sedimentary aquifers have been linked to local geology, geomorphology, hydrogeology, and geochemistry of sediments and water, as well as anthropogenic activities such as mining and land use (Kinniburgh and Smedley 2001; Smedley and Kinniburgh 2002; Weinman et al. 2008; Bhattacharya et al. 1997; Saha and Sahu 2016).

Arsenic contamination of groundwater in the Brahmaputra floodplains of Assam, in the northeastern part of India, has been reported lately (Singh 2004; Choudhury et al. 2015, 2018; Chetia et al. 2011). A recent study by UNICEF and the Public Health Engineering Department (PHED), Government of Assam, reported arsenic contamination in 76 blocks in 18 districts of Assam with 29% of wells (out of a total 56,180 sources tested) having As concentration above the World Health Organization's (WHO) permissible limits of 10 μg/L (Mahanta et al. 2015). More recently, there has been an increased attempt to understand the groundwater contamination issue in the Brahmaputra Floodplains (Choudhury et al. 2015, 2018; Verma et al. 2016). In the present study area, on the southern bank of the Brahmaputra River, Goswami et al. (2014) reported dissolved As concentrations as high as 500 μg/L. Based on their studies Verma et al. attributed high arsenic concentrations in the study site to differences in tectonic history and sediment provenance (Goswami et al. 2014).

Over three decades extensive studies on arsenic in Bangladesh and West Bengal have enriched our understanding on the arsenic menace (Ahmed et al. 2004; Ravenscroft et al. 2009; McArthur 2019; Nickson et al. 2000; Bhattacharya et al. 1997; Breit et al. 2004). In terms of spread and thickness of alluvial plains, the Brahmaputra floodplains in Assam has some close resemblance to those of Bangladesh, yet in-depth studies concerning factors contributing to arsenic enrichment within the aquifers of the region have only been taken up much recently (Mahanta et al. 2015). The current study thus aims to evaluate the geomorphic controls on the spatial *As* heterogeneity in aquifers of the Brahmaputra River floodplains in Assam.

2 Study Area

The study area forms a part of the Upper Assam foreland basin (Fig. 2.1). The land surface of the site is characterized by a depression followed by a slightly elevated area comprising of the sandy, loamy soil type characteristic of tea garden areas. A topographic gradient of 10–12 meters (m) is observed along the 35 m transect. The Brahmaputra River draining the Himalayas bounds the northern boundary while the Naga Patkai hill ranges bounds the southern boundary of the study transect. The two rivers, viz. Dhansiri and Bhogdoi, flowing from the Naga Hills mark the western and eastern extremities of the study area. The southern tributaries, the Burhi Dihing, the Dhansiri and the Kopili, drain through turbidities similar to those in Siwaliks, the Indian Plate and ophiolites (Singh 2006).

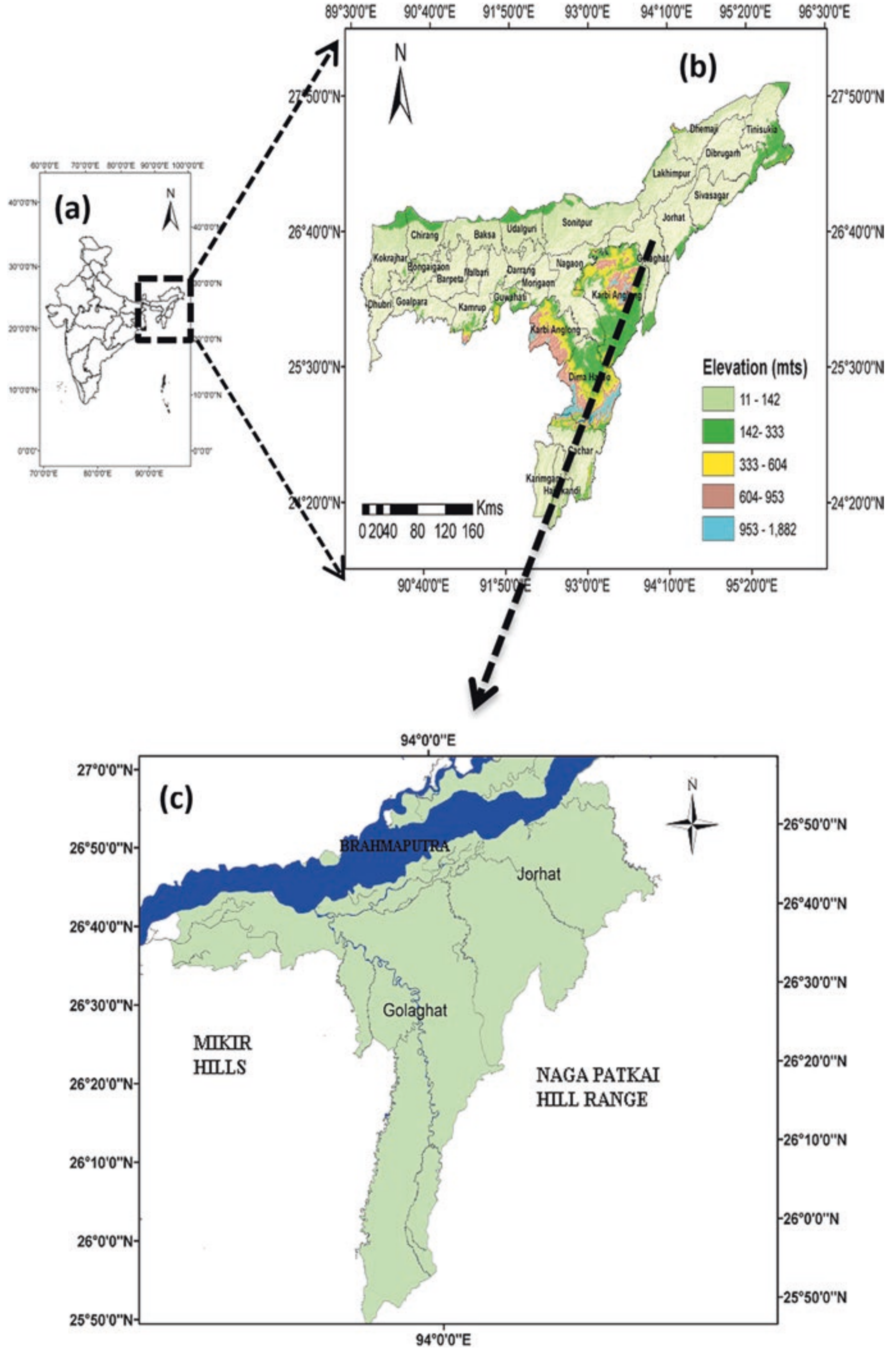

Fig. 2.1 Map showing the location of (**a**) India, (**b**) Assam and (**c**) Context map of the Study Area

2.1 Land Use and Land Cover

Agricultural lands dominate most of the study area. Forests and wetlands cover the remaining areas of the district along with numerous perennial and non-perennial water bodies. Apart from crops, tea plantation is a major industry with several hectares being used for plantations (Fig. 2.2) (CGWB 2013).

2.2 Hydrogeology

Along the study area, groundwater occurs both under water table to semiconfined conditions in the near-surface conditions, and under semi-confined to confined conditions in the deeper horizons (CGWB 2013). Depth to water level in the water table zone varies from 0.41 to 3.07 m bgl in the pre-monsoon period and 0.56 to 3.41 m bgl during the post-monsoon period. Available subsurface information indicates that three or four prolific systems exist down to an explored depth of 300 m, in the central parts. Adjacent to the Brahmaputra River, five to six aquifer systems with limited thickness exist within the depth range of 400 m (CGWB 2013). In the southern parts, the aquifer system fades or its thickness reduced due to the mixing of finer particles of sand and clay (CGWB 2013).

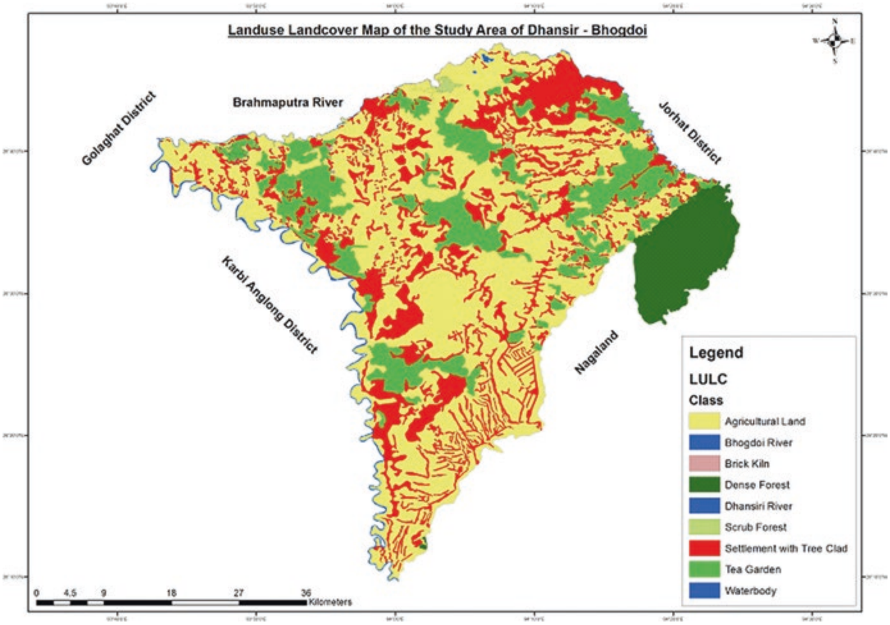

Fig. 2.2 Land use and Land cover of the study area

3 Materials and Methods

The outcome of the Arsenic Screening and Surveillance Program in Assam valley (Mahanta et al. 2015), revealed groundwater *As* distributions to be localized at discrete pockets within the Brahmaputra floodplains. Close scrutiny of some of these hotspot areas, where groundwater As concentration was much above WHO guideline values, demonstrated that significant variation in As concentrations occurred within the southern bank of the Brahmaputra River. Thus, for an in-depth understanding of this spatial variability, a 35 km representative transect perpendicular to the Brahmaputra River was selected for groundwater sampling and geomorphic mapping. The elevation difference of 10–12 m is observed along the 35-km long transect. A total of 132 groundwater samples were collected for detailed hydrochemical analysis during November 2016.

During sampling, ancillary information, such as the age of well, depth of well and drilling method adopted, was collected from the household. Prior to sampling each of the wells was purged for 5–10 min to homogenize the sample and minimize the impact of iron pipes. High density polyethylene (HDPE) bottles were used for sample collection, which were kept in 1.2 N HCl solution overnight and then washed 5–6 times with double distilled water and air dried before sample collection. 100 mL water sample was filtered using 0.45 μm pore size Whatman of syringe filters and preserved with 3–4 drops of HCl for cations and trace metals while another set of the unacidified filtrate was collected in a separate bottle without any headspace in the bottle. All the water samples were protected from sunlight during transport to the laboratory, and stored at 4 °C until analysis. They were analyzed within 2 weeks of sampling (Fig. 2.3).

ESRI ArcGIS 9.3 software was used for the preparation of maps and spatial mapping of As and other parameters in the study area, while LISS IV imagery obtained from North East Space Application Center (NESAC) was used for mapping out geomorphic units in the study area. Required image processing was done prior to the use of the LISS IV imagery. Geomorphological units were identified through visual interpretation on the basis of tone, texture and structure of the different features on the imagery.

4 Results and Discussion

Geomorphic controls on *As* distribution patterns in the Bengal basin and the Central Gangetic plains have been widely reported by many (Weinman et al. 2008; McArthur et al. 2004; Saha and Sahu 2016). To investigate the influence of geomorphic features on *As* distribution in the aquifers of the Brahmaputra Basin, LISS 1 V imagery of the southern transect was mapped. Mapping indicated that most parts of the study area are marked by depositional geomorphic units of fluvial origin (viz. Active Floodplain, Alluvial Plains and Older Alluvium), while hills of structural origin were observed towards the Naga Patkai Range (Fig. 2.4).

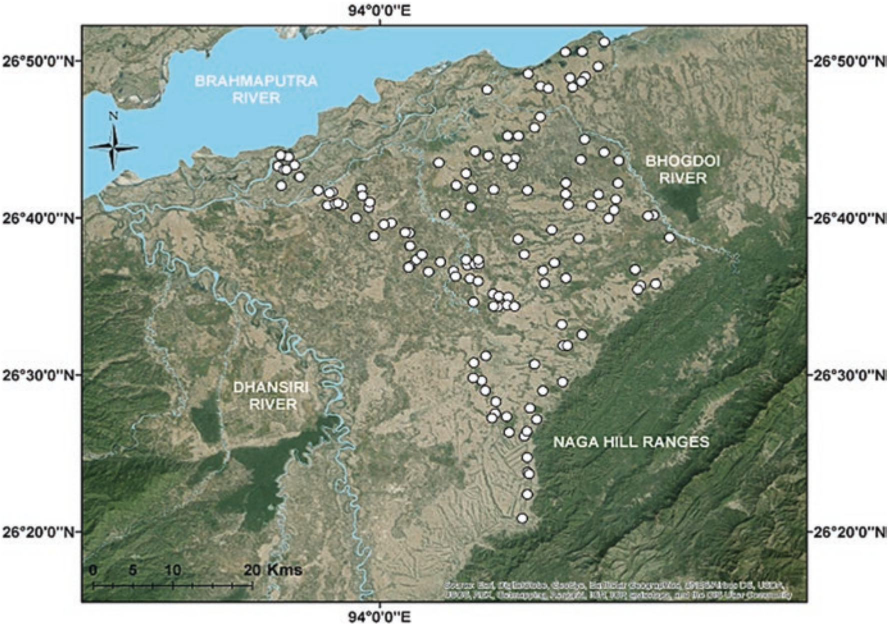

Fig. 2.3 Map showing groundwater sampling (marked as white circles, n = 132) locations along the study area transect. Brahmaputra River is flowing from east to west at the bottom. The satellite image of the study site acquired from ESRI, World Imagery (ESRI 2018)

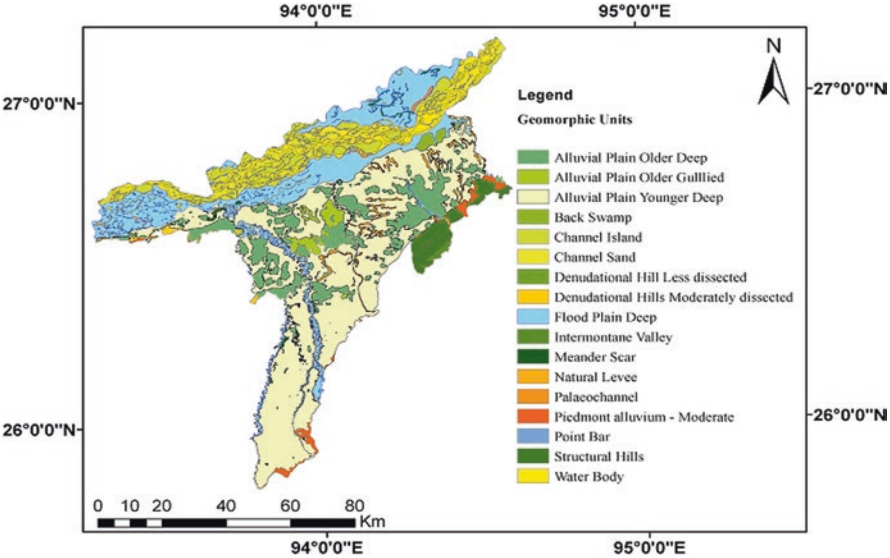

Fig. 2.4 Geomorphology map of the area along the southern transect, showing different geomorphic units mapped out from LISS IV Imagery

The study area forms part of upper Brahmaputra River floodplain, a geomorphic unit denoting the alluvial plain between Brahmaputra River in its North and Naga Patkai Hill ranges in its south. The younger alluvium is sediments deposited as an alluvial fan, and floodplain sediments brought down by the Brahmaputra and its southern tributaries, viz. Dhansiri and Bhogdoi that drain the Naga Patkai ranges (Mathur and Evans 1964). The Quaternary sediments, overlying uncomfortably the Tertiary deposits, are described as Older Alluvium or High Level Terraces, and consist of indurated yellowish or reddish clays with sand, shingle, gravel and boulder deposits. Natural levees occur as wedges all along the banks, breaching of some of which leads to the development of crevasse splay deposits (Sarma 2005). Most of the existing paleochannels are manifestations of neotectonics (Sarma 2005). These paleochannels are the remnant geomorphic unit formed through the lateral shift of the rivers. Brahmaputra plain is covered by younger alluvial sediments at most of the places deposited from the sediment load carried by the river and its tributaries (Sarma and Phukan 2004). The Brahmaputra River forms a large anabranching, multichannel and multipattern axial Tributary River in the Brahmaputra valley portion of the Himalayan foreland basin (Sarma 2005).

4.1 Variation of As *Concentrations in Different Geomorphic Features*

Overlayed map132 groundwater samples tested for *As* on the geomorphic map revealed that the primary geomorphic features, which include Younger Alluvial Plains and the Older Alluviums, contain *As* above permissible limits as opposed to the widely reported observations that aquifers in older alluviums host low As aquifers (Ahmed et al. 2004; Nickson et al. 2000; Polya et al. 2005) (Fig. 2.5). The older alluvium along the study transect are dominated by loamy sandy soils and are primarily used for tea plantations. Out of 132 samples analyzed, 77% sources in Younger alluvium and 89% sources in older alluvium are found to be affected with *As* beyond permissible limit of 10 mg/L. *As* enrichment along the different geomorphic units followed the trend as Younger Alluvium> Older Alluvium> Natural Levees> Floodplain Deep (Fig. 2.6, Table 2.1).

In Bangladesh, older alluviums, which are the Pleistocene sands with their characteristics yellow to brown hue, are reported to produce As safe aquifers; the understanding being that those aquifers have had considerable time to flush the *As* from the aquifers (Ahmed et al. 2004; Nickson et al. 2000; Polya et al. 2005). The high *As* concentration observed in the older alluvium in the study area can be attributed to the fact that the older alluvium units are depositional units with high potential for tea cultivation. Probable deep irrigation for tea plantations may perhaps facilitate drawdown of As enriched Holocene water to Pleistocene aquifers. The effect of deep irrigation on facilitating drawdown of *As* enriched Holocene water to Pleistocene aquifers has been well established in Bengal Basin and Vietnam (Berg

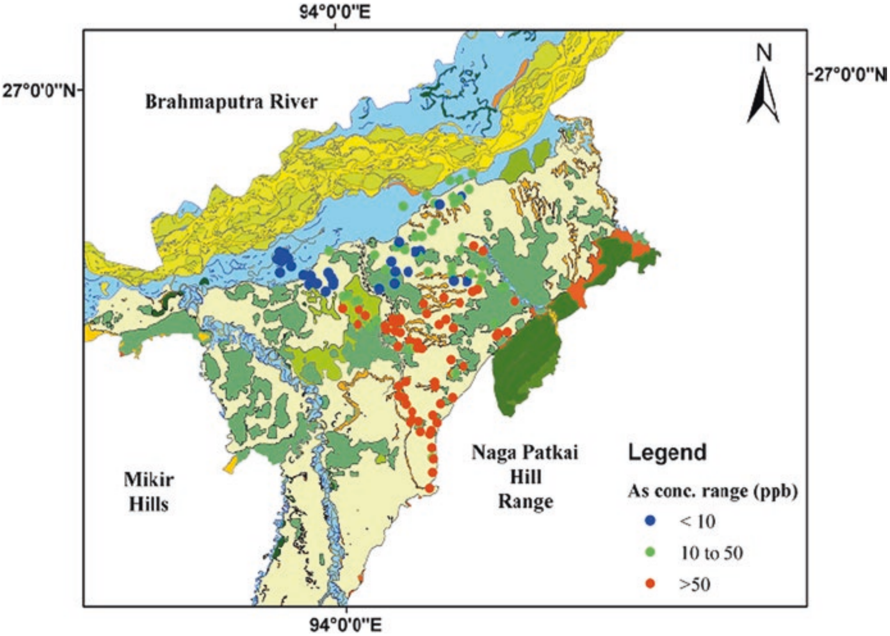

Fig. 2.5 Overlayed map of groundwater *As* concentrations on the geomorphic map along the southern transect. Classified geomorphic units are similar to those outlined in Fig. 2.4

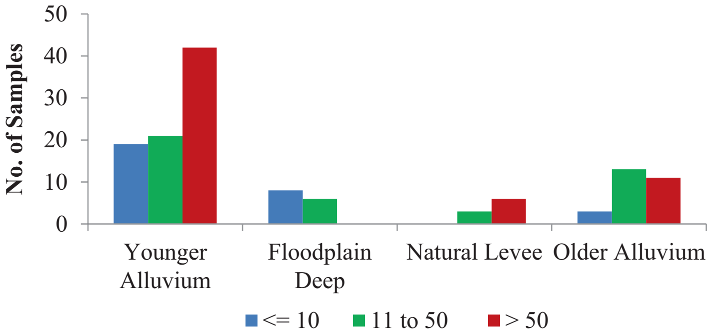

Fig. 2.6 Arsenic contamination in groundwater samples using data points extracted from four geomorphic units

et al. 2008). Thus, *As* enrichment of older alluvium is attributed to the drawdown of As enriched groundwater to older alluvium aquifers, although it should be confirmed with further studies.

Although paleochannels have good potential for groundwater, these areas ironically are reported to host high *As* groundwater (Hoque et al. 2011). Satellite mapping of LISS IV image of the southern study transect demonstrate no such paleochannels in the area close to the Naga Hills. These areas instead are part of the

Table 2.1 Arsenic concentration in different geomorphic units in ppb (parts per billion)

Geomorphic units	Min (ppb)	Max (ppb)	Average (ppb)	Std. Dev (ppb)
Younger	0.0	604.0	117.0	138.5
Floodplain Deep	0.0	35.5	9.3	11.7
Natural Levee	39.9	229.0	133.6	76.9
Older	0.5	299.0	72.4	87.1

young alluvial deposits. Aquifers dominated by thick clay cappings represent low energy environment. With little evidence of major south bank tributary system influencing the zone of high *As*, it is hypothesized that weathered materials deposited from the Naga Patkai hills get accumulated in the topographic low in the frontal zone of the mountains. With limited scope of these materials to get flushed out, the aquifers near the foothills host high arsenic groundwater (Choudhury et al. 2018).

Unlike in most *As* affected areas, areas of active floodplains in the study transect host low As groundwater, particularly along the studied transect. Recently (Stahl et al. 2016) reported two different conditions under which riverine recharge could either contaminate or flush aquifers of *As*. Active depositional sites near rivers are sites hosting high *As* owing to those being reactive riverbed sediments. On the contrary, non-depositional areas are zones which host low *As* groundwater because non-deposition will decreases the thickness of the reactive riverbed sediments. While these possibilities do exist in our study sites in the upper Brahmaputra Valley, considering that the active floodplain area near the Brahmaputra main channel is an area of active sediment deposition, we attribute the low *As* aquifers in areas of active floodplains to the recurrent flooding events which flush the aquifers of their As levels.

Natural levees in the study are observed to host high *As* groundwater, in conformity to that observed in the Bengal Basin. Weinman et al. (2008) associated high *As* groundwater with levees formed during a relatively low-energy fluvial regime. The low energy regime facilitates the accumulation of finer materials and the formation of clay/mud capped aquifers. These clay capped aquifers host high *As* groundwater. An analogy can also be drawn for the southern transect where river meandering is a dominant characteristic of the tributaries.

5 Conclusion

The present study reiterates the important role of geomorphic units in controlling As distribution. The study highlights that geomorphic features can explain As heterogeneity in the Brahmaputra Valley aquifers. Among different geomorphic features, younger alluvium, natural levees and older alluvium aquifers in the study area are observed to host high arsenic groundwater. While depositional features and levee characteristics explain high As aquifers in the levees, high As aquifers in older alluvium are attributed to leaching of *As* enriched groundwater from the uppermost contaminated shallow aquifer to the underlying to Pleistocene aquifers, although further studies would be required for certainty.

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Chapter 3
Reductive Dissolution of Fe-oxyhydroxides a Potential Mechanism for Arsenic Release into Groundwater in the Alluvial Plain of River Brahmaputra

Shirishkumar M. Baviskar and Runti Choudhury

Abstract The mobilization of dissolved arsenic (*As*) in groundwater environment is controlled by its chemodynamics associated with solid-phase arsenic. The mechanism of Arsenic mobilization in the Groundwater of the alluvial plains of river Brahmaputra were studied from aqueous and solid-phase geochemical analyses of groundwater samples and sediment cores at various depths from a borehole. The sediments cores were analyzed for parameters like total and sequentially extracted Fe and As, organic carbon content and carbonate phases. The groundwater samples collected from the close proximity of the drilled bore well were analyzed for major and trace element hydrogeochemistry. Fe oxyhydroxides were observed as the major leachable for arsenic solid phases. The presence of Fe oxyhydroxides was found in the aquifer sediments using scanning electronic microscope energy-dispersive X-ray (SEM-EDX) and X-ray diffraction (XRD) analysis. This experimental research study suggest that bacterially mediated reductive dissolution FeOOH is probably an important mechanism for releasing As from the sediments into the groundwater.

1 Introduction

Consumption of Arsenic (As) can lead to arsenicosis which is fatal as it causes cancer of skin, lung, and bladder and gastrointestinal and pulmonary conditions (Smith et al. 2009; Chakraborti 2011). Higher As concentrations in water, soil, and sediments are a major public health concern in many parts of the world (Smedley and Kinniburgh 2002; Mohammed Abdul et al. 2015). Mobilization of As from the solid

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phase into the aqueous phase is not yet fully understood (Blute et al. 2009; Dai et al. 2018). As distribution between solid and aqueous phases is determined largely by redox conditions, which govern the oxidation states of As and the thermodynamic stability of solid phases. Due to the redox condition As associated with solid phase via surface adsorption as inner and outer sphere complexes (Goldberg and Johnston 2001; Al-Sid-Cheikh et al. 2015) co-precipitates with an aqueous phase (Waychunas et al. 1993; Park et al. 2016).

Arsenite As(III) or arsenate As(V) or both, which are inorganic oxyanion forms of arsenic, dominate natural sediment and water systems with different toxicity levels. Arsenite has been considered more toxic than arsenate. However, recent studies have shown that most of the ingested arsenate is reduced to arsenite (Welch and Stollenwerk 2003). Under the oxidizing environment, pentavalent arsenic is the dominant species (Eliche 2009). In oxidizing environments at below pH 2, As occurs as arsenic acid ($H_3As_VO_4$); while in reducing environments and at below pH 9, As occurs predominantly as arsenous acid($H_3As_3O_3$). At higher pH, arsenous acid is deprotonated and forms $H_3As_{III}O_2^-$ (Bhattacharyya et al. 2003).

It is reported that maximum As concentrations are observed in the Jorhat district located on the southern bank of the river Brahmaputra in Assam, India (Chakraborti et al. 2004). Recently researchers have investigated the As release mechanism from the solid phases into the dissolved phase (Mahanta et al. 2015a, b; Baviskar et al. 2015). Chakraborti et al. (2008) had found As contamination of groundwater in the Brahmaputra floodplains of Assam, the northeastern part of India. Study by United Nations Children's Fund (UNICEF) and the Public Health Engineering Department reported As contamination in 76 blocks in 18 districts of Assam with 29% of wells (out of a total 56,180 sources tested) having As concentration above the World Health Organization (WHO) permissible limits of 0.01 mg/L (Mahanta et al. 2015a). Chakraborty et al. (2009) observed dissolved arsenic concentration values to be 0.49 mg/L. Singh (2004) suggested that sediments having clay and organic compounds in high percentage might retain and release As into the groundwater aquifers of Assam.

There have been extensive studies on the source and mobilization of arsenic in Hooghly river basin in West Bengal and Brahmaputra river basin in Assam in India and Bangladesh (Horneman et al. 2004; Anawar et al. 2004; Mahanta et al. 2015a, b; Baviskar et al. 2015). Many researchers have highlighted correlations between Iron (Fe) and As levels during the desorption from a solid phase to dissolved arsenate phase (Pedersen et al. 2006; Rowland et al. 2007; Quicksall et al. 2008). The objective of this research is to investigate the mechanism of co-dissolutions and correlations between Fe and As in Brahmaputra floodplain of Assam during mobilization of *As* from solid to aqueous phase. The approach involves simultaneous experimental investigations by studying hydrogeochemistry and mineralogy of the groundwater, and soil samples obtained from freshly dug bore well, and analyze different parameters to identify factors that influence *As* release from solid phase into the aqueous phase.

2 Materials and Methods

2.1 Study Area

The study area Jorhat district forms a part of the upper floodplains of the Brahmaputra River consisting of distributed young alluvial sediment terrains. The Jorhat district lies at 26.75°N 94.22°E in the Indian state Assam covering an approximate area of 2851 km^2 (Fig. 3.1). The area falls in a temperate climate zone marked by heavy rainfall and humidity (temperature ranges between 6 and 38 °C and mean relative humidity between 92.00% and 98.00%). The annual average rainfall in the study area is 2818 mm (Mahanta et al. 2015a, b; Baviskar et al. 2015). The Brahmaputra basin is surrounded by the eastern Himalayas in its north and east; the Naga and Patkai range in the northeastern part, with Shillong plateau on its south (Mahanta et al. 2015a). It is an integral feature of the fluvial landforms in Assam. The geological characterization of the basin sediments shows that it is from the Paleozoic and Cenozoic era, comprising of gneisses, high and low-grade schist, ultrabasic rocks, shale, sandstone, mottled clay, and conglomerates (Sharma 2005). The recent alluvial deposits have originated as the alluvial fans and floodplain sediments of Brahmaputra River and its tributaries (Mahanta et al. 2015a). The Brahmaputra River and its tributaries carry most of these alluvial sediments often disturbed by tectonism-induced seismicity and landslides (Sharma 2005). The northern tributaries are braided, and the composite flood plain made up primarily of Holocene

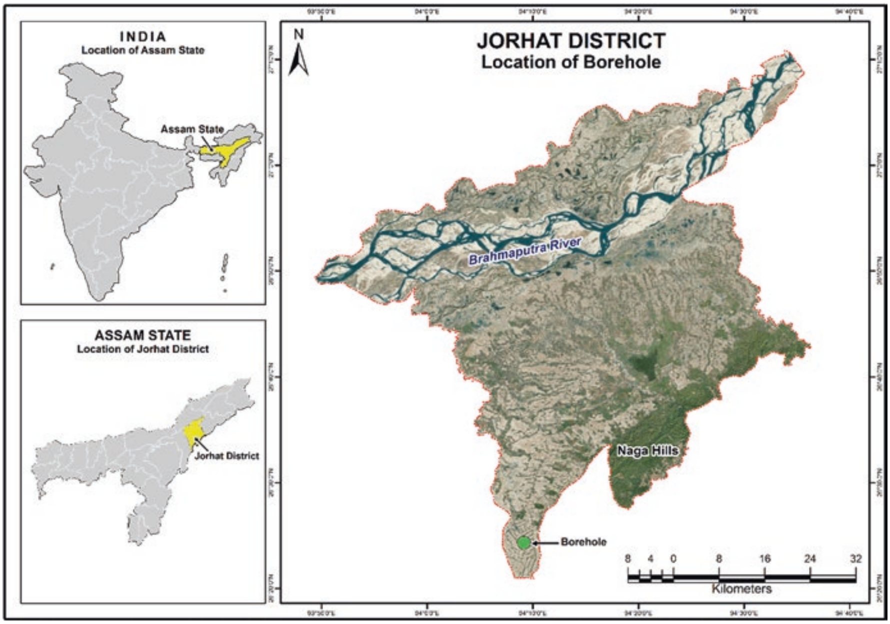


Fig. 3.1 Map of borewell location in the Jorhat district in Assam, India

sedimentary aquifers. The predominant geologic formations include unconsolidated alluvial sediments of Quaternary age characterized as older and younger alluvium. The upland areas are occupied by older alluvium with oxidized sediments which is relatively compact in nature. The younger alluvium occurs along the low-lying tracks of the area aligned to the river course. A superficial blanket of clay belonging to younger alluvium occurs further in the southern part. The study area is characterized by thickness of aquifer in the central and northern parts comprising of medium to coarse sand within an explored depth of 450 m, capped in the upper parts by 30 to 50 m of clay beds and varied thickness of clay intercalation in between the latter possibly inhibiting vertical connectivity of aquifers and thus confining the arsenic-bearing strata to a specific depth horizon (Baviskar et al. 2015; Mahanta et al. 2015a).

2.2 Sample Collection and Characterization

The borewell was drilled for the present study around a few highly arsenic contaminated wells (Fig. 3.1). Drilling was done by hand flapper technique as describe by Horneman et al. (2004). The sediments from different depths of the drilled borehole were collected in January 2010. Water table depth was found to be in the range of 2.10 to 2.60 m. The sediment samples were obtained at different depths ranging up to 35.00 m. To avoid direct contact with sunlight and oxygen the collected sediments were packed in two layers of zipped black polyethylene bags and brought and stored in the laboratory until analysis. The groundwater samples from 18 tube well sources located in the vicinity of the drilled well site were also collected. Standard procedures of groundwater sample collection and preservation were followed (Baviskar et al. 2015; Mahanta et al. 2015b). The in situ measurements for pH and electrical conductivity of the samples was done using a portable pH meter (pHTestr 10 Eutech) and conductivity meter (ECTestr 11—Eutech). The collected groundwater samples were also transferred to the Environmental Engineering laboratory in IIT Guwahati for quantification of the physicochemical parameters.

2.3 Sediment Analysis

Sediment samples were analyzed for Fe and As concentrations, organic content, carbonate content, moisture content and specific surface area. Elemental analysis was performed according to standard protocols for extraction of trace elements from sediments (as per D3974-8 (reapproved 2003) by ASTM validated by ISO 11466:1995—Soil quality). The sequential extraction method was used to evaluate the metal distribution into different chemical forms present in solid phases (Tessier et al. 1979; Filgueiras et al. 2002). One gram of dry soil sediment samples were sequentially leached using deionized water (DIW) for quantification of the water-soluble fraction of As and other trace elements. For estimating the elements bound to carbonates, the first phase residue was extracted with 40 mL of 0.11 mol/L acetic acid (Tessier et al. 1979; Filgueiras et al. 2002). The second phase residue was then extracted by using 40 mL of 0.1 mol/L hydroxylammonium chloride to evaluate the elements in reducible phase, i.e., poorly crystalline and amorphous oxyhydroxides, followed by extraction with hydrogen peroxide to evaluate the oxidizable phase, which contains the elements bound to the organic matter and sulfides. Finally, the last residue was extracted using aqua regia to estimate the residual phase which is the immobile phase (Baig et al. 2009). The extracts were filtered using a filter paper by adding 1 mol/L HNO_3 solution in small quantities on the last residue in the centrifuge tube. The determination of Fe in the extracts was performed using flame atomic absorption spectroscopy (FAAS) and As by vapor generation assemblage (VGA) attached to FAAS. The calibration process for the determination of As and Fe using FAAS was followed using methods described in Hovind (2004). Table 3.1 shows the sequential extraction steps and the mobility of the elements evaluated together.

Organic and carbonate contents were evaluated by a loss on ignition method of Bengston and Enell (1986) and Dean (1974); while particle size distribution was determined using laser diffraction principle in a particle size analyzer (Master sizer 2000, Malvern).

Mineralogy of the sediment samples was studied combining optical petrography, X-ray diffraction, and electronic scanning microscope (SEM) coupled with an energy-dispersive X-ray (EDX) analysis on the residuals obtained from each BCR protocol step. The mineralogical study (MAC Science XXP 18) was conducted on the residuals obtained from each sequential extraction protocol step by using X-ray powder diffraction with Cu Kα radiation and a position-sensitive detector. The accelerating voltage was kept at 30 kV and the current at 20 mA. A divergent slit (1°), a scattering slit (1°), and a receiving slit (0.2 mm) were used to produce the

Table 3.1 Summary of the sequential extraction procedure followed in this research

Step	Extractant	Solid Phase
1	Deionized water	Water Quantification of the water-soluble fraction of As and other trace elements
2	Acetic acid: CH_3COOH (0.11 mol/L) at pH 2.85	Exchangeable, water, and acid soluble (carbonates)
3	Hydroxyammonium chloride: $NH_2OH.HCl$, (0.1 mol/L) at pH 2	Reducible (iron /manganese oxides)
4	Hydrogen peroxide: H_2O_2 (8.8 mol/L) followed by ammonium acetate: CH_3COONH_4, (1.0 mol/L) at pH 2	Oxidizable (organics substances and sulfides)
5	Aqua regia: 3 HCL+ HNO_3	Residual (remaining silicate bound metals)

best spectrum. Scans were conducted from 4° to 80° at the rate of 1°– 2θ/min. The morphological investigation was also carried out on the residuals obtained from each phase of sequential extraction protocol by SEM-EDX, where the accelerating voltage was kept at 30 kV. The EDX was run with a lithium-drifted silicon detector at a resolution of 133 eV. The images were taken with maximum magnification. The mineral phases present in the residuals obtained from each BCR protocol step were also recognized by X-ray diffraction (XRD) analysis as described by Moeck (2008).

2.4 Groundwater Analysis

Groundwater samples (n = 18) were analyzed for their chemical constituents like calcium, magnesium, sodium, potassium, chloride, bicarbonate, sulfate, phosphate, and nitrate. This was achieved using standard methods as suggested by the American Public Health Association (APHA—20th edition). Concentrations of Ca, Mg, and Na were measured using a flame photometer (Model: 128 Systronics). Standard solutions for the above analysis were prepared from respective analytical reagent grades. Chloride was measured using the argentometric method, sulfate was measured following a turbidimetric method, and phosphate was determined using an ascorbic acid method. Major anions $HCO^-{}_3$, Cl^-, PO_4^{3-}, and $SO4^{2-}$ were analyzed in unacidified water samples (APHA—20th edition). Quantification of $HCO3^-$ was done using a titration method. Arsenic concentrations were measured using a hydride generation technique (Haavard 2004) in FAAS with VGA. The instrument was calibrated, and the sample concentrations were confirmed using a series of As calibration standards of known concentrations. The calibration standards were prepared from high purity single-element stock solutions.

3 Results and Discussions

3.1 Groundwater Chemistry

pH of the groundwater samples collected from 18 tube wells sources in the proximity of the drilled bore well was found to be ranging between 6.00 and 6.40. HCO^{3-} dominated the anion chemistry with values ranging from 116.00 to 210.00 mg/L, while Cl^- varied between 12.50 and 91.60 mg/ L. Concentrations of $SO4^{2-}$ and $PO4^{3-}$ ranged between 1.50–46.70 and 0.22–2.40 mg/L, respectively. NO^{3-} concentrations were low and ranged from 0.01 to 2.00 mg/L. Microbial degradation of organic matter in the aquifers results in the reduction of both $SO4^{2-}$ and NO_3^-(Mayorga et al. 2013; Yu et al. 2018b). Distribution of major cations in groundwater (see Table 3.2) were Ca^{2+} (7.40–25.20 mg/L), Na^+(14.00 – 48.60 mg/L), K^+ (1.00–2.00 mg/L), and Mg^{2+} (1.80–6.10 mg/L). Concentrations of dissolved As, Fe, and Mn in groundwater in the study area are given in Table 3.2. Fe concentrations

Table 3.2 Groundwater geochemistry ((mg/L), pH and electrical conductivity (EC in µS/cm) of the study area (n = 18)

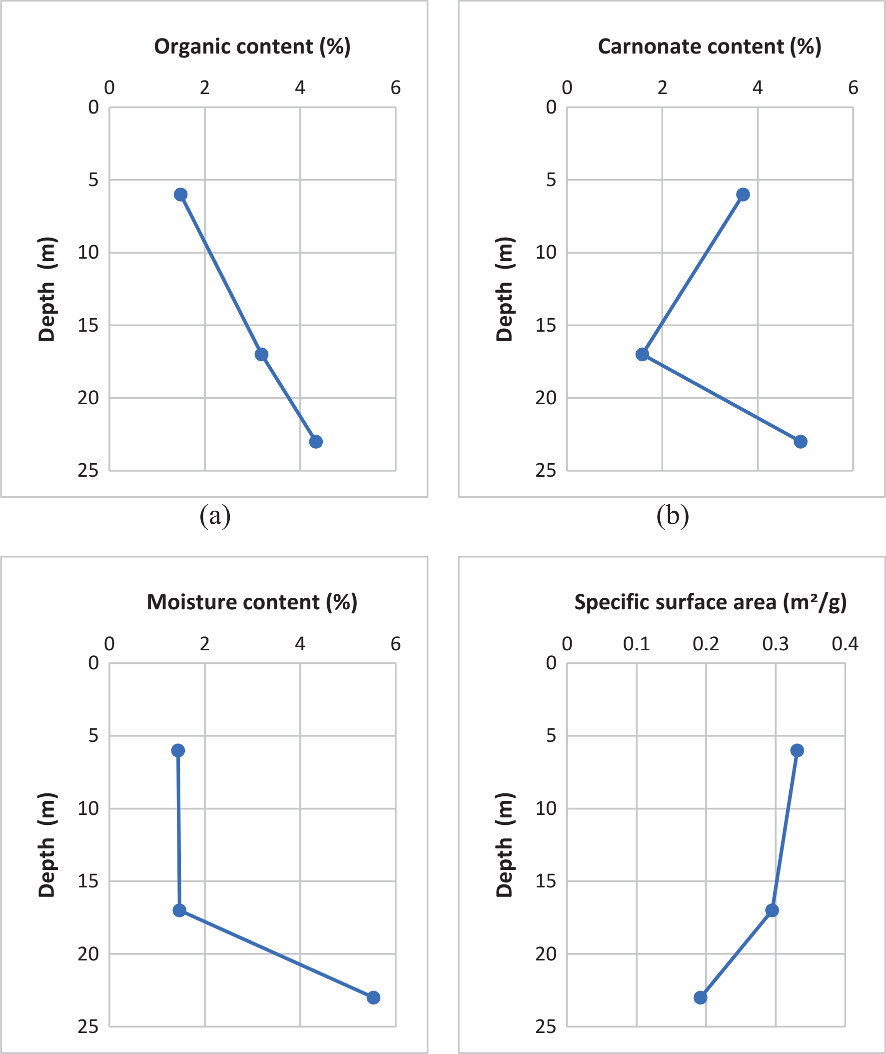
Parameters	Max	Min	Mean	Median	Std. Dev
pH	6.8	6	6.4	6.35	0.25
EC	376	197	316	318.5	46.27
Na^+	48.63	14.09	33.35	34.39	9.43
Ca^{2+}	25.2	7.4	16.45	17.2	6.27
Mg^{2+}	6.12	1.8	4	4.18	1.52
Fe	42.88	0.95	17.13	14.94	11.14
K^+	1.95	1.05	1.34	1.38	0.29
Mn	0.49	0.02	0.24	0.21	0.14
HCO_3^-	210	116	165.43	160	32.11
$SO^{2-}{}_4$	46.7	1.5	13.8	9.15	11.82
Cl^-	91.6	12.5	28.29	20.59	21.88
$PO^{3-}{}_4$	2.4	0.22	0.7	0.39	0.61
$NO^-{}_3$	2.02	0.01	0.64	0.38	0.63

in groundwater samples ranged between 0.95 and 42.88 mg/L, while Mn concentrations ranged between 0.02 and 0.49 mg/L. The concentration of As from 18 sampled tube wells varied between 0.05 and 0.44 mg/L, with all samples having concentrations above 0.01 mg/L (which is the WHO standard for safe drinking water). A recent study by Goswami et al. (2014) in the same district reported arsenic concentrations as high as 0.46 mg/L in groundwater, with 1.80% for 286 samples tested having As concentrations greater than 0.30 mg/L.

The magnitude of As contamination of groundwater in the Jorhat district seems alarming from the current study. The presence of cations and trace elements like Fe and Mn in groundwater (see Table 3.2) may relate their association with As as geogenic (Baviskar et al. 2015). Lower pH (6.35) values of groundwater due to the presence of large anionic concentrations (see Table 3.2) and microbial decomposition of organic matter can create possible reducing conditions favorable for desorption and release of As from its geogenic sources (Lawson et al. 2016; Yu et al. 2018a).

3.2 *Sediment Geochemistry*

Variable concentrations of organic content, carbonate content, moisture content, and specific surface area were observed along different depths obtained from the borehole (see Fig. 3.2). Measurable organic content was found in the fine sand fractions, with concentrations ranging between 1.49% and 4.32%. Carbonate content ranged between 1.59% and 4.9%. Since the study area is subjected to active fluvial deposition, the moisture content of the sediments was relatively high (Mukherjee et al. 2012). Geochemical results from the total digestion of sediment core samples



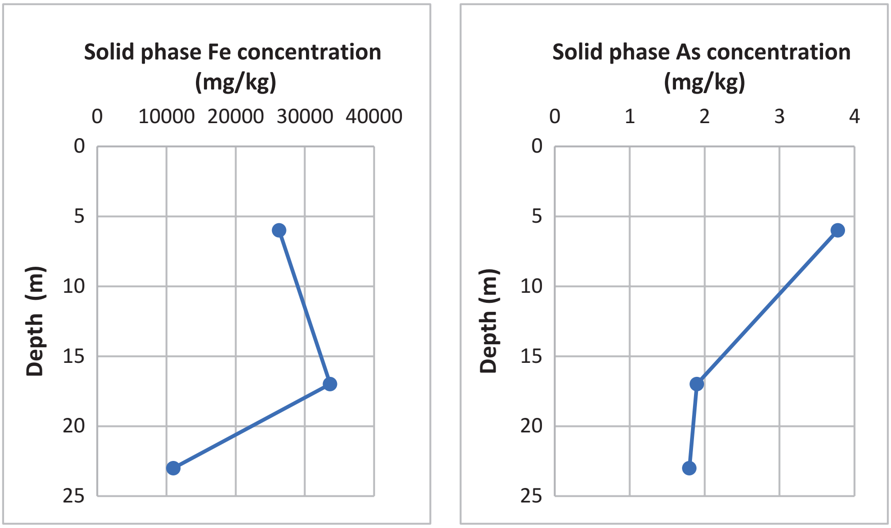
(a) (b)

(c) (d)

Fig. 3.2 Variation of organic content, carbonate content, moisture content, and specific surface area along the depth

are shown in Fig. 3.3, which shows variations in concentrations of the two different elements. Fe concentrations ranged between 26,302.65 and 11,010.47 mg/kg and arsenic concentrations in sediments ranged between 1.79 and 3.77 mg/kg.

The presence of organic content and carbonate content found along the soil depth (see Fig. 3.2) may be because of biological decomposition of organic matter, which may be an additional factor for generating reducing conditions in the soil (Lawson et al. 2016; Yu et al. 2018a). The higher amount of moisture content of soil could



(a) (b)

Fig. 3.3 Variation of Fe and As along depth

cause easy migration for the trace elements into groundwater. Figure 3.2 shows that the soil samples obtained along the depth had a higher specific surface area. Thus, As associated possibly with Fe (Mn and Al) oxide/oxyhydroxides sorbed on these finer soil particles may get desorbed in reduction conditions (Bhattacharya et al. 2013).

The observed concentrations of As in sediments are nearly within the natural range (Smedley and Kinniburgh 2002). The higher concentrations of Fe present along the soil depth as seen in Fig. 3.3 could help us to consider the presence of Fe–oxyhydroxides which may act as a sorbing surface for As (Acharyya 2002).

3.3 Sequential Extraction

Sequential extraction analysis reveals that Mn- and Fe-oxyhydroxides are the major leachable As solid phases (see Fig. 3.4). The relative percentile quantification of iron showed crystalline phase (46.00%) followed by Fe–Mn oxides (22.00%), surface and carbonate bound (20.00%), with minimum (13.00%) Fe bound to organics and sulfides (see Fig. 3.4a). The relative percentile measurment of arsenic showed that it was bound to the crystalline phase (89.00%) followed by Fe–Mn oxides (6.00%) then surface and carbonate (5.00%), with a small fraction (2.00%) bound to organics and sulfides (see Fig. 3.4b). As present in the residual phase at all depths was bound to the crystalline phase. Sequential extraction studies performed in the Bengal Delta plain have shown higher As present in the oxyhydroxide phases of Fe

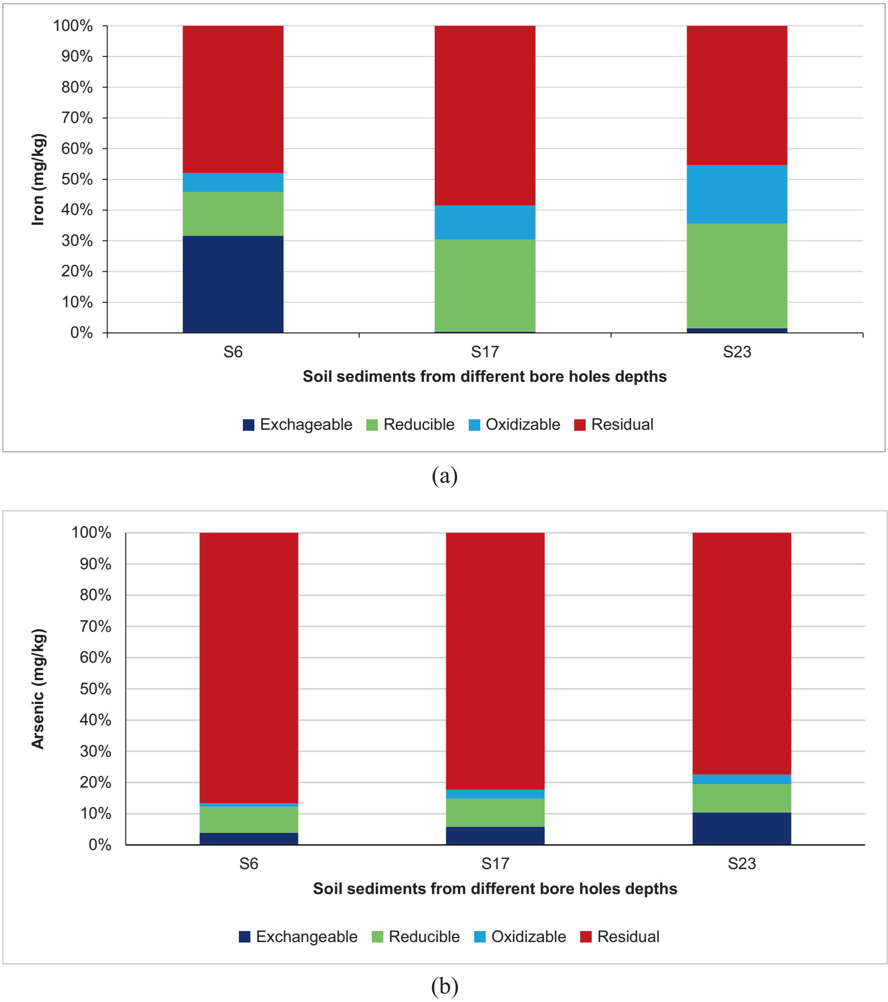


Fig. 3.4 Sequential extraction of borehole sediments at variable depths. (**a**) iron and (**b**) arsenic

(Reza et al. 2010). The study area being predominated by aluminosilicates (clay minerals) has been highlighted by the SEM/EDX and XRD analysis (Fig. 3.5); the dominance of the residual phases can be justified since As and Fe are likely to be bounded significantly to the mineral structures of the silicate minerals. These minerals being relatively stable, compared to pH and redox-sensitive Fe–oxide/oxyhydroxides, are thus not expected to be sources of As to groundwater in our study area.

Sequential extraction data demonstrate that the highest proportion of the total extractable As is primarily associated with amorphous and crystalline Fe (Mn and Al) oxide/oxyhydroxides (see Fig. 3.4). Comparing to previous studies which are consistent with the observed results demonstrates that Fe(III) oxides/oxyhydroxides

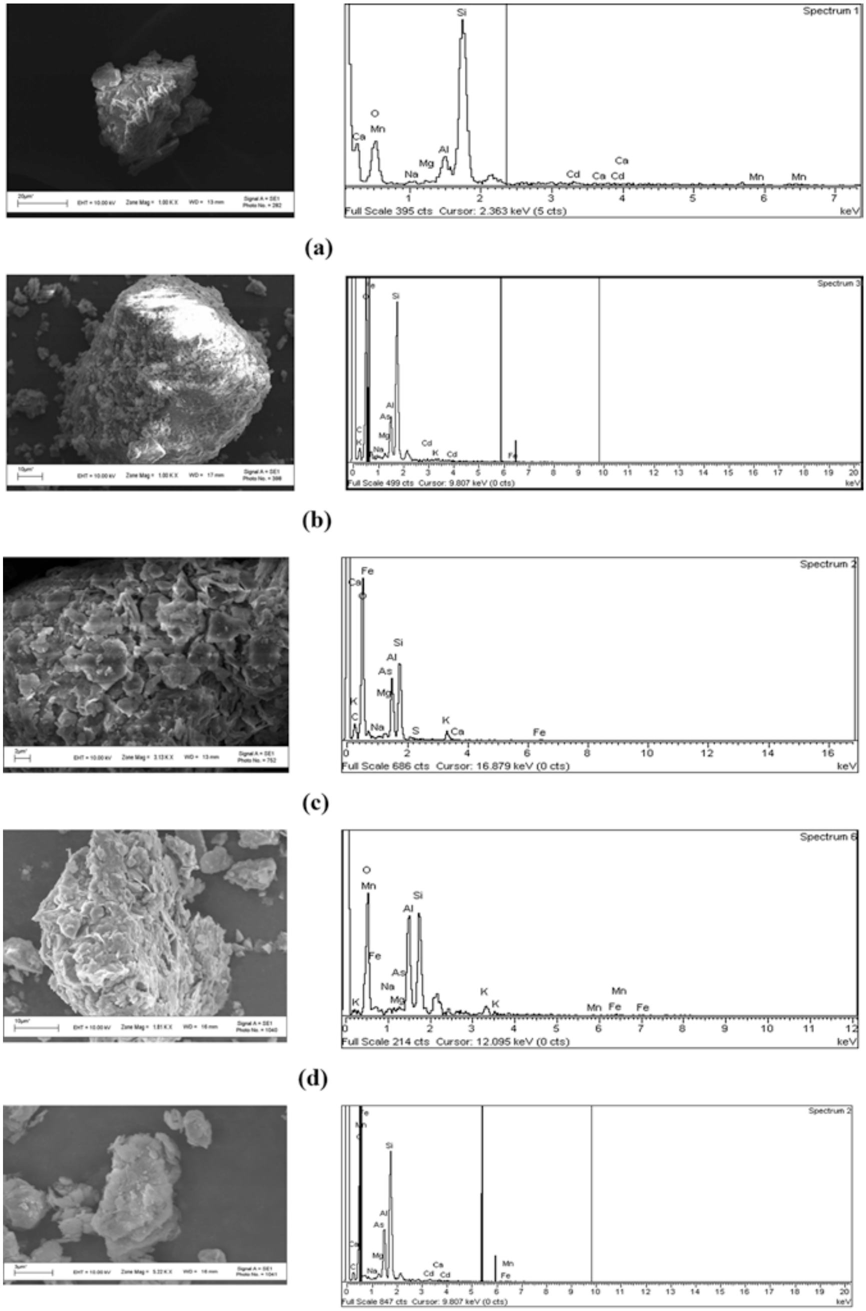


Fig. 3.5 SEM and EDX of aquifer sediments at 23 m depth. (**a**) SEM and EXD of Untreated S23 soil sediments; (**b**) SEM and EDX of Exchangeable S23 soil sediments; (**c**) SEM and EDX of Reducible S23 soil sediments; (**d**) SEM and EDX of Oxidizable S23 soil sediments; (**e**) SEM and EDX of Residual S23 soil sediments

are important hosts for arsenic (Haque et al. 2008). Arsenic commonly gets associated with Fe(III) oxide/oxyhydroxides in aquifer sediments either by coprecipitation or by adsorption (Manning and Goldberg 1996, 1997). Under reduced redox conditions, these associated minerals are released to groundwater (Haque et al. 2008). Specifically, the microbial-mediated reductive dissolution of Fe(III) oxides/oxyhydroxides which can release As into solution is hypothesized to be the primary source of arsenic in groundwater (Acharyya 2002).

3.4 Aquifer Sediment Mineralogy

The SEM/EDX and XRD analyses of the sediment samples from that particular depth were carried out in order to understand how the different mediums like the exchangeable, reducible, oxidizable, and residual fractions might have affected the sediment morphology and mineralogy. A higher percentage of Fe is observed at 23.00 m depth (sample S23) during sequential analysis for As in the sediment sample as shown in Fig. 3.5. EDX spectra further exhibited peaks for Cr in the oxidizable and residual phases of the sediment sample. The SEM/EDX analysis profiles of the aquifer sediments shows that the mineralogy was dominated by aluminosilicates of Na, K, and Mg as well as that of Fe which were obtained from the depth of 23.00 m.

The XRD analysis profiles of the sediment samples confirmed the presence of phyllosilicates (17-20-30), quartz (26.95, 26.85, and 27), NaCl (halite) (56), $CaSO_4$ (gypsum) (11.7, 46), $CaCO_3$ (calcite) (46), feldspar (20–30, 27.5), Fe_2O_3 (hematite) (24.183), MgO (42.91), Al_2O_3 (45.86), AsS, and clay minerals (illite), with the bulk proportion being dominated by quartz, phyllosilicates, feldspars, and clay minerals (see Fig. 3.6). The amplitudes of the mineral peaks gradually diminished from the untreated soil to the residual step. Several minerals were sources of iron, manganese, and arsenic in the aquifer sediments.

The EDX results acquired during sequential extraction steps indicated the presence of Na, K, Mg, and Fe bounded with silicates which could serve as adsorbing surface for As (Scott Fendorf et al. 1997). Peaks of Cr in all the steps of sequential extraction were visible in EDX graphs, but as EDX results are not sediment specific, which are insufficient results to consider Cr to be generally spread all over the aquifer. The presence of Cr in the aquifer sediments could possibly be due to the diagenetic processes involving iron oxides and aluminosilicate, which appears to be the most vital factor controlling the behavior of As and Cr in sediments (Sompongchaiyakul and Sirinawin 2004). The SEM and EDX analysis results further revealed that the magnitude of agglomeration decreased from the first to the last step, and the EDX results supported the SEM grain mineralogy, where the amplitude of the occurrence of the peaks representing respective elements decreased, suggesting a reduction of their relative concentration in the corresponding phases. The morphology of the different sequential extraction phases showed that the state of agglomeration decreased from the untreated sediment followed by the exchange-

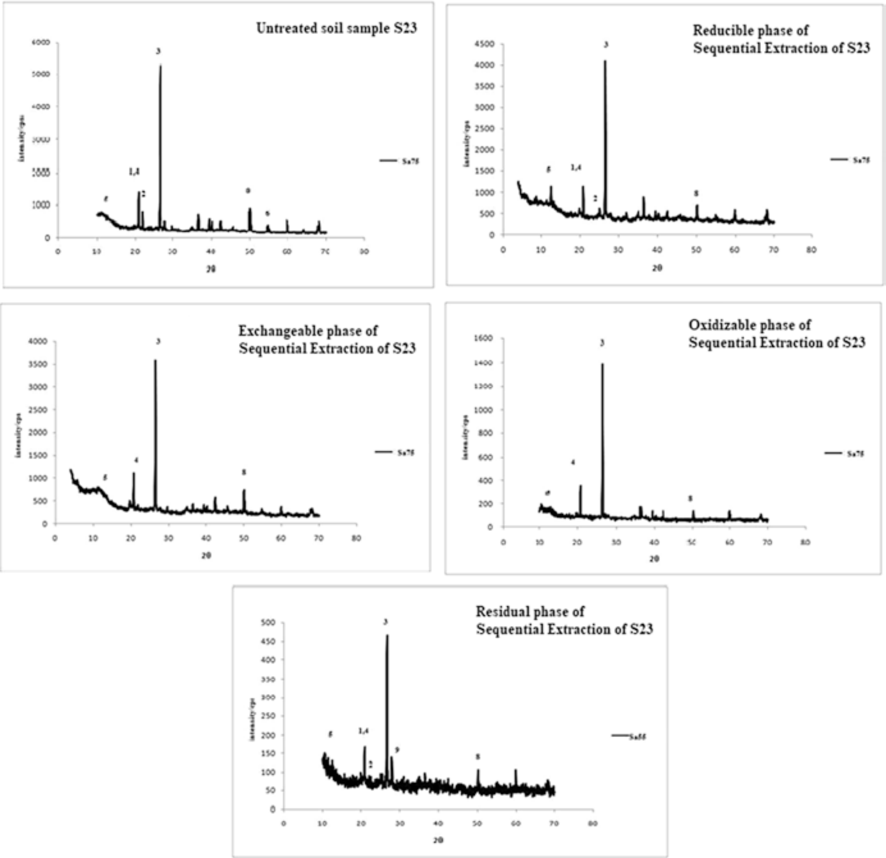


Fig. 3.6 XRD of aquifer sediments at 23 m depth

able phase to the residual state. Agglomeration of the sediment samples rendered different textures in the different phases indicating release of adsorbed elements into the groundwater when these sediments come in contact with changed chemical environment. Textural change of the sediment surfaces on which adsorption of Fe, Mn, and As may take place is also pointed out by the SEM and EDX analysis of sediment samples subjected to sequential extractions (see Fig. 3.5). Results of the SEM analysis also indicated that the sediment grains were moderately rounded to well rounded, suggesting that the sediments must have been transported over distance before deposition and the source of these sediments are not local, hinting that the Himalayas could be a possible provenance (Baviskar et al. 2015).

Results of the XRD analysis were in compliance with the sequential extraction, as the XRD analysis showed the presence of quartz in high amount, and the residual phase was mostly in crystalline phase (see Fig. 3.6). The lithology confirmed that the fine sediments composed of iron-bearing minerals and clay minerals including illite, as indicated by the XRD analysis, could be possible sources of As.

Geochemical studies using sequential extraction results and scanning electron microscopy on our sediment samples demonstrate that Fe oxyhydroxides and Fe oxyhydroxide-coated quartz and feldspar are the dominant carriers of As in the sediments. Results of sediment and groundwater analyses support the mechanism of reductive dissolution of iron hydroxides and the concomitant release of sorbed or coprecipitated As into the groundwater. This mechanism can explain As concentrations in the groundwater up to 0.44 mg/L in the proximity of the bore well.

4 Conclusion

In this research, we investigated the chemical characteristics and nature of As released into the groundwater from aquifer sediments of Brahmaputra alluvial plain. The study improved our understanding of the solid and dissolved arsenic fate in the aquifer under study. The arsenic concentrations in the groundwater samples were above the WHO guidelines of 0.01 mg/L and ranged between 0.05 and 0.44 mg/L. In the target aquifers, low concentration of NO_3^- and SO_4^{2-} coupled with a high concentration of HCO_3^-, which provided strong reducing conditions, was observed. Results of the sequential extraction and the presence of Fe oxyhydroxide phases as indicated by the SEM/EDX and XRD analysis further support the reductive dissolution of Fe oxyhydroxides as the potential mechanism of arsenic release in groundwater. With no major industrial activities in the study area, arsenic contamination in the region is indicated to be of geogenic origin. Thus, the arsenic-rich phases in the aquifer remain the only likely source of arsenic in groundwater. More studies on source and processes of arsenic release and mobilization are required to better understand the arsenic contamination of groundwater in the region.

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Chapter 4
Sources of Arsenic Exposure in Well-Nourished Children

Torsak Tippairote, Weeraya Karnpanit, and Dunyaporn Trachootham

Abstract Arsenic contamination is a global burden and arsenicosis is mostly found in poor children living in highly exposed area. However, recent evidences suggest that continuous low-level exposure of arsenic can also link with some health hazards. Therefore, this book chapter will describe the potential exposure sources of arsenic in the context of healthy, well-nourished children who lived in non-polluted area. Prevalence, biomarkers and factors affecting arsenic exposure in well-nourished children will be described based on current scientific evidences. Sources of As exposure in well-nourished children including environmental, dietary sources and the relationship between both sources of arsenic will be discussed. Reference level/ safety limit of exposure from multiple sources of arsenic will be summarized. Methods of exposure assessment will be described. Furthermore, several perspectives for future research will be addressed such as the need for establishment of cut-off level to categorize high As exposure group, data collection method for estimation of dietary exposure to As, identification of underrepresented sources of As in well-nourished children and strategies to raise awareness and create sustainable preventive measures of arsenic contamination.

1 Introduction

Arsenic contamination is widespread. Toxic levels of arsenic are well documented in natural water sources, soils, food crops, air, household environment, foods and consumer products. Arsenic is detected even in cosmetic products such as eye shadow, vaccines and house dust (Sainio et al. 2000; Hepp et al. 2014). Thus, human exposure to arsenic is almost inevitable. Based on exposure prevalence and potential impact on human health, the Agency for Toxic Substances and Disease Registry

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(ATSDR) of the Unites States of America (US) ranked arsenic as the first in the hazardous substance priority list, (ATSDR 2017). Many studies showed detectable arsenic level in various human biological tissues such as peripheral blood, cord blood, red blood cells, plasma, serum and urine (May et al. 1986; Sakai et al. 2017; Minichilli et al. 2018).

Among the possible arsenic exposure routes, contaminated groundwater is the known biggest threat to public health (WHO 2018). People who lived in the polluted area are continuously exposed to arsenic via drinking and cooking water, irrigation, the backyard soil, air and local agricultural products (Arcega-Cabrera et al. 2017). The World Health Organization (WHO), European Union (EU), and United States Environmental Protection Agency (US EPA) set the drinking water arsenic level standard at 10 μg/L (Council 1999; ATSDR 2007; WHO 2018). In 2006, seventy countries were reported to have arsenic level in drinking water above the WHO standard (Chakraborti et al. 2018). While moderate to high concentrations of arsenic in groundwater were found in 107 countries (Chakraborti et al. 2018). Approximately 296 million individuals worldwide are at risk of arsenic toxicity (Chakraborti et al. 2018). For example, a survey of municipal water supplies by the Kolkata Municipal Corporation (KMC) in India concluded that, on average, a person in the communities consumed 0.95 mg arsenic per kilogram of body weight per day (per kg bw/day) with an estimated cancer risk of 1.425×10^{-3} (Chakraborti et al. 2018).

In areas with standard complied water quality (arsenic level lower than WHO standard), people are still possibly exposed to arsenic via other different routes such as food products, wood preservatives, cigarette smoking or air pollution (Edlich et al. 2005; Hojsak et al. 2015; Molin et al. 2015; González-Castanedo et al. 2015; Hays et al. 2006). For example, a recent study in South Australia reported 53% of rice-based food samples from supermarket contain arsenic exceeding EU recommended value (100 μg/kg) for young children (Chakraborti et al. 2018). Furthermore, emerging studies reported growing concerns on cumulative low-level exposure (<10 μg/L) and potentially health risk. For example, detrimental effects on cognitive status of adults and elders were associated with long term residents in two counties of Texas, United States, where the average arsenic ground water levels were 3.0 and 7.4 μg/L, respectively (O'Bryant et al. 2011). The impacts of chronic exposure to low-level arsenic include neurodevelopmental defects, immune-maturation and others adverse events associated with alteration in genetic and epigenetic expression (Reichard et al. 2007; O'Bryant et al. 2011; Schmidt 2014). Importantly, younger children are more vulnerable than older children and adults due to their distinctive high susceptibility to toxins (ATSDR 2007).

To increase awareness about the potential health impacts of chronic exposure to low-level of arsenic, here we review the potential exposure sources of arsenic in the context of healthy and well-nourished children who lived in non-polluted area.

2 Prevalence of Arsenic Exposure in Well-Nourished Children

2.1 Biomarkers of Arsenic Exposure

To quantify the absorbed arsenic dosage in human tissues, there are a number of biomarkers to be considered. Total arsenic level in blood, urine, hair and nail samples have been used and studied as the biomarkers of exposure (Hughes 2006). Total urinary arsenic is the most common biomarker of exposure for inorganic arsenic (iAs) (Hughes 2006). Studies had already shown association between environmental arsenic levels to the levels in these tissue samples. For examples, level of arsenic contamination in water sources and daily intake levels were associated with detected arsenic levels in blood, urine and hair samples (Valentine et al. 1979; Kurttio et al. 1998). Arsenic concentrations in the air were also shown to be associated with measured arsenic levels in fingernails (Agahian et al. 1990). However, the arsenic level in these biological tissues carry their inherited differences and the measured level may be confounded by consumption of seafood containing high concentrations of organic arsenic (Karagas et al. 2000). Understandings of their unique sources of exposure are needed to accurately interpret the exposure level from different tissue samples.

In blood samples, total arsenic levels represent only short-term exposure due to its rapid clearance from the blood within a few hours after oral ingestion (Tam et al. 1979; Vahter et al. 1983). Therefore, the role of blood arsenic level as the biomarker of ongoing exposure in healthy unexposed children is quite limited. The possible application is when a physician needs to confirm the recent history of patient's exposure and acute clinical presentations with possible arsenic poisoning (True 2002).

In animal models, 75–98% of initial arsenic dosage were continuously excreted via urine for up to 3 days following single intravenous injection (Vahter et al. 1983). Hence, urine arsenic level can be used as an indicator of longer exposure duration than blood level. In a study, urinary arsenic levels were shown to be inversely correlated to the distance of children's residence from the site of copper smelter stack (Milham and Strong 1974). While the arsenic levels in environmental samples were steadily decreased with the distance, the urinary levels were constant at background level after one-half mile beyond the smelter site (Polissar et al. 1990). Therefore, total urinary arsenic level is apparently not the ideal indicator for ongoing low concentration exposure in non-exposed population. The total arsenic level can also be confounded by exposure to less toxic organic arsenic from consumption of seafood, especially seaweed (Hughes 2006). However, one can choose to measure urinary inorganic arsenic and its methylated metabolites, specifically monomethylarsonic acid (MMA), to exclusively estimate inorganic arsenic exposure (Yamauchi et al. 1989; Council 1999; Aylward et al. 2014). Inspite of its relatively short-term exposure indicator, urine arsenic level is generally used as the biomarker for conducting studies on people who lived or worked near polluted areas (Milham and Strong

1974; Pinto et al. 1976; Enterline et al. 1987; Polissar et al. 1990; Minichilli et al. 2018). A recent study showed high urinary porphyrins in arsenic exposed group; thereby, proposed urinary porphyrins as an early warning biomarker for chronic arsenic exposure especially in younger (less than 20 years old) population (Wang et al. 2002). Nevertheless, for the healthy unexposed children, urine arsenic level is still not the optimal biomarker of exposure.

In response to growing issues of chronic low-level arsenic exposure to children health, arsenic level in hair and nail samples seems to be superior biomarker to those in blood and urine samples. Many studies showed that the arsenic levels in hair and nail samples represented the long-term exposure ranging from 1 to 12 months earlier (Valentine et al. 1979; Bencko et al. 1986; Choucair and Ajax 1988; Yamauchi et al. 1989; Agahian et al. 1990; ATSDR 2007). In addition, the simplicity and non-invasive nature of sampling hair and nail make them more appealing choices in specific conditions such as repeated wide population studies or in young children group. However, subpopulation reference levels and quality control analysis are needed to ensure minimal inter- and intra-laboratory variations and accurate interpretation of results (Puchyr et al. 1998; Kempson and Lombi 2011; Namkoong et al. 2013; Wolowiec et al. 2013). To date, the most concerning issues for analysis of element in hair and nail samples are external incorporation of the elements into hair and nail. These contamination can be minimized by standardized sample collection, handling, washing and processing methods (Puchyr et al. 1998).

In general, urine arsenic levels are preferred for estimation of exposure level in individuals who lived near a polluted area (Milham and Strong 1974). Blood arsenic level is reserved for determination of acute intoxication in clinical settings such as unintentional or intentional ingestion of arsenic containing medications (ATSDR 2007). Furthermore, skin hyperpigmentation and dermatologic lesions such as palmoplantar hyperkeratosis have been proposed as a long-term biomarker of cumulative arsenic exposure (Puchyr et al. 1998). Nonetheless, in children who lived in areas with no documented source of arsenic contamination together with concerns of cumulative ongoing exposure, hair and nail arsenic levels may be the suitable biomarkers for exposure assessment.

2.2 Prevalence of Arsenic Exposure in Well-Nourished Children

According to the data of U.S. National Health and Nutrition Examination Survey (NHANES), the average urinary total arsenic levels of 6–11-year-old children gradually decreased from 8.30 μg/L in 2003 to 5.21 μg/L in 2014 (Puchyr et al. 1998). However, the average levels in high exposure groups, represented by 95th percentile rank, were not following such decreasing trend. These levels ranged from 29 to 53 μg/L, which were 5–9 times of the average levels in the corresponding survey cycles. However, the reported levels in high exposure group were still not exceeding the ATSDR's upper limit level of 100 µg/L in unexposed individuals (ATSDR 2007). The findings suggested that the national arsenic exposure control measures were effective to decrease the exposure levels in general population. However, certain groups of children were still vulnerable to potentially high exposure level.

A 2018 study from Thailand reported 36% prevalence rate of high arsenic level in hair samples of 3–7-year-old children who lived in urban area with no known arsenic polluted sites in surrounding area. Most of the children were well-nourished according to WHO BMI-for-age Z-score nutritional status categorization (Tippairote et al. 2018). The average hair arsenic level in these children was 0.18 µg/gm, while the average level in high exposure group and the maximal level were three and ten times higher than this level, respectively (Tippairote et al. 2016, 2018). Two separate studies in Russian 3–4-year-old and 2–9-year-old healthy children showed average hair arsenic levels at 0.08 and 0.03 µg/gm, respectively (Skalny et al. 2017a, b). Healthy unexposed children from Brazil, U.K, German, France, Czech Republic, and Poland had the average urinary total arsenic level at the range of 3.60–6.73 µg/L as compared to 5.21–8.30 µg/L in U.S. children from previously mentioned report (Trepka et al. 1996; White and Sabbioni 1998; de Burbure et al. 2006). Among these studies in healthy children, there were certain variations in arsenic exposure level depending on the area of studies and degree of local environmental arsenic contamination. Alarmingly, the number of healthy children with high arsenic levels seems to be higher than expected. In the Thai study, more than one out of three healthy children had high hair arsenic levels (Tippairote et al. 2018). Long term impacts of accumulated arsenic exposure on healthy development of these children warrant further investigation.

2.3 Factors Affecting Arsenic Exposure in Well-Nourished Children

Children are more vulnerable to low-level arsenic exposure than adults (Chiba and Masironi 1992). For example, the relatively low arsenic concentration in tobacco did not affect the urinary total arsenic level in cigarette smoking adults, while the urinary levels in their children were proportionally associated to number of smoking parents living in the same houses (Chiba and Masironi 1992). Younger ages are more affected because of their innate lower tolerance to arsenic exposure due to various physiological and behavioral factors. These include the lower body weight in relation to the exposure dosage, larger skin surface in proportion to children body volume, immature absorption and excretion processes, immature hepatic biotransformation enzymes and others (ATSDR 2007).

Children behaviors also affect their arsenic exposure levels. Comparing to adult, children spent more time outdoor, stay closer to the contaminated ground, less likely to discriminate hazardous or unhygienic items and sometimes eat inappropriate things (NRC 2001). Hand-to-mouth behaviors in children are common and probably are their main exposure determinants (NRC 2001). Other family, socioeconomic, lifestyle and environmental factors also determine children arsenic exposure. Family socio-economic constraints can make them less likely to get access to good quality toys with less arsenic contamination (NRC 2001). Outdated copper arsenate treated wooden playground may still exist in their neighborhood. Family traditional beliefs and medical practices may possibly expose children to arsenic containing herbs or medications in contrast to modern healthcare options with minimal exposure to toxic substances including arsenic (ATSDR 2007). At the other end, children in high income family may be exposed to arsenic due to frequent access to high arsenic containing seafood meal or seaweed snacks (Awata et al. 2017). Our previous study showed that children who lived in the big city had higher level of arsenic in hair sample than those who lived in surrounding area (Tippairote et al. 2018). Up to certain level, the cumulative arsenic exposure from different sources may eventually exceed the children's tolerance levels to toxin. This may explain the high arsenic levels found in tissue samples of well-nourished children with little or no history of exposure to arsenic.

It is worth noting that arsenic metabolism in children may differ from that of adults (Skröder Löveborn et al. 2016). For example, a previous study in Bangladesh found higher percentage of dimethylarsinic acid (DMA) and lower percentage of iAs in urine of 9-years-old children, compared to their mothers at pregnancy (Skröder Löveborn et al. 2016). Efficiency of arsenic methylation in children was strongly associated with plasma selenium and folate status (Chiba and Masironi 1992). Therefore, the level of arsenic in biological specimens may also be influenced by adequacy of these nutrients.

3 Sources of Arsenic Exposure in Well-Nourished Children

Arsenic is a ubiquitous element. Therefore, exposure to arsenic arising from both natural processes and human activities is unavoidable. Children may become exposed to various arsenic forms (inorganic and organic) depending on geochemical occurrence and the levels of human activities in their living area. Arsenic exposure can occur via ingestion, inhalation and dermal absorption (GomezCaminero et al. 2001; WHO 2011a). The primary routes of arsenic exposure are via ingestion of arsenic contaminated food and drinking water as well as accidental soil ingestion in young children (GomezCaminero et al. 2001; WHO 2011a; De Miguel et al. 2017; Cubadda et al. 2017; ATSDR 2007). Arsenic exposure through inhalation can occur in children living in areas with high arsenic-containing air such as mining and smelting sites and industrial areas. Dermal exposure happens rarely (ATSDR 2007; Hong et al. 2014).

3.1 Environmental Sources

Arsenic exists mostly in bound form. Binding to carbon, iron, oxygen, and sulfur results in a variety of inorganic and organic arsenicals in various oxidation states. Environmental arsenic comes from natural, anthropogenic and use of arsenic containing products (Hughes 2006).

3.1.1 Natural Sources

Primarily inorganic arsenic is ubiquitously found in soil, air, and water. There are over 200 mineral species containing arsenic. The background levels of arsenic in soil range from 1 to 40 mg/kg (WHO 2001). Children may get exposed to contaminated soil via touching, eating, breathing and eye contact. Soil microorganisms and volcanic activity release arsenic into the air (Hughes 2006). Arsenic air levels vary across geographical area around the world. Lower levels of air arsenic were found in rural areas (0.007–28 ng/m^3), while higher levels of air arsenic were observed in urban areas (3–200 ng/m^3) (WHO 2001). Therefore, well-nourished children living in a city may actually inhale higher level of arsenic. Since water can dissolve mineral species and release arsenic, inorganic arsenic (iAs) was found in ground water, seawater and fresh water (Hughes 2006). The range of water from sea and river was 1 to 10 μg/L (WHO 2001) while arsenic in ground water could be 1000 times greater than those ranges (Nordstrom 2002). Chakrabarti et al. wrote that about 107 countries have reported moderate to higher concentrations of arsenic in their groundwater. There were 296 million individuals potentially at risk of arsenicosis due to the consumption of arsenic through drinking, cooking, and irrigation water and food materials. Almost 59% of the 56 Asian countries have reported groundwater arsenic contamination issues (Chakraborti et al. 2018).

3.1.2 Anthropogenic Sources

Nonferrous metal smelters and coal-burning energy plants are major anthropogenic sources of iAs. These processes contaminate air, water, and soil with iAs. Arsenic in air at a copper smelter in Tacoma, Washington, USA exceeded 1000 μg/m^3 during certain periods of its operation (Enterline et al. 1987). Furthermore, manufacturing of arsenical pesticides and the improper operation of metal mining may contaminate the environment with iAs (Hughes 2006). Therefore, inhalation of arsenic contaminated air is commonly found in people working in or living near nonferrous smelters, coal burning industry and pesticide manufacturing plants. And they are generally considered as high-risk group of arsenic toxicity. In fact, a recent study in Chile reported that air As in the work-site of copper smelting were ranging from 0.0012 to 0.092 mg/m^3 (Schultze and Pastene 2018). These numbers exceeded the threshold limit of 0.01 mg/m^3, established by both American Conference of Governmental

Industrial Hygienists (ACGIH) and Occupational Safety and Health Administration (OSHA) (ACGIH 2003; OSHA 2017). Consistently, the urine sample of all workers in the copper smelting area during 2007–2010 contained high As exceeding the permissible exposure limit of 35 μg/g creatinine (Schultze and Pastene 2018).

3.1.3 Product Uses

There are a variety of commercial, medical, veterinary and pesticide products containing arsenic. For example, semiconductors, glass, toys, preservatives, pesticide, growth promoter in farm animals and chemotherapeutic agents could be source of arsenic (Hughes 2006; WHO 2001). Well-nourished children living in city may be exposed to arsenic through uses of such products. Recently, wood preservatives containing chromated copper arsenate (CCA) has been found to be a potential source of arsenic exposure in well-nourished children living in residential area. CCA-treated wood is found in many residences and playgrounds in the United States. A study found five-fold increase of water-soluble arsenic on the hands of children who played on playgrounds built with CCA-treated wood compared to those without CCA-treated wood (Kwon et al. 2004). The concern is raised when children use their hands to eat food. In 2003, the use of CCA-treated wood in homes and playgrounds was voluntarily withdrawn owing to the concern about exposure to children (Hughes 2006). In conclusion, the major sources of environmental arsenic exposure in well-nourished children are air inhalation in urban area and direct/indirect contact to the arsenic containing products. Nevertheless, the absorption of iAs after inhalation or dermal exposure is less than after oral exposure (NRC 1999; WHO 2001). Thus, dietary sources of arsenic exposure may be more important than other routes.

3.1.4 Dietary Sources

The most important source of non-occupational human exposure to arsenic is diet. In the United States, the estimated daily intake of arsenic ranges from 2 to 92 μg/day (Tao and Bolger 1999). Overall, organic arsenic is the major form of dietary arsenic. Organic arsenic (e.g. arsenobetaine) is mostly found in seafood such as shell fish (1.1 ppm to 30 parts-per-million (ppm), prawn and shrimp (42–174 ppm) (WHO 2001). In contrast, levels of inorganic arsenic (iAs) are highest in grains, fruits and vegetables. iAs contributes to 17–24% of total dietary arsenic (Schoof et al. 1999).

Drinking Water

The most important dietary medium of arsenic exposure is drinking water (Hughes 2006). Exposure to arsenic particularly inorganic forms via drinking water is an important public health issue worldwide (Ng et al. 2003; Naujokas et al. 2013;

Singh et al. 2015b). In groundwater, inorganic arsenic (iAs) namely arsenate [As(V)] and arsenite [As (III)] are the dominant forms of Arsenic (WHO 2011a; Baig et al. 2016). Low level (less than 0.3 μg/L) of organic arsenic such as methylated arsenicals can be found in natural water (WHO 2011a). In general, arsenic concentrations in drinking waters are low; however, there are some areas where people are exposed to drinking water contaminated with high arsenic levels. If water with arsenic levels exceed 10 μg/L is used for drinking and cooking, it will be the major inorganic arsenic exposure source (FAO/WHO 2011). Approximately 200 million people worldwide are affected from arsenic-containing drinking water, and around 31 million people in South Asian countries consume drinking water with extremely high level of arsenic (greater than 50 μg/L) (Kordas et al. 2016; Rahman et al. 2009). Interestingly, a study in Pakistan reported high arsenic concentrations (up to 98 μg/L) of groundwater from hand pump machines in several districts, while the arsenic in municipal treated water samples were not above the WHO recommended level of 10 μg/L (Baig et al. 2016). Risk assessment of drinking water in Pakistan children aged 6–15 years old showed that young children in certain districts of Pakistan had high risk of carcinogenic and non-carcinogenic chronic arsenic toxicity (Baig et al. 2016). Likewise, another study in Patna district, in Bihar state in India, reported an estimated high cancer risk of 19 per 1000 and 87 per 1000 for 5–10-year-old children living in Rampur Diara (RD) and Haldichapra (HC) area, respectively (Singh and Ghosh 2012). Such high-risk estimation was based on consumption of drinking water contaminated with 231 μg/L and 52 μg/L in RD and HC, respectively. These numbers exceeded both WHO guideline of 10 μg/L and the Bureau of Indian Standards (BIS) standard of 50 μg/L (Singh and Ghosh 2012).

Several studies showed that continuous exposure of arsenic via drinking water increased arsenic levels in biological specimens. A study in Bangladesh showed high levels of arsenic in hair samples (0.33–3.29 μg/g (ppm)) of children under 12 years (Uddin et al. 2006). The average level was higher than maximum acceptable level of 1 ppm (Uddin et al. 2006). The study concluded that such high levels resulted from longtime intake of arsenic-containing groundwater (Uddin et al. 2006). Interestingly, a study by Kurttio et al. concluded that an increase of 10 μg/L of arsenic in drinking water led to an increase of 0.1 μg/g arsenic in hair (Kurttio et al. 1998). Likewise, another study by Karagas et al. found strong correlation between arsenic in drinking water (greater than 1 μg/L) and in toenails ($r = 0.83$, $p = 0.0001$) (Ab Razak et al. 2015). A ten-fold increase of arsenic in well water resulted in a two-fold increase of arsenic in toenails (Karagas et al. 1996).

For well-nourished children living in areas with no geological or anthropogenic contamination of arsenic, low levels of arsenic were found in groundwater and drinking water. Therefore, those children may have low exposure of arsenic from drinking water (Kordas et al. 2016; EFSA 2014). A study in urban city of India showed that 82% of tap water samples and 100% of filtered water samples had arsenic within the accepted limit (0.01 mg/L) (Singh et al. 2015a). Therefore, filtering water should be recommended. Consistently, another study in an urban part of Iran found that water treatment facility played an important role in reducing arsenic concentration in drinking water from 39 μg/L to 9 μg/L (Mosaferi et al. 2014).

Food

Food is one of the significant sources of arsenic exposure in general population including children (WHO 2001; FAO/WHO 2011; Mosaferi et al. 2014). Arsenic contents in foods from different countries vary significantly depending on many factors such as the food type, cultivating conditions (soil, water, pesticides, and fertilizers) and geochemical activity. High arsenic levels are found in fish and shellfish, dried seaweed and rice. Lower contents of arsenic are detected in cereals, vegetables, fruits, and dairy products (GomezCaminero et al. 2001; WHO 2011a; FAO/WHO 2011; Sigrist et al. 2016). Fish and shellfish have been reported as major dietary arsenic sources; however, these food commodities mostly contain organic arsenicals especially arsenobetaine which are less toxic form of arsenic compounds. The ratio of inorganic arsenic in fish and shellfish usually does not exceed 10% of the total arsenic content (GomezCaminero et al. 2001; FAO/WHO 2011). Extremely high arsenic concentration, varying between 30 and 130 mg/kg, was reported in dried seaweed, (WHO 2011a). A seaweed species, namely Hijiki (*Hizikia fusiforme*), found in Chinese, Japanese and Korean cuisines contains high level of inorganic arsenic of 67–96 mg/kg (Rose et al. 2007). To prevent high dietary exposure to inorganic arsenic, seaweed should be avoided especially in children diet (Rose et al. 2007).

Agriculture in most Asian countries relies on groundwater for irrigation. Therefore, iAs in contaminated groundwater can be transferred to crops in the food chain. A study in India found 19% of irrigation water samples from 597 irrigation tube wells located in West Bengal, exceeded the FAO standard for irrigation water of 100 μg/L (Chakrabarti et al. 2018). Consistently, 76% of the crops irrigated by such contaminated water contained inorganic arsenic (Chakrabarti et al. 2018). Likewise, another study in Ganga River basin (GRB) area, which covers parts of India, Bangladesh, Nepal, and Tibet, reported hazardous levels of As in irrigation water (about 1000 μg/L) exceeding the FAO standard, and in food materials (up to 3947 μg/kg) exceeding the Chinese Ministry of Health's standard (0.15 mg/kg) for food in South Asia (Chakrabarti et al. 2018). Due to soil and water contact, terrestrial plants particularly rice contains higher levels of inorganic arsenic (both trivalent and pentavalent) than those of other commodities (Sigrist et al. 2016; Rintala et al. 2014; Khan et al. 2010). Rice can intrinsically accumulate high levels of inorganic arsenic compared to other cereals since it is commonly grown under flooded conditions with high arsenic mobility (Munera-Picazo et al. 2015; Zhu et al. 2008; Xu et al. 2008). The ratio of inorganic arsenic to total arsenic in rice varies from 10 to 100% (Williams et al. 2005; FAO/WHO 2011). Recently, rice and rice-based products for baby and young children are of concern as major source of inorganic arsenic exposure if they consumed it on daily basis (Zhu et al. 2008; Meharg et al. 2008; Shibata et al. 2016). Other cereal-based foods such as bread and rolls were reported as the main contributor for arsenic exposure in European children (Mosaferi et al. 2014). Although most milk and dairy products contain undetectable or low levels of total arsenic and inorganic arsenic, high consumption these products in some countries makes them major contributors to inorganic arsenic dietary exposure of infants and toddlers (FAO/WHO 2011; Mosaferi et al. 2014).

3.2 Relationship Between Environmental and Dietary Sources

Compared to children living near smelter, coal-burning energy plants or pesticide manufacturing factories, well-nourished children living in urban area may have less chance of direct arsenic exposure. However, migration of arsenic from environment along food chain is possible. Consumption of such arsenic contaminated diet may pose hidden health risk. Consumption of animal produces grown by contaminated drinking water, milk, and feeds may be considered as alternative source of arsenic exposure in well-nourished children. In fact, a recent study in West Bengal in India showed that egg, all organs of poultry, cows and milk in arsenic endemic area retains significantly high concentration of arsenic (Datta et al. 2012). This is consistent with previous reports of arsenicosis in this endemic region even though they had been supplied with arsenic-free drinking water for over 12 years (Datta et al. 2010; All India Institute of Hygiene and Public Health 1996). Likewise, a study in Bangladesh found that arsenic concentrations in rice straw, husk and grains were higher in areas with high concentration of arsenic in groundwater and soils (Kabir et al. 2016). Thus, growing crops in arsenic contaminated soil or irrigation water could be responsible for accumulation of arsenic in crops. Naturally, vegetables and fruits contain low levels of arsenic, and it was reported that arsenic was not detected in 86% and 83% of vegetables and fruits, respectively. However, high arsenic contents could be found in vegetables grown in arsenic contaminated soil or irrigated with arsenic contaminated water (FAO/WHO 2011; Smith et al. 2006; Rehman et al. 2016). Vegetables grown with arsenic contaminated water collected from Bangladeshi households had unusually high arsenic and inorganic arsenic maximum contents of 2.3 and 1.7 mg/kg, respectively (Smith et al. 2006). Of these, up to 14% was attributable to inorganic arsenic in cooked rice. High levels of arsenic (exceeding 1 mg/kg) in vegetables such as bitter melon and luffa grown with arsenic contaminated water in Pakistan were reported (Rehman et al. 2016). Importantly, a study in an endemic area of chronic arsenicosis, in Bengal Delta in India, reported high amount of As in multiple crops including 194 μg/kg in boro rice and 156 μg/kg in aman rice (Samal et al. 2011). Urine analysis of adults and children in this area consistently showed high level of As (between 154 and 276 μg/L) in urine, with overall As retention of 50–60% (Samal et al. 2011). An *in vivo* study in animals showed that 90% bioavailability of iAs from rice consumption (Juhasz et al. 2006). Furthermore, rice consumption leads to an increased excretion of iAs (Davis et al. 2012). Thus, it is highly likely that indirect exposure of As through consumption of the contaminated crops may induce arsenicosis (Munera-Picazo et al. 2015). Nevertheless, further studies are needed to demonstrate direct relationship between consumption of iAs-contaminated food and risk arsenic-related diseases.

In conclusion, consumption of arsenic contaminated grains and vegetables moves arsenic through food chain (from-farm-to-table). This could be the major sources of arsenic exposure in well-nourished children.

3.3 Reference Level/Safety Limit of Arsenic Exposure

3.3.1 Exposure Source

Water

(a) **Drinking water:** World Health Organization provisional guideline for drinking water quality recommends that iAs in drinking water should not exceed **0.01 mg/L (10 μg/L)** (WHO 2011a, 2017). It is worth noting that some highly contaminated countries such as India, a permissible limit in absence of an alternative source is 0.05 mg/L (50 μg/L) (WHO 2005; WHO India 2018).

(b) **Irrigation water:** The FAO's recommended maximum concentration level of iAs in irrigation water is up to **100 μg iAs/L** (Ayers and Westcot 1994). The maximum concentration is based on a water application rate of 10,000 m^3 per hectare per year, which is consistent with good irrigation practices (Ayers and Westcot 1994).

Food

Since the reduction of permissible inorganic arsenic in drinking water to 10 μg/L the contribution of As exposure through rice and rice-based products became an increasingly important**.** In 2014, Codex Alimentarius Commission has established **a maximum limit of 200 μg iAs/kg in rice** (Codex Committee on Food Additives and Contaminants 2017; Munera-Picazo et al. 2015). Consistently, European Union recommended **100 μg iAs/kg** and 200 μg iAs/kg of inorganic arsenic for **young children** and adult, respectively (Chakrabarti et al. 2018). For countries consuming rice as staple food, the safety standard has been set at lower level. For example, People's Republic of China set the strictest regulation, with a maximum threshold of 150 μg iAs/kg for rice (Heikens 2006; Chakrabarti et al. 2018). For countries without rice as staple food such as Australia, the maximum permissible concentration (MPC) for As in rice is 1 mg/kg (Heikens 2006; Food Standards Australia and New Zealand 2018). Besides rice, safety limit values of iAs for other foods in China are also lower than those of Australia; i.e. 100 μg iAs/kg of fish in China, compared to 2 mg iAs/kg of fish in Australia (Heikens 2006, Food Standards Australia and New Zealand 2018).

Although the permissible level of iAs in water and food had been established, no maximum safe intake values have been set up by any international food safety authority (Munera-Picazo et al. 2015). Previously, the Joint Food Agricultural Organization/World Health Organization (FAO/WHO) Expert Committee on Food

Additives (JECFA) proposed a provisional tolerable weekly intake (PTWI) of arsenic at the level 15 mg/kg bw/week in 1998. However, the PTWI was withdrawn in 2011 (FAO/WHO 2014). The reasons for withdrawal was based on the fact that such PTWI dose was higher than the range of benchmark dose lower confidence limit ($BMDL_{01}$). European Food Safety Authority (EFSA) established a $BMDL_{01}$ of iAs between 0.3 and 8 μg/kg bw/day for an increased risk of bladder, lung and skin cancer (EFSA Panel on Contaminants in the Food Chain 2009). Thus, it is difficult to establish a single value as maximum safe intake of inorganic arsenic in food.

Soil

Environment Agency, United Kingdom established the Soil Guideline Values (SGVs) for inorganic arsenic in soil according to land use; i.e. 32 mg iAs/kg dry weight of soil used in residential area, 43 mg iAs/kg dry weight of soil in allotment (gardening) area and 640 mg iAs/kg dry weight in commercial use (Martin et al. 2009).

Air

Center for Disease Control, National Institute for Occupational Safety and Health, USA set up recommendation standard for occupational exposure of inorganic arsenic in 1973 and revised it in 1975. The latest recommendation stated that no worker should be exposed to a concentration of inorganic arsenic in excess of 0.002 mg (2 μg) per cubic meter of air as determined by a 15 minutes sampling period (U.S. Department of health, education, and welfare, Center for Disease Control, National Institute for Occupational Safety and Health 1975).

3.3.2 Biological Markers

Currently, there were no international standard safety limit established for arsenic level in blood. There are some variations of reference values among countries.

Blood

According to Agency for Toxic Substances and Disease Registry (ATSDR, USA) normal levels of arsenic in low-exposed individuals are less than 1 μg/L in blood samples (ATSDR 2007). While other countries utilized RV95, the 95th percentile of the measured pollutant concentration levels in the relevant matrix of the reference population, as reference values (HBM (Human Biomonitoring) Commission 2016). Since RV95 values indicates the level of background exposure, people with amount of blood arsenic higher than RV_{95}s indicated abnormally high exposure of arsenic (Saravanabhavan et al. 2017). The reference level of total arsenic in blood for

6–19 years old and 20–79 years old Canadian population are 1.4 µg/L and 2.0 µg/L, respectively (Saravanabhavan et al. 2017). In contrast, the reference values of Brazilian and Italian population (18–65 years old) are 9.87 and 5.32 µg/L, respectively (Saravanabhavan et al. 2017). These data suggested that the reference values and background exposure of arsenic are varied among different countries. Since blood arsenic level only indicates acute exposure, RV_{95} values may be useful just for monitoring of potential acute toxicity but not for chronic health hazards.

Urine

ATSDR, USA recommended that normal levels of arsenic in low-exposed individuals should not exceed 100 µg/L in urine sample (ATSDR 2007). The reference values (RV_{95}s) of total arsenic in urine for 3–79 years old Canadian population is 27 µg/L. In contrast, the reference values of German, Belgium and Korean population (18–65 years old) are 15, 48.8 and 106.1 µg/L, respectively. These data suggested that the reference level in each country were varied.

Hair and Nail

ATSDR, USA suggested equal or less than 1 ppm (µg/g) of arsenic in hair or nail samples of a low-exposed person (ATSDR 2007); WHO maximum acceptable level is 1 ppm (1 µg/g) (ATSDR 2007). Some research studies compared the observed level of arsenic in hair with those of the certified value of arsenic in standard reference hair material (Samanta et al. 2004). For example, 0.28 ± 0.04 µg/g was the arsenic value of Standard reference materials (SRM) certified human hair from (GBW07601) from China National Analysis Center for Iron and Steel, Beijing, China (Samanta et al. 2004). The certified values were used to categorize as low or high exposed groups. A systemic review reported a variety of reference ranges for arsenic in hairs found in several studies (Mikulewicz et al. 2013). For example, an Italian study used 0.14–0.24 mg/kg as reference range for 3–5 years old children, while a Korean study used 0.05–0.2 mg/kg as reference range for 3–6 years old children (Mikulewicz et al. 2013). The systemic review pointed out that the reference range may depend on various factors including the sample preparation, analytical methods, gender and age of study populations (Mikulewicz et al. 2013).

3.4 Methods of Dietary Exposure Assessment

3.4.1 Dietary Exposure Assessment

Dietary exposure assessment aims to determine the extent of chemical substance exposure or intake via foods, beverages and drinking water consumption (WHO 1995). The estimated exposure is usually presented as either a **central tendency**

estimate (CTE) which is the mean or median exposure or a **reasonable maximum estimate** (RME) calculating from high-end exposure (e.g. 90th, 95th and 97.5th percentile). However, calculation of RME would be reliable only if the data were obtained from at least 60 human volunteers (EFSA 2014). Since arsenic compounds consist of various species which are carcinogenic and non-carcinogenic, the exposure assessment should be well-designed to achieve further risk characterization of carcinogenic or non-carcinogenic health effects. For non-carcinogenic effect, estimated exposure can be calculated as **average daily dose (ADD)**. For carcinogenic effect, dose is often calculated as **lifetime average daily dose (LADD)** with estimated lifetime exposure is taken into account (US EPA 2005). Food consumption data and concentration of arsenic are prerequisite for estimation exposure to arsenic.

Ideally, **food consumption data** should include information on factors affecting dietary exposure such as demographic characteristics (age, ethnicity, gender and socio-economic status), body weight and geographic region. Dietary patterns and susceptible groups (e.g., infants and toddlers) should be taken into account for arsenic exposure assessment of children. There are various approaches used for collecting food consumption data which can be classified as different levels of population or nation, household and individual (FAO/WHO 2005, 2009). National food consumption data of several countries worldwide used for exposure assessment were conducted based on individual consumption. Most countries have food consumption data for adults, but consumption data of children and infants is less available (FAO/WHO 2005). Several factors including cost, target population, education level, resource available and studied period time should be considered for selecting of method for food consumption data collection (FAO/WHO 2005, 2009).

One of common approaches for food consumption collection is **food record survey** or **food diary** which subjects record all foods consumed over a period. Significant limitations of this method are that parents or guardians must keep records for young children, and the amount of foods depend on the subject's ability to estimate (FAO/WHO 2005, 2009). Individual dietary records were used for assess dietary exposure to inorganic arsenic in several studies (EFSA 2014; Lovreglio et al. 2012). Another method is the **24-hour recall** which types and amounts of food consumptions are recalled from memory with the aid of a train interviewer. Although this method causes less burden for subjects, the obtained consumption data may be less reliable because it depends critically on memory of subjects. Parents or caretakers also have to respond for children (FAO/WHO 2005). The 24-hour recall were used for evaluation of arsenic exposure especially from rice in many studies (Kordas et al. 2016; EFSA 2014; deCastro et al. 2014; Davis et al. 2014; Wu et al. 2015). The other approach widely used for collecting food consumption is **food frequency questionnaire (FFQ)**. The questionnaire consists of a list of commonly consumed foods which can be completed by subjects or interviewers. The frequency of food consumed is estimated as per day, week, month, or year (FAO/WHO 2005). This approach is useful in the collection of retrospective or prospective data, and specific foods that are known to be contaminated of arsenic can be included in the questionnaires (FAO/WHO 2005). However, the intake frequency and amount of food consumed rely significantly on memory of subjects, and the validity depends on the representativeness of the foods listed in the questionnaire (FAO/WHO 2005). Several studies mostly conducted in adults applied FFQ to assess exposure to arsenic (Cascio et al. 2011; Wei et al. 2014; Cleland et al. 2009). **Duplicate diet survey** or **duplicate portion study** is a method by which the usual food portion consumed by individual is prepared in duplicate. Then, the duplicate sample is pooled and homogenized as daily intake sample. Although this approach is more precise than the others, it is not appropriate for large-scale study because of the cost and time-consuming. It is also impracticable for participants who regularly eat out, and causes heavy burden for both staffs and subjects (FAO/WHO 2005). A few studies used duplicate diet survey for estimation of arsenic exposure in small group of subjects interested (Kile et al. 2007; Ohno et al. 2007). To improve reliability and validity of food consumption data, combining of the methods for the collection of data might be carried out (FAO/WHO 2005).

3.4.2 Arsenic Concentration Assessment

In dietary exposure assessments, the selection of concentration of arsenic in food and drinking water applied for the assessment is very crucial (FAO/WHO 2005). Proposed maximum level of arsenic in particular food item and drinking water established by international and national organizations could be used for predicted exposure assessment as **theoretical maximum daily intake (TMDI)** (FAO/WHO 2005). This approach assumes that arsenic is presented at the **maximum level (ML)** in individual food commodity, and merged with average daily consumption of foods per person. This approach is suitable for pre-regulation of arsenic in food, but it is commonly overestimated (FAO/WHO 2005).

For more refined exposure assessment, actual arsenic contents in foods and drinking water should be used. A major challenge for selection of arsenic content applied for exposure assessment is that various species of arsenic exists with extensively different toxicity (WHO 2011b). As mentioned previously, some commodities contain high ratios of inorganic arsenic species which is the most toxic form and classified as human carcinogens by International Agency for Research on Cancer (IARC). Whereas several foods such as fish, seafood and dairy products had lower ratios of inorganic arsenic species (WHO 2011b; FAO/WHO 2011). Applying **total arsenic concentration** for exposure assessment to arsenic and further calculation of cancer risk could lead to misinterpretation. Since last decade, assessment of dietary exposure to arsenic were mostly conducted using inorganic arsenic concentrations together with total arsenic contents. **Speciation analysis** for arsenic contents in foods is a better approach to assess the arsenic exposure and characterize cancer risk; however, it is costly, complicated and time-consuming (Cubadda et al. 2017). Inexpensive, rapid and simple technique for arsenic species determination are still needed for monitoring and surveillance of arsenic in foods and drinking water. Alternatively, conversion factors of total arsenic into inorganic arsenic of particular commodity can be applied if the concentrations of inorganic arsenic are not available. However, the conversion factors should be selected carefully to avoid uncertain source of exposure data (EFSA 2014).

Another challenge is handling of non-detected concentration of arsenic in food samples. Some previous studies applied zero for non-detected arsenic level in foods which the exposure assessment could be underestimated depending on their limit of quantitation (LOQ) or limit of detection (LOD) of arsenic determination. While overestimated assessment could occur if non-detected values were replaced with the LOD or LOQ. Where feasible, the **substitution method** using the lower-bound (LB), medium bound (MB) and upper-bound (UB) approach is suggested to apply for arsenic concentration below LOQ or LOD (FAO/WHO 2005, 2009; EFSA 2010). For LB, MB and UB, the arsenic concentrations below the LOD/LOQ values were replaced by zero, half of LOD/LOQ value and LOD/LOQ value, respectively (FAO/WHO 2005, 2009, EFSA 2010).

3.4.3 Combination of Dietary Intake and Arsenic Concentration Data

Two approaches, namely deterministic and probabilistic, are used to assess dietary exposure to arsenic by merging food consumption and arsenic concentration. **Deterministic approach** or point estimation is a traditional approach applied to estimate an exposure assessment. It is commonly used for exposure assessment because the procedure is easy and can be done even with limited data and resource (FAO/WHO 2005, 2009). Mostly, the estimation of average exposure of deterministic approach assumes that the distributions of concentration and/or consumption data are parametric. In fact, information on chemicals contents and intake rates is mostly non-parametric distributions; therefore, using median values are more representative and less overestimated. In some cases, the normality of data was tested firstly. If it is non-parametric distribution, the data set was assumed as log-normal distribution for convenience. In fact, the data set of the concentration or consumption is not true log-normal distribution. Consequently, the risk calculated from deterministic method is likely to be overestimated (FAO/WHO 2005, 2009). Additionally, the calculation of high-end exposure should depend on the data distribution (e.g. normal, log-normal, exponential, etc.) (FAO/WHO 2005, 2009).

To overcome the exposure overestimation, **probabilistic approach** has been developed and applied widely. Probabilistic approach incorporates uncertainty and/or variability information (FAO/WHO 2005, 2009; US EPA 2014). The degree of exposure assessment certainty and the differences between exposed populations are provided. Generally, deterministic method reports risks as average and high-end exposure but probabilistic approach can explain more on the uncertainties and/or variabilities in the estimated exposure. Probabilistic method can also identify the key contributing to the uncertainties and/or variabilities (US EPA 2014). Therefore, the information from this approach is useful for risk managers to compare the risks with different management alternatives, to accomplish a science-based level of safety and to support regulatory decision makings (US EPA 2014). Commercially available software such as @RISK (Palisade cooperation) using Monte Carlo simulation can be used for probabilistic risk assessment effectively (US EPA 2014).

4 Perspectives for Future Research

4.1 Establishment of Cut-off Level to Categorize High Arsenic Exposed Group

According to Agency for Toxic Substances and Disease Registry (ATSDR, USA) normal levels of arsenic in low-exposed individuals are less than 1 μg/L and 100 μg/L in blood and urine samples, respectively, and equal or less than 1 ppm (μg/g) in hair or nail samples (ATSDR 2007). The usefulness of blood arsenic level to detect low-level exposure in healthy low-exposed children is very limited. The cut-off level cannot be established due to its narrow range and short half-life nature (Hughes 2006). The urine total arsenic levels in healthy low-exposed children without recent history of seafood consumption are generally expecting to be below 10 μg/L (ATSDR 2007; Council 1999; Hwang et al. 1997). Their baseline urinary levels can also be varied by study regions as described earlier. The exact cut-off level that warrant actions to prevent long-term health consequences in these healthy low-exposed children group was not established by any study to date. However, if one is determined to take proactive action for the issue, possible inorganic arsenic exposure sources may be worth investigating if the children urinary total arsenic levels were consistently above 10 ug/L.

The cut-off arsenic levels in hair samples of heathy low-exposed children at different ages are not yet identified. A study in Cambodia utilized data of hair arsenic and signs of arsenicosis from 616 respondents to propose the cut-off level of greater than 1.0 μg arsenic per 1 g of hair as an indicator of an arsenic exposed individual (Hashim et al. 2013). The standardization of sample collection, handling, washing, processing and analyzing procedures are still evolving. Therefore, discrepancy in the details can make the comparison of available analytic results from different studies not virtually accurate (Mikulewicz et al. 2013). However, several attempts were made to set up reference level in healthy low-exposed children from different countries. Studies in hair samples from 3–6-year-old Korean and 3–5-year-old Italian children reported arsenic levels of 0.20 and 0.24 ug/gm, respectively, at their upper and 95th percentile rank (Park et al. 2007; Senofonte et al. 1989). These hair arsenic upper range levels were much lower than those in older children. Other studies in 11–13-year-old Italian and 12–18-year-old Brazilian children yielded arsenic levels of 0.03 and 0.02 ug/gm, respectively, at the 97.5th and 90th percentiles rank (Dongarra et al. 2011; Carneiro et al. 2011b). In general, proactive actions should be considered when the detected hair arsenic levels in healthy low-exposed children exceeds two to three standard deviations above the average level of their age and location matched population. More studies are warranted to finalize the hair arsenic cut-off level to categorize high exposure group in healthy low-exposed children.

Several studies reported different ranges of normal arsenic level in nail samples but generally not exceeding the ATSDR upper limit at 1 μg/g (ATSDR 2007; Council NR 1999; Narang et al. 1987; Takagi et al. 1988). A Brazilian study in 126 healthy unexposed 12–18-year-old students reported the 90th percentile rank level at 0.17 μg/g (Carneiro et al. 2011a). However, the arsenic concentration in toenails of infants in New Hampshire Birth Cohort Study ranged from 0.000 to 1.21 μg/g (Davis et al. 2014). Such high arsenic level in infant nails were associated with their mother nail arsenic levels indicating *in utero* exposure (Davis et al. 2014). Based on EFSA scientific opinion on arsenic, dietary exposure to inorganic arsenic for children under 3 years of age is estimated to be from two to three-fold that of adults (EFSA 2014). Since young children may be more sensitive to arsenic toxicity than older children and adults, specific cut-off levels for young children should be established. Further cohort studies are warranted to establish association between arsenic levels in biological specimens of young children and long-term toxicity. This piece of information is the missing jigsaw.

An important challenge in establishing the cut-off levels is the lack of standard reference material. Nowadays only standard reference materials of total arsenic in urine, muscle and liver and speciated arsenics in urine have been certified (Hughes 2006; Yoshinaga et al. 2000). Owing to more hazardous nature of inorganic arsenic, the reference materials for speciated arsenics are needed. Unfortunately, a previous report by Hansen et al. found that an arsenic sulfur compound detected in urine had been misidentified as trivalent methylated form of arsenic (DMAIII) (Hansen et al. 2004). Another challenge of cut-off establishment is the varied susceptibility among different individuals. Polymorphism in genes encoding arsenic-metabolizing enzymes could influence variability in arsenic metabolism and susceptibility to arsenic toxicity (Vahter 2000). Genetic background should be considered when establishing cut-off levels, i.e. population with polymorphic genes may require lower cut-off levels of arsenic.

4.2 Data Collection Methods for Estimation of Dietary Exposure to Arsenic

Information on food consumption is crucial for estimation of arsenic exposure. However, food consumption data of children in many countries are not available. This could be due to the difficulty in collecting consumption data from kids. A national survey in the United States, Feeding Infants and Toddlers Study (FITS) 2016 collected dietary data by using a 24-h dietary recall telephone survey with the primary caregivers of 3235 children aged 0 to <4 years (Welker et al. 2018). Although this work is one of the largest studies for feeding behavior of young children, the authors admitted that there could be some reporting bias and measurement error inherently associated with dietary recall (Welker et al. 2018). Future studies should focus on developing new more attractive and effective tools to facilitate collecting food consumption data of children such as visual methods, online application and gamification. A new questionnaire DAFA had been recently developed and validated in 7–10 years old children. The answer choices are all in picture format (Barros et al. 2007).

Most of the previous studies on arsenic exposure in children have emphasized on collecting data based on particular food item such as rice or drinking water which are considered as major arsenic contributors. However, in order to obtain accurate arsenic exposure, collective effect of all food items consumed should be estimated. In addition, dietary exposure assessment should be carried out concomitantly with biomarker assessment.

It has been shown that different food items contain various forms of arsenic with different levels of toxicity. As a result, estimation of total arsenic exposure may not result in realistic estimation of the potential toxicity of arsenic. For example, seafood and dairy products have lower ratios of inorganic to total arsenic contents (WHO 2001). This lower ratio is associated with lower toxicity. In contrast, rice has higher ratio indicating higher toxic effects (Zhu et al. 2008). Some studies have determined the total arsenic contents and used conversion factors to estimate the inorganic arsenic contents (EFSA 2014; Sigrist et al. 2016). Although this could be better than reporting only the total arsenic contents, the result may not reflect the actual toxic effect. There are wide variations in the ratio of inorganic to total arsenic levels depending on various factors such as cultivars, locations, agricultural practices. Therefore; further studies should focus on exposure to different arsenic species commonly detected in foods (e.g. As(III), As(IV), DMA, MMA).

It is well known that types of food processing such as heating have a significant effect on chemical composition and their activities in foods (Devesa et al. 2008). A recent study showed that heat treatment exceeding 150 °C such as baking, grilling and frying not only reduced total arsenic content but also decomposed organo arsenic compounds into inorganic arsenic species (DMA and MMA) (Devesa et al. 2008). Thus, effect of processing and cooking should be taken into consideration for more effective estimation of exposure to toxic compounds. In addition, bioavailability of food components determines the actual effect on human health (Zhu et al. 2008). For example, bioavailability of organic arsenic species is much lower than that of inorganic forms (Khan et al. 2010; Zhang et al. 2016). To acquire realistic arsenic exposure highlighting the potential toxic effects, several factors should be considered as discussed.

4.3 Identification of Underrepresented Sources of Arsenic in Well-Nourished Children

Young children are one of the most vulnerable group. Being fully nourished children from a well-to-do family, they seem to have lower risk of arsenic exposure than undernourished children living in highly exposed area. Nevertheless, there are still some underrepresented sources of arsenic exposure to this seemingly low risk well-nourished group. A recent exposure assessment study in the United States showed that the largest source of inorganic arsenic for infants and toddlers between 4 and 24 months old was rice cereal (55%), followed by other infant solid foods (19%), and drinking water (18%) (Shibata et al. 2016). Hazard quotient of chronic exposure (HQchronic) at the 50 and 75th percentiles in this group exceeded 1.0 for both rice cereal and total sources (Shibata et al. 2016). In a well-nourished family, iron-rich rice cereal is the first food commonly introduced to the 4–6 months old infant (Byrd-Bredbenner 2016). Therefore, over-nourishment of young children may actually pose them to hidden risk. Interestingly, a recent national survey in the United States, Feeding Infants and Toddlers Study (FITS) 2016 reported that almost all (95%) American children aged 2–3 years old consumed grain products, and 59% consumed a whole grain–rich products (Welker et al. 2018). Since the richest sources of inorganic arsenic are drinking water and rice, the above consumption data raised the concern of high arsenic exposure in young children. Interestingly, US Food & Drug Administration (USFDA) recently proposed to set the maximum arsenic limit of 100 parts per billion (100 ppb or 100 ng/g) (Erickson 2016). Furthermore, previous studies showed accumulation of inorganic arsenic in the bran of grain and polishing did reduce the inorganic arsenic (Zhao et al. 2010). Consistently, a recent Thai study found higher inorganic arsenic content in brown rice products for infant than that of white rice product (Wongsaengnak 2018). To ensure adequate intakes of fiber and vitamins, US nutritional guideline (My Plate) recommended that half of the total grain consumed should come from whole grains (Byrd-Bredbenner 2016). However, considering the increased health risk of inorganic arsenic in unpolished grain such as brown rice, national recommendations regarding whole grains intake of each country should consider the risk of arsenic in each specific geographical area with special caution for infant and young children. Risk assessment of arsenic in whole-grain products for young children should be further investigated.

4.4 Strategies to Raise Awareness and Create Sustainable Preventive Measures of Arsenic Contamination

Social awareness is one of the most important factors for prevention of arsenic contamination and toxicity. A recent study in India reported that 63% of surveyed population in arsenic-affected communities were aware of arsenic problems (Singh et al. 2018). The significant predictors of awareness included socioeconomic factors, water and sanitation behavior factors and social capital and trust factors (Singh et al. 2018). Furthermore, lower-caste, less educated, and unemployed people were the ones with least awareness and at highest risk. To raise awareness of people in the society especially in the high-risk area, information technology could play a critical role in providing statistical information and create network of information for monitoring of the problems. Interestingly, a recent work proposed cloud-based decision support system for arsenic health risk assessment (CC-AHRA) (Singh 2017). The CC-AHRA system is designed to establish database of research information from all stakeholders including academic institution, federal agencies, non-profit organizations, private enterprises and independent researchers (Singh 2017)**.** People worldwide can put such database in cloud/internet to be accessible. The CC-AHRA system also provided digital arsenic analyzer (in mobile application format) as a tool to assess health risk of arsenic at both individual and community levels (Singh 2017). The system will definitely facilitate and accelerate decision making for any preventive/intervention measures in order to mitigate arsenic contamination and toxicity problems. More international collaborations and governmental supports are needed to create sharing and connecting network of information and to make it accessible by under-privileged communities. Then, CC-AHRA system could potentially be a sustainable tool for resolving a global health challenge of arsenic contamination.

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Chapter 5
Environmental Arsenic Exposure and Human Health Risk

Anindita Mitra, Soumya Chatterjee, and Dharmendra K. Gupta

Abstract Millions of people worldwide are exposed to arsenic through contaminated water used for drinking, cooking, and irrigation of crops. Presence of significantly elevated levels of arsenic (exceeding the World Health Organization (WHO) recommended a provisional value of 10 μg/L) in food or drinking water has been documented from many parts of the world. Chronic exposure to the element is a major global public health issue. Clinical symptoms of acute arsenic poisoning include muscular weakness and muscle cramps, nausea, vomiting, and diarrhea. Arsenic-associated chronic poisoning leads to diseases like cancers, skin lesions, diabetes, hypertension, etc. Arsenic is a potent genotoxic agent for animals and humans that can damage DNA, induces chromosomal aberrations, sister chromatid exchange and micronuclei formations. Extensive research on the biological mechanisms underlying arsenic-associated disease suggests that among a number of cellular mechanisms, epigenetic modifications (altered DNA methylation, miRNA expression, and histone modification) are underpinned by arsenic exposure. Evidence support that inorganic arsenic is an epigenetic modulator of genes as it can alter enzymatic activity of DNA methyltransferases, histone deacetylase (HDAC) and histone acetyltransferase (HAT) that are associated with cellular growth and immune response. This review aims to present a comprehensive overview of the possible sources of arsenic exposure to humans, and effects on metabolic pathways and related health issues. Additionally, epigenetic modification underlying arsenic-associated changes and their role in arsenic-induced toxicity are also discussed.

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1 Introduction

Arsenic (symbol: As; atomic number: 33) is a naturally occurring metalloid, widely distributed in the Earth's crust (Vahter 2008). This element, commonly represented by inorganic (iAs) and organic (complexed with organic matters) compounds in the environment, is considered to be one of the most toxic elements for global health (Vahter 2008). iAs is an established carcinogen and is the most substantial contaminant in drinking water globally (Cantor et al. 2006). The naturally accumulated As in earth's crust or bedrocks permeate gradually into the water sources (Chakraborti et al. 1999; Gupta et al. 2017). Arsenic is found in four oxidation states namely −3, 0, +3 and +5 and persists as elemental, sulfide and carbonate allotropic forms (Chakraborti et al. 1999; Henke 2009; Mitra et al. 2017a). Arsenic has long been recognized as an environmental hazard, raising great concern from the environment as well as human health perspectives (Iyer et al. 2016). A human may get exposed to As from various sources such as drinking of As-contaminated water, ingestion of food materials (fruits, cereals, vegetables) grown in the As-contaminated agriculture fields, arsenical pesticides, contact of contaminated industrial/agricultural wastewaters and occupational exposures to As-contaminated dust, fumes or mist (Saha et al. 1999). Reduced trivalent compounds of arsenic (As^{III}) are more polar than oxidized pentavalent arsenic compounds (As^{V}), therefore, more toxic in nature (Akter et al. 2005). Rapid and extensive absorption of arsenate and arsenite compounds occur through the gastrointestinal tract following accumulation in tissues and body fluids (Alava et al. 2014). The rate of absorption of As through inhalation entirely depends on its molecular size. e.g., molecular absorption of sodium arsenite, sodium arsenate, and arsenic trioxide through inhalation were rated higher than arsenic sulfide and lead arsenate (Abdul et al. 2015). Acute and chronic exposure of arsenic leads to a range of clinical symptoms characterized as "Arsenicosis" (Mc Carty et al. 2011). There are several experimental proofs signifying chronic exposure to As leads to the development of cancers of various organs, such as skin, lungs, liver, and urinary bladder, in humans. Arsenic exposure can also give rise to precancerous dermatological lesions like hyperkeratosis (palmar and plantar), hyper- and hypopigmentation and raindrop pigmentation (Banerjee and Giri 2016). Though skin symptoms are assigned as hallmarks of arsenic toxicity, a plethora of epidemiological surveys substantiate that chronic arsenic toxicity can also be noticeable in the form of nondermatological disorders such as peripheral neuropathy, respiratory distress, ocular problems like conjunctivitis, cardiac arrest, diabetes, and genotoxicity (Chatterjee et al. 2017a; Chakrabarti et al. 2018a). Arsenic, thus, is not only a silent killer but also responsible for reduced life expectancy and compromised life quality.

2 Sources of Human Exposure to Arsenic

2.1 Drinking and Cooking Water

Drinking water is considered to be the primary source of As for humans as evidenced from epidemiological studies in arsenic-endemic areas (Diaz et al. 2015). Drinking water, also used for cooking, is considered to be the major source of exposure to inorganic As in regions where arsenic concentration exceeded above 10 μg/L (FAO/WHO 2011) (Fig. 5.1). High levels of iAs in water in such areas used for cooking purposes additionally aggravate the level of exposure (Diaz et al. 2015). Therefore, As-contaminated water used for cooking is the source of the high total As levels detected in cooked food (Diaz et al. 2004). Groundwater in a number of countries, including Argentina, Bangladesh, Chile, China, India, Mexico, and the United States of America, is reported to contain high levels of iAS (FAO/WHO 2011). Study based on different states of India (Mukherjee et al. 2006) revealed that groundwater in West Bengal, Bihar, Uttar Pradesh, Jharkhand, Assam, Chhattisgarh, and Manipur is highly contaminated with As. The study also uncovered that As accumulation in major crops like rice, pulses, and vegetables was due to use of arsenic-contaminated irrigation water (0.318–0.643mg^{-L}) to grow these crops (Mukherjee et al. 2006).

2.2 Food

Human population is exposed to inorganic As through food and drinking water (Yorifuji et al. 2011). In fact, food is the prime source for human As intake (WHO 2001) if not exposed through drinking water. Seafood such as fish and shellfish

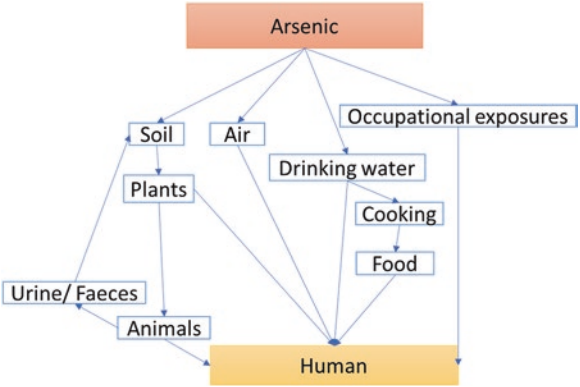

Fig. 5.1 Routes of human exposure to arsenic

consumed by a number of human populations has been recognized as the major source of As (WHO-IARC 2011) in which concentrations of the metalloid has been exceeded than those in most terrestrial foods. In marine biota, mostly organic form of As is found (EFSA 2014). Arsenobetaine (AsB), the major As species in most seafood, is presumed to be non-toxic in nature and remains unmetabolized (FAO/WHO 2011). Arsenosugars and arsenolipids, the other more complex organic forms of As present at significant quantities in some types of seafood (1–100 µg/g), are metabolized by humans (Cubadda et al. 2016). Similar to iAs species, organic As compounds produce dimethylarsinate (DMA) as a urinary metabolite. However, according to recent findings, some forms of organic As compounds and their metabolic intermediates show cytotoxicity in the cultured cell (Leffers et al. 2013; Naranmandura et al. 2011a; Meyer et al. 2014, 2015). Organic As compounds like DMA and monomethylarsinate (MMA) in trace amount is present in various foods, including rice, other cereals, and seafood. Oral exposure to DMA adversely affects fetal development as well as organs such as urinary bladder, kidneys, and thyroid; whereas primary target of MMA is the gastrointestinal tract, followed by kidney, thyroid, and reproductive system (US FDA 2016).

Current risk assessments of dietary exposure to arsenic are solely based on the inorganic species due to its extreme level of toxicity (NRC 2014; Gupta and Chatterjee 2017**.** The major route of entry of iAs in the food chain of the terrestrial ecosystem is through plant crops, which uptake As from soils through their roots depending on the concentration and phytoavailability of the metalloid in the soil. The type of the soil parent material determines the iAs concentration in soils (Abdul et al. 2015). Besides this, soil physicochemical properties such as adsorbing ability also play a key role in increasing phytoavailability of iAs to plants by preventing its mobility in soil solution (Punshon et al. 2017). Plants grown in highly As-contaminated soils have the ability to accumulate significant amounts of iAs in their edible parts like leaf and seeds (Signes-Pastor et al. 2008; Williams et al. 2007). Whereas in the animal body, iAs undergoes metabolism and excretion, and hence animal foods contain less quantities of iAs compared to plant product. Anthropogenic activities and inappropriate arsenic-rich waste disposal resulted in the accumulation of iAs up to 723 ng g^{-1} in rice grain, as reported in the Hunan province of China (Okkenhaug et al. 2012; Wang et al. 2015). A very recent study for inorganic arsenic contamination in rice-based diets including rice cakes, rice-based snacks, baby rice, and puffed rice obtained from supermarkets of South Australia, showed that the arsenic level in 53% of the samples were found to exceed the European Union recommended value (100 mg kg^{-1}) for young children and in 22% of the samples exceeded maximum level of 200 mg kg^{-1} recommended for adults (Chakrabarti et al. 2018b). Among all crops, rice was found to accumulate the highest level of iAs (0.1–0.4 mg As/kg dry weight), and rice-based products were also found to have the highest level of iAs (EFSA 2009; Meharg et al. 2008; Chatterjee et al. 2017b). The portion of As varies in the rice plant in the following order: rice straw > bran > whole grain > polished rice > husk (Carey et al. 2012). The proportion of iAs (present both as As^{III} and As^{V}) of total As in rice ranges between 10-93 %, and brown rice reported to have a higher iAs content than white rice (Torres-Escribano et al. 2008; Sun et al. 2008; Mitra et al. 2017b). The reason is that total arsenic levels attached to bran were much higher than in endosperm of white rice obtained from the same whole grain rice (Rahman et al. 2007).

2.3 Soil

Arsenic in soil exists solely in the inorganic form, except in regions where organic arsenic compounds are applied intentionally (Saxe et al. 2006). In soils, As^V species predominate due to oxidation of trivalent arsenicals (Gong et al. 2001). Soil encompasses a high level of As if contaminated with mining or smelter wastes (up to 27 000 mg/kg), and also around the waste dumping point of an arsenical pesticide manufacturing plant (more than 20100 to 35500 mg/kg) (Chatterjee and Mukherjee 1999). In agricultural soils treated with various arsenical pesticides, the mean total arsenic concentrations ranged from 50 to 550 mg/kg (Stilwell and Gorny 1997). Humans can be exposed to soil arsenic through multiple pathways including incidental ingestion, dermal absorption, and inhalation of wind-blown soil particles (i.e., fugitive dust) (Hughes et al. 2011) (Fig. 5.1).

2.4 Air

In comparison with the route of arsenic exposure from food and water, exposure through the air is generally low. In air arsenic present predominantly as particulate forms (2 μm in diameter) and as a mixture of arsenite and arsenate, with a negligible amount of organic species except in areas where arsenic pesticide are applied (Chung et al. 2014). The European Commission (2000) reported that air contributes less than 1% of total arsenic exposure, and levels of arsenic in air found lowest in remote areas (0–1 ng/m^3) than in the rural areas (0.2–1.5 ng/m^3), and, in urban areas (0.5–3 ng/m^3), and maximum (about 50 ng/m^3) adjacent to the industrial region. Humans may be exposed to a very low concentration (0.4–30 ng/m^3) of arsenic through the air (Fig. 5.1). Humans per day typically inhale approximately 40 to 90 ng of arsenic and in unpolluted areas, the value is less than 50 ng or less (US EPA 2014).

2.5 Other sources

Another potential route of As exposure is through occupational exposure to arsenic in several industries, particularly, non-ferrous smelting, electronics, copper smelters, wood preservatives, wood joinery shops, arsenic producing facilities, glass manufacturing facilities, production and application of arsenical pesticides and coal plant, etc. Exposure may occur primarily through inhalation, accidental ingestion of arsenic-containing particulates and also through dermal exposure (Fig. 5.1) (Hughes et al. 2011).

Newborn babies may be exposed to a low level of As in breast milk from mothers. For example, studies in New Hampshire, USA, showed that nursing mothers were exposed to a low level of arsenic in drinking water (Carignan et al. 2015). Infants have three times higher dietary arsenic exposure than adults as they have restricted diets (subsist on breast milk/powdered formula), lower dietary diversity (EFSA 2009) and have greater intake per unit body mass (Tsuji et al. 2007). However, studies support that As in breast milk is relatively low in comparison to drinking water (Bjorklund et al. 2012; Islam et al. 2014). The species of As present in breast milk depend on the source or magnitude of exposure. For example, breast milk samples from Bangladeshi mothers exposed to high level of As in drinking water contained iAs (Vahter et al. 2006); whereas organic species of As was found in breast milk samples from Swedish mothers who had an unknown source of As exposure from drinking water (Bjorklund et al. 2012).

3 Metabolic Pathway of Arsenic in Humans

After being exposed to a toxic chemical, its dispersal throughout the human body and the rate of excretion or metabolic alteration to less or more toxic forms varies according to the chemical species and also between individuals, which ultimately governs the toxicological outcome of exposures to the chemical (Banerjee and Giri 2016). Arsenicals to which humans are most likely to be exposed in the environment are inorganic arsenic compounds, and metabolism and disposal of the inorganic compounds largely depend on their valence state (ATSDR 2007). Inorganic As^{III} and As^{V} species are readily absorbed from the intestine in humans as well as rapidly excreted as methylated metabolites in urine than the parent compound (Lee 1999). Methylated forms of arsenic in the trivalent oxidation state (MMA^{III}) are stable intermediates formed during biomethylation as evidenced by different studies. For example, the bile of rats treated with arsenate or arsenite contains monomethylarsonous acid (Gregus et al. 2000). Similarly, urine samples from individuals who have chronically consumed iAs-contaminated drinking water contain monomethylarsonous and dimethylarsinous acid (Thomas 2016). Some of the methylated derivatives of inorganic arsenic are more reactive and toxic than arsenite, and therefore, have more potential to induce cytotoxicity and genotoxicity (Naranmandura et al. 2011b; Rehman et al. 2012) (Fig. 5.1). Besides the variations in the level of arsenic exposure between individuals, genetic polymorphism of the methyltransferase (AS3MT) gene is considered to be a major determinant for inter-individual variation in arsenic methylation patterns (Eichstaedt et al. 2015). Other factors playing a regulatory role in arsenic metabolism profile includes age, sex, body mass index and nutrition status of the individuals (Jansen et al. 2016; Kuo et al. 2017).

The classical pathway of As metabolism proposed earlier by Frederick Challenger (1945) and later substantiated by Cullen and Reimer (1989) describes that inorganic As^V is first reduced to As^{III}, and then oxidative methylation occurs to form monomethyl arsenic acid (MMA^V) in which methyl group is contributed by S-adenosyl-l-methionine (SAM). It is typical for methyltransferases, in which AdoMet is the methyl donor and the catalytic reaction, to be interlinked to a multiple-step process regulating the availability of this prime methyl group donor (Drobna et al. 2009). MMA^V is further reduced to the trivalent form monomethyl arsonous acid (MMA^{III}), that undergoes a second round of oxidative methylation (again in the presence of SAM) to form dimethyl arsinic acid (DMA^V) (Banerjee and Giri 2016). The final product of arsenic metabolism in human is dimethyl arsinous acid (DMA^{III}) formed by reducing DMA^V and is excreted through urine (Steinmaus et al. 2007). The regulatory genes involved in this pathway include those encoding arsenate reductases GSTO, methyltransferase (AS3MT) and purine nucleoside phosphorylase (PNP). According to a recent report by Zhang et al., AsB, less toxic organoarsenic derivatives, was produced in marine herbivorous fish (*Siganus fuscescens*) following biotransformation of both As^{III} and As^V (Zhang et al. 2016). Although methyltransferases (AS3MT) play a key role in methylation of As^{III}, the liver and urine samples of arsenate-exposed AS3MT knockout mice still show mono- and dimethylated arsenicals, but to a lesser extent than wild-type mice, suggesting that either the presence of other genes, besides AS3MT, encoding methyltransferases or anaerobic intestinal microbiota can methylate orally administered arsenicals (Watanabe and Hirano 2013).

Hayakawa et al. (2005) proposed an alternative pathway for arsenic metabolism involving glutathione. Accordingly, inorganic arsenite is first conjugated to cellular glutathione to form arsenic triglutathione (ATG) and subsequently methylated to monomethyl arsonic diglutathione (MADG). This MADG can then either be oxidized to MMA^V via the intermediate formation of MMA^{III}, or it can be further methylated and subsequently oxidized to yield DMA^V. Therefore, both MMA^V and DMA^V are the end products of arsenic metabolism and excreted through urine.

A third model of arsenic metabolism was proposed by Rehman and Naranmandura (2012), in which, iAs first binds to protein moieties in the liver, followed by reductive methylation by conjugating with thiol groups (contributed either by cysteinyl residues of proteins or glutathione) involving S-adenosyl-methionine. Several authors have suggested that due to a higher affinity of arsenic toward cysteine-rich proteins than toward glutathione, the As^{III} species mostly remain in the protein-binding forms (Bogdan et al. 1994; Naranmandura et al. 2006; Khairul et al. 2017). The methylation of DNA also requires SAM and arsenic exposure may cause DNA hypomethylation due to continuous methyl depletion following depressed levels of SAM which concurrently facilitates aberrant gene expression that results in malignant transformation (Roy and Saha 2002). The end products of arsenic metabolism are MMA^V and DMA^V, similar to the alternative pathway hypothesized by Hayakawa et al. (2005).

4 Acute Arsenic Poisoning to Humans

Symptoms of acute As toxicity usually occurs approximately 30 min following exposure or ingestion, and includes reddish rashes in the skin, extreme thirst but a difficulty in swallowing, and dryness inside the mouth with breath having a slightly garlicky odor (Hughes et al. 2011; Chakrabarti et al. 2018a). An appearance of transverse white lines across the fingernail called Mee's lines specifies acute arsenic poisoning and the victim may die due to cardiovascular collapse and hypovolemic shock (Chakrabarti et al. 2018a). Early acute As intoxication is clinically manifested by symptoms including weakness, muscular cramps, and abdominal pain, nausea, vomiting and severe diarrhea (Hughes et al. 2011). Degenerative effects on mucosal vascular supply lead to capillary damage, generalized vasodilation, sloughing of tissue fragments, and vasogenic shock (Chatterjee et al. 2017a). In severe poisoning, skin turns into cold and clammy, due to circulatory failure beside decreased urine output and kidney damage. Development of psychosis associated with hallucinations, paranoid delusions, and delirium with seizures, coma, and death might follow (Saha et al. 1999 ; Hughes et al. 2011 ; Naujokas et al. 2013). Clinical manifestation also includes a diffuse skin rash, toxic cardiomyopathy, seizures, hematological abnormalities, renal failure, respiratory failure, and pulmonary oedema; peripheral neuropathy or encephalopathy are also common in neurological manifestations (Ratnaike 2003).

5 Chronic Arsenic Poisoning to Humans

Chronic nature of poisoning is more tricky as making a precise diagnosis of symptoms is very much challenging. Arsenic-related dermatosis is often confused with other related symptoms. Mostly affected systems are skin, liver, lungs, and blood; conjunctival congestions are infrequent in arsenical dermatosis (ASD) (Walkes et al. 2014; Argos et al. 2014). Anemia and leukopenia, as well as frequent occurrence of thrombocytopenia, are very common with chronic arsenic exposure (Chakrabarti et al. 2018a). Nonspecific but typical cutaneous mutability with early obstinate erythematous flush leading to melanosis, hyperkeratosis, and desquamation is apparent (Hughes et al. 2011). Palms and soles are also affected by diffuse desquamation. Unending skin problems lead to the development of multicentric basal cell and squamous cell carcinomas (Saha et al. 1999). The most noticeable chronic manifestations of As exposure involves the skin, lungs, liver and blood systems and leads to a predisposition to varied disease of all the major organ/system as summarized in Table 5.1.

Table 5.1 Summary of different diseases or clinical manifestation associated with As exposure.

System/Organ affected	Clinical manifestations	References
Integumentary System	Skin abnormalities are considered as a hallmark of chronic As exposure. Clinical manifestation includes melanosis, hyperkeratosis, hyperpigmentation on the neck, face and back, thickening of palm and sole, skin cancer	Guha Mazumder et al. (1998), Argos et al. (2011), and Chakrabarti et al. (2018a)
Respiratory System (lung, trachea)	Inhalation of As dust or fume during mining of rocks or ores manifests respiratory complications such as a chronic cough, laryngitis, tracheobronchitis, and rhinitis. Other complications are a sore throat, voice harshness, hemorrhage at the mucosal and submucosal and alveolar region, reduced survival from pulmonary tuberculosis, lung cancer	Parvez et al. (2010), Dauphine et al. (2011), Naujokas et al. (2013), Steinmaus et al. (2014), and Chakrabarti et al. (2018a)
Cardiovascular System	Increased risk of cardiovascular diseases (CVD), injury to vascular system and heart; Myocardial injury and arterial thickening, increasing the risk of CVD and death.	Medrano et al. (2010), Gong and O'Bryant (2012), Wiwanitkit (2015), and Chakrabarti et al. (2018a)
Hepatic System	Ascites, jaundice or enlargement of the liver, increased level of liver enzymes; hepatic fibrosis; non-cirrhotic portal fibrosis, cirrhosis, central necrosis, liver cancer	Yuan et al. (2010), Chen et al. (2010), Gibb et al. (2011), Jomova et al. (2011), and Chakrabarti et al. (2018a)
Renal System	Oligourea, a higher level of serum creatinine, albuminuria and proteinuria, mutilation to capillaries and glomeruli of the kidney (As induced) with partial or total renal failure, bladder and other urinary cancer	Yuan et al. (2010), Gibb et al. (2011), Naujokas et al. (2013), Zheng et al. (2014), Peters et al. (2015), and Chakrabarti et al. (2018a)
Nervous System	Arsenic species can cross the blood-brain barrier and found to distribute throughout the brain with the highest accumulation in hypophysis. Common clinical symptoms comprise paraesthesia, pain and numbness in the soles of the feet, myopathy, encephalopathy, headache, lethargy, restless sleep, loss of libido, increased urinary urgency, mental retardation, delusion, compromised intellectual function in children and adults; loss of reflexes, seizures and coma, impaired motor function	Sanchez-Pena et al. (2010), Vahidnia et al. (2007), Hamadani et al. (2011), Gong et al. (2011), and Parvez et al. (2011)

(continued)

Table 5.1 (continued)

System/Organ affected	Clinical manifestations	References
Immune System	A major target of iAs is macrophages and As mediated immunotoxicity is accomplished by inhibiting the function of macrophage unfolded protein response (UPR) signaling pathway. iAs also impairs the maturation, activation, proliferation and function of lymphocytes. Variation in cytokine expression and immune-related gene expression, inflammation and frequency of infant morbidity was found to increase due to the predisposition of infectious diseases.	Srivastava et al. (2013), Martin-Chouly et al. (2011), Spivey (2011), Ahmed et al. (2011), and Kile et al. (2012)
Endocrine System	Arsenic is a well-known endocrine disruptor of the thyroid, pancreas (inhibition of insulin activation; diabetes), gonads and also upsets hypothalamo-pituitary- adrenal axis. Impaired glucose tolerance in pregnant women, and glucocorticoid receptor pathways, inhibition of a number of important enzymes; Phenylarsine oxide (PAO) hinders glucose transport, impedes NAD-linked oxidation of pyruvate or α-ketoglutarate.	Ciarrocca et al. (2012), Naujokas et al. (2013), Lu et al. (2011), Davila-Esqueda et al. (2012), Shen et al. (2013), Goggin et al. (2012), Rahman et al. (2010), Del Razo et al. (2011), Islam et al. (2012), and Jovanovic et al. (2013)
Development and Reproductive System	Dose and duration dependent arsenic exposure have an impact on fetal growth and fetal death. Pregnancy complications along with premature delivery have been reported. In males, arsenic-induced gonadal dysfunction was found through reduction of testicular weight, reduced level of testosterone and gonadotrophins, apoptosis and necrosis.	Golub et al. (1998), Tabocova et al. (1996), Chakraborti et al. (2003), Davila-Esqueda et al. (2012), Shen et al. (2013), Kim and Kim (2015), and Chakrabarti et al. (2018a)

6 Arsenic as a Genotoxic Agent

Arsenic exposure promotes chromosomal aberrations, micronuclei formation, DNA strand breakage, sister chromatid exchanges, and oxidative DNA damage in humans at a sub-cytotoxic concentration (Fig. 5.2) (Pozo-Molina et al. 2015; Salazar and Ostrosky-Wegman 2015; Ganapathy et al. 2016; Alahmari et al. 2017). Arsenic exposure has been associated with cancer, diabetes, cardiovascular and neurodegenerative diseases (Ruiz-Ramos et al. 2009; Tokar et al. 2010; Parvez et al. 2011; Syed et al. 2012; Maull et al. 2012). Though methylation of As is a prerequisite for the detoxification process, the trivalent methylated metabolites (MMA^{III} and DMA^{III}) have been shown to be more genotoxic than inorganic arsenic (Gosse et al. 2014). Naranmandura et al. (2011a) reported that methylated arsenicals treated RLC-16 cells from rat liver generates reactive oxygen species (ROS) in mitochondria and causes ARE-mediated activation of thioredoxin reductase gene expression in WI-38

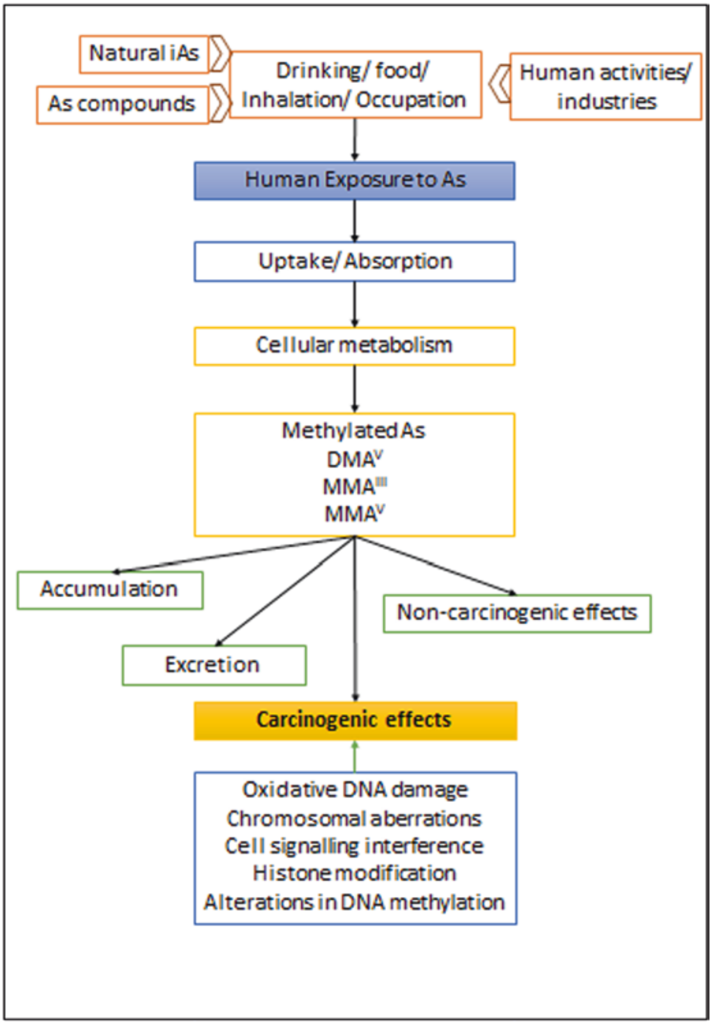

Fig. 5.2 Schematic representation of arsenic-induced toxic effects in humans. (Modified after Hubaux et al. 2013)

lung fibroblasts (Meno et al. 2009). Similar results found in human epidermal keratinocytes after exposure to monomethyl arsonous acid with augmented expression of genes involved in skin carcinogenesis, stress response, synthesis of growth factors, and in a reversal of cell signaling pathways (Bailey et al. 2010). The activation of P13K/AKT signaling pathway, the key regulator for many physiologic processes, may aid for cancer development as evidenced from cell proliferation, migration, invasion, and anchorage-independent growth following chronic As exposure (Chen and Costa 2018). Similarly, activation of EGFR (epidermal growth factor receptor), a tyrosine kinase transmembrane receptor has been reported following arsenic exposure in bladder cells regulating important processes in carcinogenesis, including cell survival, cell cycle progression, tumor invasion, and angiogenesis (Carpenter and Jiang 2013). Several studies have shown that the genotoxicity of pentavalent organoarsenic compounds (MMA^V and DMA^V) is limited to millimolar dose range (higher concentration), while trivalent methylated species (MMA^{III} and DMA^{III}) are genotoxic at micromolar doses (lower concentration), therefore, more toxic in nature (Colognato et al. 2007). However, the genotoxic potential of another organoarsenical known as roxarsone (4-hydroxy-3-nitrobenzenearsonic acid), widely used as a chicken fodder, was shown to induce micronuclei formation and DNA damage in mammalian cells (Zhang et al. 2012).

Arsenic-mediated DNA damage is indirect, occurs mainly due to oxidative stress (Liu et al. 2001a; Ganapathy et al. 2016), and leads to chromosomal aberrations, DNA strand breakage, DNA adducts, and DNA- protein crosslinks (Wang et al. 2001). Sister chromatid exchange (SCE) assay is a widely used method for genotoxicity testing of any compound as it proves double-strand breaks, oxidation, and alkylation lesions or cross-links in DNA strand (Battal et al. 2015). The treatment of human lymphocyte culture with sodium arsenite showed dose-dependent increase in the frequency of SCE as recently reported by Bal et al. (2018a, b). Oxidative stress is produced due to the production of several reactive oxygen species (ROS), such as superoxide anion (O_2–), hydroxyl radical (•OH), hydrogen peroxide (H_2O_2), singlet oxygen ($^1O_2$). Superoxide anion is the primary species (Valko et al. 2006) produced by endothelial cells following exposure to As. Arsenite (As^{III}), the most toxic inorganic As species can affect the expression of several genes such as stress-responsive genes, DNA repair, and damage-responsive genes, and the activation of transcription factors such as the AP-1 complex (Salazar and Ostrosky-Wegman 2015). Arsenite can also enhance the release of proinflammatory cytokines, influencing the response to acute arsenic toxicity (Liu et al. 2001b).

The p53 antioncogene encodes a transcription factor and key regulator for sustaining genome integrity. p53 is triggered in response to several stress signals such as UV induced DNA damage and promotes the transactivation of a number of downregulatory genes such as p21/CDKN1A (prevents cell cycle progression and let the cell for DNA repair before entering into S phase) (Kastan et al. 1991; Derheimer and Kastan 2010), GADD 45, MDM2 or Bax (causes apoptosis via downregulation of Bcl-2 in cells with severely damaged DNA) (Miyashita et al. 1994). Mutation of the P53 gene is very common in cancerous cells (Greenblatt et al. 1994) and suggested to be a potential target of arsenic (Salazar and Ostrosky-Wegman 2015). Exposure to high doses of arsenic increases the cellular level of p53 and induces DNA damage (Martinez et al. 2011a, b; Muenyi et al. 2015), which is followed by G1 cell cycle arrest, allowing DNA to repair or trigger apoptosis (Liu et al. 2003; Louria-Hayon et al. 2003). Following As-induced DNA damage, p53 promotes cell cycle arrest through activation of p53 target gene CDKN1A/p^{21} (Vogt and Rossman 2001; Yih and Lee 2000). The p53 protein also plays a crucial role in reducing intracellular ROS to non-toxic levels through transactivation of antioxidant genes (namely Sestrin 1 (SESN1), Sestrin 2 (SESN2) and glutathione peroxidase-1 (GPX1)) (Sablina et al. 2005). Mutation in the P53 gene significantly decreases the basal transcription of these antioxidant genes, leading to an upsurge of ROS and successive oxidative damage of DNA. In the absence of p53 surveillance ROS may exert major changes in cell proliferation patterns from regulated toward malignancy in response to a low dose of arsenic exposure (Ganapathy et al. 2016). Unusual increases of ROS and combined oxidative stress caused by arsenic exposure leads to an increase in mitochondrial transmembrane potential and activation of unfolded protein reaction (UPR) crucial for tumor development (Srivastava et al. 2013; Hunt et al. 2014). In response to arsenic exposure, p53 appears to be compromised before cells progress to malignant transformation (Ganapathy et al. 2014, 2015). A recent report by Ganapathy et al. (2016) also supports that low dose of arsenic exposure suppresses p53 function and upregulates ROS which induces tumorigenesis via triggering endoplasmic reticulum (ER) stress and further promoting unfolded protein reaction (UPR) as observed in human keratinocytes and lung epithelial cells.

7 Arsenic-Mediated Epigenetic Modulation of Gene Expression

Perturbation of cellular oxidative balance by generating excess reactive oxygen species (ROS) (Bailey and Fry 2014) subsequent to As exposure appears to be one of the epigenetic regulators of As toxicity in exposed aerobic cells (Cerda and Weitzman 1997). Besides Arsenic, several studies demonstrated that other metals such as nickel, cadmium, and lead induce epigenetic alterations (McVeigh et al. 2001; Dolinoy et al. 2007; Bleich et al. 2006). Metals aid to increase the synthesis of reactive oxygen species resulting in oxidative DNA damage that can alter the ability of methyltransferases to interact with DNA and leads to generalized altered methylation of cytosine residues at CpG sites (Baccarelli and Bollati 2009). The epigenetic modification does not comprise changes in the nuclear DNA sequences, but it modifies the processes that govern heritable changes with chromosomal markers in genomic expression (Feinberg and Tycko 2004; Cortessis et al. 2012; Ray et al. 2014). Epigenetic alteration, therefore, has potential to make a reversible variation in DNA and a prime mechanism for next generations to better sustain the biotic and abiotic stresses (Ren et al. 2011; Yong Villalobos et al. 2015; Talukder 2017). Three major epigenetic mechanisms, DNA methylation, covalent posttranslational modifications of a histone protein, and noncoding RNA-mediated gene silencing, are known each playing specific roles in the regulation of gene expression (Paul and Bhattacharjee 2016). A variety of environmental toxicants or xenobiotics can cause disease manifestation through epigenetic deregulation. Being an environmental toxicant and carcinogenic agent, As acts as an epimutagen and can induce epigenetic modifications that lead to different types of diseases including cancer.

7.1 DNA Methylation by Arsenic Exposure

Although the role of DNA methylation in inducing different types of cancers have been extensively studied, very few studies have been made in arsenic-induced cancers. Chronic exposure to arsenic exhausts *S*-adenosylmethionine (SAM), the main methyl donor required for arsenic detoxification (during biotransformation of iAs to MMA and DMA), that sometimes reduces the consumption of methyl groups by a cellular device for different methylation activities leading to overall 'global hypomethylation' (Hubaux et al. 2013). Epigenetic alteration associated with As mainly occurs due to the scarcity of methyl ($-CH_3$) groups within the cell following the altered availability of SAM, the chief provider of a methyl group for DNA methyltransferases (DNMT) (Chatterjee et al. 2017a). This hypomethylation leads to a decrease in the activity of DNMT i.e., loss of CpG methylation status of promoters for some methylated-silenced genes which ultimately lead to activation of specific genes along with oncogenes (Deleted in Bladder Cancer 1 (DBC1), Hras1 and c-myc) (Cui et al. 2006; Chen et al. 2001; Takahashi et al. 2002). Furthermore, hypomethylation leads to a decrease in LINE1 methylation and total 5-methyldeoxycytidine content in lymphoblastoid cells (Intarasunanont et al. 2012).

However, hypermethylation of promoters of several tumor suppressor genes like p15, p16, p53, and death associated protein kinase (DAPK) by As exposure have also been reported in a dose-dependent manner (Chanda et al. 2006; Zhang et al. 2007; Fu et al. 2010; Ren et al. 2011). Hypermethylated DAPK gene was found in urothelial carcinoma patients living in As-contaminated areas (Chen et al. 2007). Thus reports show that both epigenetic mechanism, i.e., hypomethylation and hypermethylation, leads to deregulation of gene expression and malignancy by upregulating oncogene expression and downregulating tumor suppressor genes, respectively (Reichard and Puga 2010; Ziech et al. 2011; Ren et al. 2011).

7.2 Histone Modification by Arsenic Exposure

Nucleosomal histone modifications provide another mechanism for As mediated epigenetic gene regulation. The Nucleosome core particles, the basic structural unit of chromatin, consists of stretches of DNA (~146 bp) wrapped around a histone octamer consisting of two copies of each core histone (H2A, H2B, H3, and H4) (Luger et al. 1997). Histone H1 is not part of the nucleosome core, but it stabilizes the linker DNA strand wrapping the "bead," forming the "pearl necklace" structure (Luger et al. 1997). The N terminal tails of the core histone protein (particularly those of histones H3 and H4) contains positively charged amino acids like lysine, arginine and serine (Bannister and Kouzarides 2011) that are subjected to frequent posttranslational and covalent modifications including acetylation, methylation, phosphorylation, citrullination, ubiquitination, sumoylation, ADP ribosylation, deamination, and proline isomerization (Kouzarides 2007; Bannister and Kouzarides

2011; Ray et al. 2014). Arsenic-mediated histone modifications include methylation and acetylation (Paul and Bhattacharjee 2016).

The key enzymes catalyzing the dynamic and reversible histone acetylation are histone acetyltransferases (HATs) and histone deacetylases (HDACs) (Glozak and Seto 2007). Evidence for As mediated epigenetic alteration through altered histone acetylation has been reported by a number of authors (Jensen et al. 2008). Alteration in histone H3 acetylation has been observed in As^{III}- and MMA^{III}-exposed human urothelial cells (in vitro) undergoing malignant transformation (Jensen et al. 2008). As^{III} induced histone acetylation is responsible for the up-regulation of genes involved in apoptosis or in stress response and probably is mediated by HDACs (Li et al. 2002, 2003). As^{III} was found to hinder HDAC gene activity that can be correlated with increased global histone acetylation (Ramirez et al. 2008). Another report of reduced histone acetylation (H4K16) and increased cytotoxicity was found in a dose and time-dependent manner by both As^{III} and MMA^{III} treatment in human bladder epithelial cells. MYST1 mediated H4K16 acetylation is necessary to protect the cells against arsenicals (Jo et al. 2009).

Di-and trimethylated forms of lysine (K) residues within the histones are imperative to chromatin conformation and maintaining its supercoiled structure. In vitro studies revealed that As can modify the methylation of these residues and have been associated with the advancement of several types of cellular dysfunctions and even tumorigenesis (Paul and Bhattacharjee 2016). e.g., an increased rate of methylation of H3K4me3 and H3K9me2 was noticed due to increased levels of histone methyltransferase G9a protein under As^{III} stress (Zhou et al. 2009). Arsenic-mediated epigenetic regulation of gene expressions can be accompanied by histone methylation and recruitment of transcription factors to the binding site of the promoter as was found in ferritin gene expression, an arsenic-responsive gene (Huang et al. 2013). Ferritin gene, under oxidative stress, is transcriptionally upregulated through activation of the antioxidant responsive element (ARE) by the transcription factor Nrf, thereby, providing an important cellular defense mechanism by chelating excess intracellular free iron, thus lessening hydroxyl radical formation. The study of Huang et al. (2013) showed that arsenic induces histone H4R3 and H3R17 methylation in human keratinocytes, along with the nuclear accumulation of PRMT1 and PRMT4, which is accompanied by Nrf2 recruitment to the ARE in the promoter site and upregulates the expression of antioxidant gene ferritin.

7.3 miRNA Mediated Gene Silencing by Arsenic Exposure

Potential roles of miRNAs in metal stress responses in plants have been studied extensively (Sunkar et al. 2006 ; Zhou et al. 2010 ; Li et al. 2012). Studies support that miRNAs mediated gene regulation is another epigenetic mechanism that affects development, growth, and response to stress in humans as well (Zhou and Xi 2018). Marsit et al. (2006) reported that exposure of human lymphoblasts to 2 μM NaAsO for 6 days altered the expression of several miRNAs including miR22, miR34a, miR210, miR221, and miR222. While the expression of mi R210 was found to be repressed, the expression of miR22, miR34a, miR210, miR221, and miR222 was increased (Marsit et al. 2006). Another report shows miR19a and miR29a mediated apoptosis in the human bladder (T24) and hepatocellular carcinoma (HepG2) cells, respectively (Cao et al. 2011 ; Meng et al. 2011). Reduced expression of miR181b and miR9, and expression of Nrp1 resulted in cell migration, tube formation, and angiogenesis, which are otherwise under control in a normal cell (Wang et al. 2011 ; Cui et al. 2012 ; Hubaux et al. 2013). A study by Wang et al. (2011) showed that chronic arsenite exposure leads to repression of miR-200 which is mediated by EMT-inducing transcription factors ZEB 1 and ZEB 2 (zinc-finger E-box-binding homeobox factor) and progressive methylation of miR-200 promoters ultimately leads to malignant transformation of p53-knocked down human bronchial epithelial cells. Very recently it is proved that differential expression of miRNAs and target genes at the early stage of arsenite exposure may contribute to arsenic-induced carcinogenesis (Al-Eryani et al. 2018). The potential role of miRNA in mediating the response to arsenic exposure was evidenced from the study of Chen et al. (2017) which showed that inhibition of miR-155 by its specific inhibitor could remarkably mitigate the malignant growth and promotes apoptotic cell death in arsenite-transformed cells. A similar report was found in lung adenocarcinoma cells, where, inhibition of miR-155 succeeded in reduced cell proliferation, and triggered cell death (Lv et al. 2016).

8 Conclusion

Arsenic naturally occurs in the earth's crust. However, the toxicity of the metalloid has created human misery in many parts of the world. Several researchers are working to develop technologies for As toxicity mitigation and uplift millions of people from this dreaded situation. Arsenic enters into the human body either through contaminated drinking water, food or inhalation. Both at genetic and epigenetic levels, As biotransformation into As^{V}, As^{III} and its methylated conjugates play an important role in its carcinogenicity. Arsenic may interfere with the cellular process through the production of reactive oxygen species, which can induce or suppresses metabolically active proteins/enzymes related to DNA damage repair, cell cycle regulation, epigenetic abnormalities, etc. Chronic arsenic exposure leds to alteration in EGFR, PI3K/AKT pathways visibly linked to the development of oncogenicity. Molecular mechanisms of tumorigenesis and malignant transformation in the human body due to As toxicity should be investigated to produce more favorable translatable results for early mitigation. It is also essential to make As-related research and information readily available in a common platform for assessing risks and decision supports. Cloud-based computing and the Internet of things (IoT) may help to congregate data to support affected communities in evaluating risks due to arsenic exposure (Singh 2017). Therefore, an integrated study on various aspects like environmental monitoring, health surveillance, exposure data, individual risk characterization, and human biomarkers are required to provide in-depth perception into the pathophysiological complexity for the genesis of As related toxicity and diseases processes for developing specific therapeutic and risk management strategies against it.

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Chapter 6
Sustainable Arsenic Mitigation: Problems and Prospects in India

Nandita Singh and Om Prakash Singh

Abstract Arsenic contamination of groundwater has emerged as a significant challenge for the human right to water in India because of over 80% dependence on groundwater for drinking, particularly in the rural areas. In order to mitigate the arsenic menace and provide safe drinking water to the masses, government and non-governmental agencies have initiated several interventions. These interventions can be categorized under one of the following approaches, namely, treatment of contaminated water; supply of groundwater with acceptable level of arsenic; surface water supply; and rainwater harvesting. While it is a reality that all populations estimated to be 'at risk' of arsenic exposure have not yet achieved 'coverage' under one or more of these approaches, there exist more pertinent issues. These can be summarized under the rubric of 'sustainability' of the interventions, primarily from environmental, social, cultural and economic perspectives. This chapter evaluates the different arsenic mitigation interventions in India from these different sustainability perspectives, and proposes that rainwater harvesting offers the most promising way forward for sustainable arsenic mitigation.

1 Introduction

High concentration of arsenic in groundwater is reported from several states of India. Among earliest known instances is West Bengal, first reported in 1983 (Garai et al. 1984), followed by Chattisgarh in 1999 (Chakraborti et al. 1999), in Bihar and Uttar Pradesh in 2003 (Chakraborti et al. 2003; Ahamed et al. 2006), in Jharkhand in 2004 and Rajasthan in 2012 (Chakraborti et al. 2018). According to recent

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research publications, arsenic above the WHO standard of 10 μg/l is presently reported in the groundwater in eight states: in 25 out of 75 districts of Uttar Pradesh, 22 out of 38 districts of Bihar, three out of 24 districts of Jharkhand, three out of 33 districts of Rajasthan, 14 out of 22 districts of Haryana, two of 27 districts of Chhattisgarh, one of 11 districts of Delhi, and 14 of 19 districts of West Bengal (Chakraborti et al. 2018). However, according to Government of India sources, more than 50 μg/l of arsenic in ground water is reported from parts of ten states, namely, West Bengal, Assam, Bihar, Jharkhand, Uttar Pradesh, Punjab, Haryana, Chhattisgarh, Karnataka and Manipur (MOWRRDGR 2017). Moreover, arsenic concentration more than 10 μg/l is reported from patches in additional 11 States/Union Territories. In all, about 239 million people across 153 districts in 21 states are estimated to be drinking water that contains unacceptably high levels of arsenic. This is almost one-fifth of the country's population. Further, according to information lately tabled by the government in the Parliament, the total number of arsenic-affected persons in the country is about 14.8 million, with more than 10.4 million in West Bengal, 1.7 million in Bihar, and 1.5 million in Assam (Anonymous 2017). Calculations based on the recent government information reveal that 65% of Assam's population (about 21 million), 60% in Bihar (about 59 million) and 44% in West Bengal (about 40 million) are exposed to arsenic-contaminated drinking water. In terms of absolute numbers Uttar Pradesh has the largest population at risk – over 65 million, while the total figure for the country could be up to 239 million (Jadhav 2017).

In addition to drinking water, arsenic contaminated water also affects large populations through the food chain (Singh and Ghosh 2011). Vegetable crops grown in arsenic-rich groundwater adds manifold to daily arsenic intake, reported to be as high as 560 and 393 μg of arsenic for adult and children, respectively (Santra et al. 2013). Highest arsenic accumulation has been reported in potato, brinjal, arum, radish, lady's finger and cauliflower, while lower level has been observed in beans, green chilli, tomato, bitter gourd, lemon and turmeric. Besides, mustard, pea, Mung bean and rice are also substantially affected (Santra et al. 2013), and even milk has been found to be seriously affected in areas where groundwater is contaminated with arsenic (Rana et al. 2007). An additional concern is that food crops grown using arsenic contaminated water are sold off to other places, including uncontaminated regions nationally as well as internationally where the inhabitants may end up suffering from arsenic toxicity from consumption of contaminated food (NIH and CGWB 2010).

Exposure to arsenic-contaminated water and food rings alarm bells from the perspective of the human rights of the masses (Kumar et al. 2016). According to the General comment No. 15 on the Right to Water adopted by the UN Committee on Economic, Social and Cultural Rights in 2003, "the water required for each personal or domestic use must be 'safe', therefore free from … chemical substances, … that constitute a threat to a person's health" (CESCR 2002). Moreover, the International Covenant on Economic, Social and Cultural Rights (ICESCR), 1966 under article 12 recognizes the human right to health as "the right of everyone to the enjoyment of the highest attainable standard of physical and mental health", for which access to 'safe' water is the foremost "underlying determinant" (CESCR 2000). Prolonged ingestion of water containing arsenic concentrations higher than 10 μg/l causes multiple health problems, and so does high doses of arsenic through the food chain. More than 5 years of exposure can result in carcinogenic and non-carcinogenic health issues such as melanosis, keratosis, skin lesions, neurological disorders, liver damage, respiratory complications, and various kinds of cancers (Jain and Chandramani 2018; Kumar et al. 2016; Abdul et al. 2015; Ravenscroft et al. 2009).

According to the Universal Declaration of Human Rights proclaimed by the UN General Assembly in 1948, "everyone has the right to a standard of living adequate for the health and well-being of himself and of his family" (Article 25) as well as the right to work (Article 23) which includes "the opportunity to earn a livelihood by work freely chosen or accepted", towards which States are obliged to take appropriate measures to develop an enabling environment for productive employment opportunities (UNGA 1948). The impact of arsenic exposure on health can progressively interfere with the enjoyment of these rights by affected populations since an unhealthy body leads to unhealthy minds and appreciably lowers physical capacity at work, in some cases also leading to death, plunging dependent families into deeper circles of deprivation and poverty. Arsenic contamination of groundwater and hence drinking water sources and food are thus also a great impediment to sustainable development (Johnston 2016).

In light of this, mitigation of arsenic in water consumed for drinking and irrigation becomes a significant obligation for the government in India which is a signatory to all the major human rights instruments of the United Nations. India is a federation of states and water is a 'state' subject under the Constitution. Consequently, it is the State governments which are directly responsible for adopting policies and implementing interventions for provision of safe drinking water to the affected populations. The Government of India has also taken an active role in guiding and supporting the State governments in terms of policy as well as financial resources in undertaking appropriate action towards mitigation of the arsenic menace in drinking water, though the government appears to be less attentive to arsenic in the food chain.

It is a fact that though large populations as noted above are 'at risk' of exposure to excess arsenic through drinking water and that the number of people already known to be affected is large, not all affected pockets are yet 'covered' under arsenic mitigation interventions. While a good number of locations are quite recently identified, a greater concern is whether people living in habitations already 'covered' through safe water interventions are able to enjoy their human right to water and the concomitant rights as described before? Evidence suggests that many of the arsenic mitigation interventions are unable to provide arsenic-safe drinking water to the target populations due to a multitude of reasons. For example, some become dysfunctional soon after installation, some are abandoned by the users, while still others fall inadequate in fulfilling the demand (SOES 2004; Singh 2012b; Hossain et al. 2015; Kumar et al. 2016). In light of this, the pertinent questions to arise are:

Are the various arsenic mitigation interventions already in place in the arsenic-affected areas of India sustainable? What kinds of challenges confront sustainability of these mitigation solutions? How can arsenic mitigation be made more sustainable so that the human rights of the ‘at risk’ populations are protected?

This chapter attempts to evaluate the different arsenic mitigation approaches and interventions in India from sustainability perspective, identifying the factors that influence their sustainability, and finally, proposing a way forward. The main focus is on arsenic mitigation interventions in India with respect to drinking water, but implications for the food chain are also considered. The chapter is organized into six sections. In the second section, the conceptual framework used for analyzing the sustainability of arsenic mitigation interventions will be presented. The interventions adopted for mitigating arsenic in drinking water in different States encompass a variety of strategies and technologies. The third section will describe the approaches and interventions adopted for arsenic mitigation in India. In the fourth section, an overview of the approaches adopted by the different arsenic affected States in India will be presented. In the fifth section, the sustainability of the different arsenic mitigation interventions will be outlined, as seen from social, cultural, environmental and economic perspectives. Finally, the sixth section will summarize the sustainability perspectives on arsenic mitigation and identify a way forward for addressing the current limitations that can enable the women, men and children ‘at-risk’ to enjoy their human right to water, human right to health and other related rights.

2 Methodology

The findings presented in this chapter are based upon long-term ethnographic research on arsenic mitigation undertaken by the authors in the states of West Bengal and Bihar. First-hand field research was undertaken in a total sample of 46 villages in these two states. Primary data was collected from these villages through intensive residential fieldwork in connection with aspects like sufficiency, appropriateness (and thereby acceptability), accessibility, and affordability of the arsenic mitigation interventions. Data collection primarily rested upon qualitative techniques like participant observation, key-informant and household level interviews, case studies and focus group discussions (FGDs). Besides, visualizing tools, like social and resource mapping, were also used. The informants included women as well as men from different segments of local community. Data analysis with respect to these criteria was primarily qualitative; first, the data pertaining to the ongoing interventions and their sustainability was classified under different criteria, and then compared to arrive at generalizations and identify trends. Detailed interviews and FGDs that deal with implementation of technological interventions for arsenic mitigation were also organized with key representatives from – governmental, non-governmental and international agencies. Also, scientists active in research on the issue were consulted. This chapter further draws upon findings of similar research undertaken by the authors in Rajasthan and Gujarat where water quality has been an important historical concern and local communities have adopted traditional nature-based solutions to address the problem. The learnings from studies on these nature-based solutions in these quality-affected areas, that also offer the benefit of climate resilience as an added value, have been incorporated in attempting to define a way forward.

3 Conceptual Framework for Analyzing Sustainability of Arsenic Mitigation

The word *sustainable* comes from the Latin *sustenare* which means “to hold up”, “to maintain” or “to support”. ‘Sustainability’ therefore implies ‘the ability to be maintained at a certain rate or level’[1] or “the ability to keep something going for a long time” (Sutton 2004). The notion of sustainability has been applied to a diversity of contexts, including development, technology, business, agriculture, livelihoods, education and many others, each bringing forth its own perspective (Giovannoni and Fabietti 2013).

The usage of the term ‘sustainability’ in the current form was established at the 1972 UN Conference on the Human Environment held in Stockholm. It was further developed and holistically described by the World Commission on Environment and Development (WCED), also called the Brundtland Commission, in 1987 through the concept of ‘sustainable development’ (Sutton 2004). Sustainable development denotes “development that meets the needs of the present without compromising the ability of the future generations to meet their own needs” (WECD 1987). This concept considers three pillars of sustainability, namely, ecological, social and economic, recognizing that all must be considered together to find lasting prosperity in society. Since then, these three pillars have been widely applied to identify, pursue and describe sustainability of efforts and actions in different contexts. In recent years, cultural sustainability has been considered as the fourth pillar, based on the argument that culture is a necessary foundation for the transition to a truly sustainable society (Soini and Birkeland 2014).

Sustainability can be described as a shared quality concerning a valued system, object, process or attribute in society and a question concerning sustainability may arise whenever there is a perception of threat or risk about its persistence or excellence (Sutton 2005). The shared quality connected to the valued system, object, process or attribute could be defined along diverse axes, primarily social, environmental, economic, and cultural. In the context of arsenic mitigation, the goal of sustainability is important in two senses: first, in terms of sustainability of the interventions implemented for mitigating the arsenic menace, and second, in terms

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[1] URL: https://en.oxforddictionaries.com/definition/sustainability. Accessed on 12 Feb 2018.

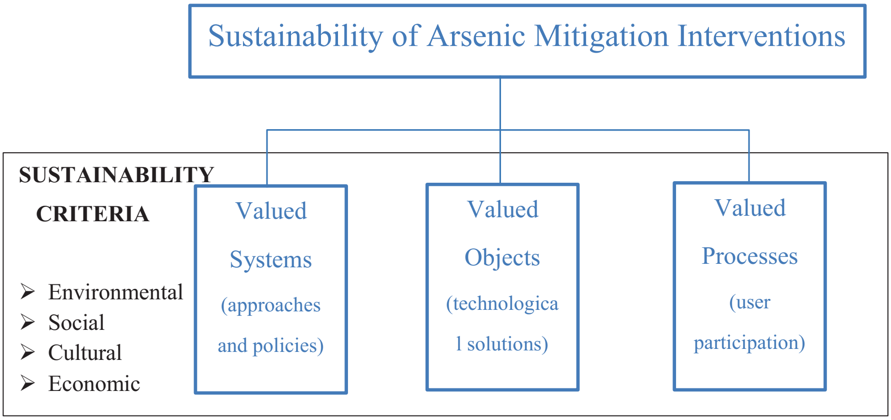

Fig. 6.1 Conceptual framework for evaluating sustainability of arsenic mitigation interventions

of their implications for sustainable development and promotion and protection of the basic human rights within a context of large-scale arsenic contamination in groundwater. It is obvious that failure to reach sustainability in the first sense would imply failure in fulfilling the second one too. It is therefore important to assess the sustainability of different arsenic mitigation interventions. These interventions may be seen as valued systems (approaches and policies), objects (technological solutions) or processes (user participation).

Sustainability of the arsenic mitigation interventions could be conceptualized as "the ability to continue to be implemented /used/managed for prolonged periods of time and delivering the same output in the same way as at the beginning, so as to help meet the needs of the present without compromising the ability of the future generations to meet their own needs of accessing arsenic safe water for drinking, livelihood and other uses". Sustainability of arsenic mitigation interventions could be evaluated against each of the four parameters of sustainability: social equity, cultural acceptability, environmental sustainability and economic efficiency. The conceptual framework for evaluating sustainability of arsenic mitigation interventions is represented in Fig. 6.1.

4 Approach for Arsenic Mitigation in India: An Overview

The basic approach in arsenic mitigation in India has been provision of 'safe' drinking water to the affected populations, whether as immediate relief or as a long-term solution. The approach for supply of safe drinking water in turn consists of four different strategies, namely, (a) treatment of contaminated water, (b) supply of groundwater having acceptable level of arsenic, (c) surface water supply, and (d) rainwater harvesting. Under each of these strategies, different technologies have been developed and provided as options to water users in the local communities, particularly in the rural areas. These are briefly summarized below.

4.1 Treatment of Contaminated Water

A number of technologies have been developed for the removal of high concentrations of arsenic from drinking water. Some of these are developed in India while some others represent imported technologies. In general, the technologies are designed based upon the following processes: (a) oxidation and sedimentation, (b) coagulation, (c) sorptive filtration, (d) ion exchange, and (e) membrane techniques. Other potential approaches include phytoremediation or the use of bacteria, which can play an important role in catalyzing biological arsenic removal processes. All the arsenic treatment technologies ultimately concentrate arsenic in the sorption media, the residual sludge or in a liquid media. The treatment technologies are very commonly installed as community water supply points on hand pumps or sometimes through public tap stands. This option was widely adopted in West Bengal in the beginning and has since been seen as a short-term immediate solution in India. A major limitation of this approach is the high risk of environmental pollution from the used media due to leaching of arsenic into the ground or water resources and soil (Panda 2015).

4.2 Supply of Groundwater with Acceptable Arsenic Levels

For this purpose, deep aquifers separated from shallow contaminated aquifers by impermeable layers are believed to be a dependable source. Consequently, installation of deep tube wells for tapping such aquifers is the common technology (WB/WSP 2005). Such water can be further provided conveniently at community level through household-based piped water schemes or through community points. Dug wells that accumulate groundwater from the top layer of a water table, which is replenished each year by arsenic-safe rain and percolation of surface waters through aerated zone of the soil, are also found to represent a technological option even in areas where tube wells are contaminated. The water can be used for drinking and cooking after minor treatment using chlorination and lime (WB/WSP 2005). Converting them into sanitary wells is proposed as a way forward for protection against microbial contamination.

4.3 Surface Water Supply

Surface water supply has been recently seen in India as the best long-term option for arsenic mitigation (MDWS 2013). This approach presents several additional advantages such as water delivery near home, protection from external contamination, better quality control through monitoring and convenience in collection of water. Water can be made available through house connection, yard connection, or stand post, depending on affordability of each option to the consumer. However, despite the benefits, cost effectiveness of the option has been an important issue in question, especially where the settlements to be served are scattered. Also, in this option, the problem of biological and chemical contamination is to be addressed through treatment and disinfection if surface water is to be used for drinking. The supply source is more commonly from rivers, but even local ponds, lakes and tanks can be considered.

4.4 Rainwater Harvesting

Rainwater harvesting is recently being promoted as a viable option in arsenic affected areas. Rainwater harvesting has been seen as a solution for arsenic problem in two ways: individual household-based rooftop rainwater harvesting which can provide potable water for families or harvested surface water run-off which can be used to recharge high arsenic groundwater resources, thereby diluting the concentrations (DDWS undated; Hossain et al. 2015).

However, while rainwater is usually seen as a much cleaner water source and may provide a low-cost solution, among technological limitations for the rooftop variety identified in rural areas are limitations of roof area, storage capacity and cost effectiveness that may actually prove to be disadvantageous as an option for the poorer segments. Also, appropriate maintenance of the system is seen as presenting several difficulties (WB/WSP 2005; Hossain et al. 2015).

5 Arsenic Mitigation in India: State Perspectives

Of the 21 states affected by high arsenic in groundwater, some have initiated interventions for its mitigation. In this section, the policies and strategies adopted in the four worst affected states, namely, West Bengal, Bihar, Uttar Pradesh and Assam will be outlined. West Bengal, the first state to identify arsenic as a public health concern, has designed several interventions for assessing the threat and mitigating it for over a period of more than three decades. Supported by Rajiv Gandhi Drinking Water Mission from the Centre, the first Steering Committee was constituted by the State Government in 1988, which conducted a multi-centric study to find out the cause of the problem. Thereafter a number of Task Forces have been constituted, with representation of all the academic and research institutes working on arsenic and groundwater quality-related problems. Lately, the State Government has developed a Master Plan for long term mitigation for the problem, which is presently in the process of implementation with financial support from the Centre (Planning Commission 2007).

Arsenic toxicity is known in nine districts of West Bengal and the State has received substantial financial support from the Government of India for undertaking action for mitigation (Santra 2017). In 2005, the Planning Commission constituted a Task Force to work out modalities of combating the menace of arsenic pollution with identification of appropriate technologies. On the whole, in West Bengal, arsenic removal technologies have been given considerable priority especially as a short-term remediation measure, with their installation starting at the end of 1998. By 2005, the West Bengal government and other organizations invested about three million USD in installing arsenic removal plants (ARPs) purchased from both national and international manufacturers (Hossain et al. 2005). 1900 units were set up at an average price of 1500 USD for each in five out of nine arsenic affected districts of West Bengal. Several community-based units installed in the State are based on sorptive filtration technique using activated alumina, such as the units manufactured by Alcan, Apyron, Oxide (India), and RPM Marketing. Granular ferric oxide is used as the media in units produced by Pal-Trockner, while the READ-F arsenic filter from Japan uses hydrous cerium oxide as the adsorbent. Besides, coagulation-sedimentation-filtration and ion-exchange techniques have also been used (Hossain et al. 2005; Santra 2017). In fact, a Technology Park was set up in Baruipur block of South 24 Parganas district where 19 ARPs from 11 different national and international manufacturers were installed for evaluation in terms of their arsenic removal efficiency, cost-effectiveness and user-friendliness to help the State Government and other agencies in selecting the appropriate technologies. The 'Technology Park Project' was implemented by All India Institute of Hygiene and Public Health (AIIH&PH), Govt. of India, Kolkata, in partnership with a number of NGOs under the financial support from India-Canada Environment Facility (ICEF) (Hossain et al. 2005). Based on the findings of the report from this park, the government installed over 2000 ARPs in different arsenic affected habitations, but very few countable ones have proved to have sustained the test of time (Planning Commission 2007). A good number of domestic filter units based on different techniques have also been developed and distributed in a number of districts, the operational life of which are varied (Singh et al. 2009).

Among medium- and long-term mitigation alternatives, surface water-based piped water supply has been implemented in some areas. For example, water from River Hooghly is being treated and made available to the villagers through piped distribution system in Nadia and North 24 Parganas districts. Where piped water supply schemes are based on groundwater, arsenic removal plants are attached to the supplying tube well, and in some places, pond-based schemes where rainwater is harvested and then treated for supply through pipes or standpoints. On the whole, the policy for arsenic mitigation adopted in West Bengal shows a gradual shift from dependence on arsenic removal technologies to use of surface water. In some places, water supply based on deep tube wells in the range of 150–300 m has also been initiated but questions over sustainability of the option have been raised (Ghosh and Singh 2009).

In Bihar, arsenic was first reported in 2002 in Bhojpur district and since then it has come to be reported from 17 out of 38 districts (Kumar et al. 2016). With support from UNICEF, the Public Health Engineering Department (PHED) of the State government, which is the nodal agency for arsenic mitigation, has adopted a multi-pronged approach for arsenic mitigation. According to senior PHED officials, for safe water supply in the affected areas, piped water schemes based on surface water is the first approach in Bihar's mitigation policy. Since the arsenic-affected areas lie close to the Ganges, a number of such schemes based on Ganges water are being executed. The first of these was executed in Bhojpur district, which is designed to serve affected population in 39 villages for the next 30 years, with a capacity of 40 litres per capita per day (lpcd) and supply time of 2 hours a day. Here water is first drawn from the Ganges, thereafter, treated through conventional methods and then disinfected using sodium hypochlorite, to be finally delivered to villagers primarily through community water points (Kumar et al. 2016).

The second approach in the PHED policy in Bihar State has been sanitary wells since the dug well water is free from arsenic contamination and also fairly acceptable to the people as an age-old practice. The water can be used for drinking and cooking after minor treatment using chlorination and lime. At many places, dug wells which had mostly fallen out of use have been rejuvenated and converted into sanitary wells. UNICEF has constructed 50 sanitary wells in 11 districts. According to government reports, more than 300 new wells have also been constructed and fitted with hand pumps, including many at schools (NIH and CGWB 2010). The third approach has been mini-piped water schemes based on groundwater abstraction from arsenic-safe deep aquifers. The first of these was in Semaria Ojhapatti village in Bhojpur district, with more being planned in other affected districts. The fourth approach is based on rooftop rainwater harvesting, actively promoted by UNICEF in Bihar. ARPs are the last priority in Bihar's mitigation strategy, promoted in isolated pockets where any other option is not viable. The most common techniques are based on adsorption/ion-exchange process (Kumar et al. 2016). According to PHED plans, arsenic removal units are to be installed in 700 schools with two filters in each school to supply arsenic-safe treated groundwater. Extensive IEC activities and awareness programmes are also being organized at District, Block and Panchayat level all over the state to educate the rural masses (Kumari and Maurya 2016).

In Uttar Pradesh State, arsenic contamination of groundwater was first recognized in 2003. According to recent studies, arsenic in groundwater above 50 μg/l is now identified in 31 out of 70 districts (Namrata et al. 2015). For providing safe water to the affected communities, the initial strategy adopted in 2006 and 2007 was to install deep tube wells by tapping a deeper third layer beyond 70–100 m below ground level, which was found to be arsenic-free. Under this strategy, 475 affected habitations of Ballia and Lakhimpur Kheri districts were provided with hand pumps on deep tube wells (UPJN 2018).

The above approach was however discontinued in 2008, following a decision from Government of India (MDWS 2011). Then the alternative mitigation strategies left were: (a) use shallow wells as sanitary wells for providing arsenic safe water, which also is not a safe source as discovered by later testing in Bahraich district, (b) install community-based filters and provide home-based filters with proper community orientation for arranging backwashing and proper disposal of filtered arsenic which is highly concentrated in nature, (c) providing surface water source by lifting water from rivers such as Ganga and Ghagra, which is highly capital intensive and long term mitigation measure, and (d) community awareness. After planning was made for sanitary wells in 492 habitations, threat of occurrence of arsenic in shallow aquifers put a halt on this mitigation measure too (MDWS 2011). Meanwhile, UNICEF advocated the efficacy of approach of treatment of arsenic contaminated water in the absence of alternate mitigation measure and supported a pilot project for providing community-based arsenic removal filters at seven locations in Lakhimpur Kheri, Bahraich, Gorakhpur and Ballia districts (MDWS 2011). Subsequently, the State government approved installation of 310 such community-based filters in Ballia and 500 more in Gorakhpur, Bahraich and Lakhimpur Kheri districts. Seven piped water supply schemes in district Ballia and five piped water supply schemes in district Lakhimpur Kheri utilizing surface water from rivers, lakes or ponds, which are free from arsenic contamination, have also been approved by the state government (UPJN 2018). Besides UNICEF, WaterAid has also contributed to the mitigation efforts especially through awareness generation and community management models for ARPs (MDWS 2011).

In the worst affected Bahraich district in Uttar Pradesh, the University of Miyazaki from Japan, together with Japan International Cooperation Agency (JICA) and the government of Uttar Pradesh, implemented an arsenic mitigation project during the period 2008–2012 (Jaiswal et al. 2010). The integrated mitigation approach implemented in the project included awareness-raising of villagers, installation of ten alternative water supply units (three filters for dug wells to treat the fecal coliform bacteria and seven arsenic removal plants) and healthcare for arsenicosis patients (Yano et al. 2012). For monitoring the progress of all mitigation interventions in the State and providing further guidance, a Central Team was set-up in 2011 to study the problem in its totality and suggest to the State Government a clear roadmap for surveillance of arsenic related problems and provide arsenic free drinking water to the populations of Ballia and other affected districts (MDWS 2011).

In Assam State, the mitigation plan encompasses several components, namely, extensive awareness generation in the arsenic endemic areas, tapping of alternate surface sources wherever feasible, promotion of rainwater harvesting system, use of domestic arsenic removal filters, hand pump attached community-based arsenic removal units, promotion of traditional ponds, and upgradation of existing dug wells to sanitary ring wells (APHED 2018).

Apart from the specific interventions adopted in the States, the centrally sponsored scheme National Rural Drinking Water Program (NRDWP), launched in 2009 and modified in 2012, aims at providing adequate and safe drinking water to the rural population of India. According to NRDWP guidelines, every rural citizen should have access to a basic minimum service level of 40 lpcd of safe drinking water, including inhabitants in the arsenic affected areas. This program has enabled the provision of significant additional resources to the sector and created an environment for the development of infrastructure and capacity for safe drinking water supply in rural areas, including arsenic-affected ones (MDWS 2017). With respect to water quality concerns, and hence arsenic mitigation, the Ministry of Drinking Water and Sanitation has asked all the States to commission surface water-based piped water supply schemes in all water quality affected habitations as a long-term sustainable solution. However, since commissioning of Piped Water Supply projects may take 3–5 years to complete and the rural people cannot be put to risk of consuming contaminated water during this period, all States were advised to install community water purification plants in reported arsenic affected habitations by March 2017 (MDWS 2017).

6 Sustainability of Arsenic Mitigation Interventions in India: Some Reflections

In this section, sustainability of the interventions described above is examined using the conceptual framework defined earlier, evaluating these in terms of social equity, cultural acceptability, environmental sustainability and economic efficiency. The reflections are based primarily upon evidence collected from the field through different methodological tools as described before, namely, first-hand ethnographic studies in arsenic-affected local communities, interviews and discussions with representatives of the government at Central and State levels and some of the non-governmental organizations active in the areas of study, and communication with scientists active in research on the issue.

6.1 Environmental Sustainability

Environmental sustainability entails a position whereby the arsenic mitigation interventions should have minimal negative impact on the environment in the short- as well as long terms. The policies and approaches adopted in the affected states of West Bengal and Uttar Pradesh, and also promoted under the NRDWP by the Central government, as examined above, show preference for ARPs, either as the ‘only feasible solution’ or ‘immediate relief’.

The ARPs, whether as community-level units or domestic filters, are based on complex chemical processes which produce arsenic-rich sludge on a day-to-day basis. Even regeneration of activated alumina and ion exchange resins used in the treatment units result in semi-liquid wastes that may be too arsenic-rich for simple disposal (Ghosh and Singh 2009; Mandal et al. 2016). According to the concept of Integrated Water Resources Management (IWRM), the basic components of the ecosystem, namely, groundwater, surface water, land and other natural resources, are closely interconnected and hence can deeply impact each other (GWP 2000). Therefore, while the drinking water may become arsenic-safe after treatment, the implications of the treatment process for the environment may be enormous. Because of inadequate attention to proper disposal of the sludge and other waste generated, arsenic keeps leaching back into soil, which may further get washed down into groundwater, and also directly contaminate surface water resources. Thus, ultimately drinking water supplies and water for other uses such as irrigation and animal husbandry in the affected communities get contaminated. This can also lead to poisoning of aquatic organisms such as fishes and of dairy products, thereby making arsenic an even greater public health and environmental threat (Mandal et al. 2016). This gigantic problem has unfortunately not yet been made an integral part of arsenic mitigation policies (Singh et al. 2008a; Singh 2012b).

None of the other three strategies, namely, surface water supply, supply of groundwater with acceptable arsenic levels and rainwater harvesting appear to possess such large-scale qualitative impacts on the environment. However, the strategy for drinking water supply based on surface water, as prioritized in Bihar and also promoted in Assam, and identified as the long-term solution within the NRDWP framework, can pose limitations of environmental sustainability in quantitative terms. About one-fifth of the country's population is estimated to be exposed to arsenic-contaminated drinking water. If each of these persons should be served through a piped water system progressively based on surface water sources as envisaged in the NRDWP, then such systems could soon become unsustainable. The water available in rivers, dams and lakes is limited, and due to several changes in the built environment of their catchments and over-abstraction of water, their water volume over the years has been progressively declining. This in turn leads to destruction of the aquatic ecosystems, with further negative impact on the terrestrial ecosystems nearby. Ultimately even the water needs of the innumerable dependent communities may remain unfulfilled on a long term. Several examples exist in the country where rivers and lakes have dried up due to over-abstraction of water and lowered catchment flows (Singh et al. 2016; Singh 2012a; Saleth 2011).

Similarly, supply of groundwater from deeper aquifers through deep tube wells, as promoted in West Bengal and Bihar, poses limitations from environmental sustainability perspective. For example, wasteful over-withdrawal and usage of larger quantities of precious arsenic-safe groundwater reserves for various domestic purposes besides drinking such as washing and bathing as observed in affected villages in West Bengal, can lead to substantial lowering of the water table in the long term, particularly if adequate attention to groundwater recharge is not paid (Saleth 2011). Furthermore, qualitatively, as discovered in Ballia district of Uttar Pradesh, absence of adequate capping between arsenic-containing shallow aquifer and arsenic-safe deep aquifer can lead to leaching of arsenic, converting an arsenic-safe source into a contaminated one (MDWS 2011). Use of contaminated water from this source can lead to not only public health issues but also environmental issues when considered in the long-term perspective. Such a concern has also been raised in the context of West Bengal (Chakraborti et al. 2001).

From the above observations, it is obvious that currently most of the different arsenic mitigation technologies suffer from one or another limitation especially when considered from the long-term environmental perspective, either qualitatively or quantitatively. These in turn have spill-over effects on other water use sectors besides drinking water. For environmental sustainability, it is important that technologies be aligned to local environmental conditions since they are part of the repertoire that help local communities adapt to the specific environmental stresses (Singh 2016b). It is also obvious that most of the technological solutions implemented for arsenic mitigation have been designed by 'experts' from outside as blanket 'one-fit-all' options and not been specifically tailored to 'fit' into the local milieu, developed 'jointly' with the local users (Singh 2008). Rainwater harvesting is the only technological option that local communities in the Gangetic basin have been traditionally associated with; but even in this case, the new version of rooftop rainwater harvesting does not integrate the perspective and knowledge of the local users. This has resulted in failure of this mitigation option (Kumar et al. 2016).

6.2 Social Equity

Social equity in the context of arsenic mitigation is rooted in the question of 'accessibility' to arsenic-safe drinking water sources. None of the policies and approaches presented above for arsenic mitigation have considered and incorporated the social realities of the arsenic-affected communities for which mitigation interventions are implemented. Policies and programs are based on 'headcounts', and a common assumption underlying installation of mitigation technologies is that once made available within a habitation, it will be used equitably by all targeted beneficiaries. Thus, program guidelines may contain details about identifying appropriate 'public' points in vicinity of the user community, but no monitoring is included in the program guidelines for verifying all beneficiaries have equitable right of access to mitigation technologies (Singh 2006a).

Physical proximity is just one of the factors determining the effectiveness of or accessibility to mitigation technologies. For example, given a choice between a contaminated hand pump at home and fetching water from a safe source at some distance, villagers interviewed in West Bengal during this study expressed inability to at least 'always' fetch water from a safe source. This was true for women who have several other domestic chores to attend to as well as men who 'work' outside home on a daily basis. This was further linked to three conditions: 'sensitivity' towards the outcome of arsenic exposure, the availability of help at home to procure water, and the daily water demand (i.e., the number of household members). Where the sensitivity was high enough (as expressed by the users themselves), efforts to fetch safe water from the mitigation intervention was prioritized despite other bottlenecks. It was observed that the sensitivity varied considerably across similar educational backgrounds (Singh et al. 2009; Singh 2006a).

Besides physical accessibility, social accessibility is an important concern in Indian villages, since social heterogeneities prevalent in the recipient communities influence the question of 'access' to the publicly placed arsenic mitigation technologies (Singh et al. 2005). A safe drinking water source may be physically located close to home, but it may not be accessible equally to all residents due to social rules of access to public resources prevalent in the community. In general, the principles of caste and social dominance define rules for sharing resources, including space and water. Castes are ranked, endogamous (in-marrying) groups, in which membership is determined by birth. Each caste is part of a local system of interdependent groups, linked through economic, ritual and social relationships (Beteille 1996). Caste-based principles in user communities stipulate that public spaces in the village are accessible to and usable by only those castes that reside in the immediate vicinity; and decision-making regarding such access and use often rests with the dominant elite. The dominant elite is often a caste that is highly placed in caste hierarchy, and enjoys numerical preponderance and sizeable ownership of village land (Srinivas 1959). Thus, castes lower down in the hierarchy may not be able to fetch water from arsenic-safe sources located in a social space identified as belonging to the dominant elite (Srinivas 1959; Singh 2006a).

Social patterns based on the principle of dominance also have an important bearing on inter-ethnic situations in mixed villages regarding access to and management of spaces and resources in the village by other ethnic groups like the tribals and in predominantly Hindu villages by other religious groups like Muslims and Christians (Singh et al. 2008b). In West Bengal, religion was found to be a basis for lack of social access to community-based arsenic removal units. In a sample of 35 units studied, at least eight instances were recorded where despite the target group being originally heterogeneous, the actual users of the unit belong only to the dominant religious segment. In one instance in Nadia district where the target group had equal distribution of followers from two different religions, initial conflicts over access to the unit led to the settlement of a time schedule chart whereby both the user factions could enjoy equitable access (Singh et al. 2008b).

The question of social accessibility may also arise in case of the piped water supply networks because despite the connections provided, water may fail to be delivered to all the villages/hamlets, and the tail-end villages are most common to suffer. This may happen because of unattended broken taps, misuse of the treated water for purposes like washing animals and cleaning of utensils, and much water wastage in the communities closer to the head or in the middle section of the network. They may even pilfer water for irrigation or disrupt the connections towards the lower end of the system, so that either the pressure drops too much or no water remains in the system. This was reported as a significant problem by villagers at the tail end of the first piped water supply scheme based on Ganga river in Bhojpur district of Bihar (Kumar et al. 2016).

Gender can also be an important factor influencing social equity in access of the mitigation technologies. Focus group discussions in two villages in Bihar revealed that women from upper castes such as Brahmin and Rajput are not expected to fetch water from public points. Therefore, even if arsenic-safe water is made available through interventions at public points, their utilization by upper caste households in general and by their women in particular may remain limited. Treated water from such public points will need to be procured by men or children, which may imply that for cooking and even for self-consumption, women may continue to use water from their contaminated domestic hand pumps.[2]

Another concern in social sustainability is the aspect of user participation in management and maintenance of the mitigation interventions, which also has a gender dimension. In West Bengal under an Indo-Canadian Project, arsenic removal units have been installed with community participation. Here management committees consist of men and women or sometimes only women (Singh et al. 2009). In reality, however, it was found that even where women are listed as members, the actual responsibilities with respect to activities such as collection of monthly contributions, organizing the timings of use, etc., are actually shouldered by men. Further, not all targeted households are interested in participating, as time and finances may be involved. With a lower level of sensitivity to arsenic, participation may be avoided, even if time and money are available (Singh 2016a).

Regarding the maintenance of arsenic removal units at community level, regular backwashing and periodic change of the filter media are essential in many of the designs installed. Though women have been especially motivated and even trained to engage in technical maintenance of the units, their participation was found to be low. Backwashing of the media is considered by women to be a time-consuming and cumbersome task and therefore they instead expect men, who are traditionally looked upon as responsible for technology maintenance, to take up this task. Similar views were encountered in relation to the maintenance of the domestic filters, the use and management of which is more strictly seen as related to women (Singh 2012b).

6.3 Cultural Acceptability

The relationship between society and natural resources may be comprehended as “environmental orientations”, within which ideological constructs about water for different uses are defined in any community (Singh 2006b). Cultural acceptability of water for drinking and cooking in West Bengal and Bihar is strongly built upon four different physical properties, namely, color, smell, taste and freshness (Singh et al. 2009). Good quality water for these purposes is expected to be colorless, odorless, sweet and fresh and these qualities, in turn, are seen as closely related to the local concepts of purity, health and hygiene. Freshly drawn water is regarded as pure as well as safe on health grounds. In fact, these properties are also the grounds

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[2] Source: Field Data (2016).

on which arsenic contaminated water continues to be used widely for drinking and cooking, since arsenic imparts no smell, taste or odor to the water (Singh 2012b; Singh et al. 2009).

The water provided by the arsenic mitigation interventions must be culturally 'acceptable', fulfilling the above-mentioned criteria. However, the cultural constructs described above fail to be recognized and included within the scope of arsenic mitigation interventions, whether at policy/program level or as technologies. Wherever the arsenic mitigation options have failed to deliver the four water qualities stated above, their acceptability has suffered. With respect to surface water supply provided in rural areas of two affected districts in West Bengal, observations made in 15 villages revealed that though women find the technology easy to operate, they have not adopted the tap water for drinking and cooking. They exhibit a preference for water from deep tube wells. The most commonly cited reason is the difference in taste and odor due to "chlorine treatment of the water before supply". Also, the piped water supplied is said to be sometimes dirty and muddy and therefore unhygienic. Consequently, the piped water which is supplied as arsenic-safe water for drinking ends up being used commonly for washing, cleaning and bathing. Regarding the domestic filter, the reservations about taste and odor persist. Besides, the entire procedure is perceived by women as an alien 'add-on' procedure to their existing cultural practices of domestic water management as a result of which, even when adopted at first, it fails to become sustainable at household level (Singh et al. 2008a, 2009).

In Bihar, rooftop rainwater harvesting alternative promoted by UNICEF also has been rejected on cultural grounds (Kumar et al. 2016; Singh et al. 2012). Though rainwater is widely recognized as the purest form of water, freshness and cleanliness of the collected rainwater is a problem. Observations made in some of the villages showed that children are averse to using the roof-top rainwater for drinking. In fact, in one of the primary schools, the village health worker herself refused to drink water from the rainwater harvesting tank on the grounds of freshness and cleanliness and thereby the implicit health risks. The children preferred to drink water from the nearby hand pump which is contaminated with arsenic (Kumar et al. 2016; Singh et al. 2012).

Piped water supply based on Ganga river has however proved to be a success in terms of cultural acceptability (Kumar et al. 2016). Since Ganga water is widely regarded as holy, and the treatment process adopted at the first supply plant installed in Bihar has been found to produce water with which complaints of odor, color or taste are not associated, there is large-scale adoption of this water for drinking in the supplied villages. Similarly, deep tube well water in West Bengal was found to have high cultural acceptability since women find this water colorless, odorless and sweet, besides providing the advantage of making the cooked rice 'white' and keeping it fresh for a longer time in the hot and humid climate. These advantages are reportedly missing when using water from the domestic hand pump or from the ARPs (Singh 2012b).

Another cultural factor that affects sustainability of the mitigation interventions is the alignment of the new technology with the traditional or local forms already in use since long. The arsenic removal technologies, irrespective of their chemical process, form and size, suffer a huge disadvantage on this ground. Most of these represent complex, alien technologies for the masses which is beyond the capacity of the communities to use and manage despite training. Even monitoring of the water quality is not easy despite training of personnel from the community (Singh 2012b).

On cultural acceptability grounds, dug wells may have a better acceptability in Bihar as reflected at a focus group discussion conducted by the authors in one of the affected villages where even presently, dug wells fitted with hand pump are used.[3] Similarly, use of ponds for drawing water for drinking and cooking was observed in West Bengal during this study, which attests the continuing value of this rainwater harvesting-based traditional water source in rural Bengal. Ponds provided with water treatment system and supply points may thus be developed as a culturally acceptable solution for arsenic mitigation in West Bengal (Singh 2012b). However, at present, the arsenic mitigation policy and approaches in India do not have in place the possibility of including user perspectives in designing and implementing mitigation strategies in affected communities. The user participation generally comes at a later stage after the selection of technology and its implementation has started (Singh 2008).

6.4 Economic Efficiency

Economic efficiency can be examined from the perspective of affordability which can be an important issue in some cases, in turn impacting the economic sustainability of the mitigation interventions. Piped water supply schemes are an attractive intervention that may appear to be a solution for all drinking water challenges in quality-affected habitations. However, these schemes are expensive to install, whether single village-based or multi-village, and even expensive to maintain, more so if the users are not motivated and engaged effectively from the beginning. In one case in Bhojpur district in Bihar, it was found that transfer of running costs for operating a tube well-based village piped water scheme from the agency to the community after its completion led to its total failure. Since regular electricity supply was a problem, a diesel-operated motor was installed to run the tube well. However, the village is numerically dominated by poor peasants who were not capable of contributing to the diesel costs from the beginning. Even the few wealthier families could not find it affordable to bear the cost for the entire village on a long-term basis. Consequently, the water supply scheme failed to be operated to supply safe water to the villagers, even after laying down of the entire piped water network (Singh 2012b).

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[3] Source: Field Data (2012).

Another instance of a problem of affordability was noted in Bihar regarding private efforts to supply arsenic-safe water to affected communities which has been recently started on small scale by private entrepreneurs.[4] These entrepreneurs deliver safe water to the community on a daily basis at a price which becomes difficult for the poorer households to afford. One woman from a village, which so far has received no agency-based mitigation intervention but is served by a private entrepreneur, expressed her inability to afford the cost of the safe water bottles on a monthly basis. This woman has been herself diagnosed with arsenicosis, and her daughter too shows some of the symptoms of the disease, but they continue to drink the contaminated water from their domestic hand pump despite the awareness and willingness to switch to safer sources.[5]

In most other instances, it was found that the monthly charges for community-based arsenic removal units appeared to be low enough to be affordable for all. But social and cultural factors discussed earlier impact the motivation of intended users to contribute financially (Singh et al. 2009).

7 Discussion and the Way Forward

In this section, a brief analysis of the reflections on the sustainability of arsenic mitigation interventions is presented. This is followed by an attempt to define a more effective way forward. From the reflections above, it is evident that problems of sustainability exist with all the current strategies. A number of factors embedded within environmental, social, cultural and economic contexts influence the process, resulting in one or more of the following consequences at the community scale: (a) a given intervention fails to be equitably accessible, with at least some intended users left out; (b) the intervention fails to be adopted wholly or partly, particularly with respect to use for drinking and cooking; (c) it becomes dysfunctional because of problems of maintenance (which can be rooted in the complexity and alienness of the technology) or the cost; (d) fails to serve the purpose of safe water provision in a holistic manner due to "slipping back" in quality or causing contamination in the environment that can "backflow" into community's water and natural resources. Also, none of these are able to provide protection against arsenic contamination beyond drinking water.

Considering the basic nature of the four different mitigation strategies mentioned before, it is more than obvious that the arsenic treatment technologies appear to be the least sustainable, as these may be confronted with sustainability challenges from environmental, social, cultural as well as economic perspectives. The strategy of surface water supply from rivers and lakes through piped water schemes is ostensibly sustainable due to lesser cultural and quality-based environmental challenges, but when scrutinized from social, quantity-based environmental or economic per-

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[4] Source: Field Data (2016).

[5] Source: Field Data (2016).

spectives, the sustainability may fail to be upheld. The sustainability of the arsenic-safe groundwater-based supply alternative may also be challenged by environmental constraints primarily because groundwater availability in the country is not only limited but progressively declining, and there may be risk of contamination at a later stage. Besides, there may be the possibility of economic or even social challenges. The rainwater harvesting alternative holds tremendous scope of environmental sustainability, but social, cultural or even economic challenges may exist and these would need to be addressed at the outset.

On the basis of perspectives obtained from different stakeholders ranging from users to agencies and the scientific community and the analysis presented above, it is obvious that for sustainable arsenic mitigation, environmental sustainability is the most basic concern that must be upheld in order to ensure that arsenic mitigation interventions can lead to safe water access to affected communities on a long-term basis, preferably also considering the water used within agriculture and other livelihood sectors. An intervention that does not contribute to environmental sustainability in qualitative and quantitative terms cannot be upheld for long, even if other conditions, namely, social, cultural and economic sustainability are fulfilled. For this, first and foremost, a long-term policy perspective is required. Even if any measure for immediate relief should be adopted, the environmental costs should be analysed and addressed adequately before implementation. Second, considering the qualitative and quantitative challenges posed by the current mitigation interventions, it can be argued that an environmentally sustainable and holistic way forward for arsenic mitigation would require reduction or halting of the groundwater extraction, simultaneously allowing the aquifer and water tables to regenerate sufficiently, which in turn would help dilution of arsenic levels. Finally, towards this end, 'nature-based' solutions – i.e., "actions which are inspired by, supported by or copied from nature" (WWAP/UN-Water 2018) can offer the best possibilities. A matrix of criteria that emerges from the analysis and discussion presented in this chapter is summarized in Table 6.1. This set of criteria must be considered in order to design and implement sustainable arsenic mitigation in India and elsewhere.

In light of this, rainwater harvesting is a potential sustainable solution which can provide quantitative as well as qualitative advantages over any of the alternatives analyzed above. Rainwater is the purest form of water and research in the arid and semi-arid zones of Rajasthan and Gujarat shows that where the average annual rainfall is low, ranging from as little as 100 mm to about 700 mm, and where salinity of groundwater has been a huge problem since centuries, the traditional wisdom of rainwater harvesting has sustainably provided sweet water to communities for drinking as well as agriculture (Agarwal and Narain 1997; Singh 2016b). Even in the Thar Desert and the Rann of Kutch communities have survived over generations through rainwater harvested and used for drinking through ponds and shallow wells. Groundwater from deep aquifers, which is saline, was not used for drinking purposes (Singh 2016b). On the contrary, after government introduced piped water schemes based on groundwater supply, fluorosis is becoming rampant in these States due to newly discovered fluoride contamination of groundwater (Annadurai et al. 2014). Rajasthan alone accounts for 51% of fluoride and 42% of saline affected

Table 6.1 Matrix of criteria for sustainable arsenic mitigation

	Component		
Evaluation criterion	Approaches and policies	Technological solutions	User participation
Environmental sustainability	Long-term approach rooted in 'nature-based' solutions	Locally-viable 'nature-based' technologies e.g. rainwater harvesting, use of surface water	Adapted to the local context of environment, resources, climate etc.
Social equity	Monitoring of equitable access and use of the facilities	Equitable, in terms of 'social access' of the mitigation technology, and not only 'physical access'	'Inclusive' management of arsenic-safe water delivery at local level – involving both genders, all classes and ethnic groups
Cultural acceptability	Decentralized approach with local communities as 'partners' in the mitigation process	'Appropriate', 'simple', 'user-friendly' technology rooted in local science, local resources, and respecting cultural norms and values	Inclusion of user perspective in design, location and management
Economic efficiency	High cost-effectiveness	Low cost in installation and maintenance	Affordable by the poorest

areas in the country (Reddy 2010). The cultural practices of the local communities in these areas provide living examples of how the arsenic menace can be managed more sustainably through rainwater harvesting (Agarwal and Narain 1997; Singh and Singh 2017a, b).

In Bihar, the average annual rainfall is 1224 mm, 84% of which is received during the monsoon season, while in West Bengal, the figure is 1439 mm (Mondal et al. 2015). Only a part of this rainfall is preserved and made available for use through dams and other kinds of storage structures. Majority of it is lost as surface runoff that finally flows into the sea or ocean. A substantial part of this runoff can be preserved at the level of local catchments in the arsenic-affected communities through different kinds of rainwater harvesting structures, including traditional village ponds (called 'pukur' in West Bengal) as well as through the Ahar-Pyne system where the monsoon rainfall is collected as surface run-off within a series of embankments from upper to lower catchments that are interconnected through a network of drainage channels. The water reservoir so created by the embankment is called *ahar* while the drainage channels carrying the water to or away from it are called *pynes*. Apart from other functions such as irrigation and flood control, the system helps sustainably fulfil drinking water needs of the local communities through groundwater recharge (Agarwal and Narain 1997; Koul et al. 2012; Singh and Singh 2018).

Uses of surface water sources and adoption of rainwater harvesting practices have been recommended for arsenic mitigation since long (Ghosh and Singh 2009; Giri et al. 2011; Planning Commission 2007). Also, rainwater harvesting has been recognized as one of the solutions for arsenic and other water quality problems in drinking water within the NRDWP framework (MDWS 2017). However, it has not yet been systematically adopted in policy and practice as the most significant and sustainable way forward that offers a bundle of environmental benefits over others in the context of arsenic mitigation. Not only it provides arsenic-free drinking water but also has the potential of addressing the problem of arsenic in the food chain, because harvested rainwater can be used for recharging groundwater (which can contribute to reducing the arsenic concentration) and can also be directly used for irrigating agricultural fields. It also takes care of the problem of over-withdrawal of groundwater which can further intensify the arsenic problem. Besides, it offers the benefit of building climate resilience by enhancing water storage in a climate change-induced drought regime.

However, it is to be noted that interventions based on rainwater harvesting will need to be adapted to local contexts in order to enhance their sustainability from cultural, social and economic perspectives. Since rainwater harvesting has been a tradition in most of the arsenic affected areas, though in different forms, there is need to rejuvenate the traditional practices, sensitize masses about the need and benefits, and 're-build' their skills and capacities to create and manage these resources. There is also need to provide communities with options for microbial protection which may be a limitation with the harvested rainwater collected in different kinds of structures ranging from household tanks to community-level ponds. On the whole, there will need to sensitize and mobilize the concerned communities for adopting community- and/or household-based rainwater harvesting practices, with substantial time allocated for this phase before community participation is initiated or individual households are invited to install rainwater harvesting based systems. There may be need to help communities design appropriate structures and systems that match the given geological, hydrological and cultural contexts, besides solving any financial issues that may arise. On the whole, it can be said that while rainfall in the country is substantial at more than 3000 billion cubic meters per year, only about 5% gets stored in dams and reservoirs, and another small part retained as river water flows (Bhattacharyya et al. 2015). The rest flows away into the Arabian Sea, Indian Ocean and Bay of Bengal and therefore becomes a lost resource. Decentralized use and management of this rainwater offers a sustainable prospect for tackling the problem of arsenic contamination in drinking water, enabling women, men and children in the affected communities enjoy their rights to water, health, work and overall development. Arsenic mitigation policy in the country should therefore incorporate rainwater harvesting as the most feasible and sustainable alternative.

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Chapter 7
Arsenic Contamination of Soil in Relation to Water in Northeastern South Africa

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Abstract Little is known about the arsenic contamination of soil in relation to water in South Africa. In fact, there is a gap in knowledge about the topic as far as Africa as a whole is concerned. This chapter addresses the limited information on the presence and threat of arsenic in South Africa's environment. The focus of this chapter is on soil (and indirectly water) contamination in the former Venda tribal area in northeastern South Africa where for many decades the apartheid government used arsenic-based dip solutions to treat East Coast Fever among cattle. Soil samples taken at 5-m, 20-m and 100-m at a depth of 300-mm from 10 old dip tanks revealed 11 readings above 2.0 mg/kg and 2 readings above 30 mg/kg. We found that these old contaminated dip sites were not rehabilitated and that houses are now being built as close as 50-m from the centers of contamination. It is clear that the problem of arsenic contamination of soil and water in South Africa, a water scarce country, deserve more attention from researchers and the various levels of government.

1 Introduction

Arsenic (As) as a chemical compound was discovered in the eighth century, but it was not until the grand-scale commercial production of arsenic trioxide (As_2O_3) from 1850 onwards, that the danger inherent to this substance became clear (Piracha et al. 2016). Arsenic is a natural hard metal found most of the time in low levels in the environment (Musingarimi et al. 2010). It is viewed as one of the most toxic natural elements found on earth (Smith et al. 1998; USEPA 1999; Singh 2017).

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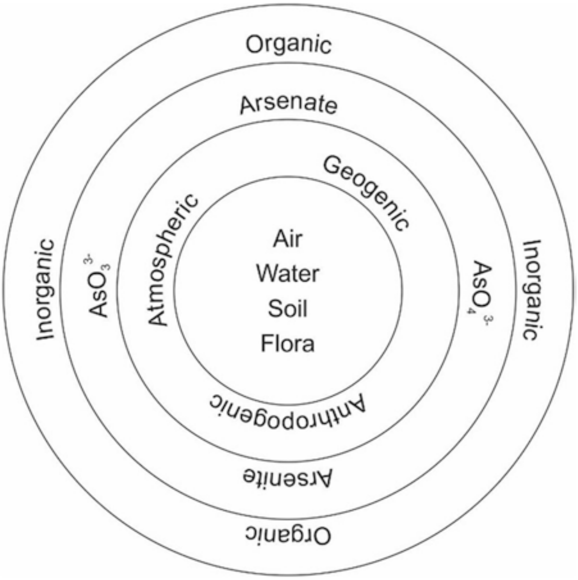
Chronic exposure to arsenic-contaminated materials such as soil, water, food, and atmospheric gasses may lead to arsenicosis, a chronic illness that produces skin disorders, gangrene and various forms of cancer (Ryan et al. 2001; UNICEF 2008; Roychowdhury 2010). The ingestion of, and exposure to, arsenic also may cause cardiovascular and respiratory diseases and neurological malfunctioning (Korte and Fernando 1991). According to the World Health Organization (WHO 1981), the fatal human dose for ingested arsenic is between 70 and 180 mg.

An explanation and discussion of arsenic in the environment should involve four interactive strata: (1) arsenic compounds, (2) chemical configurations, (3) source materials, and (4) contact/transfer media (see Fig. 7.1). The first stratum of the arsenic complexity is the differentiation between the compounds organic arsenic, inorganic arsenic, and arsine gas. Of these three, arsine gas is the most toxic compound (Sami and Druzynski 2003). According to the World Health Organization (WHO 2000), organic arsenic and inorganic arsenic include a trivalent assemblage and a pentavalent assemblage. In both organic and inorganic compounds, the trivalent assemblage is more toxic than the pentavalent assemblage. The second stratum of the arsenic complexity consists mainly of two different chemical composites, namely arsenite (AsO_3^3) and arsenate (AsO_4^{3-}), with arsenite being more toxic than arsenate (Smith et al. 1998). In the third stratum of arsenic complexity, three main groups of arsenic source materials, namely geogenic arsenic materials, arsenic materials of anthropogenic origin, and atmospheric source materials (see, amongst others, ATSDR 2000), are taken into consideration. Geogenic arsenic material is usually found with background geological materials (soil and rocks) and is specifically associated with minerals such as antinomy, copper, gold, iron, lead, nickel, silver and uranium (Sami and Druzynski 2003). Arsenic in the envi-

Fig. 7.1 Arsenic strata



ronment that is the result of anthropogenic activities is amongst others related to mining, the treatment of wood, the pharmaceutical and glass industries, fertilizers and pesticides (Piracha et al. 2016). Atmospheric source materials can be of both natural origin (e.g., volcanic activity) and anthropogenic origin (see below). At the core of the arsenic, complexity are interactive contact/transfer mediums such as air, water, soil, and flora.

Because of the mobility of arsenic, the direct and indirect interaction of its compounds, chemical configurations, source material, and contact/transfer mediums are so complex that one of the mediums cannot be researched in isolation. This chapter presents a case study of soil contamination in a particular region in South Africa, but is, at the same time sensitive to the entire complexity of arsenic in the environment.

2 Global Locations of High Arsenic Concentrations

Supported by a considerable volume of sources, Mukherjee et al. (2006) identified 42 main arsenic contamination locations across the globe. Most of the evidence concerning high levels of soil and water arsenic contamination relates to Asian countries such as Bangladesh, Cambodia, China, India, Japan, Myanmar, Nepal, Pakistan, Sri Lanka, Thailand and Vietnam (see, for example, Guha Mazumder et al. 2010; Roychowdhury 2010; Singh et al. 2016). High concentrations of arsenic (largely due to geogenic events and anthropogenic activities) also have been reported in European countries such as Bulgaria, the Czech Republic, Finland, Germany, Greece, Hungary, Romania, Spain, Sweden, and Switzerland. Further concentrations of arsenic-related to one medium or another have been detected in Australasia (Australia, New Zealand and Tasmania), Central America (Mexico), the Middle East, North America (Alaska, Canada and the USA), South America (Argentina, Chile, Brazil), and the United Kingdom (Nordstrom 2002; Sarkar et al. 2007; Singh 2017).

Ahoulé et al. (2015) and Singh (2017) referred to the limited amount of research on arsenic contamination in Africa. Mukherjee et al. (2006) mentioned only two important arsenic locations in Africa: Egypt and Ghana. In both these countries, the detected arsenic is of natural origin. Fatoki et al. (2013) referred to a few reports on arsenic contamination in Botswana, Burkina Faso, Ghana, Nigeria, the Rift Valley of Ethiopia, and South Africa. Singh (2017) mentioned that only 15 countries in Africa have so far been identified as having challenges of arsenic contamination. In this regard, Ahoulé et al. (2015) referred to specific work conducted by researchers.[1]

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[1] Higy and Cordey (2011) (Benin); Huntsman-Mapila et al. (2006), and Mladenov et al. (2013) (Botswana); Smedley et al. (2007), Somé et al. (2012), Nzihou et al. (2013), and Ouédraogo and Amyot (2013) (Burkina Faso); Abdel-Moati (1990) (Egypt); Reimann et al. (2003), Rango et al. (2010, 2013), and Dsikowitzky et al. (2013) (Ethiopia); Amonoo-Neizer and Amekor (1993),

Although Ahoulé et al. (2015) refer to several research efforts conducted in Africa,[2] Fatoki et al. (2013) believe that the lack of information on arsenic contamination in Africa is, firstly, the result of limited research on the topic related to the continent and, secondly, the lack of international attention to reports on arsenic contamination on the continent.

The presence of arsenic in the South African environment is, in the first place, related to the residues of geogenic source materials (Hammerbeck 1998) and various types of anthropogenic actions (see below). Large parts of the South African environment is geologically dominated by the Karoo Supergroup and the Witwatersrand Basin that have been distorted by, amongst others, geological events, meteoric incidents, climate variations, and the change of sea levels over an extended period. The crust material is rich in minerals, including, cobalt, copper, gold, lead, manganese, nickel, platinum, silver, uranium and zinc, as well as coal that, together with anthropogenic actions, are directly or indirectly associated with arsenic in the environment. Secondly, anthropogenic activities such as the burning of coal and fly ash, stockpiling of residual material from gold mines on waste heaps, fossil fuel power plants, the production of fossil fuel petroleum products, the timber industry, and the use of pesticides, insecticides and stock dips further contribute to the presence arsenic in the South African environment (see, for example, Botes et al. 2007; McCarthy 2011; Niyobuhungiro et al. 2013).

Despite the limited evidence on South African waters mentioned by Ahoulé et al. (2015), authors such as Sami and Druzynski (2003), Dzoma et al. (2010), Ogola et al. (2011), and Akinsoji et al. (2013) indicated the presence of arsenic of more than 0.05 mg/L in South Africa's waters. Such occurrences are five times the maximum safe permissible value (SMPV) for drinking water recommended by the World Health Organization (WHO 2012).

Based on data collected over many years by the South African Department of Water Affairs and Forestry (DWAF), Kempster et al. (2007) reported on several readings of arsenic above 1 mg/L. At the same time, Kempster et al. (2007) highlight the lack of a formal national monitoring programme of arsenic levels in South Africa's water resources.

Despite the relationship between arsenic in soil and water, this topic – with reference to South Africa – has received only little attention from researchers (see Dzoma et al. 2010; Ogola et al. 2011), and it is to this matter that the attention of this contribution now shifts.

Smedley (1996), Smedley et al. (1996), Serfor-Armah et al. (2006), Asante et al. (2007), Baumah et al. (2008), Kortatsi et al. (2008a, b), Akabzaa et al. (2009a, b), Rossiter et al. (2010), Akabzaa and Yidana (2012), Bhattacharya et al. (2012), and Kusimi and Kusimi (2012) (Ghana); Pritchard et al. (2007, 2008) and Mkandawire (2008) (Malawi); El Hachimi et al. (2005, 2007) (Morocco); Asubiojo et al. (1997) and Gbadebo (2005) (Nigeria); Dzoma et al. (2010), Ogola et al. (2011), and Akinsoji et al. (2013) (South Africa); Bowell et al. (1995), Taylor et al. (2005), and Kassenga and Mato (2009) (Tanzania); Rezaie-Boroon et al. (2011) (Togo); and Jannalagadda and Nenzou (1996) (Zimbabwe).

[2] See footnote 1.

3 Arsenic in the Soils of NE South Africa: A Case Study

The general occurrence of low levels of arsenic in soil is a natural occurrence. According to the WHO (1981), arsenic concentrations in uncontaminated soil are in the range of 0.2–40 mg/kg. Stock dipping in the past that caused arsenic contamination received attention in Australia, New Zealand and the southern states of the USA (McLaren et al. 1997; Mukherjee et al. 2006; Sarkar et al. 2007; Piracha et al. 2016). Here, we report on arsenic contamination of soil related to past cattle dipping processes in the Vhembe district (Limpopo province, South Africa). This study has also been reported on in other, different formats by us (see Ramudzuli 2014; Ramudzuli and Horn 2014).

3.1 *The Study Area and Its People*

Venda was the ethnic 'homeland' of the Venda people during the South African apartheid era (1948–1994), but remained under the overarching governance of the Republic of South Africa during this period (Horn 1998). In 1979, Venda decided to accept the offer from the South African government to become an 'independent' homeland (Horn 1998). In 1994, after the first democratic election in South Africa that included all the country's people, Venda, the study area, became part of the country at large as the Vhembe district of the Limpopo province, the northernmost of the nine provinces of South Africa (Fig. 7.2).

Traditionally, cattle farming occupied a central role in Venda society. Notwithstanding their monetary value, cattle pulled ploughs, sledges and wagons, and carried baggage. More importantly, cattle ownership defined the economic and social status of its owner. Consequently, *lobola* (payment for a bride), was traditionally determined in 'cattle currency' (Ramudzuli 2014).

3.2 *East Coast Fever*

Towards the end of the South African War in (1898–1902), there was a shortage of cattle in southern Africa (Norval et al. 1992). This shortage resulted in the importation of cattle from Australia and India. On the way, cattle were offloaded at the harbor of Mombasa (Kenya), an endemic area of a cattle disease known as East Coast Fever (ECF) (Cranefield 1991). On arrival in South Africa, this at the time unknown disease quickly spread through the cattle herds in entire Southern Africa (Cranefield 1991).Therefore, from around 1900 to the 1960s, Southern Africa was in the grip of ECF (Norval et al. 1992). The veterinarian Arnold Theiler discovered in 1910 that ECF is a cattle disease caused by an intracellular protozoan of the genus *Theileria parva* transmitted by ticks of the species *Rhipicephalus appendiculatus*

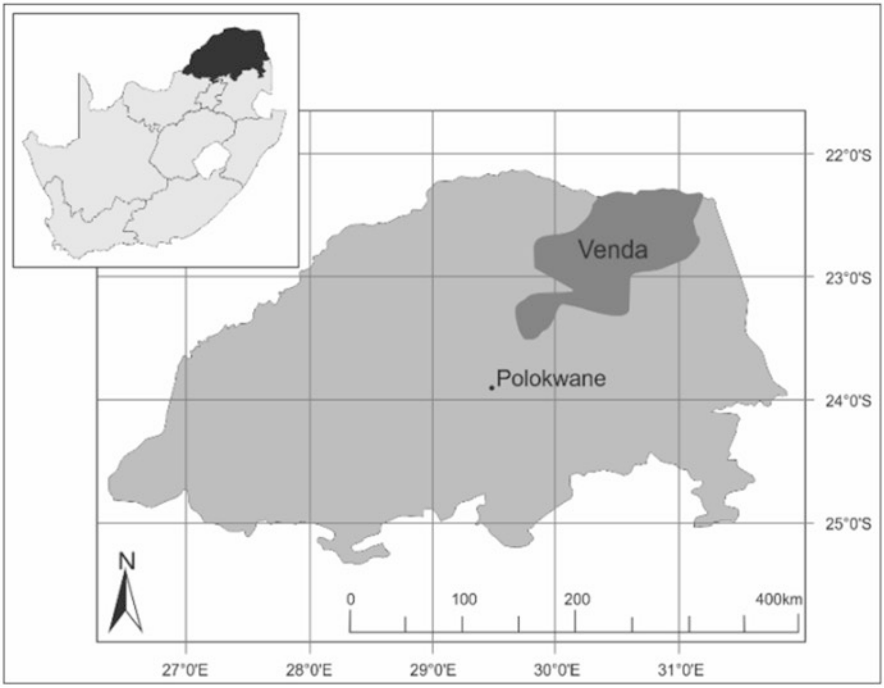
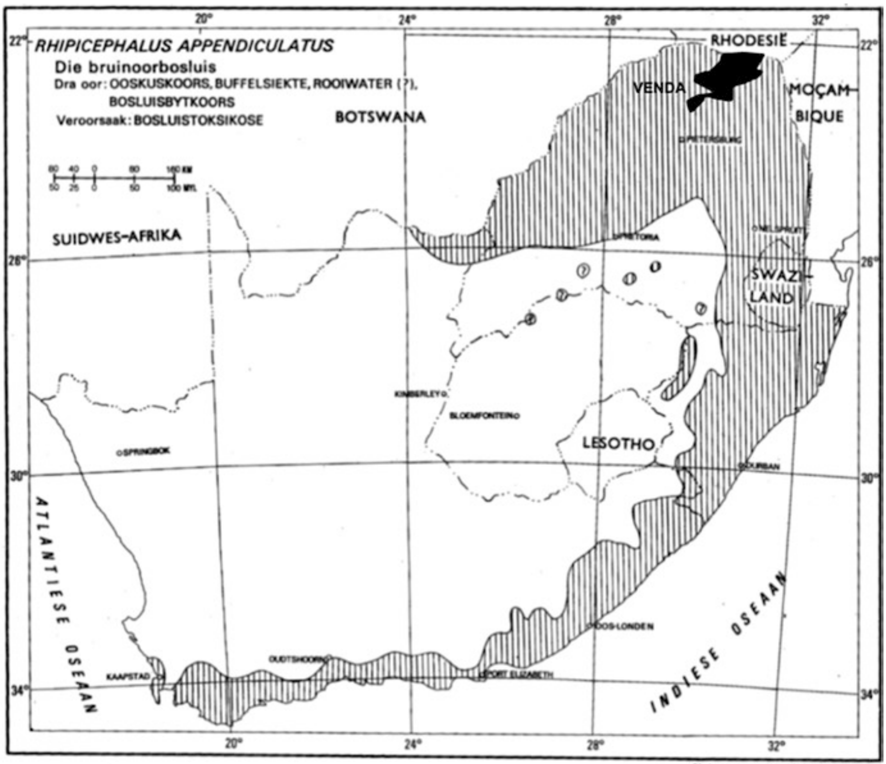


Fig. 7.2 Study area (Venda) in Limpopo province, South Africa

(brown ear tick) (Theiler 1971; Davies 2004). Ordinance No. 38 of 1904 (NASA LC442/05 1904; NASA TAB-A341/15 n.d.) resulted in an attempt to curb the disease in parts of South Africa using arsenic-based animal dipping compounds, already introduced in South Africa in 1893 (Norval et al. 1992).

The Disease of Stock Act 14 of 1911, as well as the Dipping Tanks (Advances) Act 20 of 1911, facilitated the implementation of a limited dipping program (Marole 1967). Thereafter, the Pretoria Conference of 1929 introduced a countrywide ECF Control Programme of regular cattle dipping with the use of mainly arsenic oxide (As_2O_5) and trioxide (As_2O_3) compounds (Ramudzuli and Horn 2014). The programme expected white (European) commercial farmers, assisted by state subsidies, to administer their dipping, whereas the Native Affairs Department (NAD) carried the responsibility to provide the service to black African communal areas such as the later Venda homeland (Mbeki 1964; Beinart 2003). The distribution of the brown ear tick in South Africa determined the location and building of thousands of dip tanks (Fig. 7.3).

According to Fletcher (2000) and Turton (2004), plunge dipping was a common method of tick control. The first plunge dip tank in Venda (Fig. 7.4) was built in 1915 (Marole 1967; Nemudzivhadi 1985). In spite of these attempts, approximately 1.4 million ECF affected cattle died in South Africa between 1902 and 1945.



Direkteur - generaal van Opmetings, 1976.

Fig. 7.3 Distribution of the brown ear tick in South Africa. (Reproduced from South Africa 1983)

At the time of the eradication of ECF in South Africa around 1954, when compulsory cattle-dipping and cattle-control restrictions were lifted in the private agricultural sector, it became clear that the populace in the tribal areas (homelands) did not have the required resources to continue cattle dipping on their own (Tomlinson 1955). As a result, the compulsory dipping program continued in these areas under new provisions of the Animal Disease and Parasite Act 13 of 1956, in order to control other tick-borne diseases and to provide a general state-led veterinary service to these communities. Initially the NAD, in cooperation with the respective tribal councils, was responsible for the erection and maintenance of cattle-dipping tanks in the tribal areas. The Native Affairs Department's Division of Veterinary Services in liaison with Native Affairs Commissioners and tribal chiefs and territorial councils provided technical services (NASA TAB-A341/15 n.d.; Linington 1949; P.J. Sefara, InterVet consultant, personal communication, 4 August 2011). Ten million cattle were dipped every 7–14 days (Norval 1983).

Around 1960, ECF was eradicated in Southern Africa, including Basuthuland/Lesotho, but compulsory cattle dipping with arsenic-based cattle dipping com-

Fig. 7.4 The second author (MR Ramudzuli) standing at the ruins of the first dip tank built in Venda in 1915. (Reproduced from Ramudzuli 2014)

pounds continued in South Africa's homelands. There were two reasons for this practice: Firstly, the South African government felt obliged to provide veterinary services to the black African homelands; and secondly, some of the black African tribal areas such as that of the Venda, Shangaan, and Swazi people bordered on the Kruger National Park (KNP). Because of the danger of diseases such as Foot and Mouth Disease (FMD), Corridor Disease, Anthrax, and Tuberculosis carried by game (J. Nethengwe, Vhembe District Veterinary Section, personal communication, 13 December 2013), the area next to the KNP was divided into an infected zone, a protection (buffer) zone and an FMD surveillance zone, almost parallel to the border of the KNP (Fig. 7.5). Although the diseases indicated above are not treatable through dipping, the compulsory scheduled dipping events provided excellent opportunities to observe the cattle for possible symptoms.

After claiming independence from South Africa in 1979, Venda established its own Directorate of Veterinary Service residing under the Venda Department of Agriculture and Forestry. However, seconded officials from the South African Government were still overseeing the application of the service (The Republic of Venda 1979). Dipping took place once a week during summer and fortnightly during winter (N.E. Mafhara, Limpopo Province Department of Agriculture, Veterinary Division, personal communication, 2 July 2004). Although declared illegal in 1983, the unofficial use of stocked arsenic dipping solutions continued for some time (Norval et al. 1992).

Dipping under the supervision of local officials assisted by officials seconded by the national government of South Africa continued until 1994 when the post-apartheid government took over the government of the country. Since then, the

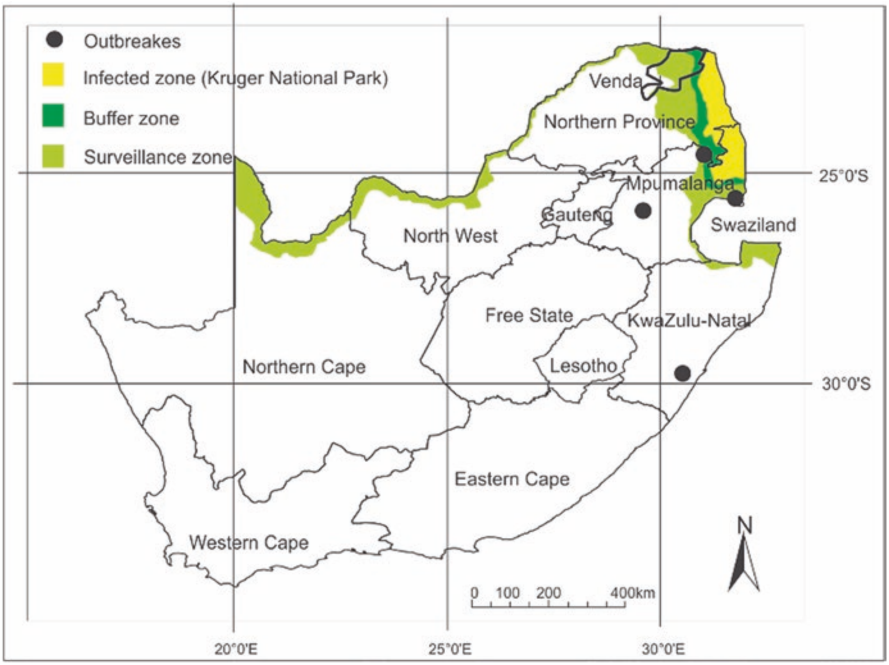


Fig. 7.5 Cattle disease control zones in South Africa

'100%' (composite) dipping program was scaled-down. Eventually, the official dipping system in Venda (now the Vhembe district of the Limpopo province) collapsed (M. Mafhara, Limpopo Province Department of Agriculture, Veterinary Division, personal communication, 13 June 2015).

New outbreaks of FMD since 2001 resulted in a re-evaluation of the direct and indirect role of cattle dipping to control cattle diseases (J. Nethengwe, Vhembe District Veterinary Section, personal communication, 13 December 2013). Although the gradual introduction of a new government-driven dipping programme through the Veterinary Section of the National Department of Agriculture (NDA), it is estimated that only 50% of the dip tanks in Vhembe is still operational (J. Nethengwe, Vhembe District Veterinary Section, personal communication, 5 March 2015).

Even though the post-apartheid South African government has not as yet published a new policy on stock dipping, it has established a national task team to draft a new policy (Mampane 2004, 2011). In the meantime, the Animal Diseases Act 35 of 1984 remains enacted, although the Animal Diseases Regulations under this Act was amended as recently as 2014.[3]

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[3] See: Animal Diseases Act 35 of 1984, Animal Diseases Regulations as published by Government Notice No. R. 2026 (1984) and amended by Animal Diseases Regulations: Amended, by Government Notice No. R. 865 (2014).

4 ECF and the Contamination of Soil

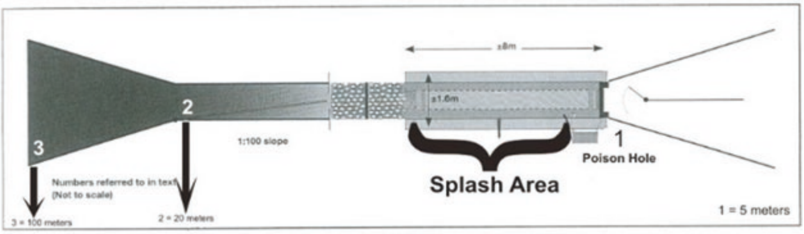
The second author, Ramudzuli, supervised by the first author of this contribution, collected soil samples from 10 dip sites in the Vhembe district in South Africa (for detail see Ramudzuli 2014). The dip sites were selected based on the dates of construction of the various dip tanks, soil characteristics, and eco-regions. The dip tanks were situated at the villages of Khubvi, Mukula, Rambuda, Sambandou, Thengwe, Tshandama, Tshifudi, Tshikuwi, Tshituni and Tshivhulani (Table 7.1).

The level of arsenic concentration was measured at distances of 5-m, 20-m and 100-m from the respective dip tanks (Fig. 7.6). The 5-m collection site enclosed the splash area (point 1 on Fig. 7.6) and was usually close to the poison trench where solution waste accumulated when the tanks were cleaned. The 20-m distance covered a draining pen in which the cattle were huddled whilst still wet with dip solution (point 2 on Fig. 7.6). The 100-m distance (point 3 on the figure) covered a radius around the tanks from where the cattle dispersed and served as the control point.

Single, linear point soil samples following the contours of the terrain were taken at a depth of 300-mm and placed in clean, labeled plastic bags. The packaged soil samples were chemically analyzed for arsenic by an accredited soil laboratory of

Table 7.1 Location of dip tanks selected based on ecological and soil characteristics of surroundings. (Reproduced from Ramudzuli and Horn 2014)

Site	Latitude	Longitude	Date built	Eco-regions and soil characteristics
Established before 1948				
Tshivhulani	22°55.35′ S	30°30.12′ E	Early 1920s	ER 2.01 (Central Highland): Deep red clays predominate
Khubvi	22°49.52′ S	30°34.03′ E	1923	ER 2.01 (Central Highland): Heavily weathered, compacted red clay
Rambuda	22°47.05′ S	30°27.06′ E	1940	ER 5.04 (North-Eastern area): Red loam with a high level of organic matter
Tshikuwi	22°53.83′ S	29°58.91′ E	1940	ER 5.03 (Western area): Heavily weathered, compacted red loam
Tshituni	22°56.82′ S	30°02.57′ E	1940	ER 5.03 (Western area): Gravelly with traces of brown clay
Established from 1948				
Sambandou	24°49.59′ S	30°39.33′ E	1948	ER 5.04 (North-Eastern area): Sandy loam with a very high level of organic matter
Tshifudi	22°48.24′ S	30°43.27′ E	1948	ER 2.01 (Central Highland): Sandy loam with prevalent organic matter
Makula	22°51.00′ S	30°36.59′ E	1948	ER 2.01 (Central Highland): Weathered, compacted red clay
Thengwe	22°49.59′ S	30°32.58′ E	1950	ER 5.04 (North-Eastern area): Sandy with little organic matter
Tshandama	22°30.07′ S	30°45.05′ E	1950	ER 5.04 (North-Eastern area): Sandy with little organic matter



Schematic diagram of a dip tank layout

Fig. 7.6 Sampling points of soil at 5-m (1), 20-m (2), and 100-m (3) distances from dip tanks. (Reproduced from Ramudzuli and Horn 2014)

the Agricultural Research Council (ARC) of South Africa (Ramudzuli 2014). The analysis performed involved a scan of an ammonium EDTA extract. An ammonium EDTA solution was added to soil samples, and the solution was filtered to isolate the chemicals. The findings are reported in Table 7.2.

The two highest As readings were at Sambandou (46.76 mg/kg at 5-m) and Tshivhulani (30.18 mg/kg at 5-m). The highest mean As values for the three distances was 18.24 mg/kg at Sambandou and 10.13 mg/kg at Tshivulani. Readings above 0.2 mg/kg occurred at Tshifudi, Khubvi, Rambuda, and Mukula at the 5-m distance, at Sambandou, Khubvi, Rambuda, Mukula, and Tshifudi at the 20-m distance, and at Rambuda, Khubvi and Sambandou at the 100-m distance. The decline of contamination values varied. The values of Sambandou (−39.88 mg/kg) and Tshivhulani (−29.99 mg/kg) revealed big differences between the 5 and 20-m sampled points. Results further indicated a rapid decrease in the arsenic concentration values between 5-m sample points and 100-m sample points, with the exception of Rambuda. The explanation of the increased level of contamination at 100-m at

Table 7.2 Arsenic levels and soil qualities at sample points

	As concentration (mg/kg)				Change		
Dip sites	5-m	20-m	100-m	Mean	5 to 20-m	20 to 100-m	Comments
Sambandou	46.76	6.88	1.09	18.24	−39.88	−5.79	Sandy loam with very high level of organic matter
Tshivhulani	30.18	0.19	0.01	10.13	−29.99	−0.18	Predominating deep red clays
Tshifudi	3.85	0.23	0.15	1.41	−3.62	−0.08	Sandy loam with prevalent organic matter
Khubvi	3.65	3.69	3.60	3.65	+0.08	−0.09	Heavily weathered, compacted red clay
Rambuda	3.53	3.63	3.70	3.62	+0.10	+0.07	Red loam with high level of organic matter.
Mukula	2.30	1.20	0.08	1.20	−1.10	−1.12	Weathered compacted red clay; steep slope.
Thengwe	0.14	0.07	0.09	1.19	−0.07	+0.02	Sandy with little organic matter
Tshikuwi	0.08	0.12	0.02	0.07	+0.04	−0.10	Heavily weathered, compacted red loam
Tshituni	0.02	0.06	0.01	0.06	+0.40	−0.05	Gravelly with traces of brown clay
Tshandama	0.00(2)	0.00(3)	0.00(2)	0.00(2)	+0.00(1)	−0.00(1)	Sandy with little organic matter
Mean	**9.05**	**1.46**	**0.88**	**3.96**	**−7.44**	**−0.73**	

Notes: In the ‘Change’ columns the symbol ‘−’ indicates a decreasing and the symbol ‘+’ indicates an increasing arsenic levels between sampling points

Rambandou may be related to the drip of dip solution from the treated cattle, which congregate in the unpaved drip yard before they are dispersed, and the overland flow of dip solution and overflow of dip sites as during flooding (Ramudzuli and Horn 2014).

To explain the differences in As concentrations between various dip sites, three factors were taken into account: soil properties, the presence of organic matter, and locational influences. In the case of the highest reported reading at Sambandou (46.76 mg/kg at 5-m), the soil property is a sandy loam with a high level of organic matter. At Tshivulani (30.18 mg/kg at 5-m), the soil is a deep red clay, but it should also be taken into account that the dip tank is located in a topographical depression. A reading of 3.85 mg/kg was obtained from Tshifudi at a 5-m distance where the soil is a sandy loam with prevalent organic matter. The fourth highest reading at a distance of 5-m from the dip tank occurred at Khubvi (3.65 mg/kg) where the soil is a heavily weathered, compacted red clay. However, a clear explanation of the environmental circumstances for the variations between the measurements at these four sites is not possible with the information available.

Arsenic in soil is an accumulative and non-degradable substance and presents a danger to humans and animals (Smith et al. 1998; Ramudzuli and Horn 2014). The real danger of arsenic in the soils of the study area that resulted from cattle dipping with arsenic-containing liquids is the direct contact with contaminated dipping tanks and dipping remnants, and the use of polluted surface water and groundwater.

In 2018, we revisited the sites where we conducted our original research and came to the following conclusions:

1. Several contaminated dip sites are still in use at present and cattle often have immediate access to surface water, such as streams and ponds, after dipping.
2. Old dilapidated sites have not been disinfected and properly fenced-off.
3. The design of dipping sites, such as those in Rambuda constructed in 1940, is still dangerous for humans and animals. The poison hole at Rambuda is situated approximately 20-m away, at a level lower than the dip tank, and is connected to the tank by a narrow furrow.
4. Present day human activities increase the danger of contact with the old dip sites. Khubvi tank is now situated in the middle of a maize field. The Rambuda site is utilized for mud brick making. Continuous ploughing and brick making may shift the soil downslope, and thereby assist in the migration of arsenic. Moreover, houses have been built next to some unenclosed dip sites (see Fig. 7.7). At a larger scale, the proximity of old dip tanks to rivers and streams in the study area poses great concern (see Fig. 7.8).

Fig. 7.7 Houses built next to an unenclosed dip tank at Tshandama

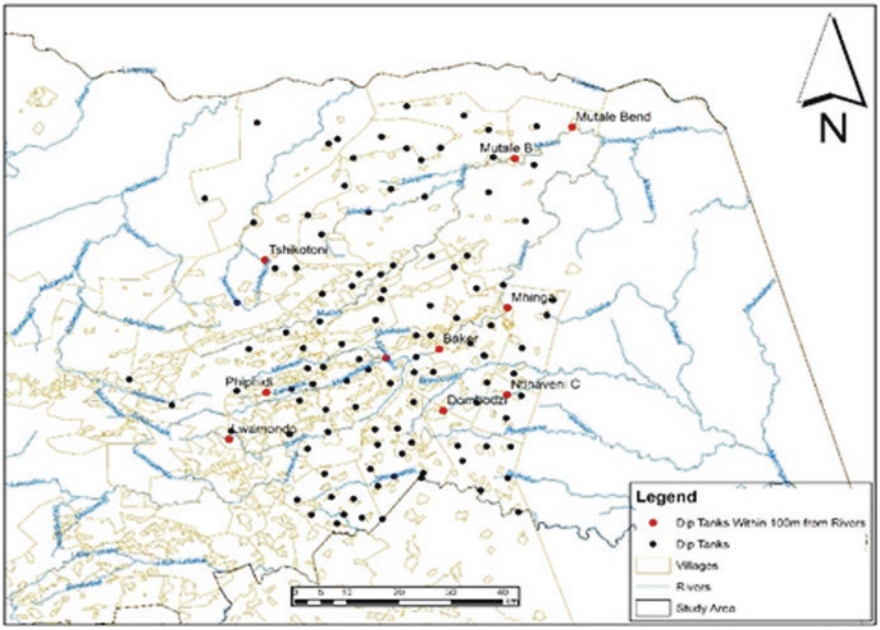


Fig. 7.8 Map showing the proximity of rivers and streams to historical cattle dip sites in Northeastern South Africa

5 Conclusion

According to the available literature, South Africa is not a main arsenic contamination location in the world. This chapter reflected upon a number of reasons that may spearhead a different mindset. We have evaluated the geology[4] and human activities associated with the presence of arsenic in the local environment in South Africa. However, the case study presented in this chapter focused on soil contamination caused by past cattle dipping with arsenic-based solutions to treat East Coast Fever in the country, and the continued use of stockpiled arsenic-based solutions for cattle dipping even after the use of it became illegal in 1983. We collected 30 soil samples at 10 cattle dipping sites at a depth of 300-mm in the former Venda homeland of South Africa and tested the samples for the presence of arsenic remnants. Only two samples contained levels of arsenic exceeding median readings in

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[4] More detail on arsenic related geological formations in Northeast South Africa includes the Mount Dowe Group of the Beitbridge Complex; the Nzhelele, Sibasa and Tshifhefhe Formation of the Soutpansberg Group; the Schiel Complex; the Phalaborwa Complex; the Rooiwater Complex, and the Gravelotte Group (Kempster et al. 2007). The location of these formations are closely connected with the South African greenstone belt starting at the border with Swaziland, reaching northwards to the study area and then turns westwards, covering both sides of the Limpopo river towards the Beitbridge linking South Africa and Zimbabwe.

the United States of America and Australasia where stock dipping is also the cause of concerning levels of arsenic in soil. However, the uncoordinated planning and development of post-apartheid South Africa's former 'homeland' areas is a serious concern. For example, the rehabilitation or enclosure of old arsenic contaminated dip tank sites in these areas have not received the required attention. At a larger scale, South Africa is prone to high levels of arsenic contamination in the future because of the reasons mentioned above. South Africa is a water scares country, and its water requires proper protection. The arsenic contamination of water is researched from many angles with different intentions. However, the close link between arsenic in soil and arsenic in water, and the reciprocal impact of arsenic in these two mediums remains an under-researched topic in South Africa and Africa in general.

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Chapter 8
Arsenic Contamination of India's Groundwater: A Review and Critical Analysis

Aslesha Kaur Dhillon

Abstract Over the past years, the extent and magnitude of arsenic contamination of India's groundwater has been growing in enormous proportions. However, there has not been any integrated and holistic approach, or any concrete action taken to combat the issues and threats of arsenic contamination, specifically by the Indian Government. This paper aims to critically analyze the scope of arsenic contamination in India and the efforts taken by the Central Government of India to address this issue. First, this paper will introduce arsenic, examine its transportation and distribution in the environment and the human population, and discuss the impact of arsenic exposure on humans. Second, the paper will discuss the scope of arsenic contamination in India and how it is screened and identified. Third, this paper will critically review the water governance structure of India. Fourth, this paper will discuss the challenges in mitigating and combating arsenic contamination of groundwater in India. Finally, the paper will conclude by providing recommendations for risk-mitigation and local management of groundwater arsenic contamination.

1 Introduction

Groundwater is used for domestic purposes in India by 80% of the rural population and 50% of the urban population (Ministry of Water Resources 2014). While the majority of the ground-water in the country is potable, water quality issues arise from geogenic activity such as salinity, nitrate, iron, fluoride and arsenic (Ministry of Water Resources 2014). In India, the area and population of arsenic endemic states are 529,674 km^2 and 359 million (approximately), respectively (Chakraborti et al. 2016b). The Indian Government's Parliamentary Committee report on the occurrence of high arsenic content in groundwater found that more than 70 million

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people in over 90 districts are at risk of arsenic contamination because of its presence in groundwater above WHO's guideline value of 0.01 mg/L (10 μg/L) (Ministry of Water Resources 2014).

The geogenic contamination of groundwater through arsenic is spatially assorted and confined to specific regions. In India, arsenic affected states are mainly located in the Ganga Brahmaputra Plain (Chakraborti et al. 2016b). Chronic arsenic exposure and inorganic arsenic are responsible for myriad illnesses, and can cause various health effects such as dermal, cardiovascular, respiratory, gastrointestinal, endocrinological, neurological, reproductive developmental, cancerous and cutaneous effects (Chakraborti et al. 2017). The appearance of skin lesions are a critical warning signal as they indicate severe internal damage (Chakraborti 2011). Studies have shown in Punjab, chronic exposure to arsenic through its presence in the groundwater has likely increased mortality rates and, cardiovascular diseases and cancers of liver, lungs and bladder (Singh 2016a). This has also been linked to infant mortality and it has compromised motor and intellectual function in children (Singh 2016a). At this time, no known medicine can cure arsenic toxicity (Chakraborti et al. 2018). Only a few preventive measures such as consuming arsenic-safe water and nutritious foods, including vitamins are recommended (Chakraborti et al. 2018).

Arsenic contamination of groundwater and the corresponding health effects were first reported in 1976 in Chandigarh and some villages in Punjab and Haryana (Datta and Kaul 1976). Subsequently, in 1983, cases of arsenic contamination and arsenicosis emerged in West Bengal (Garai et al. 1984). The School of Environmental Studies (SOES), Jadavpur University, Kolkata, India brought this issue in focus through organizing the International Conference on Arsenic (Chakraborti et al. 2018). Elevated levels of arsenic in the groundwater were discovered by SOES in Madhya Pradesh (now Chhattisgarh) in 1999, Bihar and Uttar Pradesh in 2003, in Jharkhand in 2004 and along the Allahabad-Kanpur track in 2009 (Chakraborti et al. 2018). Moreover, this was also reported in Assam and Manipur, in the Brahmaputra Plains between the years 2004 and 2006 (Chakraborti et al. 2018). According to Mishra et al. (2016) the following districts have arsenic-contaminated groundwater above the WHO standard of 10 μg/L: 25 of 75 districts of Uttar Pradesh; 14 of 19 districts of West Bengal; 14 of 22 districts of Haryana; 22 of 38 districts of Bihar; 3 of 24 districts of Jharkhand; 2 of 27 districts of Chhattisgarh; 3 of 33 districts of Rajasthan; and 1 of 11 districts of Delhi.

Over the past years, India's groundwater arsenic contamination has been growing at an exponential rate. The Indian government has not taken any holistic or integrated approach in mitigating and combating the challenges and threats of arsenic contamination. The aim of this paper is to conduct a critical analysis on the scope of arsenic contamination in India and the Central Government of India's efforts in addressing this issue. First, the paper will introduce arsenic and discuss how it spreads within the environment and human population. It will also examine the impact of arsenic exposure on human health. Second, the paper will assess the extent of arsenic contamination across various states of India and its screening and identification methodology. Third, the paper will critically analyze the water governance structure of India through reviewing its policies and institutions. Fourth, the paper will discuss the key challenges in addressing this issue. Finally, this paper will provide risk-mitigation, local management and, partnerships and advocacy recommendations to effectively address arsenic contamination of India's groundwater.

2 What is Arsenic?

Arsenic is a metalloid and its compounds occur in crystalline, powder, amorphous or vitreous forms (Gomez-Caminero et al. 2001). It is a natural component of the Earth's crust, present at an average concentration of 2 mg/kg (Gomez-Caminero et al. 2001). Arsenic is typically found in trace quantities in rock, soil, water and air. However, in certain areas, arsenic can be found in higher concentration because of natural conditions and/or anthropogenic activities such as metal mining and smelting, fossil fuel combustion and pesticide use (Tchounwou et al. 2012).

2.1 Natural Sources of Arsenic

The natural source of arsenic is found abundantly in the earth's crust as it is present in more than 200 different mineral species. The most common form of arsenic is called arsenopyrite. It is estimated that one-third of the arsenic present in the environment is originated naturally, usually through volcanic action and low-temperature volatilization – which means arsenic containing vapor generated from solid or liquid forms of arsenic salts (Tchounwou et al. 2012). While organic arsenic which contains carbon is usually found in marine organisms, it is also sometimes found in territorial species (Tchounwou et al. 2012).

The greatest range and highest concentrations of arsenic in natural conditions are found in groundwater. This is because of two reasons: first, the strong impact of the water-rock interactions and second, aquifers have a higher tendency to favor arsenic mobilization in physical and geochemical conditions, especially in reducing conditions (Sharma et al. 2014). Thus, most of the arsenicosis cases reported globally are because of arsenic's exposure through groundwater and not surface water. Moreover, large parts of India, Bangladesh, Taiwan and Vietnam rely on arsenic contaminated groundwater for drinking purposes and irrigation of staple crops and vegetables (Sharma et al. 2014).

2.2 Anthropogenic Sources of Arsenic

Arsenic trioxide is a by-product of metal smelting operations and is used to commercially produce elemental arsenic. It is estimated that 70% of the world arsenic production is used in timber treatment, 22% in agricultural chemicals and the remaining in the glass, metallic alloys and pharmaceuticals (Tchounwou et al. 2012). The main industrial processes that are responsible for arsenic contamination are metal smelting, burning of fossil fuels and mining (Mandal and Suzuki 2002). Another source of arsenic contamination is the use of arsenic in the preservation of timber. Moreover, pesticides that contain arsenic have contaminated agricultural lands (Singh et al. 2015b).

2.3 Transportation and Distribution of Arsenic in the Environment and Human Population

2.3.1 Environment

Arsenic's transportation and distribution in the environment is a complex process because of the various chemical forms arsenic may be present in, and its different forms are constantly cycled through the soil, water and air (Gomez-Caminero et al. 2001). The main sources of arsenic in the atmosphere are high temperatures processes such as the coal-fired power plants, volcanic activity and burning vegetation (Gomez-Caminero et al. 2001). Arsenic trioxide is the most dominant form of arsenic released into the atmosphere, and it readily adheres onto the surface of the particles. These particles are distributed through wind, and they fall back on the ground because of either rainfall or their own weight (Gomez-Caminero et al. 2001).

Low temperature biological and natural reactions that involve microbes also release arsenic into the environment. Microbes that react with arsenic in solid and sediments generate arsine gas or other volatile arsenic compounds (Gomez-Caminero et al. 2001). However, once arsine gets oxygenated by reacting to air, it is converted back into non-volatile forms of arsenic and settles back on the ground (Gomez-Caminero et al. 2001). If arsenic is present in well-oxygenated water and sediments, mostly all arsenic is present in the stable form of arsenate, but some arsenate and arsenite forms are interchangeable and less stable contingent on the biological and chemical conditions.

Additionally, selected chemical forms of arsenic adhere strongly to organic and clay matter, and this further impacts how they behave or react in the environment (Gomez-Caminero et al. 2001). While, arsenic can potentially be released through sediments and water, this is also contingent to the biological and chemical conditions. Soil and weathered rock that has arsenic can also be transported in the environment through wind or water erosion. However, as arsenic compounds usually adhere strongly to soils, water percolation is only able to move the arsenic a short distance within the soil (Gomez-Caminero et al. 2001).

2.3.2 Human Population

Amongst the human population, contamination of drinking water is the main source of arsenic. However, populations that are not exposed to high concentration of arsenic in their drinking water can consume arsenic through food that was grown in arsenic-contaminated soil or irrigated with arsenic contaminated water (Singh et al. 2015b).

In most South-Asian countries such as India, Bangladesh, Nepal and Pakistan, groundwater is used for irrigation. Due to lax regulations and implementations, irrigation units are installed in shallow depths; and thus, farmers use arsenic-contaminated water for agriculture irrigation. As a result, arsenic has entered people's food chain and livestock. Chakrabarti et al. (2018), conducted an analysis in North 24-Parganas, West Bengal, India of 597 irrigation tube wells, where they found 6.4 tons of arsenic dumped in the crop fields. Also, approximately 19% of the samples exceeded the 100 $\mu g/L^{-1}$ standard of irrigation water set by the Food and Agriculture Organization (FAO). This study also found that 76% of the arsenic present in the crops was inorganic, and domestic animals were also vulnerable to arsenic as they consumed arsenic contaminated straw and water. In India, elevated levels of arsenic within the range of 13 and 800 $\mu g/kg^{-1}$ were found in the following food materials: luffa, lentils, brinjal, maize, cucumber, wheat, gourd, rice husk, ladyfinger, rice and green gram (Chakrabarti et al. 2018).

2.4 Impact of Arsenic on Human Health

Humans can consume arsenic through two ways: first, arsenic contaminated water and second, arsenic contaminated food. The immediate symptoms of acute arsenic poisoning include diarrhea, abdominal pain and vomiting that can be followed by muscle cramping, tingling of the limbs, numbness and in lesions and formation of hard patches on the soles of the feet and hands (WHO 2010). There are specific dermal effects that have been attributed to chronic arsenic toxicity, such as melanosis (pigmentation) followed by keratosis and hyperkeratosis (Bhowmicka et al. 2018). According to Mazumder (2008), arsenic keratosis appears as diffuse thickening either alone or in combination with nodules symmetrically distributed, on the palms of the hands and soles of the feet.

Further, high concentrations of arsenic can cause several disorders that can be both carcinogenic and non-carcinogenic. These can occur in the respiratory system, cardiovascular system, digestive system, reproductive system, endocrine system, neurological system, hematopoietic system and renal system (Singh et al. 2015b). According to a study conducted by Mazumder et al. (2000), where they examined 7683 participants in arsenic contaminated areas of West Bengal, India, persistent exposure and consumption of arsenic through drinking water caused many respiratory complications amongst humans such as shortness of breath, cough, bronchitis and wheezing. Likewise, Ahmed et al. (2017) found that prolonged exposure of arsenic amongst children resulted in inflammation of airways and alteration of liver function. Moreover, studies have found that arsenic exposure can induce hypertension and increase the risk of diabetes (Mazumder et al. 2012; Rahman et al. 1998, 1999).

The consumption of inorganic arsenic through drinking-water has been shown to cause cancer in humans (WHO 2010). The International Agency for Research on Cancer (IARC) has classified arsenic and its compounds as carcinogenic to humans (Group 1) (IARC 2004). On the basis of a number of studies in various countries such as Taiwan, Bangladesh, India, USA, Chile and others, the IARC concluded that chronic exposure to arsenic can cause cancer in skin, urinary bladder and lungs. A study conducted by Saha (2003), in the arsenic contaminated areas of West Bengal, India, found 5.14% incident rates for malignancy, wherein 4.34% of the cases were regarding skin malignancy and 0.78% about internal malignancy.

Chronic arsenic exposure can also have an adverse impact on pregnancy. Milton et al. (2005), conducted a study where they found arsenic in the tube well water in the range from undetectable to 1710 μg/L and reported excess risks for spontaneous abortion and stillbirths among the 533 women participants in Bangladesh. Ahamed et al. (2006a) also found spontaneous abortions, stillbirths and premature births in women living in high arsenic affected areas in Bangladesh, where the range of arsenic was between 201 and 1200 μg/L. Studies have also shown that there is a strong connection between arsenic exposure and infant mortality (Rahman et al. 2007).

Currently, there is no effective treatment for arsenic toxicity (Bhowmicka et al. 2018). Prevention from arsenic exposure is the only assured way to limit the effects of arsenic poisoning. Humans are exposed to arsenic through various ways as discussed above and demonstrated through the figure below (see Fig. 8.1). The intake of micronutrients such as vitamin E, C, A, zinc, selenium and folic acid have proven

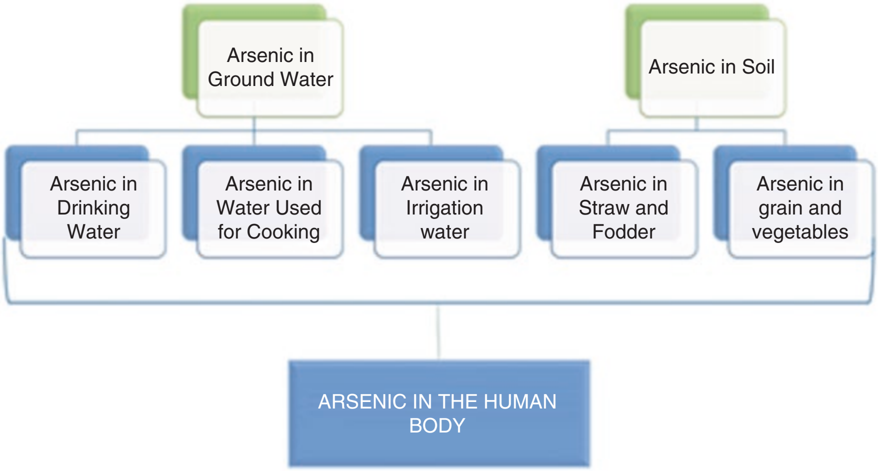

Fig. 8.1 Pathway of arsenic to human body (Shakoor et al. 2017)

to be effective in relieving the symptoms of arsenicosis, particularly skin lesions and helping in fast-tracking the natural secretion of arsenic from the body (UN Children Fund 2010). UNICEF's Arsenic Primer Study notes that it is critical to ensure that these vitamins are not over-prescribed as they can lead to toxic dosages, and mostly these compounds require time between 6 and 12 months to be effective, with recurrence rates being high (UN Children Fund 2010). It is important to find alternate solutions and therapies that work in a short period of time and have minimum side-effects.

3 Scope of Arsenic Contamination in India

As discussed above, arsenic occurs naturally in the environment and it is converted into inorganic arsenic compounds when it combines with other elements such as oxygen, chlorine and Sulphur. It is widely used in agriculture, electronics, medicine, metallurgy, livestock feed and chemical warfare agents. As a contaminant, arsenic is substantial with respect to its toxic nature and its extreme diverse signs of poisoning. The major source of arsenic ingestion in the human system is drinking water. According to the Bureau of Indian Standards (BIS) 2012, the permissible limit of arsenic in groundwater is 0.01 mg/L. In the absence of an alternate source, the permissible limit is 0.05 mg/L. WHO's provisional guideline for arsenic in drinking water is 10 µg/L (0.01 mg/L) (WHO 2011). While significant concentrations of arsenic in ground water are reported throughout India, the Ganga-Brahmaputra Plains are most largely affected.

The Central Ground Water Board (CGWB) (2015) noted that, "the aquifers in the alluvial plains embedded within the Late Quarternary deposits are also reported to be affected, with a few exceptions where the Hard rock aquifers are also affected as in Chhattisgarh and Karnataka states." The Central Ground Water Board Authority of India's survey found high concentration of arsenic above the relaxed limit of 0.05 mg/L in groundwater in 86 districts of 10 states, namely, West Bengal, Assam, Bihar, Jharkhand, Uttar Pradesh, Punjab, Haryana, Chhattisgarh, Karnataka and Manipur. The concentration of arsenic in groundwater is marked by a wide spatial variability. With respect to the depth, the contaminated water is usually confined within 100 m in the alluvial aquifers. Furthermore, a recent survey by CGWB has shown concentrations of arsenic in excess of 0.01 mg/L from additional 11 states and the occurrence of arsenic in these regions are reported from only limited samples (Ministry of Water Resources, RD, and GR 2015). The Ministry of Water Resources (2015) has noted:

> "While there have been suggestions that the groundwater arsenic contamination is predominantly restricted to the alluvial aquifers of the Ganges delta that consists of sediments transported from the sulfide-rich mineralized areas of Bihar and other surrounding areas of the basin of deposition; recent studies have however revealed that the 'vast tract of Indo-Gangetic alluvium extending further to the west and the Brahmaputra alluvium have elevated concentrations of Arsenic in wells placed in the late Quaternary and Holocene aquifers."

There have been reports of arsenic concentration greater than 50 ppb within the North-eastern states of India. Thus, millions of people are at serious risk of being poisoned from arsenic. Furthermore, arsenic levels in various parts of Assam, Manipur, Tripura and Arunachal Pradesh were reported to be above 300 parts per billion (ppb) in 2007 (Ministry of Water Resources 2015). According to a 3-year survey conducted by the Assam State PHE Department in collaboration with UNICEF, Jorhat district was found to be one of the worst arsenic hit areas amongst the 17 districts in the state with high groundwater arsenic contamination (Chakraborti et al. 2016b). Titabor, a subdivision of the Jorhat district is reported to have lethal levels of arsenic in its groundwater (Ministry of Water Resources 2015). The following part will examine the extent of arsenic contamination in the most affected states of India to better understand the enormity of this issue.

3.1 West Bengal

In West Bengal, 79 blocks in 8 districts had arsenic above the permissible limit of 0.05 mg/L in their groundwater (Kunar 2009; Chaurasia et al. 2012). Chakraborti et al. (2009) conducted a study over 20 years and analyzed water samples from 140 to 150 tube wells in all the 19 districts of West Bengal for arsenic. They found that 48.1% had arsenic above the permissible limit of WHO of 10 μg/L, 23.8% had arsenic above the Indian Government limit of 50 μg/L and 3.3% had arsenic above 300 μg/L (Chakraborti et al. 2009). Based on the arsenic concentration, this study divided West Bengal into three zones, namely, highly affected, mildly affected and unaffected zones. The highly affected zone included nine districts along the eastern side of the Bhagirathi river i.e. Maldah, Murshidabad, Nadia, North-24 Parganas, South-24 Parganas, Bardhaman, Howrah, Hooghly and Kolkata. Some tube wells in this zone reported arsenic above 300 μg/L. Moreover, 95% of the blocks in Murshidabad, Nadia, North-24-Parganas, South-24 Parganas and Maldah are affected with arsenic contamination. The districts mildly affected with arsenic concentrations mostly below 50 μg/L include Koch Bihar, Jalpaiguri, Darjeeling, Dinajpur-North and Dinajpur-South. While 16.2 million people live in the highly affected areas of arsenic contamination (Chaurasia et al. 2012), more than 26 million people are actually at risk of consuming arsenic-contaminated water. Therefore, the arsenic contamination of water is a huge public health crisis and a national and international concern. Thus, several international aid agencies such as the WHO and UNICEF are providing support and assistance to the state government for analyzing the magnitude of this problem and finding potential measures to mitigate it (Bhowmicka et al. 2018).

3.2 Bihar

Bihar is divided into two physiographic units by the Ganga River, namely the North and South Ganga Plains (Singh 2015). Bihar has a two-tier aquifer system that is formed through its geological stratification into the Holocene newer alluvium and the Pleistocene older alluvium (Saha 2009). A 15 to 32-m-thick aquitard divides the deeper aquifer systems and the shallow aquifer systems (Singh 2015). It is interesting to note that the shallow aquifer was contaminated by arsenic, but on contrary the deeper aquifer was not contaminated by arsenic.

In Bihar, groundwater arsenic contamination first appeared in two villages, Semaria Ojhapatti and Barisban in the Bhojpur district in the Middle Ganga Plain, in 2002. This area is located in the flood-prone belt of Sone-Ganga inter-fluvial region. The Central Ground Water Board and Public Health Engineering Department have conducted investigations and found arsenic contamination as high as 0.178 mg/L in the nearby villages, thereby impacting the hand pumps that are located at 20–40 m below ground-level (Ministry of Water Resources 2015). Chakraborti et al. (2016a) conducted a study to investigate the extent and severity of arsenic contamination in five blocks of Patna District, Bihar. They collected 1365 hand tube well water samples and found that 61% of the samples had arsenic above 10 μg/L, 44% of the samples had arsenic above 50 μg/L and the maximum concentration of 1466 μg/L. The study also found that all the biological samples collected had arsenic above the normal levels, and 69 out of 712 people screened had arsenic skin lesions (Chakraborti et al. 2016a).

In Bihar, 17 out of 37 districts and 87 of 532 blocks have been studied to investigate the extent of arsenic contamination in groundwater. While only three community blocks were safe from arsenic contamination (Ghosh et al. 2009), 12 community blocks had arsenic below the WHO standard of 10 μg/L (Gupta et al. 2014; Singh 2015). Though, ten community blocks had arsenic above the WHO guideline standard, they were below or equal to the BIS standard of 50 μg/L (Singh 2015). Meanwhile, 62 community blocks had arsenic contamination above the BIS standard of 50 μg/L (Singh 2015; Ghosh et al. 2009; Nickson et al. 2007; Saha 2009; Singh and Choudhary 2010). Approximately 13 million people live in these community blocks that are contaminated by arsenic (Singh 2015). It is important to note that these districts are predominantly distributed along the course of the Ganga River, with the exception of Darbhanga, Purnea and Kishangarj (Ministry of Water Resources 2015).

3.3 Uttar Pradesh

In the state of Uttar Pradesh (UP), the first case of groundwater arsenic contamination was reported in 2003 during a survey of 25 villages in the Ballia district (Ministry of Water Resources 2015). It was suspected that arsenic would be found in UP because of the presence of the Holocene sediments of the active river systems, like in West Bengal and Bangladesh (Scharp et al. 2018). This was confirmed in the Ballia District and then used to focus further testing in selected “arsenic risk” blocks (Scharp et al. 2018).

Arsenic contaminated tube well water was identified in 20 out of 70 districts through a study conducted by the UP government with assistance of UNICEF (Yasunori et al. 2012). However, this study was only performed on the government tube wells and did not include the numerous privately-owned tube wells. Ahamed et al. (2006b) conducted a 2-year survey to examine arsenic contamination of groundwater in three districts of UP namely, Ballia, Varanasi and Gazipur. This study analyzed water samples from 4780 tube wells and found that 46.5% had arsenic concentration above 10 μg/L, 26.7% had arsenic concentration above 50 μg/L and 10% had arsenic concentration above 300 μg/L. The maximum arsenic levels observed by this study were 3192 μg/L. Critically, this study noted that older tube wells had a greater chance of arsenic contamination (Ahamed et al. 2006b).

3.4 Jharkhand

In the state of Jharkhand, the first case of arsenic contamination was reported in the Sahibganj district in 2003–04 but was only confirmed by CGWB through a detailed investigation in 2006–07.

Mukherjee et al. (2006) surveyed four blocks in Jharkhand i.e. Shahibganj, Mandaro, Taljhari, and Rajmahal from the Shahibganj district and discovered that the while, all blocks had arsenic concentration above 10 μg/L, Shahibganj block had arsenic concentration above 50 μg/L. They analyzed 1024 water samples from 17 villages and found that 30% of the samples had arsenic above 10 μg/L and 19.4% has arsenic above 50 μg/L. Nayak et al. (2008) also conducted a detailed study in the district of Sahibganj in Jharkhand and found that 178 tube wells in three villages were highly contaminated by arsenic. Specifically, 91% of the tube wells had arsenic concentration above 10 μg/L, 79.8% above 50 μg/L and 42% above 300 μg/L. In these three villages of the district of Sahibganj, this study found that out of 522 people examined, 71 people were reported with arsenical skin lesions, and out of the 40 children examined, nine were also reported with arsenical skin lesions. Additionally, this study noted various ‘clinical and electrophysiological neurological features and abnormal quantitative sensory perception threshold’ (Nayak et al. 2008).

3.5 Manipur

Manipur is one of the seven North-Eastern Hill states of Indi and has severe arsenic contamination of its groundwater, specifically in its valley districts. Manipur has nine districts, and four of them are located in the Manipur Valley which constitutes only 10% of the State’s land but is home to 59% of the people. In 2004–05, CGWB reported arsenic in four samples of Thoubal and Bishnupur districts (Scharp et al. 2018). Chakraborti et al. (2008), analyzed water samples from 628 tube wells out of expected total of 2014 tube wells and found that 63.3% contained arsenic concentration above 10 µg/L, 23.2% had arsenic concentration between 10 and 50 µg/L and 40% had arsenic concentration above 50 µg/L. This study also found that in Manipur there was no co-relation between the depth of the tube well and arsenic contamination as opposed to other arsenic affected states of India in the Ganga–Meghna–Brahmaputra (GMB) Plain. Since the state water supply primarily depends only on surface water, the wells have been abandoned and their water is not being used for drinking, cooking or agricultural processes (Ministry of Water Resources 2015).

3.6 Punjab

According to Thakur et al. (2016), a study conducted in 2007 reported arsenic contamination in the alluvial aquifers of Punjab between 3.5 and 688 µg/L. This study found that in southwestern Punjab, 11% of the aquifers had arsenic between 10 and 25 µg/L, 54% had arsenic concentration between 25 and 50 µg/L and 35% had arsenic concentration above 50 µg/L (Thakur et al. 2016).

Singh et al. (2015a), also reported that arsenic concentration above 10 µg/L appear in various places along the Muktsar-Malout belt in Talwandi Sabo area that is located in the southern-western parts of Punjab. Singh et al. (2015a) also found that arsenic contamination was more dominant in the newer alluvium. Moreover, most of the districts contaminated by arsenic are situated along two rivers that originate from the Himalayas, Ravi and Beas.

The Department of Water Supply and Sanitation of the Government of Punjab reported in 2012 that five districts had arsenic concentration above the permissible limit. Namely, Amritsar District had arsenic concentration of 0.099 mg/L, Taran Taran had arsenic concentration of 0.083 mg/L, Firozepur District had arsenic concentration of 0.055 mg/L, Gurdaspur District had arsenic concentration of 0.058 mg/L and Rupnagar District had arsenic concentration of 0.091 mg/L. The CGWB also reported that the ground water in 13 out of 22 districts in Punjab were contaminated with arsenic within the range of 0.01–0.39 mg/L. The following districts according to CGWB had arsenic above the permissible limit of 0.05 mg/L: Amritsar, Taran Taran, Mansa, Kapurthala, Fazilka and Rupnagar district (Ministry of Water Resources 2015).

3.7 Karnataka

The Department of Mines and Geology of the Government of Karnataka has reported the presence of arsenic in ground water in two districts, namely Yadgir (old Gulburga) and Raichur (Ministry of Water Resources 2015). It is theorized that gold mining and associated activities are a contributing factor to arsenic contamination

(Kozhisseri 2008; Bhattacharya and Lodh 2018). Moreover, studies conducted by UNICEF and the Government of Karnataka found the water well supplies of many villages contaminated by arsenic, above the WHO guideline of 10 μg/L (Government of Karnataka 2008, 2010).

In a study conducted by Chakraborti et al. (2012), 181 individuals of Kiradalli Tanda village were screened for arsenical symptoms and 58.6% i.e. 106 of the total individuals had at least one skin lesions caused bychronic arsenic toxicity. In this study, 94.5% of these individuals volunteered skin and hair samples for analysis, and it was found that 100% of the samples had elevated levels of arsenic (Chakraborti et al. 2012).

4 Methods to Measure Arsenic Contamination Levels in Water Sources

There are two methods to measure the amount of arsenic contamination in water: the laboratory chemical analysis and field test kit. The measure of the field test is more qualitative and the choice of method for analysis is contingent on specific criteria, which includes the precession of the measurement required (Talbi et al. 2005).

There are two kinds of field tests, first, the one that provides a 'yes or no' answer and second provides a range of concentration. The field test that provides a 'yes or no' is not beneficial to conduct further analysis or aide the implementation and mitigation measures. Moreover, quality control is required to ensure the reliability of analysis within a specific laboratory and also to ensure consistency of the measurement between different laboratories (Talbi et al. 2005). In India, West Bengal is the only site where arsenic is screened through entirely using laboratory spectrometer analysis; thus, reducing the risk of a misclassifying a sample of water as contaminated. It is important to note that there is a higher risk of well misclassification when using field tests, and this risk can be reduced through using multiple tests (Talbi et al. 2005). However, field testing may be sufficient for the preliminary screening tests of arsenic contamination.

A World Bank Report found that for the purpose of testing whether the water source exceeds a standard of 10 or 50 ppb, a field test kit does not really need to be able to distinguish between 200 and 300 ppb to further identify if the water is contaminated (Talbi et al. 2005). The report also found that in India, surveys have used field test in a semi-quantitative way to categorize well groundwater as above or below the national standard of 50 ppb. Furthermore, it was noted that the field tests prove to be very reliable when testing if the arsenic is well above or below the national limit, and found that the field kits are likely to have errors in the 25–100 ppb range (Talbi et al. 2005).

5 India's Water Governance

India's water sector is witnessing challenging times. With increasing scarcity and pollution of water, there are significant challenges that plague this sector. In order to address the challenges of the Indian water sector and to ensure water security, it is critical to bring a paradigm shift in water management. Water governance is critical in addressing the challenge of arsenic contamination at the central level. Therefore, this section will review the main central institutions responsible for managing water in India.

5.1 Water: A Human Right

The General Comment No. 15 of the Committee on Economic, Social and Cultural Rights (2002) defined the right of water as:

> "The right of everyone to sufficient, safe, acceptable, physically accessible and affordable water for personal and domestic uses."

The General Comment No. 15 (2002) also noted that the right to water is critical and indispensable for leading a dignified life. Moreover, in 2010 and through the Resolution 64/292 the United Nations General Assembly also officially recognized the 'right to safe and clean drinking water and sanitation as a human right that is essential for the full enjoyment of life and all human rights' (United Nations Resolution 64/292, 2010). For instance, the right to water and sanitation is a prerequisite to enjoy other human rights such as the right to food, the right to health and life, and the right to housing.

In India, the human right to water and sanitation is not explicitly mentioned in its constitution. However, both the state courts and the Supreme Court have interpreted article 21 of the Indian Constitution i.e. the right to life as inclusive of the right to safe and clean drinking water, and sanitation (Catarina de Albuquerque 2014). Furthermore, Article 39 (b) of the Directive Principles of State Policy (DPSP) acknowledges the principle of equal access to the material resources of the community and mandates that "the State shall, in particular, direct its policy towards securing that the ownership and control of the material resources of the community are so distributed as best to sub serve the common good" (Catarina de Albuquerque 2014).

The recognition of water as a human right is critical for the following reasons: first, it ensures that the necessary standards are established so that countries can be held accountable; second, it provides clarity for the role of states in providing access to water; and third, it prioritizes access to water for the vulnerable and marginalized people without access (Singh 2016b, c).

5.2 India's Water Governance Structure

Arsenic contamination of groundwater is a multi-dimensional problem and cannot be treated as a silo. This is a huge issue that has been prevalent in the country for the past four decades, across 96 districts in 12 states with enormous impact on the health of human and animal populations in those regions. Currently, there are seven ministries and several organizations at the central level that are responsible for handling this issue, exclusive of the state governments and their agencies, namely: Ministry of Drinking Water & Sanitation, Ministry of Water Resources, Ministry of Health & Family Welfare, Department of Agricultural Research and Education, Ministry of Science & Technology, Department of Agriculture and Cooperation, Department of Animal Husbandry, Central Ground Water Board, Central Ground Water Authority, Water Quality Assessment Authority, Council of Scientific & Industrial Research, and Indian Council of Agricultural Research.

The legal and governance structure of India to manage groundwater is a complex, multilayered framework of constitutional and statutory provisions at both the central and state levels. While groundwater is part of the jurisdiction of the states, the central government had issued a model groundwater bill in 1970, which has been revised and circulated many times (Garduño et al. 2011).

However, only selected states have formally adopted this bill. World Bank's study on India's groundwater governance found two main legal drawbacks namely, water being assumed to follow the right to land and the absence of groundwater legislation at the central level (Garduño et al. 2011). These legal drawbacks were addressed through case law affirming the human right to water and the government's right and obligation to protect ground water under the right of life stated in the Indian Constitution (Garduño et al. 2011). India's Planning Commission's Groundwater Expert Group have argued that the priority should be on enforcing and implementing existing measures as the current legislative framework is reasonably strong for effective groundwater management.

One of the main policy affecting groundwater management are the 1998 National Water Policy (NWP) and the 2002 amended version (Ministry of Water Resources 2012). While these policies were a result of intense political discourse, they cannot be legally enforced because they have no statutory status. Nevertheless, state governments can find them valuable for developing their own water policies. Furthermore, the governance framework for groundwater protection is very complex because while, the Water Act of 1974 and the Environmental Protection Act of 1986 address main pollution issues in India, there are at least 13 other related policies and acts that address this issue (Garduño et al. 2011).

In India, water is managed under two separate categories, namely surface water and groundwater. The Central Water Commission (CWC) is responsible for governing surface water, while the Central Ground Water Board (CGWB) is responsible for groundwater. The following section will critically review both the CWC and CGWB.

5.2.1 Central Water Commission (CWC)

In 1945, the Central Waterways, Irrigation and Navigation Commission was established. In 1974, the water sector was separated and the CWC became an individual entity. Today, the CWC is the technical division of the Ministry of Water Resources and is responsible for matters relating to flood control, irrigation and multipurpose projects. The CWC's scope of work also includes the planning, development and management of the surface water resources of the entire country.

The commission has significant challenges with respect to its mandate and means of implementation. The responsibility of the commission is restricted to only surface water resources, and it fails to include other components of water resources in the hydrological cycle, specifically groundwater. Since water is interconnected, it is imperative that the governing agencies work in a holistic and comprehensive manner. A case in point: the river water quality monitoring function of CWC overlies and duplicates to a certain extent the work of the Central Pollution Control Board (CPCB) (Talbi et al. 2005). Further, the hydro-meteorological data collection overlays the role of the India Meteorological Department (IMD) and the hydrological research and studies actually lie within the function of the National Institute of Hydrology (NIH) (Talbi et al. 2005).

Furthermore, the CWC does not focus or produce any expertise on issues relating to the socio-economic impact of water and the environment. Further, it does not have sufficient knowledge on efficient irrigation management and water utilization. A significant function of the CWC is to provide training to in-service engineers from the central and state organizations in diverse aspects of water resource development. However, the CWC only focuses on the engineering side of water projects and does not provide a whole understanding and training in water management (Talbi et al. 2005). The CWC in its current form is only able to address a small percentage of the water management issues, with several significant components of the hydrological cycle and its processes remain untouched (Talbi et al. 2005). Furthermore, the commission's current focus is on the supply-side management of water, and as it is primarily staffed with engineers, it fails to include any knowledge of any other discipline that interacts with water resource management (Talbi et al. 2005).

5.2.2 Central Ground Water Board (CGWB)

The CGWB was created in 1950 and is responsible for exploring and developing the groundwater resources of the country. During the commencement, the board was part of the agriculture division, however in 1971, under the Ministry of Water Resources it became its own entity. Initially, the mandate of the CGWB was very specific wherein they had to drill exploration wells to assess groundwater resources and further instruct the method and time to harness the resource through drilling. Gradually, the CGWB took on more responsibility to monitor the groundwater resources and ultimately became the top national organization that deals with groundwater resources in the country. The CGWB also supports the State Groundwater Boards (SGBs) who are responsible for these activities on the state level.

The exhaustive data on India's groundwater that was generated through years of field studies is one of the biggest contributions of the CGWB. Nevertheless, the scope of the CGWB's work on groundwater is limited because of the lack of cooperation and institutional will between the central and state governments. Further, the deployment of outdated equipment also severely limits the work of the board and its focus on the supply side solutions (Talbi et al. 2005).

Hydrologically, the two categories of water, i.e. surface water and groundwater, are interconnected and cannot be governed as silos. Thus, it is important to create a shift in the institutional framework of the CGWB and CWC to ensure a more holistic and comprehensive water management framework.

5.3 National Water Commission: A Viable Option?

The Government of India set up the Mihir Shah Committee, a seven-member committee headed by Dr. Mihir Shah, to provide recommendations for the reform of the country's water sector. The Committee created a report called, *A 21st Century Institutional Architecture for India's Water Reforms* (Committee on Restructuring the CWC and CGWB 2016). The report noted that infrastructure projects related to water resources should be enhanced through enabling technology, assessment, planning and monitoring capabilities. The projects should be deployed effectively through a demand-based approach and a partnership between the central and state governments. The Mihir Shah report further noted that the water resource governance requires a high-level organization that is forward-looking, transdisciplinary, strategic and responsive in its skill set. Furthermore, the report asked for a more action-oriented organization and not just a data collection and analysis organization. The report proposed to establish the National Water Commission (NWC) to replace CWC and the CGWB and become the highest organization managing and governing issues of water policy, data, and governance.

The structure proposed by the Committee for the NWC is as follows: first, they proposed that the NWC should be an adjunct office of the Ministry of Water Resources and function with essential accountability and full autonomy. Second, the committee proposed that the NWC should be led by the chief national water commissioner, a senior administrator who has a solid tenure and extensive background in the public and development administration, and finally have representatives of the hydrology (present chair, the CWC), hydrogeology (present chair, the CGWB), hydrometeorology, river ecology, ecological economics, agronomy (with focus on soil and water) and participatory resource planning & management. Third, the NWC must have an extensive presence in all the regional and major river basins of India. Fourth, the committee proposed that the NWC should enable and construct an architecture of partnerships with educational and research-based institutions and practitioners in the water sector, specifically in areas where internal expertise is lacking. The proposed responsibilities and mandate of the NWC by the committee are divided into several divisions for effective implementation, namely: Irrigation reform, river rejuvenation, aquifer mapping and participatory groundwater management programme, water security division, urban and industrial water management division, water quality division, data management and transparency division and the knowledge management system and capacity building division (Committee on Restructuring the CWC and CGWB 2016).

The main assumption of the report was that a centralized institution could promote integrated water resources management planning at the basin level for both surface and groundwater. However, lack of coordination and data sharing are not the reasons for the shortage of integrated planning at the ground level; but it is the inability of the concerned state departments, that control the water resource, to foresee how the future development of the water resource will impact its quality, supply and demand. Thus, can the two central agencies CWC and CGWB be held responsible for the poor state of affairs in the water sector? This report also fails to identify a significant problem that the state level water agencies such as the water resources department, water supply department, watershed management agency, and state pollution control board, do not have the incentive and the will to perform effectively because of the inherent problems in the institutional design (Water Policy 2017). These problems include: sectoral, disintegrated supply-side approach to water management, inadequate water resource monitoring and assessment, a centralized approach of institutions and lack of well-defined water rights (Water Policy 2017).

Furthermore, the report recommends participatory aquifer mapping versus groundwater management. The two main reasons for the over-exploitation of aquifers are the absence of well-defined water rights in groundwater and the inefficient pricing of electricity that is supplied through the farm sector (Water Policy 2017). Lack of data and insufficient information regarding the groundwater levels and its flows, do not mitigate the fact that the resources are depleting and getting contaminated at a rapid speed, something farmers and official agencies are well aware of (Kumar et al. 2016). Participatory aquifer mapping is not a solution to the ongoing challenges. Certainly, investing more resources in refining the current assessment methodology does not cause any harm, but the information needed to take concrete and action-oriented steps is already prevalent. The actions needed are institutional in nature, and the lack of political will from the state governments to implement those actions is the main determent. This lack of will is clearly demonstrated through the lack of enforcement of groundwater legislations in various states, with the Maharashtra Groundwater Development and Management Act (Bombay High Court 2009) being a prime example.

Precedence form developed countries such as the US and Australia, in addressing groundwater management challenges clearly demonstrates that the solution is in creating more robust institutions, working on practical implementation and have interventions that clearly define water rights of every citizen. It is imperative that these institutions are supported through an appropriate legal framework that is able to define, recognize and enforce the water rights of every citizen.

This paper finds that the NWC can provide a top-down approach and the required strategic, political, technical and governance guidance for the water sector in India. However, it is also critical that each individual state takes more responsibility in reforming their institutions and taking the required bottom-up steps to ensure more efficiency in water policy, management and governance.

5.4 Water Governance: Women, Communities and Civil Society

The Ministerial Declaration adopted during the International Conference on Freshwater (2001) stated that:

> “Water resources management should be based on a participatory approach. Both women and men should be involved and have an equal voice in managing the sustainable use of water resources and sharing the benefits. The role of women in water-related areas needs to be strengthened and their participation broadened.”

The rural water supply program in India has been designed and enforced within the capacity of the international and national policy frameworks. The National Water Policy (Ministry of Water Resources 2012) prioritized safe drinking water facilities in rural areas, and the National Policy on Empowerment of Women also stressed that women in rural areas need access to safe drinking water to households (Singh 2006b). These policies are formed by the national and state level ministries and departments that are accountable for the management of the country’s water, rural development and women’s development (Singh 2006b). The most extensive program was the ‘Accelerated Rural Water Supply Program (ARWSP), launched in 1972–73. The aim of this program was to assist states in enforcing and implementing schemes that supplied safe drinking water to villages. In 1986 this program was transformed into a technology mission and called Rajiv Gandhi National Drinking Water Mission’ (RGNDWM) (Singh 2006a). The main agencies responsible for implementing this program at the local level included the Ministry of Rural Development, the Department of Social Welfare, the Department of Public Health Engineering (PHED) and the Panchayati Raj Institutions (PRI).

It is important to note that the principal beneficiaries of the ARWSP were considered to be women because it was assumed that providing access to safe drinking water would reduce their burden of water-fetching and enable better health and improve socioeconomic opportunities (Singh 2006a). Moreover, the participation and role of women has improved as they are increasingly becoming members of the PRIs and participating in the decision-making process.

The ARWSP program defines access to safe water in accordance with its coverage and has the following provisions: the daily requirement of water for human beings is 40 L per capita; for every 250 people, there should be one hand-pump or stand-post; and independent habitations that don't have any potable sources can have one source provided that the population is less than 250 and there are either 20 households or 100 people. If the provisions mentioned above are not fulfilled, then the habitations are not considered covered. According to this program, areas that have a water source but have water quality issues, is considered a 'no safe source' habitation. To address the issues of water quality under the ARWSP, states can install treatment plants for arsenic removal, defluoridation and iron removal, organize awareness camps and create water testing laboratories (Singh 2006a).

Most of the water supply programs are designed by bureaucratic organizations that follow a blanket approach and use common international and national design frameworks, the same maintenance, management and financing systems regardless of the local social and economic conditions (Smet and van-Wijk 2002). Moreover, Singh (2006a) conducted a first-hand ethnographic study in selected rural areas of the following states in India: Madhya Pradesh, West Bengal, Bihar and Jharkhand. This study found that while improved supply sources of water were provided to various rural areas in India under the ARWSP, women in particular did not wholly adopt the new source and abandon the traditional sources. According to the Planning Commission (2002), approximately 3.5 million hand-pumps in 100,000 piped water schemes have been installed in villages. Singh (2006a) found that majority of the women had not adopted the new source for the most important purpose i.e. drinking. It is critical to assess the level and nature of the use of the technology being exported to the affected rural areas. Singh (2006a) found in their study that the new technologies provided were not able to meet the needs and aspirations of women, who are the decision makers in selecting the source water and thus, the technologies. Consequently, the assumed advantages of implementing new water technologies and sources such as better health and enhanced socio-economic opportunities are not fully achieved.

Therefore, it is imperative that the local people participate in the water governance process through forming local committees and groups regarding water quality management, irrigation and watershed. The civil society and non-governmental organizations (NGO) can prove to be a huge asset in this respect. A case in point: An NGO initiated program, funded by a Canadian agency, installed 182 arsenic removal plants in arsenic affected areas of West Bengal. This program found that improved participation of women as members of water quality management committees, at the local level, was critical to the sustainable use and management of the arsenic removal plants (Singh 2006a).

6 Challenges in Mitigating Arsenic Contamination in India

High arsenic contamination in groundwater is of great concern because it impacts human, animals, soil and plant ecosystem and has caused over 100,000 deaths and at least 200,000 confirmed cases of illness. The '*Occurrence of High Arsenic Content in Ground Water*' report noted that at least 96 districts in 12 states had been affected by groundwater arsenic contamination. Over 70.4 million people are affected by this issue in only 35 districts. Furthermore, abnormalities have been detected in at least 40% of animals living in arsenic contaminated areas. It is startling to find that in spite of the severity of this issue and its rapid growth over the last three decades, no concrete action has been taken by the central government to coordinate activities or address the challenges faced because of arsenic contamination. Though, in 2010, the National Institute of Hydrology (NIH) and the Central Ground Water Board (CGWB) created a vision document called *Mitigation and Remedy of Ground Water arsenic menace in India*; no concerted action is really visible to implement the vision. There is no reference to arsenic contamination in the 'National Water Policy 2012' and there is no separate budgetary allocation that addresses arsenic concerns or any water quality issue. It is important to note that while the 'National Rural Drinking Water Programme' under the Ministry of Drinking Water & Sanitation received funding for water quality, it's critical that this program allocates a separate budget for water quality concerns and clearly distributes them for specific issues such as arsenic contamination.

6.1 Arsenic Data Collection Challenges

Despite the fact that the first case of arsenic contamination was reported more than five decades ago, the government does not have comprehensive data about the affected states and districts, and more importantly the number of people affected by this issue. Moreover, there are different numbers and data reported by the various ministries and departments of the Indian Government. While the Ministry of Water Resources, River Development and Ganga Rejuvenation (M/o WR, RD & GR) have reported arsenic contamination exceeding the permissible limits in 86 districts in 10 states, the Department of Agricultural Research and Education (DARE) has reported on 71 districts in 9 states with arsenic contamination above permissible limits. Moreover, the Department of Science and Technology (DST) has reported a different list of affected states and districts. The '*Occurrence of High Arsenic Content in Ground Water*' report states that 96 districts in 12 states suffer from groundwater arsenic contamination. The Council of Scientific and Industrial Research (CSIR) has noted that 70.4 million people are affected from arsenic contamination in only 36 districts, and this number will be significantly higher once people in all 96 districts are accounted for. The discrepancy of the data presented above clearly demonstrates the lack of will by the central government to collect reliable data on the extent of arsenic contamination and the people affected by it. Reliable and accurate data are critical to provide effective policy solutions to issues of public health, water quality management, agriculture, food security, irrigation and other purposes.

6.2 Source of Arsenic and Mitigation Challenges

The Indian Council of Agricultural Research (ICAR) found that more than 90% of arsenic contaminated water is used for irrigation purposes. According to the vision document of the NIH and CGWB, the major exposure pathway of arsenic was the water-soil-crop-food transfer. Naturally, exposure to arsenic from food and water increases the health risk of arsenic exposure.

No real effort has been made by the central government to actually find the root cause of arsenic in groundwater that can further help the mitigation process. The Ministry of WR, RD & GR have noted that elevated level of arsenic in groundwater is caused largely by natural geogenic processes and partly due to anthropogenic activities such as the use of fertilizers, burning of fossil fuels, and mining activity. UNICEF and the Ministry of Water Resources have noted that the sediments in the Ganga Brahmaputra plain contain arsenic compounds. However, there is still more clarity needed to understand the release of the arsenic process in groundwater because it is contingent on various physio-chemical conditions, hydro-geological characteristics of aquifers, the presence of arsenic compounds in sediments and the dynamic nature of aquifers. Therefore, it is critical for the government to conduct a time-bound study that establishes the sources of arsenic to mobilize the mitigation process with respect to groundwater arsenic contamination.

The Water Quality Assessment Authority (WQAA) was created in 2001 to address "any environmental issue concerning surface and groundwater quality and reviewing the status of quality of natural water resources." It is important to note that initially WQAA's mandate excluded quality issues that arose from geogenic activities, but it was later revised and included. While this institution has not achieved much in its 16 years since inception, it can play a critical role in monitoring and assessing the water quality of both surface and ground water in a comprehensive and holistic manner. To ensure this, the government of India must provide the necessary budgetary allocations to provide the institution with sophisticated equipment and trained professionals.

6.3 Central Leadership Challenges

Arsenic contamination is a multi-dimensional problem and cannot be treated as a silo. This is a huge issue that has been prevalent in the country for the past four decades, yet the Government of India has failed to create a strong institution or policy at the central level to deal with this issue. It is often argued that water is a state issue and therefore the central government is not entirely responsible for addressing this issue. However, this is a threat faced by at least 96 districts across 12 states with enormous impact on the health of human and animal populations in those regions. Therefore, the central government must act on an urgent basis. Currently, there are seven ministries and several central organizations at the central level that are responsible for handling this issue, exclusive of the state governments and their agencies. However, as noted before, these organizations have failed to coordinate their efforts and implement a strong policy to address this issue. This paper recommends that a specific body should be created at the central level to coordinate all efforts on arsenic contamination in groundwater. Further, this agency and all the other ministries mentioned above should ensure that a cohesive policy framework, effective mitigation program and an efficient coordination mechanism is in place to address all arsenic-related issues and concerns.

7 Recommendations

Long-term actions are needed to reduce arsenic exposure from anthropogenic activities such as mining, metal smelting and refining, burning fossil fuels, use of pesticides and timber treatment. Specifically, urgent action is needed to reduce the consumption of arsenic from drinking water and food in areas with a high level of arsenic due to geological factors. The following section of the paper will first provide recommendations for risk-mitigation, then for the water, health and agriculture sectors and finally stress on the importance of partnerships and advocacy.

7.1 Risk-Mitigation Recommendations

It is imperative that governments ensure that alternate source of drinking water are provided to local population where the arsenic concentration in the groundwater is higher than 10 μg/L by taking the following steps: rainwater collection (but ensure preventative measures are in place to avoid microbial contamination and breeding of mosquitoes); install surface based pipe water system; installing domestic or centralized arsenic removal systems and ensure the appropriate disposal of arsenic; and painting hand pumps to highlight the difference between high arsenic and low arsenic water source (Talbi et al. 2005). Further, it is important to note that, arsenic contaminated water does not have severe health effects if used on skin, so it can still be safely used for laundry, bathing and handwashing purposes (WHO 2010). It is also critical to make both the public and health professionals aware of the negative impacts of arsenic consumption and the methods to avoid it (WHO 2010).

7.2 Local Management Recommendations

The local management recommendations put grave emphasis on the local leadership to manage arsenic presence within the following three sectors: health, water and agriculture.

The following are the recommendations for the water sector. First, provide the already identified arsenic contaminated regions with the required financial and technical support to provide an alternate source of safe drinking water. Second, a strong arsenic monitoring system should be established and maintained at the local level. This should be done by registering all water sources in the area and also paint the status of whether the water source has been checked or not e.g. unpainted for unchecked, red for arsenic contaminated and green for safe. Based on this information, determine the areas of safe water for the people and also in this process create more public awareness on this issue. Third, it is imperative to strengthen the water quality monitoring capacity in both the private and public sectors to ensure that the chemical and biological quality standards are consistent. Fourth, the local governments should regulate the indiscriminate use of deep groundwater (UN Children Fund 2010).

This paper provides the following recommendations in the health sector. First, local medical and health organisations should establish an arsenicosis patient registry to enable early identification of such patients. This should be supported through a robust surveillance program managed by the local authorities. Second, the district hospitals should have the required resources and expertise to manage arsenicosis patients in most affected areas (UN Children Fund 2010). Third, there should be constant communication between the hospitals and authorities wherein they must report any new case of arsenic contamination and share the location of the patient. This is to ensure that new water points can be targeted to those villages. Forth, the local health authorities should create more awareness about the health risks of arsenic exposure. Fifth, invest in the training, screening facilities and the required equipment in the local hospitals. Finally, promote school-based education about arsenic to increase the awareness of this issue and ensure that factually correct information is being disseminated in the public (UN Children Fund 2010).

The following are the recommendations for the agriculture sector. First, optimize the combined usage of surface water and groundwater to minimize the exposure of the crops to arsenic and ensure that the sustainability of groundwater for domestic water supply is not compromised. Second, monitor and regulate agriculture practices to prevent arsenic contamination from fertilizers in the groundwater and also to prevent arsenic contamination in people from food sources. This can be done through promoting and developing agriculture practices such as arsenic resistant varieties, using only surface water for irrigation purposes, and practicing raised-bed cultivation. Finally, create more awareness about the linkages of arsenic and agriculture amongst the relevant stakeholders.

7.3 Partnerships and Advocacy

Arsenic exposure and contamination is a multi-sectoral problem that involves a number of different stakeholders. While national, state and local governments have the overall responsibility for arsenic mitigation, other relevant stakeholders can play a significant supportive role. UNICEF's Arsenic Primer Report has noted that countries where arsenic mitigation programs have been going on for a long period of time demonstrate that involving key stakeholders and developing strong partnerships are critical to the success of the program (Scharp et al. 2018). Some of these potential stakeholders include: the central and state government water resource and supply ministries and departments; central and state government health ministries and departments; water resource agencies, technical departments of academic institutions and think tanks i.e. in the field of civil engineering, chemistry, environmental science, hydrogeology, water policy, public health, and medicine; international and United Nations agencies such as UNICEF, WHO, Water and Sanitation Programme, UNDP, World Bank; and other international and local NGOs such as WaterAid (Scharp et al. 2018).

Advocacy is a critical step to encourage and promote action by the respective government authorities and other stakeholders, including the civil society, relevant international organizations, funding agencies and UN agencies. It is imperative to effectively present the relevant information to different audiences such as local communities, water professional, health professional, government officials, the private sector and relevant people. According to UNICEF, successful advocacy in arsenic mitigation programs includes raising awareness through international, national and local conferences and media events, producing articles in local media outlets, briefing local educators, leaders, medical and water professional, and developing local working groups with representatives from the government, civil society and other relevant organizations (Scharp et al. 2018).

8 Conclusion

Arsenic contamination of groundwater in India and Bangladesh and its devastating impact on human health is reported to be one of the biggest natural groundwater catastrophes in the world. This paper has provided a comprehensive overview of the scope of arsenic contamination in India and the efforts taken by the Central Government of India to combat it. While the main source of arsenic is natural geogenic activities, anthropogenic activities such as metal smelting, fossil fuel combustion and mining also contribute to the problem significantly. According to the Government of India, over 96 districts in 12 states have reported arsenic over the permissible limit of 0.01 mg/L.

The paper concludes that arsenic contamination is a multi-dimensional problem and cannot be treated as a silo. The government of India has failed to create any central solution or policy or an institution to address this enormous groundwater and health catastrophe. Furthermore, there is a clear discrepancy of the data presented by various governmental ministries on the extent and scope of arsenic contamination in India. This clearly demonstrates the lack of will by the central government to collect reliable data on the extent of arsenic contamination and the people affected by it. Moreover, reliable and accurate data are critical to provide effective policy solutions to issues of public health, water quality management, agriculture, food security, irrigation and other related purposes. This paper recommends that a specific body should be created at the central level to coordinate all efforts on arsenic contamination in groundwater.

The first part of the paper critically analyzes India's efforts to combat arsenic contamination and clearly stresses on the importance of leadership and support of the Central Government in ensuring that a cohesive policy framework, effective mitigation program and an efficient coordination mechanism are in place to address all arsenic-related issues and concerns. However, when critically examining the water governance structure and institutions of India, the paper concludes that while leadership and support at the center is critical to address this grave issue, it is not the only solution. Thus, the paper highlights and stresses on the roles of the local and state governments, and institutions in effectively implementing, mitigating and coordinating solutions to combat arsenic contamination. Finally, the paper provides recommendations for risk-mitigation; local management of water, health and agriculture sectors and stresses on the importance of partnerships and advocacy to deal with arsenic contamination at the local level.

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Chapter 9
Biosorption of Arsenic: An Emerging Eco-technology of Arsenic Detoxification in Drinking Water

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Abstract Arsenic (As) contamination of drinking water and its severe human health impacts have been a global concern during the last few decades. In order to control the problem of As in drinking water, various chemical based treatment methods, such as coagulation, ion-exchange, adsorption, and reverse osmosis, are used in removing As from water. Most of these methods are not eco-friendly and have several limitations (high material cost, high energy requirements, generation of sludge, etc.) in large-scale practical applications. To overcome these limitations, several studies considered various biological agents as potential low-cost and eco-friendly sound biosorbents in treating As-contaminated water. It has been found that a wide range of biomass such as algae, fungi, bacteria, plant parts, fruit wastes, and agricultural wastes are low-cost, recyclable, no sludge generating and highly effective biosorbents in removing arsenic from water. Thus, biosorption has emerged as an eco-friendly and cost-effective technique in arsenic remediation. The present chapter provides a review of recent literature on As biosorption technologies. The utilization of various biosorbents including their optimum treatment conditions also is extensively summarized to get a better concept about the future scope of As remediation using biosorption method.

1 Introduction

Arsenic (As), a colorless and odorless element found in air, water, and soil in trace quantities (Matschullat 2000; Prashant et al. 2009; Bhakta et al. 2016, 2017), is one of the priority hazardous pollutants posing severe environmental and human health hazardous impacts worldwide. According to the International Agency for Research on Cancer, As and its compounds are Group 1 carcinogen to human beings (Bhakta et al. 2016; Bhakta and Munekage 2009; IARC 1987).

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Arsenic contamination of precious water resources especially groundwater has emerged as a significant public health concern mostly in the developing countries (Chakrabarti et al. 2018; Bhakta et al. 2016; Mamun et al. 2009; Ranjan et al. 2009). Both natural process (i.e., deposition of As-containing minerals) and anthropogenic activities (such as, industrial discharge of fertilizers and pesticides) are responsible for causing As contamination in surface and groundwater resources (Campos 2002; Bohrer et al. 2006). Interestingly, most of the As related environmental problems arise due to the mobilization of arsenic under natural conditions rather than the anthropogenic sources (Chakraborty et al. 2015). About 245 species of arsenic-enriched minerals have been identified. However, arsenic disulphide or realgar (As_2S_2), arsenopyrite or ferrous arsenic sulphide (FeAsS) and orpiment or arsenic trisulphide (As_2S_3) are considered as the main mineral sources of As (Hossain 2006, Jiang et al. 2013). The As carrying main ores are iron arsenate, iron sulphate and calcareous soil as calcareous arsenolite. Anthropogenic sources of As are mainly from different industrial wastes arises from fertilizer, insecticides, herbicides, coal, oil, cement, smelting, mine tailing, ore processing, metal extraction, metal purification, glass, chemicals, textiles, leather, petroleum refineries, acid mines, alloys, and catalysts industries (Ioannis and Anastasios 2002; Jackson et al. 2012).

Arsenic seldom occurs in a free state and is largely found in combination with iron, sulphur, and oxygen (Jain and Ali 2000; Kamala et al. 2005; Kumari et al. 2005). It is commonly found in −III, 0, +III and +V oxidation states in the environment, whereas in groundwater it predominantly exists in two oxidation states as arsenite (As III) or arsenate (As V). Due to more mobility and solubility in water, As(III) is 25–60 times more toxic than As(V). The most important factors which control speciation of As are redox potential and pH (Ratna et al. 2004; Chiban et al. 2012). It can be mobilized in ground and surface waters at pH values 6.5–8.5 under both oxidizing and reducing conditions (Baeyens et al. 2007). Trivalent As is found more than pentavalent As in reducing groundwater conditions (Bard et al. 1985). At neutral and slightly acidic conditions As(III) exists as non-dissociated and at pH higher than eight considerable amount of anionic species is found. In the case of As(V), it is almost completely disassociated as it is present in the form of monovalent, divalent, and trivalent anions (Ali and Aboul-Enein 2002).

Concentrations of As in natural waters and food samples are identified as a global problem and often referred to as a 21st-century calamity (Anamika 2014; Shanmugapriya et al. 2015). Worldwide more than 296 million people in over 100 countries have been affected by As contamination in groundwater; among them India, Bangladesh, China, Chile, Mexico, Taiwan, Poland, Argentina, Hungary, and the USA are the most affected (Mohan and Pittman 2007; Uluozlu et al. 2010; Mudhoo et al. 2011; Chakrabarti et al. 2018). The most devastating arsenic calamity has been reported in Bangladesh and West Bengal in India, where most of the population are dependent on groundwater as the primary drinking water source (Chowdhury et al. 2000). Around 70 million people in India, 57 million people in Bangladesh, 60 million people in Pakistan are estimated to be in danger with arsenic-contaminated drinking water (Bhakta et al. 2016; Sanjrani et al. 2017; Chakrabarti et al. 2018).

The World Health Organization has revised the provisional guideline value of As concentration in drinking water and changed the limit from 50 to 10 µg/L. However, this limit has not been adopted or implemented by all countries. For example, in India and Bangladesh due to the unavailability of the safe alternative source of water the former guideline of 50 µg/L is still in use (WHO 1993; Kumar and Puri 2012; Abhinav et al. 2017). Prolonged intake of As-contaminated drinking water can increase health risks like dermal (Dermatitis, Melanosis, Vesiculation), gastrointestinal (abdominal pain, nausea, dehydration, dysphagia),cardiovascular (hypotension, heart failure, irregular heartbeat), neurological (neuritis, hyperpyrexia, paralysis), renal (hematuria, leukocyturia, glycosuria), reproductive abnormalities and cancers of various organs (Mukherjee et al. 2003; Ghosh et al. 2007; Saqib et al. 2013; Chakrabarti et al. 2018). Arsenic has been classified as a group one carcinogenic substance by the World Health Organization. It is known to be toxic to all living organisms (Halem et al. 2009; Singh et al. 2015). Chronic exposure to excessive arsenic results in changes in skin pigments and hyperkeratosis, ulcerations of the skin, and quickens the risk of cancer of various organs including skin, kidney, liver, and bladder (Farmer and Johnson 1990; Alam et al. 2002).

On account of the above, therefore, the decontamination of As is the utmost priority to provide safe drinking water to the common people in affected areas. Scientists are continuously trying to develop practically feasible techniques to remove As from the aqueous phase. Recently, the focus has been to use easily available and low-cost biological materials or biosorbents. However, comprehensive literature useful to gain detailed knowledge about the adsorption characteristics of different biosorbents is not widely available. Therefore, the objective of the present review is to draw a brief account on the application of different biosorbents in treating As contaminated water along with the other conventional As removal techniques.

2 Conventional Arsenic Removal Techniques

Prior discussion on As biosorption, a brief concept concerning some existing conventional aqueous As removal techniques have been summarized herein. The conventional technologies applied for As removal are mainly based on the process of coagulations, ion-exchange reactions, adsorption and reverse osmosis (DeMarco et al. 2003; Urík et al. 2009). Most of these As removing techniques are also capable of removing other undesirable compounds like color, odor, turbidity, bacteria, hardness, phosphate, nitrate, fluoride, manganese, etc. (Johnston and Heijnen 2001; Singh 2007). Previously, the most common technique for As removal was coagulation with metal salts or lime softening (Duarte et al. 2009). The permissible limit for As in drinking water was reduced to 10 µg/L, although the former limit of 50 µg/L has been retained in India, Pakistan, Bangladesh on the basis of treatment performance and analytically achievability (WHO 2011). These methods were able to reduce As the level in drinking water close to the former WHO guideline of

50 µg/L. Therefore, various advanced technologies were introduced to remove As and reduce it to trace levels. Although these technologies are most effective in laboratory or pilot studies, their large scale implementation is still not feasible especially in the developing countries (Nicomel et al. 2016).

2.1 Oxidation

Oxidation process involves the conversion of soluble arsenite to arsenate (Table 9.1). Therefore, oxidation alone does not remove As; it requires some other techniques such as coagulation, adsorption, and ion exchange for completing the As decontamination process (Johnston and Heijnen 2001). Arsenite can be directly oxidized by various chemical agents like atmospheric oxygen, hypochlorite, ozone, permanganate, hydrogen peroxide, etc. Among them, oxidation of arsenite by oxygen is a very slow process to complete than the other chemicals (Ahmed 2001). However, consideration of interfering substances present in water is very important in the selection of proper oxidant as these substances greatly affect the oxidation process (Singh et al. 2015).

2.2 Coagulation and Precipitation

Coagulation and precipitation are the most common methods used in As removal (Table 9.1). The positive charge on the coagulants neutralizes the negative charge present on the colloidal particles and flocculates it. Due to the inexpensive nature and relative ease of operation, iron and aluminum-based coagulants are mostly used (Andrianisa et al. 2008; Dadwal and Mishra 2017). Dissolved As is transformed into insoluble solid by the chemicals and later on processed for precipitation. In another way, soluble As species can be co-precipitated by incorporation into a metal hydroxide (Mondal et al. 2013). The efficiency of different coagulants varies as a function of pH. In removing As, $Al_2(SO_4)_3$ and $FeCl_3$ found to be effective below the pH of 7.6. Arsenate is more efficiently removed by coagulants compared to arsenite (Cheng et al. 1994; Garelick et al. 2005). The major disadvantage of this process is the production of As-contaminated sludge in large amounts. Sludge management is required to prevent the consequence of secondary pollutant (Singh et al. 2015).

Table 9.1 Comparison of some conventional As removal techniques

Technology	Description	Agents/types	Advantages	Limitations	Reference
Oxidation	As(III) oxidized to As(V) by chemical oxidizers through air/pure oxygen, photochemical or microbiological oxidation.	O_3, Cl_2, NaClO, ClO_2, $KMnO_4$, H_2O_2	Relatively simple process, in situ As removal, oxidizes other impurities	Slow process, mainly remove As(V)	Bissen and Frimmel (2003); Vasudevan et al. (2006); Sorlini and Gialdini (2010)
Precipitation/ coagulation	Uses chemicals to transform dissolved As into an insoluble solid that is precipitated (precipitation) or to adsorb As onto another insoluble solid to precipitate. The arsenic enriched solid is then removed from the liquid phase by filtration.	$KAl(SO_4)_2{\cdot}12H_2O$, $MnSO_4$, $TiCl_4$, $Fe_2(SO_4)_3$, $Ti(SO_4)_2$, $FeCl_3$	Simple in operations, chemicals are commonly available	Produces toxic sludge, secondary treatment required	Kocar and Inskeep (2003); Nicomel et al. (2016)
Sorption/ adsorption	Concentrates solutes at the surface of an adsorbent, thereby reducing their concentration in the bulk liquid phase. As-contaminated water is passed through the column which is packed with adsorption media and contaminants are adsorbed from the water.	Synthetic and commercial activated carbons or low–cost adsorbents	Easily available; simple in operation; effective for a household treatment plant	Needs replacement after four to five regeneration	Mohan and Pittman (2007); Yao et al. (2014); Gisi et al. (2016)
Ion exchange	Ions are exchanged between similar charged compounds which are held electrostatically on the surface of solid. Water is passed through a media which is packed into a column.	Strongly acidic ($-SO_3^-$), weakly acidic ($-COO^-$), strongly basic [$-N^+(CH_3)_3$] · weakly basic [$-N(CH_3)_2$]	Medium well defined, pH-independent	High cost of operation and maintenance; sludge disposal problem; difficult to remove As(III); low resin life	Issa et al. (2010); Barakat (2011); Ramos et al. (2014)

(continued)

Table 9.1 (continued)

Technology	Description	Agents/types	Advantages	Limitations	Reference
Membrane filtration	Separates contaminants from water by passing it through semi–permeable barrier or membrane. The membrane allows some constituents to pass, while blocks others.	Microfiltration, ultrafiltration, nanofiltration, reverse osmosis	Well-defined and high removal efficiency	High–tech operation and maintenance, produces a high amount of rejected water which is toxic	Choong et al. (2007); Pendergast and Hoek (2011); Fu and Wang (2011)

2.3 Ion Exchange

Ion exchange is a commonly used water treatment process (Table 9.1). This includes a physical or chemical process where ions are held electrostatically on the surface of a solid phase and exchanged for ions of a similar charge in a solution (Table 9.1). It is a reversible interchange as there is no permanent change of structure of solid is noticed. The solids used here are typically a synthetic anion exchange resin. For removal of As, usually a chlorine loaded ion exchange resin is placed in the vessel (Chiban et al. 2012). In this process, the presence of sulphate, competition with other anions, contact times, the presence of total dissolved solids are some important factors in determining the efficiency of As treatment. Below pH 7, sulphates do not influence the As(V) sorption by ferrihydrite (Jackson and Miller 2000). Application of ion exchange in the removal of As is primarily limited to small to medium scale due to its higher treatment cost compared to other conventional treatment technologies. This process is also less attractive due to its low As selectivity in the presence of other competing anions (Tsuji 2002).

2.4 Membrane Filtration

Membrane filtration is a very useful high throughput process for treating water (Table 9.1). Membranes are composed of synthetic materials with pores which act as selective barriers. It does not allow some elements in water, such as microorganisms, particulates, natural organic materials, to pass through (Shih 2005; Uddin et al. 2007). A driving force in terms of the pressure difference between the feed and the permeate side is required for the transformation of water through the membrane (Bruggen et al. 2003). There are two types of pressure oriented membrane filtrations: low-pressure membrane processes, i.e. ultrafiltration and microfiltration, and high-pressure membrane processes, i.e. nanofiltration and reverse osmosis. Membranes having pore sizes between 0.1 and 10 μm are not sufficient to remove As (Shon et al. 2013). Therefore, coagulation and flocculation are required prior to membrane filtration. The pH of the water and the presence of other ions of water are the main factors determining the efficiency of this process. Arsenite is difficult to be removed by this process as it has neutral charge in the 4–10 pH range. In this pH range, arsenate is negatively charged and has the ability to bind with surface complexion and resulting in high As removal. Therefore, complete oxidation of arsenite to arsenate is required to achieve high As removal efficiency (Shih 2005; Nicomel et al. 2016).

2.5 Sorption

Sorption is a mass transfer process in which a substance travels from an aqueous phase to a solid phase and brings itself to the solid phase by physical or chemical interactions(Table 9.1). In physical adsorption, Van der Waals forces are observed whereas in chemical adsorption no rearrangements of the ions are found. Most of the adsorbents are required to have a large surface area, and generally, they are extremely porous substances (Petersen et al. 2005). The efficiency of this technique depends on various factors such as contact time, pH, temperature, and chemical properties of adsorbate and adsorbent. Various types of activated carbons are suitable for adsorption, but due to their high cost they are not utilized in large-scale (Chiban et al. 2012). Many low-cost adsorbents like zeolite, calcium peroxide nanoparticles, magnetite nanoparticles, chitosan beads, and other activated carbons are used as in the removal of As (Mohan and Pittman 2007; Jovanovic et al. 2011). Various studies showed that iron-based adsorption is very effective in As remediation. This is due to the high affinity between iron and inorganic arsenic (Gupta et al. 2012). Iron, by acting as a sorbent, co-precipitant or by behaving as a reductant, can remove As from water (Mondal et al. 2013). Most commonly applied adsorbents are metal oxides, polymer resins and activated carbon. Although, in recent years, an intensive investigation has been going on for unconventional adsorbent materials, such as, biological materials and process residues, which have some unique advantages compared to that of the conventional adsorbents (Islam et al. 2007).

3 Arsenic Biosorption as Emerging Detoxification Technology

3.1 Biosorbent

Biosorption is a process by which one biological medium uptakes some substances (Fomina and Gadd 2014). It is also a well-known process for removing various metals from the aqueous phase. Heavy metal sorption by biological materials from water is done through metabolically mediated and/or physico-chemical pathways of uptake (Ahalya et al. 2003). It is a process which generally utilizes inexpensive dead biomass to sequester toxic heavy metals (Kratochvil and Volesky 1998). Biosorption is an effective tool in removing metal ions from contaminated solutions at very low cost and environment-friendly manner (Sulaymon et al. 2013). Biosorption is mainly used to treat wastewater where more than one type of metal ions are present. The removal of one metal ion may be influenced by the presence of other metal ions in the treatment system.

Biosorption process utilizes natural materials such as agricultural wastes, microorganisms (bacteria, algae, fungi, etc.), fruits, and vegetables as biosorbents (Table 9.2). These biosorbents have a high potential for heavy metal(loid)s

Table 9.2 Different biosorbents used and their optimum treatment conditions described in different studies

Biosorbent	Oxidation state	pH	Temp	Contact time	Biosorbent dose	Arsenic concentration	Biosorption efficiency/capacity	Reference
Cupressus female cone	As(III)	10	30 ° C	160 min	3.04 g/L	800 mg/L	260.4 mg/g	Murugan and Subramanian (2004)
Tea fungus	As(III)	7.2	40 ° C	30 min	20 g/L	1.3 mg/L	100%	Murugesan et al. (2006)
	As(V)			90 min		0.9 mg/L	77%	
Tea fungal biomass	As(V)	6–8	40 ° C	90 min	0.1 g/L	1 mg/L	0.00398 mmol/g	Mamisahebei et al. (2007)
Banana peel	As(III)	ND	25 ° C	30 min	0.1 g/L	1 g/L	74 mmol/g	Memon et al. (2008)
	As(V)						107 mmol/g	
Rice polish	As(III)	6.84	20 ° C	150 min	20 g/L	1 mg/L	41.18 μg/g	Hasan et al. (2009)
	As(V)	4.29					49 μg/g	
Momordicacharantia	As(III)	9	28 ° C	45 min	5 g/L	0.5 mg/L	0.88 mg/g	Pandey et al. (2009)
Inonotushispidus	As(III)	6	20 ° C	30 min	ND	ND	51.9 mg/g	Sari and Tuzen (2009)
	As(V)	2					59.6 mg/g	
Staphylococusxylosus	As(III)	7	ND	30 min	1 g/L	7 g/L	54.35 mg/g	Aryal et al. (2010)
	As(V)	3		150 min	2 g/L		61.34 mg/g	
Stem of *Acacia nilotica*	AS	7.5	ND	15 min	4 g/L	200 μg/L	50.8 mg/g	Baig et al. (2010)
Xanthoriaparietina	As(III)	6	20 ° C	60 min	10 g/L	25 g/L	63.8 mg/g	Sari and Tuzen (2010)
	As(V)	2					60.3 mg/g	
Potato peels	As(III)	7	30 ° C	24 h	20 g/L	1 mg/L	0.1039 mg/g	Godboley and Dhoble (2011)
Maize leaves biomass	As(III)	8	40 ° C	4 h	1 g/L	50 mg/L	84.9%	Kamosonlian et al. (2011)

(continued)

Table 9.2 (continued)

Biosorbent	Oxidation state	pH	Temp	Contact time	Biosorbent dose	Arsenic concentration	Biosorption efficiency/capacity	Reference
Rhodococcus sp.	As(III)	7	30 ° C	30 min	1 g/L	100 mg/L	84.92%	Prasad et al. (2011)
Mougeotiagenuflexa	As(III)	6	20 ° C	60 min	4 g/L		57.48 mg/g	Sari et al. (2011)
Wheat husk, rice husk, banana peel, pea peel, peanut peel, sohanjana leaf	As(V)	7.8	25 ° C	60 min	0.1 mg/L	1 mg/L	Wheat husk 98.9%, rice husk 98.9%, banana peel 99%, pea peel 99.4%, peanut peel 99.1%, sohanjana leaf 99.3%	Akhtar and Shoaib (2012)
Java plum bark, mango bark, neem bark	As(III)	6	ND	150 min	6 g	100 μg/L	Java plum 87%, mango 78%, neem 67%	Mumtazuddin and Azad (2012)
*Zeamays*cob powder	As(III)	7.5	ND	40 min	20 g/L	25 mg/L	70%	Raj et al. (2012)
	As(V)	2.5					85%	
Bacillus sp.	As	6	40 ° C	ND	ND	6 mg/L	1.6 mg/L	Saravanan et al. (2012)
Saccharomyces cerevisiae	As(III)	5	35 ° C	240 min	5 g/L		62.91 μg/g	Wu et al. (2012)
Colpomeniasinuosa	As(III)	6	ND	30 min	ND	100 mg/L	95.6 mg/g	Abtahi et al. (2013)
	As(V)	2					59.9 mg/g	
Bacillus cereus	As(III)	7.5	30 ° C	30 min	6 g/L	ND	32.42 mg/g	Giri et al. (2013)
Mosambi peel	As(III)	4	40 ° C	240 min	30 g/L	100 mg/L	2.12 mg/g	Kamsonlian et al. (2013)
	As(V)	6			40 g/L		3.32 mg/g	
Arthrobacter sp.	As(III)	7	28 ° C	30 min	1 g/L	100 mg/L	74.91 mg/g	Prasad et al. (2013)
	As(V)	3					81.63 mg/g	
Leucaenaleucocephala	As(III)	7.5	ND	40 min	4 g/L	25 g/L	81.88%	Raj et al. (2013)
	As(V)	2.5					92.61%	
Paecilomyces sp.	As(III)	6	30 ° C	24 h	1 mg/L	1 mg/L	64.5%	Rodriguez et al. (2013)

Blue pine, walnut	As	8.5	20 ° C	20 min	Blue pine 20 g/L, walnut 40 g/L	100 µg/L	Blue pine 90%, walnut 80%	Saqib et al. (2013)
Chitosan prepared fromshrimp shells	As(V)	4	ND	120 min	25 mg/L	1 mg/L	2.458 mg/g	Jaafarzadeh et al. (2014)
Citrus paradise peel	As(V)	4	25 ° C	15 min	13.3 g/L	50 mg/L	37.77 mg/g	Khaskheli et al. (2014)
Sugarcane bagasse	As(III)	9	ND	120 min	25 g/L	65 mg/L	11.59 mg/g	Tajernia et al. (2014)
Aspergillus niger	As(III)	5	ND	100 min	20 g/L	800 µg/L	75%	Xue and Peng (2014)
Ulva fasciata	As	6	27 ° C	90 min	30 g/L	6 mg/L	84.6%	Christobel and Lipton (2015)
Sargassumwightii		6		90 min	20 g/L	10 mg/L	87.1%	
Gracilariacorticata		8		60 min	20 g/L	10 mg/L	85.4%	
Orange peel	As(V)	6.5	20 ° C	900 min	4 g/L	200 mg/L	32.7 mg/g	Abid et al. (2016)
Coconut fiber	As(III)	4	34 ° C	8 h	10 g/L	2 mg/L	95.56%	Nashine and Tembhurkar (2016)
Orange waste	As	6	ND	30 min	1 g/L	250 µg/L	90%	Irem et al. (2017)
Azadirachta indica (neem) bark	As(III)	6	40 ° C	20 min	0.3 g/L	100 µg/L	89.96%	Roy et al. (2017)
Turbinaria vulgaris	As(V)	4	30 ° C	60 min	4 g/L	100 mg/L	88.31%	Sumalatha et al. (2017a)
Modified fungal biomass	As(V)	6	30 ° C	24 h	10 g/L	1 mg/L	97.1%	Gonzalez et al. (2017)

(continued)

Table 9.2 (continued)

Biosorbent	Oxidation state	pH	Temp	Contact time	Biosorbent dose	Arsenic concentration	Biosorption efficiency/capacity	Reference
Citrus limonium	As	6	30 ° C	60 min	0.1 g/L	20 mg/ L	ND	Sumalatha et al. (2017b)
Yersinia sp. strain	As(III)	7	30 ° C	80 min	ND	6.5 mg/L	159 mg/g	Haris et al. (2017)
Orange waste	As	6	ND	30 min	1 g/L	250 μg/L	90%	Irem et al. (2017)
Pseudomonas alcaligenes	As(V)	7	ND	120 min	ND	1 mg/L	96.2 μg/g	Banerjee et al. (2018)
Water melon rind	As(III)	8.2	20 ° C	120 min	1 g/L	4 mg/L	99%	Shakoor et al. (2018)
	As(V)	4.6					98%	
Sargassum glaucescens	As(III)	5.9	ND	ND	0.47 g/L	120.34 mg/L	116.6 mg/g	Tabaraki and Heidarizadi (2018)
	As(V)						207.3 mg/g	

ND Not defined

decontamination in water, such as As removing bacteria (Bhakta et al. 2017). Metal adsorbent properties of these biosorbents are assigned to the presence of phosphate, sulfate, carboxylic, amino, amide and hydroxyl groups, which are commonly found in the cell wall (Veglio and Beolchini 1997). The efficiency of biosorption depends on the substrate and involves the processes of coordination, complexation, ion exchange, adsorption and inorganic micro precipitation (Volesky 1995). Several types of research use various kinds of biosorbents in treating As contaminated water (Table 9.2). For example—banana peel (Memon et al. 2008) and rice polish (Hasan et al. 2009) was employed to remove As(III and V). Sumalatha et al. (2017a), Irem et al. (2017) and Nashine and Tembhurkar (2016) applied *Turbinaria vulgaris*, orange waste and coconut fiber as biosorbents of As. Chitosan prepared from shrimp shells showed good As removal efficiency (92.458 mg/g) (Jaafarzadeh et al. 2014). Recently, some other biosorbents, such as different fungal biomass (Gonzalez et al. 2017), modified orange peel waste (Meng et al. 2017), lettuce flour (Vieira et al. 2018), *Cucumis pubescens* peels (Gazi et al. 2018), soyabeen (Gaur et al. 2018), and watermelon rind (Shakoor et al. 2018), have also been effectively tested.

3.2 Advantages of Biosorption

Several advantages of biosorption method, over the conventional methods, have been identified (Spinti et al. 1995; Ramachandra et al. 2005; Norton et al. 2004; Volesky 2007; Kumar and Oommen 2012; Nandal et al. 2014; Agyapong et al. 2015), and are listed below:

1. **Cost effective**: biosorbents are usually derived from abundant or waste materials which are readily available at a very low cost.
2. **Recyclable**: biosorbents can be reused after the metal is recycled.
3. **Supplementation**: no additional substances are required in biosorption process.
4. **Zero sludge generation**: no secondary problems associated as there is no sludge generated in the biosorption process compared to other techniques.
5. **Competitive performance**: biosorption is capable of high performance compared to the other technique in an efficient and economical way.
6. **Short operational time**: it is a very quick method; most of the biosorbents are capable of removing As in a short time.

3.3 Factors Affecting the Biosorption of Arsenic

Biosorption of any metal depends on some important factors, such as temperature, pH, biomass dose, contact time and initial concentration of the metal, that influence the efficiency of the biosorption process. To achieve the optimum removal condition, knowledge of these factors is very important, and thus, they are reviewed below in relation to As removal.

3.3.1 Temperature

Temperature plays an important role in the biosorption process of As. However, there are different opinions obtained from different experiments on the influence of temperature in As biosorption process. Most of the biosorbents have optimum adsorbing capacity in the temperature range of 20–40 °C (Table 9.2). According to a review by Ahalya et al. (2003), no significant change is noticed in sorption efficiency with an increase or decrease in temperature. In some cases, As biosorption is initially increased with increasing temperature, whereas at high temperature, the biosorption process may be slow down due to the breaking down of arsenic ions bond on the surface of the biosorbent (Khormaei et al. 2007). The sorption medium temperature is very important for energy-dependent mechanism in this process. Most of the experiments revealed that biosorption has been effective in nearly room temperature condition (Memon et al. 2008; Rodriguez et al. 2013; Haris et al. 2017)

3.3.2 pH

The pH is the most important parameter in the biosorption process (Table 9.2) as it affects the solution chemistry of the metals, governs the activity of the functional groups in the biomass, and introduces the competition of metallic ions (Friss and Myers-Keth 1986; Galun et al. 1987). The optimum water pH for maximum removal of As may vary in different biosorbents (Table 9.2) since diverse biosorbents contain a wide range of biocomponents/biomolecules, which may regulate pH of the water solution.

The pH dependence of metal sorption was influenced by two factors: the distribution of metal ions in the solution phase, and the overall charge of the sorbent (Srivastava et al. 2006; Kamsonlian et al. 2012). The chemical species of As and pH of solvent are important in removing As from water. Arsenic exists in both monovalent and divalent anionic species in the pH range of 2–9. The As(III) removal is favourable in the pH ranged from 6 to 9, whereas pH ranged from 2 to 6 is suitable for As(V) removal (Budinova et al. 2009). The positively charged groups of amino acids of the biosorbents can hold the negatively charged monovalent As species (Nigam et al. 2013). An intensive study, considering the various types of biosorbents and their favourable pH for maximum removal of As from water, should be conducted to identify the best biosorbents for As removal.

3.3.3 Biomass Dose

Biomass dose is another important factor in As removal process of biosorbent. The optimum dose of various biomass for maximum removal of As is different in (Table 9.2). For lower values of biomass concentrations, the specific uptake or uptake capacity per biomass is higher because more pores and surface area are exposed to available metals; with an increase in the biomass dose, the total removal of As increases (Kamsonlian et al. 2012), which is attributed to the increased percentage of surface area and the availability of more functional biosorption sites (Rajeswari and Agrawal 2014). However, after a certain dose, there will be no significant increase in As removal because overlapping biomass hinders availability of active biosorption sites. In fact, it is a common phenomenon that metal uptake capacity is inversely correlated with increasing biosorbent beyond a certain optimal dosage. Therefore, beyond the optimum dosage, a drop in removal capacity is noticed due to the saturation of pores at the surface of the biosorbent (Sumathi and Alagumuthu 2014). Depending upon the initial metal concentration, the biosorbant dose may vary from 0.1 to 40 mg/L (Mamisahebei et al. 2007; Kamsonlian et al. 2013).

3.3.4 Contact Time

The time of contact between adsorbent and metal ions has significant importance in the treatment of metals contaminated water. The contact time for optimum removal of As is varied with various biosorbents, because various biosorbents have different types of biomolecules for binding As (Table 9.2). During the initial stage, a rapid As uptake can occur. However, the uptake capacity became slow and stabilized after a specific period of time when equilibrium is achieved. This happens due to the accumulation of As species and saturation of filled active sites (Borah et al. 2009). The variation of contact time has been observed in different studies. The equilibrium can be achieved as fast as in 15 min (Baig et al. 2010; Khaskheli et al. 2014). In most of the earlier works contact time of 1 h was enough to achieve maximum adsorption. In some cases, the process takes a long time of 24 h depending on the saturation capacity of the biosorbents (Godboley and Dhoble 2011; Rodriguez et al. 2013).

3.3.5 Initial Arsenic Concentration

The initial concentration of As in solution is one of the most important factors influencing the sorption process of biosorbent. The maximum sorption capacity may vary in different biosorbents under specific initial As concentration (Table 9.2). Arsenic sorption efficiency is usually high at higher As concentrations due to the presence of an adequate number of As ions in highly concentrated solutions. (Mehta and Gaur 2001; Godboley and Dhoble 2011; Saqib et al. 2013). So far, a wide series of initial As concentration ranging from 100 μg/L to 800 mg/L has been effectively tested (Mumtazuddin and Azad 2012, Murugan and Subramanian 2004).

3.4 Biosorption Mechanism

Biosorbent process involves a vast array of biomolecules. It is a difficult task to know which compounds and which mechanisms are exactly playing a crucial role in the binding and removal process of As in treating the As-contaminated water. The As removal of biosorbent is dependent on the constituents of biosorbents used along with various environmental factors. The main three processes—physical, chemical and biological are commonly involved in biosorption of all kinds of metal(loid)s. Biosorption of As by living plants occurs in two-stage processes, rapid and slow. However, plant materials adsorb As in three steps: surface adsorption either physical or chemical, diffusion of particles and fixation (Fig. 9.1). There are two types of biosorption mechanisms, metabolism-dependent, and metabolism-independent. Transportation across the cell membrane and intracellular precipitation and accumulation are found in metabolism-dependent biosorption. In metabolism-independent biosorption, the process involves precipitation, ion exchange, physical and chemical adsorption and complexation (Srivastava and Dwivedi 2016). Biosorption mechanisms are rapidly reversible and are not dependent on cell metabolism (Hoffman et al. 2004). It is also well known that various functional groups such as; $COOH^-$, HN_4^-, PO_4^-, etc. are involved in the As binding process of biosorbents (Fig. 9.1) (Rana and Bhakta 2017).

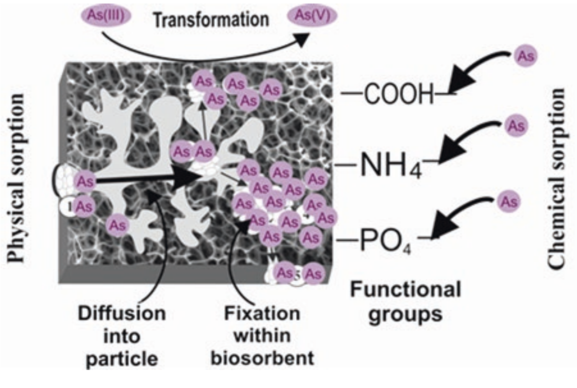

Fig. 9.1 Arsenic removal mechanisms of biosorbents

4 Conclusions and Future Prospect

The current review explored literature and critically presented the As removing biosorbents and biosorption process by comparing with conventional techniques for the treatment of As-contaminated water. Different types of biosorbents (biomass) for example—leaf, stem, root, and fruits of plants, vegetables, etc. and various aspects of biosorption techniques including advantages and limitations have been critically evaluated in the present work. It has been illustrated herein that biosorption process of As decontamination in water is significantly emerging area due to its cost-effective and eco-friendly properties. Various influencing parameters for maximum As removal process of various types of biosorbents have been discussed, which indicates the need to identify the optimum conditions of important process parameters for different biosorbents. Study concerning the As binding biomolecules present in biosorbent has not yet been performed. Therefore, it is not possible to explain the exact As removal mechanism of biosorbents. Thus, vigorous research to identify the most potential biosorbent from a vast array of natural bioresources having As affinity, optimization of its maximum As removing process parameters and elucidation of biomolecule constituents driven As binding mechanisms is inevitable in this respect to find out a novel low-cost and eco-friendly practical solution for severe As contamination problem.

However, several studies have evident that some of the biosorbents, such as biomasses of a number of plants, *Xanthoria parietina, Inonotus hispidus, Acacia nilotica, Xanthoria parietina, Zea mays, Colpomenia sinuosa, Leucaena leucocephala, Citrus paradise,* Sugarcane bagasse, orange peel, *Turbinaria vulgaris and Sargassum glaucescens,* etc. are genuinely capable of removing As in substantial quantities from contaminated water. These biosorbents are low cost easily available, therefore can be adopted by the arsenic affected/exposed communities especially in developing countries where alternative treatment technologies are durable to implement. Moreover, it can be concluded that the above-mentioned biosorbents based As sorption process could be employed as an economically and environmentally feasible eco-technology for decontamination of As in water, and hence it has significant prospects in this regard.

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Chapter 10
Assessing and Mapping Human Health Risks Due to Arsenic and Socioeconomic Correlates for Proactive Arsenic Mitigation

Sushant K. Singh and Robert W. Taylor

Abstract This study provides an environmental management approach to a global public health challenge of groundwater arsenic contamination. The studied arsenic-exposed population lives in three villages (Suarmarwa, Rampur Diara, and Bhawani Tola) within the Maner block of Patna district, in the middle-Ganga Plain in the Bihar state, India. The health risks due to the consumption of arsenic contaminated water were derived through quantifying the hazard quotient (HQ) and cancer risks followed by calculating the relative risks and odds ratio of visible arsenicosis and other diseases symptoms. A hotspots and coldspots map of the HQ was produced using Arc Geographic Information System for targeting the most vulnerable population for arsenic mitigation. In the study area, the arsenic concentrations in drinking water exceeded the limits set by the World Health Organization and the Bureau of Indian Standards. The HQ and cancer risks for children in all the three villages were high and very high, respectively. However, the hotspots of HQ were confined to Bhawani Tola. Suarmarwa experienced relatively higher risks of arsenicosis and other health challenges because of the poor socioeconomic and demographic conditions of the inhabitants. Therefore, since Suarmarwa is the most vulnerable village, it should be given priority in arsenic mitigation and health intervention programs. The HQ mapping could be an important decision-making tool for identifying the most vulnerable population for prioritizing arsenic mitigation and other health intervention activities.

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1 Introduction

Geogenic groundwater arsenic contamination, a major global public health challenge, affects nearly 300 million people in more than 100 countries (Murcott 2012; Chakraborti et al. 2016; Chakraborti et al. 2017b). The impacts of arsenic hazards on human health are aggravated by socioeconomic and demographic conditions of the population exposed (Curry et al. 2000; Singh and Vedwan 2015). Usually, underprivileged inhabitants are the foremost victims and the most vulnerable to elevated levels of arsenic in drinking water and in the food materials (Curry et al. 2000; Singh 2015a; Singh and Vedwan 2015). Among all the arsenic contaminated countries, India and Bangladesh have been reported to be the worst affected regions where over 100 million people are potentially at risk due to elevated levels of arsenic in groundwater (Singh 2015a; Chakraborti et al. 2016, 2017a; Singh and Stern 2017).

The World Health Organization (WHO) has set the drinking water standard for arsenic at 10 μg/L, five times lower than the previous standard of 50 μg/L (WHO 2011). Though this reduced level is recommended based on the socioeconomic conditions of the exposed regions, most of the developing countries, including India and Bangladesh, still follow the older standard of 50 μg/L (WHO 2011; Singh 2015a; Singh and Vedwan 2015). Considering the highly carcinogenic effects of arsenic on human beings, the United States Environmental Protection Agency (USEPA) has categorized arsenic as a group “A” human carcinogen (USEPA 1999; IRIS-USEPA 2015). Arsenicosis, a diseases caused by arsenic, may occur at various concentrations, depending on the duration of exposure, age, gender, nutrition level, and even on the genetic predisposition of the person (WHO 1981; Flanagan et al. 2012; Singh et al. 2014; Quansah et al. 2015). Poor socioeconomic conditions and malnutrition significantly contribute to developing various arsenicosis symptoms (Flanagan et al. 2012; Quansah et al. 2015). Among all the age groups, children are the most vulnerable group and are at risk to the accumulation of heavy metals as they need more energy and water per body weight than adults (Schrey et al. 2000). At higher levels, arsenic can cause various immediate symptoms of acute arsenic poisoning such as vomiting, abdominal pain and diarrhea, followed by numbness and tingling of the extremities, muscle cramping and even death in extreme cases (Flanagan et al. 2012; Quansah et al. 2015). In addition, prolonged exposure to arsenic through drinking water and food can induce skin pigmentation, skin lesions, keratosis (hard patches on the palms and soles of the feet) and skin cancer (Flanagan et al. 2012; Quansah et al. 2015). Besides skin cancer, long-term exposure to elevated levels of arsenic may also cause cancers of the bladder and lungs, developmental defects, neurotoxicity, diabetes, pulmonary disease, and cardiovascular diseases including heart attack (Flanagan et al. 2012; Quansah et al. 2015; Chakraborti et al. 2017b). Adverse pregnancy outcomes, infant mortality, deleterious impacts on children’s cognitive development, cognitive impairment in adults, and even DNA damage have also been reported in arsenic contaminated areas (Flanagan et al. 2012; Dutta et al. 2015; Quansah et al. 2015; Liu et al. 2017; Chakraborti et al. 2017b).

Deriving the Hazard Quotient (HQ) or Hazard Index (HI) of a chemical pollutant help estimating the potential for non-carcinogenic and carcinogenic health risks due to prolonged exposure to a particular contaminant, in this case, arsenic (IRIS-USEPA 2015). Various studies have reported the HQ/HI assessment in the arsenic contaminated areas as a health risk assessment tool. For example, the HQ/HI values have been derived for the arsenic-exposed communities due to water (Singh and Ghosh 2012; Singh et al. 2014), food materials (Singh and Ghosh 2012; Singh et al. 2014; Liang et al. 2016), and animal milk consumption (Kazi et al. 2016). All these studies report the HQ/HI values but lack in spatial modeling of the derived HQ/HI values. A recent study presented a composite vulnerability index and Geographic Information System (GIS)-based vulnerability maps for the arsenic contaminated areas and the communities living in those areas (Singh and Vedwan 2015). This map could be helpful for policy-makers to prioritize areas for arsenic-mitigation programs and predict the probability of success of an arsenic-mitigation policy in an area. Similarly, quantifying and mapping the HQ/HI and the cancer risks due to the consumption of arsenic contaminated water would help health professionals to prioritize the at-risk areas, select communities for epidemiological studies, and for policymakers to prioritize arsenic mitigation policies. Health surveys in arsenic contaminated areas would help identify people exposed to elevated levels of arsenic, and those who are susceptible to other diseases, contributing to their overall vulnerability (Singh and Vedwan 2015).

In line of the above research gaps and insights, this study aims to (a) derive HQ and cancer risks due to arsenic exposure and develop a spatial HQ model; (b) conduct socioeconomic, demographic and health assessments in the surveyed villages; and (c) assess relationships between socioeconomic and demographic factors with arsenic-induced symptoms and other diseases.

2 The Study Area-Bihar State of India

The study area, Bihar state of India, sits within the arsenic-contaminated region comprising the Bangladesh, Nepal, and the Indian states of West Bengal, Jharkhand, and Uttar Pradesh. The state still follows the WHO old standards, set in 1963, of 50 μg/L of arsenic for drinking water (Yamamura 2001; Singh and Vedwan 2015). However, the Bureau of Indian Standards (BIS) has set an acceptable limit of 10 μg/L of arsenic in treated drinking water (BIS 2012). Groundwater arsenic contamination in Bihar was first detected in 2002 in the Bhojpur district (Chakraborti et al. 2003), followed by Patna, Vaishali, and Bhagalpur districts in 2004 (Ghosh et al. 2009), and has now been investigated and detected in more than 50% of the districts in the state (Singh et al. 2014). The remaining districts have either not been investigated, or studies from these districts are not yet reported in the mainstream scientific literature. There exists a wide spatial distribution of arsenic contamination in groundwater in the region with arsenic levels of greater than 1000 μg/L, 20 times higher than the BIS standards, in four major districts, all within 10 km of the River

Ganga (Ghosh et al. 2009; Singh and Vedwan 2015). Approximately 12 million inhabitants have been reported as at-risk in the Bihar state (Singh et al. 2014; Singh 2015a; Singh and Vedwan 2015). However, the total exposed population and the consequences of exposure are still unknown. Previous studies have addressed health impacts due to consuming arsenic-contaminated water and food (Chakraborti et al. 2003; Singh 2011; Singh and Ghosh 2011, 2012; Singh et al. 2014). Only a few studies have addressed the demographic and socioeconomic conditions in the arsenic contaminated areas in the region (Singh and Ghosh 2012; Singh et al. 2014; Singh and Vedwan 2015). Moreover, household-level socioeconomic and demographic surveys in this region are limited (Singh 2015a). Easily interpretable presentation of results in statistical models or GIS maps for policy-makers is lacking (Singh and Vedwan 2015). Therefore, we conducted household-level socioeconomic and demographic surveys in three villages, Suarmarwa, Rampur Diara, and Bhawani Tola, in the Maner block of Patna district of Bihar, India (Fig. 10.1). Maner block is one of the highly arsenic affected blocks in Bihar (SOES 2004; Singh 2011, 2015a, b; Singh and Ghosh 2012; Chakraborti et al. 2016).

3 Materials and Methods

3.1 Water Samples Collection and Arsenic Testing

A detailed method of water samples collection and arsenic testing is described in Singh et al. (2016). In brief, groundwater samples were collected from 57 sources in Suarmarwa, 50 sources in Rampur Diara, and 50 sources in Bhawani Tola (total 157). All the samples were analyzed using arsenic field test-kits (FTK) developed by the National Chemical Laboratory (NCL), Pune, India (Singh 2015a; Singh et al. 2016). A detailed report on arsenic in the studied villages are presented elsewhere (Singh et al. 2016). In this study, the arsenic concentration data is used for deriving the HQ/HI and cancer risks.

3.2 Health Risk Assessment

3.2.1 HQ/HI and Cancer Risk Assessment Due to the Consumption of Arsenic

Arsenic-related cancer risks to children and adults in the study area and HQ values were calculated by applying the USEPA's method (USEPA 1999). Applying this method to arsenic-affected communities requires arsenic concentration data, per capita consumption of water, the average body weight of children and adults, potency factor and the reference dose for arsenic (USEPA 1999; Singh and Ghosh

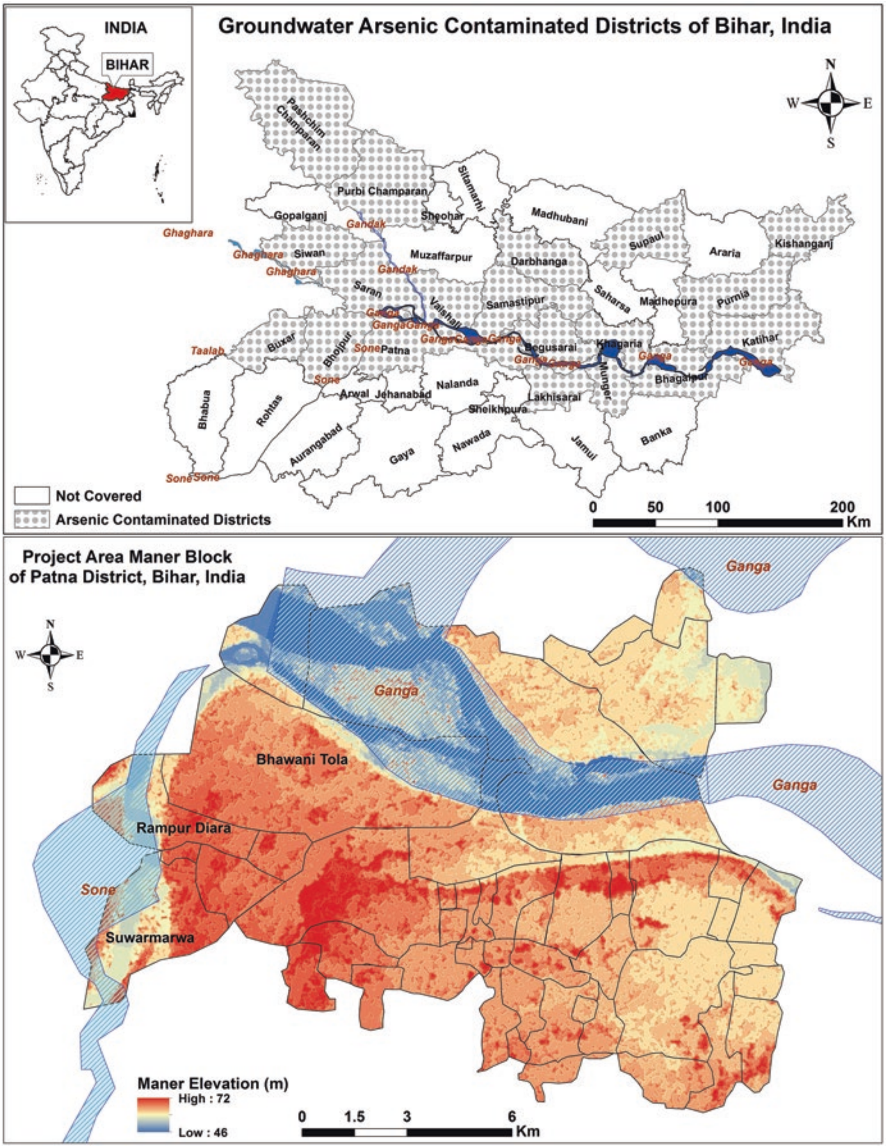

Fig. 10.1 The three study villages, Suarmarwa, Rampur Diara, and Bhawani Tola, in Maner block of Patna city, Bihar state India

2012; Singh et al. 2014). General expressions of the equations used to derive cancer risk (Eqs. 10.1–10.3) and HQ (Eq. 10.4) are:

$$\text{\textit{Average total dose}}\left(\text{mg}\right) = As_{water} \times \text{\textit{Ingestion rate}}\left(\frac{\text{L}}{\text{day}}\right) \tag{10.1}$$

where As_{water} is the arsenic concentration in drinking water in units of mg/L.

$$\text{\textit{Chronic daily intake}}\left(\left(\text{mg / kg}\right)/\text{ day}\right) = \frac{ATD\left(\text{mg}\right)}{\text{\textit{Bodyweight}}\left(\text{kg}\right)} \tag{10.2}$$

where ATD is the average total dose from Eq. (10.1).

$$\text{\textit{Cancer risk}} = \text{\textit{Chronic daily intake}} \times \text{\textit{Potency factor}} \tag{10.3}$$

The potency factor or slope factor is defined as “the result of the application of a low-dose extrapolation procedure to estimate cancer health risk due to consumption of carcinogenic substance” by the USEPA’s Integrated Risk Information Systems (USEPA-IRIS) (USEPA 1999; IRIS-USEPA 2015). The potency factor or slope factor for arsenic through oral route is 1.5 and the unit is (mg/kg)/day (USEPA 1999; IRIS-USEPA 2015).

$$\text{\textit{Hazard Quotient}}\left(HQ\right) = \text{\textit{Chronic daily intake}} \times \text{\textit{Reference dose for arsenic}} \tag{10.4}$$

The USEPA-IRIS derived oral reference dose of arsenic for health risk assessment and it is defined as “an estimate of a daily exposure to the human population, including sensitive subgroups, that is likely to be without an appreciable risk of deleterious effects during a lifetime” (USEPA 1999; IRIS-USEPA 2015). The unit measurement of the reference dose is mg/kg body weight/day (IRIS-USEPA 2015).

3.2.2 Socioeconomic, Demographic and Health Surveys

A detailed survey administration procedure is described in Singh (2015a). In brief, 340 households, 111 in Suarmarwa, 119 in Rampur Diara, and 110 in Bhawani Tola, comprising about 2500 adults were surveyed using a random stratified sampling technique (Singh 2015a). The survey elicited information on population demographics, socioeconomic factors, water and sanitation status, and health status. Information on health status was noted based on observations, photographic documentation with the individuals’ consent, and symptoms reported by the respondents. Indicators of signs and symptoms of arsenicosis include persistent itching, skin pigmentation and cracking (WHO 1981). The respondents reported symptoms of diseases other than arsenicosis and observable symptoms were recorded. However, no clinical investigations were performed to identify the underlying causes.

3.3 Statistical Analysis

Survey data was recorded using Microsoft Excel spreadsheet version 2010 and then transferred into SPSS for statistical analysis (IBM 2012). A bivariate analysis was executed to derive correlations between the variables. Relative risks (RR) and odds ratio (OR) of arsenicosis and other diseases symptoms were derived for the surveyed villages using the Risk Estimate tool in SPSS. Generic equations to calculate relative risks and odds ratio are presented below:

$$Relative\ Risks = \frac{\pi_1}{\pi_2} \tag{10.5}$$

$$Odds\ Ratio = \frac{\pi_1 / (1 - \pi_1)}{\pi_2 / (1 - \pi_2)} \tag{10.6}$$

The RR is the ratio of probabilities of events will occur, event 1 (π_1) vs. event 2 (π_2) (Warner 2012; Agresti 2013). Odds of an event, in this case odds of arsenicosis or other diseases, are the probability that an event will occur to the probability it will not occur, and OR is the ratio of two odds of different events (π_1 and π_2) (Warner 2012; Agresti 2013).

3.4 GIS Mapping and Data Analysis

Arc Geographic Information Systems (ArcGIS) Desktop version 10.4.1 was used to create maps and for hotspot analysis (ESRI 2012). Geographic coordinates for 88 of 157 water sources were collected using a RICOH Caplio 500 SE Global Positioning System (GPS) Enabled Digital Camera (Singh 2015a; Singh et al. 2016). However, because of poor satellite signals, we could not record the coordinates for 69 sources (Singh 2015a; Singh et al. 2016). So only 88 water samples were used for all GIS mapping and analysis. Other information such as arsenic concentration and elevation of the water sources was entered into the attribute table. This data was used to generate a contoured surface of arsenic concentrations within the wells. The Hot Spot Analysis Tool in ArcGIS was used to create a map of statistically significant spatial clusters of "hotspots" (high values) and "coldspots" (low values). This tool uses the Getis-Ord Gi∗ statistic and produces z-scores and *p*-values. A positive z-score represents a hotspot and a negative z-score represents a coldspot (ESRI 2012). The *p*-values are the probability of obtaining the expected results at 90%, 95%, and 99% significance levels.

4 Results and Discussion

4.1 *Groundwater Arsenic Contamination in the Surveyed Villages*

The NCL-FTK tested results for the groundwater samples revealed that 60% of the drinking water sources in Bhawani Tola had arsenic greater than the WHO limit of 10 μg/L, followed by 34% in Rampur Diara and 32% in Suarmarwa (Table 10.1) (Singh et al. 2016).

Out of 157 groundwater samples, 22 samples, randomly collected from Rampur Diara and Bhawani Tola villages, were analyzed using Atomic Absorption Spectrophotometry. The maximum recorded arsenic concentration was 300 and 250 μg/L in Rampur Diara and Bhawani Tola groundwater samples, respectively (Singh et al. 2016).

4.2 *Hazard Quotient and Cancer Risks Assessment and Mapping*

The HQ/HI estimation revealed that the average HQ values in all the three villages were high at the HQ scale established by the USEPA, with the average highest value (37) in Bhawani Tola (Table 10.2). The calculated HQ values were greater for children than adults in the three surveyed villages (Table 10.2), which is in line with the previous studies in the same geographical region (Singh and Ghosh 2012; Singh et al. 2014).

The very high cancer risk values for children and adults in the three studied villages indicate the vulnerability of the communities for developing cancer due to exposure to arsenic-contaminated water (Table 10.2). Out of 1000 inhabitants, 4 children and 1 adult in Suarmarwa, 5 children and 2 adults in Rampur Diara, and 11 children and 4 adults in Bhawani Tola could develop cancer with continued consumption of arsenic-laced drinking water (Table 10.2) (Singh and Ghosh 2012; Singh et al. 2014). The higher HQ values suggest that the residents of the surveyed

Table 10.1 Groundwater arsenic contamination in three villages in the Bihar state of India

	Villages (Water sources)		
Arsenic	Suarmarwa (57) (%)	Rampur Diara (50) (%)	Bhawani Tola (50) (%)
BDL	68	62	36
10 μg/L	0	4	4
11–50 μg/L	23	22	20
≥51 μg/L	9	12	40

BDL Below the detection limit of NCL-FTK

Table 10.2 Chronic risk and cancer risk due to arsenic-contaminated drinking water in three villages in the Bihar state of India

Name of village	HQ			Chronic risk level (HQ): US EPA, 1999[a]	Average Cancer risk/1000 inhabitants	Cancer risk: US EPA, 1999[b]
	Min	Max	Avg.			
Suarmarwa (children)	0	90	13	High	4	Very high
Rampur Diara (children)	0	70	17	High	5	Very high
Bhawani Tola (children)	0	90	37	High	11	Very high
Suarmarwa (adults)	0	32	5	High	1	Very high
Rampur Diara (adults)	0	25	6	High	2	Very high
Bhawani Tola (adults)	0	32	13	High	4	Very high

[a] *HQ/HI: <0.1 = Negligible; $\geq 0.1 < 1$ = Low; $\geq 1 < 4$ = Medium; and ≥ 4 = High*

[b] *Cancer risk: <1 person/1000,000 inhabitants = Very low; >1 person/1000,000 and <1 person/100,000 inhabitants = Low; >1 person/100,000 inhabitants and <1 person/10,000 inhabitants = Medium; >1 person/10,000 and <1 person/1000 inhabitants = High; and >1 person/1000 inhabitants = Very high*

villages might also experience more significant adverse but non-carcinogenic health issues (Table 10.2) (Singh and Ghosh 2012; Singh et al. 2014).

The hotspot analysis revealed that hotspots of HQ for adults were confined to Bhawani Tola (Fig. 10.2). There were no coldspots in Bhawani Tola. However, the coldspots in Rampur Diara and Suarmarwa were widely distributed (Fig. 10.2).

Sixty-five percent (65%) of the hotspots were statistically significant (GiP between <0.001 and 0.0665) and 46% coldspots were statistically significant in all cases (Fig. 10.2). The hotspots analysis produced approximately 15% places with GiZ score ranging between 0.0897 and 0.8103, which were statistically not significant and more prevalent in Suarmarwa and Rampur Diara than Bhawani Tola (Fig. 10.2). The hotspots HQ map suggests that the adults in Bhawani Tola are susceptible to arsenic-induced cancers, and requires further investigation prior to implementing arsenic-mitigation interventions (Fig. 10.2).

4.3 *Socioeconomic and Demographic Characteristics of the Communities*

Majority of the respondents in all the three surveyed villages were married male, older than 40 years (Fig. 10.3a–c). Respondents' age ($p = 0.049$) and gender ($p < 0.001$) across the three surveyed villages were significantly different. The participation of women was the minimum in Rampur Diara village. Whereas the

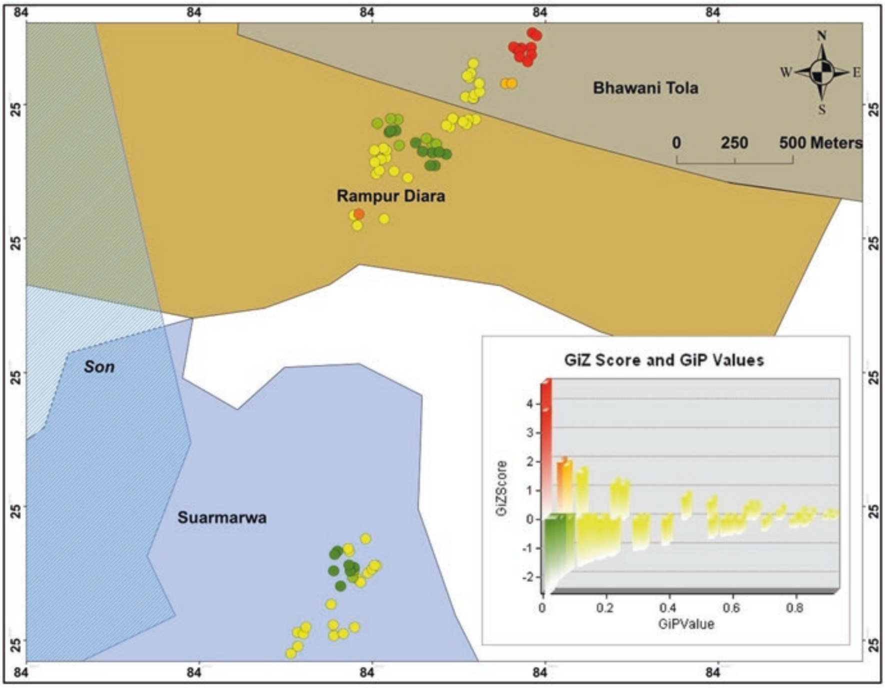

Fig. 10.2 Hotspots of HQ with GiZ and GiP values for Adults in three surveyed villages (Suarmarwa, Rampur Diara, and Bhawani Tola) in the Bihar state of India. (*GiZ represents the standard deviation, i.e. z-score of the spatially clustered hotspots, or coldspots and p values represent significance levels*)

participation of married individuals was highest in Bhawani Tola and significantly different (p = 0.015) from the other two villages. The highest number of single respondents was in Rampur Diara. Respondents' caste[1] was also significantly different (<0.001) in the three surveyed villages. In Suarmarwa, backward caste (BC) (77%) was prevalent over the scheduled caste (SC) (33%) (Fig. 10.3d). However, in Rampur Diara and Bhawani Tola villages, the population was dominated by the forward caste (FC) over the backward caste and scheduled caste populations (Fig. 10.3d). The education level of respondents in the three villages was categorized into four as illiterate, primary education, secondary education or college education. Fifty percent of the respondents from the Suarmarwa village were illiterate (people who cannot read or write), 41% of the respondents had primary education, and 9% of the respondents had secondary-level or college education (Fig. 10.3e). The respondents of Rampur Diara and Bhawani Tola villages had similar patterns in education level, with most respondents having a primary education, followed by a

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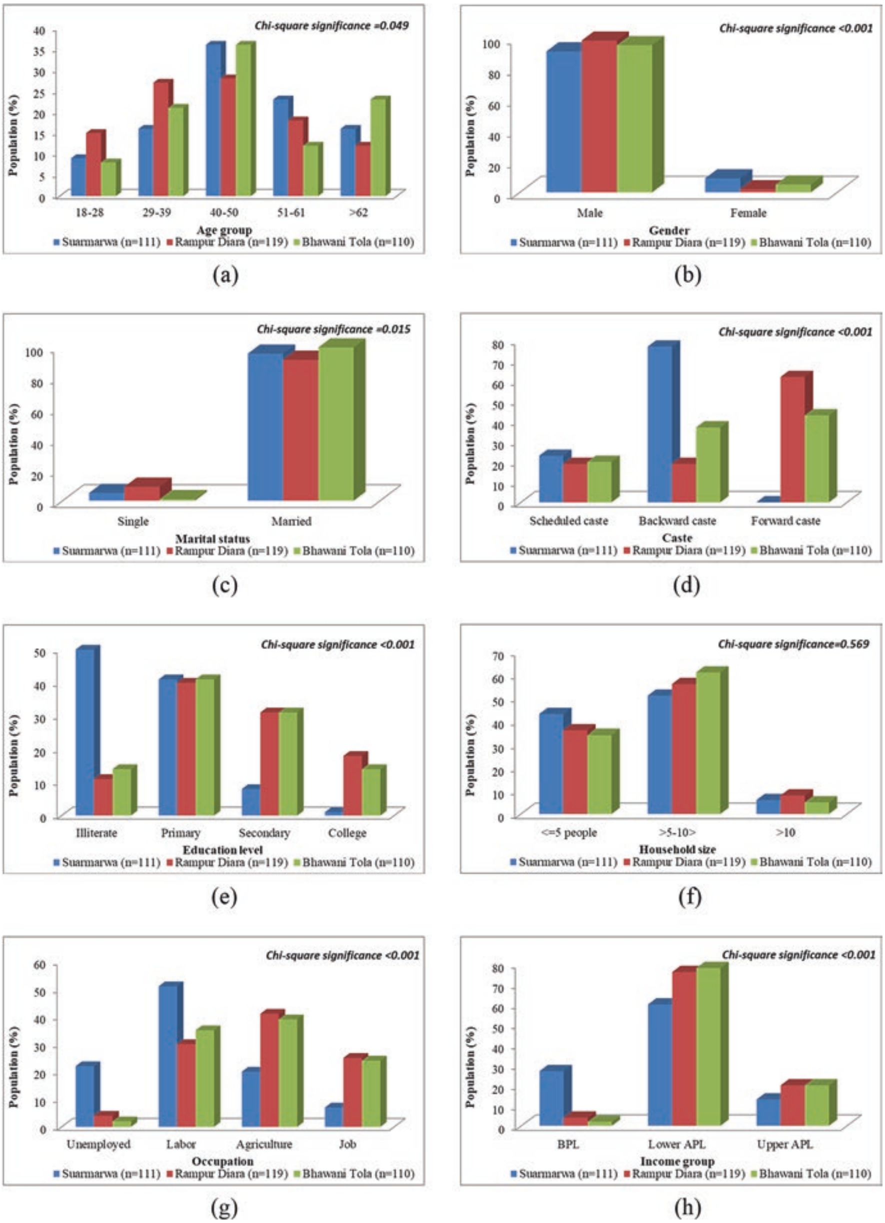
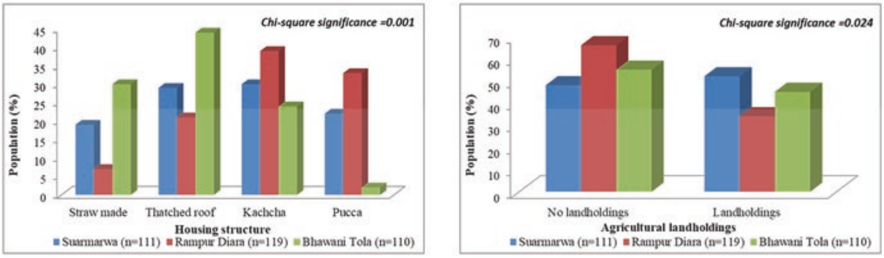
[1] The caste system is a rigid social hierarchy in Hinduism, introduced by Manu more than 1000 years BC, where the forward castes (upper caste) are at the top, followed by the backward castes, and scheduled castes and tribes at the bottom of the social stratification.



(a) (b) (c) (d) (e) (f) (g) (h)

Fig. 10.3 Socioeconomic and demographic characteristics of the communities surveyed in three villages (Suarmarwa, Rampur Diara, and Bhawani Tola) in the Bihar state of India (**a–j**). (**a**) Age group of the respondents; (**b**) Gender of the respondents; (**c**) Marital status of the respondents; (**d**) Caste of the respondents; (**e**) Education level of the respondents; (**f**) Household size; (**g**) Occupation of the respondents; (**h**) Income group of the respondents; (**i**) Housing structure of the respondents; (**j**) Agricultural landholdings of the respondents



(i) (j)

Fig. 10.3 (continued)

secondary and a college education. Only 11% and 15% population in Rampur Diara and Bhawani Tola, respectively, were illiterate (Fig. 10.3e). The literacy was also significantly different ($p < 0.001$) across the three surveyed villages.

The average family size in the study area was six people per household (Fig. 10.3f), which is higher than the average household size of the state (5.5 per household) and the country (4.8 per household) (Census-India 2011). The occupation of the respondents across the three surveyed villages was significantly different ($p < 0.001$). The majority of the population of Suarmarwa village was unemployed, or they worked as daily wage laborers (Fig. 10.3g). In Rampur Diara and Bhawani Tola village, the majority of the respondents were involved in agricultural activities, running a business for their livelihood, or employed in some other job (Fig. 10.3g). Suarmarwa had the highest population (27%) below the poverty line (BPL) (Fig. 10.3h). In all the three villages the majority of the population was just above the poverty line (APL), with income ranging in rupees (Rs.) between Rs.500 (~$9) and Rs.10,000 (~$170) per month (Fig. 10.3h). The gross monthly income across the three surveyed villages was significantly different ($p < 0.001$).

Bhawani Tola had the highest population (74%) living in poorer housing structures (thatched roof and straw made roof houses), followed by Suarmarwa village (48%) (Fig. 10.3i). Suarmarwa, which is dominated by the lower caste group, has a 52% population with agricultural landholdings. Rampur Diara and Bhawani Tola had 34% and 45%, respectively, of their populations with agricultural landholdings (Fig. 10.3j). Housing structures and the agricultural landholdings were also significantly different ($p = 0.001$ and 0.024) across the three surveyed villages. This further indicates that having or not having agricultural land could be a better indicator of vulnerable communities in those areas where the majority of the population belongs to backward and scheduled castes.

4.4 Health Surveys

4.4.1 Arsenicosis

The occurrence of arsenicosis symptoms in the three surveyed villages was significantly different (p = 0.039) with the highest (10.8%) in Suarmarwa, followed by Rampur Diara (5.2%) and the lowest in Bhawani Tola (2.7%). The RR of arsenicosis was 2 times more likely to occur in Suarmarwa than Rampur Diara (p = 0.095), and 4 times in Suarmarwa than in Bhawani Tola (p = 0.015) (Table 10.3). The likelihood of occurrence of arsenicosis in Rampur Diara was 1.9 times greater than in Bhawani Tola (p = 0.272) (Table 10.3).

Although we found higher arsenic levels in Bhawani Tola (Table 10.1), the lower occurrence of arsenicosis symptoms in this village could be because of healthier food consumption by the higher income residents of this village. The other possibility could be that the residents use deep boring tube wells for drinking purposes, which are arsenic-free in most cases. Moreover, comparatively higher income in Bhawani Tola supports this hypothesis that with higher income people tend to eat better and nutritious foods and can afford installing expensive deep boring tube wells (Fig. 10.4h).

Based on the cross-tabs analysis, it appeared that except for agricultural landholdings, the cases of arsenicosis were not different when compared across the other socioeconomic factors (Fig. 10.4a–o). Males reported to have more arsenicosis symptoms than females (Fig. 10.4a) as testified in other studies (Ahmad et al. 1999; Hadi and Parveen 2004). We hypothesize that males are more at risk of arsenicosis than females because they tend to spend more time outside their house and drink water from arsenic contaminated sources in the villages. Individuals of age group 29–39 years had more arsenicosis symptoms followed by the individuals of the age group of 18–28 years and older than 62 years, which is in line with a similar finding in Bangladesh (Hadi and Parveen 2004).

Married people showed more arsenicosis symptoms than single individuals (Fig. 10.4c). Scheduled caste individuals were found to have more arsenicosis symptoms than BC and FC respondents (Fig. 10.4d). Illiterate and the individuals with only primary education were found to have arsenicosis symptoms than the respondents who had a secondary level or a college education (Fig. 10.4e). Similar findings were reported in another study (Hadi and Parveen 2004). Households with

Table 10.3 Relative risks (RR) of arsenicosis and Odds ratio (OR) in three surveyed villages (Suarmarwa, Rampur Diara, and Bhawani Tola) in the Bihar state of India

Relative risks (RR) and odds ratio (OR)	Suarmarwa vs. Rampur Diara	Suarmarwa vs. Bhawani Tola	Rampur Diara vs. Bhawani Tola
RR of arsenicosis (95% CI)	2.072 (0.806–5.329)	3.964 (1.150–13.662)[a]	1.913 (0.490–7.461)
OR of arsenicosis (95% CI)	0.454 (0.164–1.256)	0.231 (0.063–0.844)[a]	0.509 (0.124–2.089)

[a]*Significant at $\alpha = 0.05$*

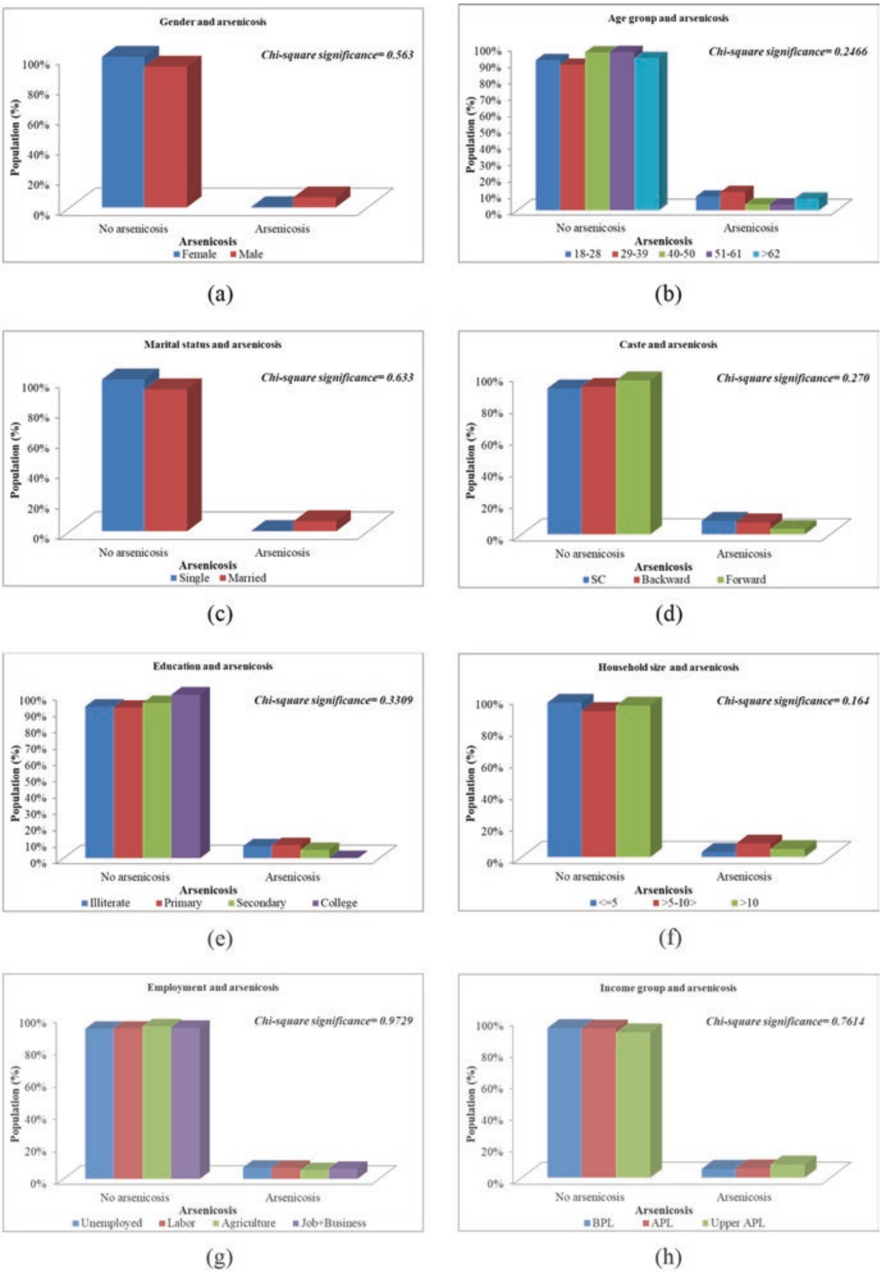
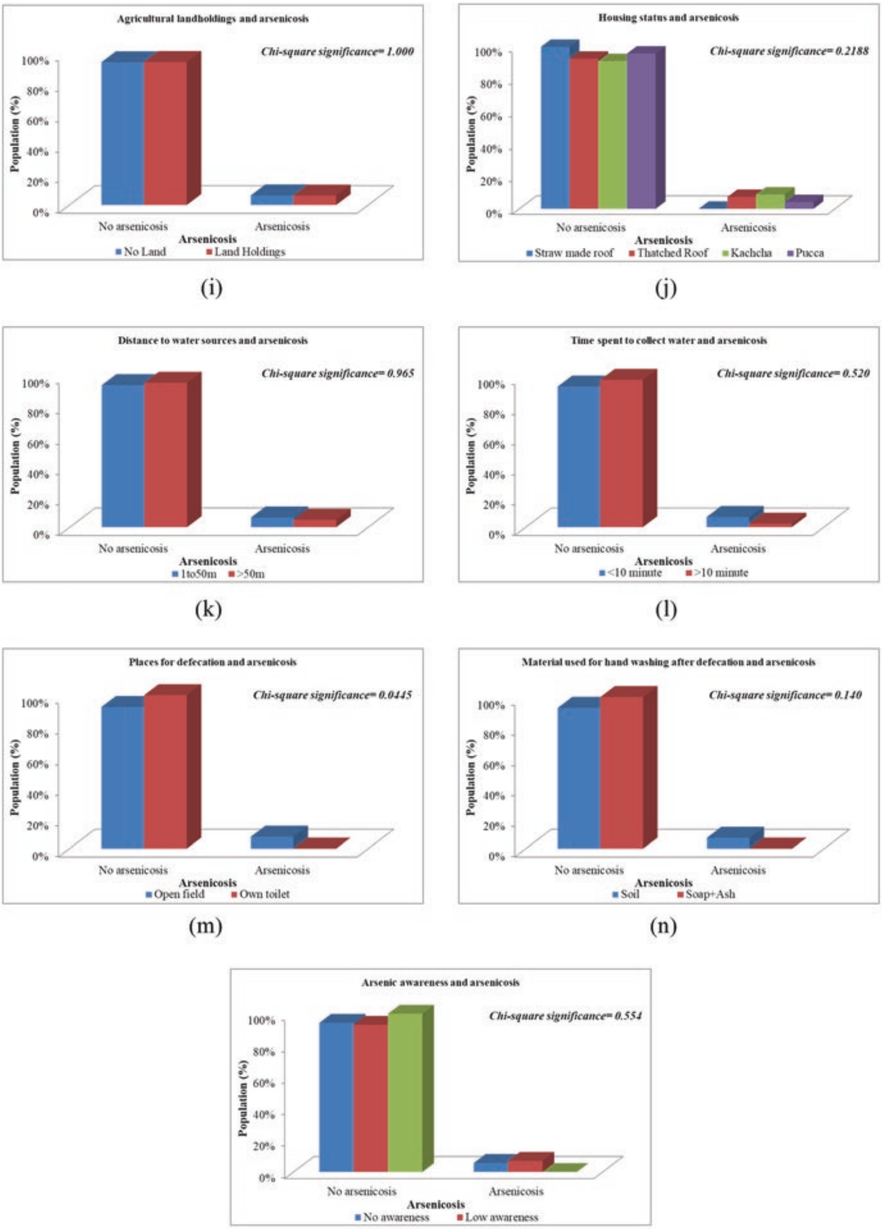


Fig. 10.4 Socioeconomic and demographic characteristics, lifestyle and risk of arsenicosis of communities in three villages (Suarmarwa, Rampur Diara, and Bhawani Tola) in the Bihar state of India (**a–o**). (**a**) Gender and arsenicosis; (**b**) Age group and arsenicosis; (**c**) Marital status and arsenicosis; (**d**) Caste and arsenicosis; (**e**) Education and arsenicosis; (**f**) Household size and arsenicosis; (**g**) Employment and arsenicosis; (**h**) Income group and arsenicosis; (**i**) Agricultural landholdings and arsenicosis; (**j**) Housing status and arsenicosis; (**k**) Distance to water source and arsenicosis; (**l**) Time spent to collect water and arsenicosis; (**m**) Places for defecation and arsenicosis; (**n**) Materials used for hand washing after defecation and arsenicosis; (**o**) Arsenic awareness and arsenicosis



(i) (j)

(k) (l)

(m) (n)

(o)

Fig. 10.4 (continued)

an average family size of 5–10 people showed more arsenicosis symptoms than households with less than five members in a family (Fig. 10.4f). Arsenicosis symptoms were almost similar among the entire employment category (Fig. 10.4g). Collectively, BPL and lower APL had the highest arsenicosis symptoms, however not significantly different across the income group (Fig. 10.4h). Other study has also reported that arsenicosis is more prevalent among the low-income group (Hadi and Parveen 2004). Arsenicosis symptoms were more prevalent among the individuals living in the thatched roof or Kachcha house[2] (Fig. 10.4j). People who traveled less and spent less time to collect water had more arsenicosis symptoms (Fig. 10.4k, l). A bivariate analysis was performed to see whether socioeconomic-demographic variables (age, income, caste, housing status) and sanitation habits (place for defecation and materials used to wash hands after defecation) are correlated with the occurrence of arsenicosis symptoms in the villages. Only sanitation habits (place for defecation) was inversely correlated with arsenicosis symptoms ($R^2 = -0.125$). It further explains that the inhabitants who use a toilet for defecation were less likely to have arsenicosis symptoms than the inhabitants who defecate in open fields. It is vital mentioning here that most of the arsenic affected communities in India and in Bangladesh live in a very unhygienic environment and have the least access to clean water, sanitation and hygiene facilities. Likewise, the individuals who use soil to wash their hands after the defecation had arsenicosis symptoms than who use soap or ashes (Fig. 10.4m, n). Incidence of arsenicosis was higher among respondents with no awareness or low awareness than respondents with arsenic awareness (Fig. 10.4o). A detailed report on arsenic awareness in the study can be found elsewhere (Singh et al. 2018).

While evaluating the expenses on the arsenicosis treatment, we found that only nine people in Suarmarwa, four in Rampur Diara, and three in Bhawani Tola spend up to Rs.1000 per month. Three people from Suarmarwa reported that they spend over Rs.1000 on arsenicosis treatment. The expenses on arsenicosis treatment across the three villages were significantly different from each other ($p = 0.033$).

Although a direct link between arsenicosis signs and symptoms and the consumption of arsenic contaminated water can only be established by epidemiological studies, we discovered a higher incidence of skin discoloration (both black and white patches) (Figs. 10.5, 10.6, 10.7), cracked skin (Fig. 10.8), fragile and discolored nails (Fig. 10.9), and persistent itching among inhabitants who were drinking water contaminated with 40–70 μg/L of arsenic.

4.4.2 Health Issues Reported by Respondents in the Study Area

Suarmarwa scored the highest population (70%) with symptoms of various diseases, followed by Rampur Diara (24%) and Bhawani Tola (13%) (Table 10.4). The Chi-Square test indicates that the occurrence of other symptoms among the three surveyed villages was different ($p < 0.001$). A list of other symptoms and diseases

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[2] Kachcha houses are the houses with temporary roofs made with cemented floor and/or wall.

Fig. 10.5 Skin discoloration (white patches) on hands (both sides) and feet of an elderly woman in Rampur Diara village, Bihar state of India. (Photograph by Sushant Singh, 2013)

Fig. 10.6 Persistent white patches on the ankle of a 10-year-old girl in Rampur Diara village, Bihar state of India. (Photograph by Sushant Singh, 2013)

Fig. 10.7 Persistent white patches on the back of a 15-year-old boy in Rampur Diara village, Bihar state of India. (Photograph by Sushant Singh, 2013)

Fig. 10.8 Persistent white lines at the bottom of the middle and ring fingers of a 14-year-old girl in Rampur Diara village, Bihar state of India. (Photograph by Sushant Singh, 2013)

Fig. 10.9 Fragile fingernails with black spots on a 25-year-old woman in Rampur Diara village, Bihar state of India. (Photograph by Sushant Singh, 2013)

is presented in Table 10.4. These are self-reported diseases symptoms, which we verified by observing the victims. However, we performed no clinical tests.

The RR of other disease symptoms was 3 times and 5.5 times more likely to occur in Suarmarwa than in Rampur Diara ($p < 0.001$) and Bhawani Tola ($p < 0.001$), respectively (Table 10.5). And the likelihood of occurrence of other disease symptoms in Rampur Diara was 1.8 times greater than in Bhawani Tola ($p = 0.027$) (Table 10.5).

In contrast to arsenicosis related symptoms, the chi-square test revealed that the symptoms for other diseases were higher in male than in female respondents (Fig. 10.10a). Individuals of age above 51 years had more non-arsenicosis symptoms than the younger respondents (Fig. 10.10b). Unmarried individuals had more non-arsenicosis symptoms than married individuals (Fig. 10.10c). Among the three caste group, a higher number of individuals in the SC and BC than in the FC had health issues; in addition, the number of individuals with health issues between the three castes was significantly different ($p < 0.000$) (Fig. 10.10d). Like arsenicosis, the

Table 10.4 A village wise list of health issues reported by respondents in three villages (Suarmarwa, Rampur Diara, and Bhawani Tola) in the Bihar state of India

Sl. No.	Diseases	Suarmarwa	Rampur Diara	Bhawani Tola
1	Stomach problem	11	2	2
2	Wound	3	0	0
3	Uterus problem	1	0	0
4	Body itching	9	4	3
5	Asthma	5	1	1
6	Cold	15	1	0
7	Fever	21	4	1
8	Pregnancy related problems	4	0	0
9	Sciatica	4	1	0
10	Skin disease	5	2	0
11	Hemorrhoids (Piles)	4	0	1
12	Pain in hands	1	1	0
13	Headache	3	2	1
14	Ear problem	1	0	0
15	Pneumonia	3	2	0
16	Intestine problem	1	0	0
17	Chest pain	4	1	0
18	Filaria	2	0	0
19	Dental problems	3	0	0
20	Jaundice	3	1	0
21	Diarrhea	2	0	0
22	Hydrocele	1	0	0
23	Sterilization	1	0	0
24	Appendicitis	2	0	0
25	Typhoid	2	1	0
26	Handicap	4	1	1
27	Atopic dermatitis (Eczema)	3	0	1
28	Body aches	1	1	0
29	Cracked heels	1	0	0
30	Eye problems	1	0	0
31	Tuberculosis	2	2	0
32	Cancer	1	0	0
33	Dysentery	1	0	0
34	Pain in legs	3	0	3
35	Tumors on body	1	1	0
36	Leg fracture	0	0	0
37	Gastric problem	1	0	0
38	Hernia	1	0	0
39	Hemophilia	0	1	0
40	Throat problem	0	1	0
41	Back pain	0	3	1

(continued)

Table 10.4 (continued)

Sl. No.	Diseases	Suarmarwa	Rampur Diara	Bhawani Tola
42	Diabetes	0	2	3
43	Sickness	0	1	0
44	Thyroid	0	1	0
45	White patches on the body	0	2	0
46	Paralysis	0	3	0
47	Blood impurity	0	0	1

Table 10.5 Relative other diseases risks and Odds ratio in three villages (Suarmarwa, Rampur Diara, and Bhawani Tola) in the Bihar state of India

Relative risks (RR) and odds ratio (OR)	Suarmarwa vs. Rampur Diara	Suarmarwa vs. Bhawani Tola	Rampur Diara vs. Bhawani Tola
RR of other diseases (95% CI)	2.993 (2.106–4.253)[a]	5.521 (3.335–9.140)[a]	1.845 (1.022–3.3329)[b]
OR of other diseases (95% CI)	0.130 (0.072–0.235)[a]	0.062 (0.031–0.123)[a]	0.475 (0.234–0.964)[b]

[a] *Significant at* $\alpha = 0.01$
[b] *Significant at* $\alpha = 0.05$

symptoms of non-arsenicosis diseases were more prevalent among illiterate, or people with minimal education and they were significantly different from each other ($p < 0.000$) (Fig. 10.10e). The incidences of symptoms of non-arsenicosis diseases across the household size were similar to the incidence of arsenicosis related symptoms (Fig. 10.10f). It was evident that more people with no employment or who were manual workers including farming reported more health issues than people employed in jobs that require higher education, and the difference was statistically significant ($p = 0.0041$) (Fig. 10.10g). The BPL and the LAPL individuals had more non-arsenicosis symptoms than the UAPL, but the difference between the income groups was not statistically significant (Fig. 10.10h). Although the frequency of symptoms of diseases unrelated to arsenic toxicity was higher among individuals with landholdings, it was not significantly different from individuals with no landholdings ($p = 0.182$) (Fig. 10.10i). The non-arsenicosis symptoms across the housing structures were significantly different ($p = 0.0074$) with the highest symptoms in straw made roofed houses followed by a thatched roof, Kachcha, and pucca houses[3] (Fig. 10.10j). The individuals who traveled more than 50 m and spent greater than 10 minutes to collect water suffer from non-arsenicosis health issues more than those who have easy access to water sources and the difference was statistically significant ($p < 0.000$) (Fig. 10.10k, l). The individuals with poor sanitation facilities or habits suffer from diseases unrelated to arsenicosis more than those who use the toilet for defecation and wash their hands with soap or ash. The symptoms of non-arsenicosis diseases among individuals' sanitation behaviors were significantly different ($p < 0.00$ and $p = 0.002$) (Fig. 10.10m, n). People who were less aware of arsenic

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[3] Pucca houses are the houses with cemented flooring, walls, and roof.

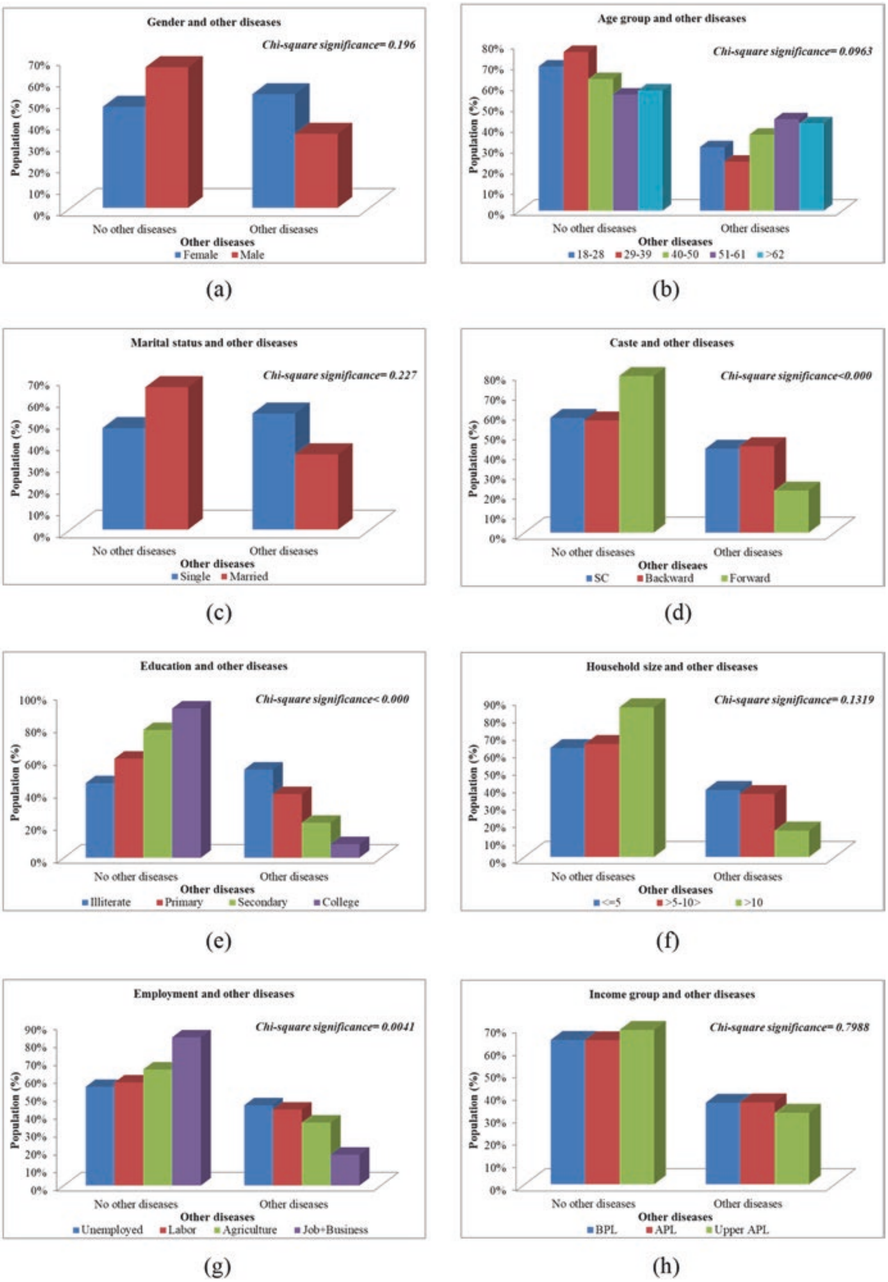
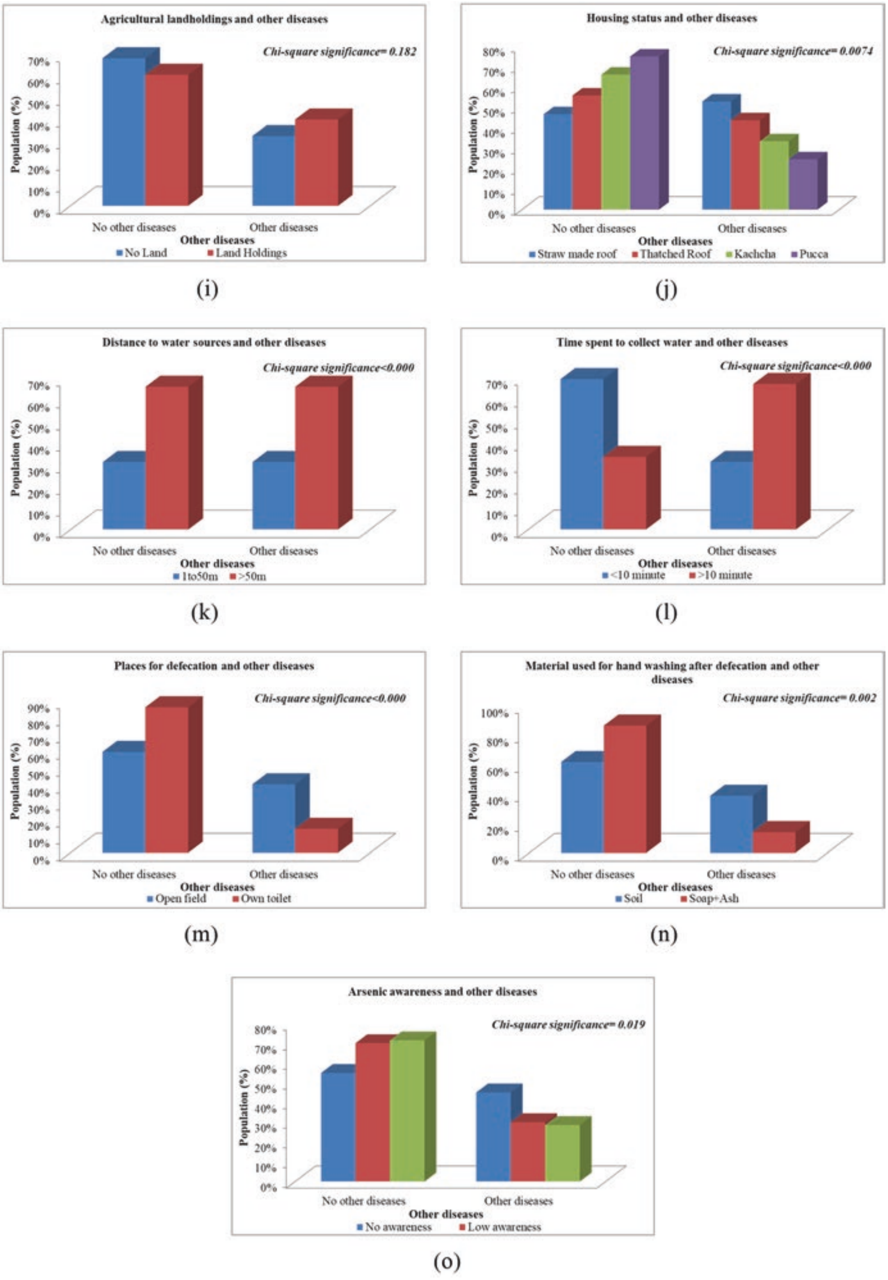


Fig. 10.10 Socioeconomic and demographic characteristics and non-arsenic related diseases of communities in three villages (Suarmarwa, Rampur Diara, and Bhawani Tola) in the Bihar state of India (**a–o**). (**a**) Gender and other diseases; (**b**) Age group and other diseases; (**c**) Marital status and other diseases; (**d**) Age group and other diseases; (**e**) Education and other diseases; (**f**) Household Fig. 10.10 (continued) size and other diseases; (**g**) Employment and other diseases; (**h**) Income group and other diseases; (**i**) Agricultural landholdings and other diseases; (**j**) Housing status and other diseases; (**k**) Distance to water source and other diseases; (**l**) Arsenic awareness and other diseases; (**m**) Places for defecation and other diseases; (**n**) Materials used for hand washing after defecation other diseases; (**o**) Arsenic awareness and other diseases



(i)

(j)

(k)

(l)

(m)

(n)

(o)

Fig. 10.10 (continued)

found to be more affected by non-arsenic related diseases, and the difference was statistically significant across the awareness levels ($p = 0.019$) (Fig. 10.10o).

We also observed that up to seven households in Rampur Diara village relied on dirty water from an unhygienic open dug well infested with insects. The households filter the water through cotton cloths prior to using for drinking and cooking purposes. The households using this dirty water are exposed to water-borne diseases that cannot be avoided by filtering through cotton cloths. For example, we found a 10-month-old child with persistent rashes and black patches on the backside of his leg, which the parents report started at 3 months (Fig. 10.11).

We suspect that there are other similar hidden pockets of households living under extremely unhygienic conditions with no alternative options for potable water sources. Being marginalized communities, they consider themselves an 'isolated community' with a world-view of an 'isolate.' The communities live in an uncertain world dominated by environmental issues and social instability and accept whatever they experience as their destiny or their bad luck (Douglas and Wildavsky 1983).

The incidence of non-arsenicosis symptoms in the communities are inversely correlated with Caste ($R^2 = -0.190$, $p < 0.01$), occupation ($R^2 = -0.183$, $p < 0.01$), housing status ($R^2 = -0.187$, $p < 0.01$), place for defecation ($R^2 = -0.217$, $p < 0.01$), and materials used to wash hands after defecation ($R^2 = -0.172$, $p < 0.01$), and positively correlated with age group ($R^2 = 0.134$, $p < 0.05$). That the results indicate older inhabitants have a greater likelihood of experiencing non-arsenic related diseases.

While evaluating the medical expenses for non-arsenic related diseases, we found that nearly 36% of the surveyed population spend money, and 27% of them spend over Rs.5000 (~$90). Among the three villages, Suarmarwa has the highest population (78 of 336 individual/households) spending on treatments for non-arsenic related diseases and the spending was significantly different ($p < 0.000$) across the surveyed villages. Expenses for the treatment of arsenicosis and other diseases were significantly different among the respondents across the three villages; they were higher in Suarmarwa than in Rampur Diara and Bhawani Tola villages.

Fig. 10.11 Persistent black rashes and patches on the backside of the leg of a 10-month-old child in Rampur Diara village, Bihar state of India. (Photograph by Sushant Singh, 2013)

5 Conclusion

The three surveyed villages, Suarmarwa, Rampur Diara and Bhawani Tola, in the Bihar state of India witness moderate to high levels of arsenic contamination in their drinking water sources that put the exposed communities at high cancer risk. The arsenic-exposed communities could develop cancer and non-carcinogenic symptoms if they continue consuming the arsenic contaminated water. Symptoms known to be associated with arsenicosis were documented in the three surveyed villages, and it was higher in the Suarmarwa village because of the poor socioeconomic conditions of the respondents in this village. It was evident that not having agricultural landholdings could be a better indicator of vulnerable communities in those villages where the majority of the population belongs to backward and scheduled castes. The HQ hotspots were confined in the Bhawani Tola village and needed immediate investigations of the health status of the communities in this village for arsenic mitigation interventions. Various health symptoms that might be linked to arsenic exposure such as skin and eye problems, pregnancy-related including sterilization, cancer, and diabetes were also identified in the surveyed villages and further clinical investigation is needed to confirm if they are due to arsenic toxicity. Poor sanitation and hygiene practices may increase the incidence of arsenicosis. Therefore, proper sanitation and hygiene practices should be encouraged in the arsenic-affected areas. Elderly people and lower caste groups are also more susceptible to non-arsenicosis health problems. Arsenicosis symptoms and symptoms of other diseases were more prevalent in Suarmarwa than in Rampur Diara and Bhawani Tola. The poor socioeconomic and demographic conditions of the communities in these villages make them vulnerable to arsenicosis and other diseases.

Arsenic-induced health issues are multidimensional challenges where the socioeconomic and demographic conditions play a vital role. Therefore, an in-depth epidemiological investigation is an urgent necessity, and health interventions should be designed by incorporating the communities' socioeconomic and demographic characteristics. Performing a cost-effectiveness analysis and developing socioeconomic models of arsenic mitigation would help in identifying the sustainable arsenic mitigation technologies in the target areas (Singh 2017a, b; Singh et al. 2017). HQ mapping can be useful for pre-planning such interventions and for targeting the most vulnerable communities. Arsenic-free water and nutritious food should be provided at a subsidized rate for the poor communities. Moreover, spreading arsenic awareness (Singh et al. 2018) would be required and information technology tools (Singh 2017b) can be used to cover a larger population in less time. This way, a healthy community in arsenic-risk society can be established.

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Chapter 11
Arsenic-Contaminated Drinking Water and the Associated Health Effects in the Shahpur Block of Bihar: A Case Study From Five Villages

Barun Kumar Thakur and Vijaya Gupta

Abstract The current paper estimates the arsenic-contaminated drinking water and its associated effects on human health. The result is based on a primary survey of 173 households from five arsenic contaminated villages from Shahpur block in the Bhojpur district from Bihar state, India. A field test kit was used to test the concentration levels of arsenic in households' drinking water. The results of the water tests revealed that more than 60% of the drinking water has excess arsenic concentration levels (more than 0.01 mg/L of WHO standards). The incidence rate due to arsenicosis (arsenic poisoning) was more among females than males. From the survey, it was also found that skin-lesions and other health issues due to arsenic induced-problems were more acute among children and women compared to men. More than half of the respondents (51%) did not hear about arsenic poisoning, and around 13.29% of the surveyed respondents know the serious health issues due to arsenic-contaminated drinking water.

This chapter is based on the first author Ph.D. work carried out at National Institute of Industrial Engineering (NITIE), Mumbai. Presently the first author is working as an Assistant Professor (Economics) at FLAME University Pune. Second author is Ph.D. supervisor of the first author and currently is a professor of Economics and Strategy at National Institute of Industrial Engineering (NITIE), Mumbai.

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1 Introduction

Drinking contaminated water is one of the most direct routes of exposure to pollutants in water. Naturally occurring metals such as arsenic, lead, nitrate, and fluoride contaminate groundwater (MoWR 2015). Arsenic in drinking water, and the associated health effects, is a major problem of global concern (Kapaj et al. 2006; Thakur et al. 2013; Chakraborti et al. 2017). Around 296 million people residing in more than 100 countries are suffering due to geogenic arsenic contamination of groundwater (Chakraborti et al. 2018). If we consider the people in India, Bangladesh, and Pakistan altogether, more than 187 million inhabitants are exposed to excess arsenic in drinking water (Hossain et al. 2013; Chakraborti et al. 2018). Inorganic arsenic of geological origin is recognized as the main form of arsenic in groundwater (MoWR 2014). The extensive problem of arsenic contamination of groundwater prevails in Argentina, Bangladesh, Chile, China, India, Nepal, Pakistan, Taiwan, and parts of Latin American countries, parts of Eastern Europe and the United States of America (Ravenscroft et al. 2009; WHO 2012; Murcott 2012). The mean level of arsenic in the continental crusts is 1–2 mg/L and in indigenous rocks 1.5–3 mg/L, whereas, in sedimentary rocks, it is in the range of 1.7–400 mg/L (MoWR 2010b). Volcanic action has arsenic-containing vapour which contributes about one-third of the natural source of arsenic, and two third comes from anthropogenic sources (MoWR 2010a). Geogenic sources for arsenic occur through geothermal or volcanic activities or through weathering of rocks and minerals (Smedley and Kinniburgh 2002). Biogenic sources of arsenic include plants and agricultural organisms or micro-aquatic biota, while anthropogenic sources of arsenic occur due to human activities (Smedley and Kinniburgh 2002).

The impact of arsenic poisoning is severe in many developing countries. India, Bangladesh, and Taiwan are among the most affected developing countries. Around 60 districts of Bangladesh have more than 0.05 mg/L of arsenic in drinking water (which is higher than the recommended limit BIS Bangladesh), and the situation is devastating. More than 12 states from India reported arsenic in drinking water. The arsenic prone area is located in the flood-prone belt of Sone-Ganga interfluvial region and is mainly distributed in the river Ganga and Brahmaputra region (MoWR 2014). Among those States, West Bengal, Bihar, Assam, and Uttar Pradesh are severely affected. Darbhanga, Purnea, and Kishanganj districts are exceptions because they are not affected by arsenic and are scattered and isolated places showing no distinct root of connection to one another (MoWR 2015). A study by Central Ground Water Board (CGWB) in India identified that the districts lying in the area of Ganga and other tributaries originating from the Himalaya shifted over a period of time and became arsenic contaminated (MoWR 2014).

Bihar is rich in groundwater resources. In 2002, two villages (Barisban and Semaria Ojhapatti) of Bhojpur district of Bihar, in the middle Ganga plain, reported more than 0.05 mg/L arsenic contamination (Chakraborti et al. 2003). As of 2017, out of 38 districts of Bihar, more than 1600 habitations of 67 blocks from 15 districts with a total population of more than ten million have been reported to have arsenic groundwater contamination above 0.05 mg/L (MoWR 2010a, b; Thakur and Gupta 2016). Ghosh et al. (2007, 2012) pointed out the spreading of arsenic in drinking water and estimated that in 2011, 18 districts had arsenic more than 0.05 mg/L, and around 24% of the population was directly consuming drinking water with an arsenic level higher than 0.01 mg/L. Saha (2009) in his study found that the southern bank of the river Ganga is more arsenic prone than the northern bank of river Ganga and affected more than 40% of the Bihar's population. It is estimated that more than 13.85 million people out of around 50 million total population in the area could be under the threat of arsenic contamination above 0.01 mg/L, and out of those at risk, for 6.96 million people the level of arsenic could be above 0.05 mg/L (Thakur and Gupta 2016).

High arsenic in drinking water over the long run has been associated with many diseases such as skin lesions (Argos et al. 2011; Pierce et al. 2011) and arsenicosis. Human health and arsenic-induced diseases in Bihar and West Bengal are raising public health concern in India, where 30 million people living in the area are potentially at risk of health problems (Hossain et al. 2013) and more than 100 million people are affected in India and Bangladesh as per World Health Organization (WHO) guidelines of a limit of 0.01 mg/L in drinking water (Hossain et al. 2013; Chakraborti et al. 2009, 2010). Previous research indicates that over ten million people in northern Bihar Gangetic plane are drinking water containing arsenic more than 0.01 mg/L (Thakur and Gupta 2014). High level of arsenic in drinking water over prolonged period has also been associated with skin lesions (Ahsan et al. 2006; Chakraborti et al. 2003; Argos et al. 2011; Pierce et al. 2011; Lindberg et al. 2008), Melanosis and hyperkeratosis (Guha Mazumdar 2003, 2008), reproductive effects (Hopenhayn et al. 2003a, b), cardiovascular disease (Chen et al. 2013; Wu et al. 2012) and diabetes (Pan et al. 2013). Besides various health effects, arsenic contaminated water also leads to economic costs such as medical cost, loss of wage, and reduction in productivity and efficiency of the affected people (Roy 2008; Khan et al. 2014; Pitt et al. 2015; Mahanta et al. 2016).

With the above-given background, this chapter examines the arsenic-contaminated drinking water and the associated health effects on the inhabitants drinking the arsenic-containing water in the region of Bihar. Following the introduction, the chapter is organized in the following way. Section 2 deals with the literature on arsenic-induced health effects due to arsenic in drinking water globally. Section 3 is on the methodology used in the chapter. Section 4 presents results and discussions. Section 5 concludes the findings.

2 Literature Review

Arsenic in drinking water and the associated health effects have been extensively researched in both developed and developing countries. Various studies examined the relationship between arsenic concentration in drinking water and its associated impact on human health (Canter 1997; Smith et al. 2000; Rahman et al. 2006;

Santra et al. 2013). The literature mainly discussed on issues such as technical (Smedley and Kinniburgh 2002; Saha et al. 2007; Amanambu and Egbinola 2015), mitigation and adaptation issues (MoWR 2010b; Gani and Scrimgeour 2014; MoWR 2015; Singh et al. 2016 b, 2017, 2018), food chain contamination (Brammer and Ravenscroft 2009; Bhattacharya et al. 2012; Huq et al. 2006; Santra et al. 2013; Jackson et al. 2012), health and economic cost (Ahmad et al. 2005; Pitt et al. 2015; Roy 2008; Maddison et al. 2005; Mahanta et al. 2016), and health impacts (Smith et al. 2000; Argos et al. 2011; James et al. 2015; Chakraborti et al. 2016a, 2017, 2018).

The positive association between arsenic in drinking water and cancer has been established by many researchers (Smith et al. 2000; Morales et al. 2000; Heck et al. 2009; Pan et al. 2013). If high concentrations of arsenic in drinking water continue for more years, arsenic gangrene and cancer are likely to appear (Clarke 2001; Rahman et al. 2005; Chakraborti et al. 2010, 2015). The clinical manifestation of arsenic also includes different forms of cancer such as skin cancer (Luster and Simeonova 2004; Rossman et al. 2004; Col et al. 1999; Guo et al. 1998), bladder cancer (Morales et al. 2000; Steinmaus et al. 2003; Bates et al. 2004), lung cancer (Hopenhayn-Rich et al. 1998; Chen et al. 2004; Chiu et al. 2004; Xia and Liu 2004; Wu et al. 2004; Marshall et al. 2007; Heck et al. 2009), as well as other non-cancer forms (Ahmed et al. 2006; Guha Mazumdar 2003; Tseng et al. 2005; James et al. 2015).

Chakraborti et al. (2016a) studied the arsenic-induced health problems in the Shahpur block and found that the population is at higher risk. In their study in a rural population of Darbhanga district of Bihar, Abhinav et al. (2016) found a strong correlation between arsenic level in groundwater and the arsenic in blood samples. Chakraborti et al. (2016b) studied arsenic-induced health problems in the Patna districts and found that cases of Arsenicosis and other skin lesions are increasing in the area. Singh and Ghosh (2011) reported the entry of arsenic into the food chain and its disturbance. Thakur and Gupta (2014) studied the arsenic in groundwater and its health effects in two blocks of Bihar and found that arsenic cases are more among children and women compared to men. Singh and Ghosh (2012) reported that children in the Maner block of Bihar are suffering due to arsenic in drinking water. Recently, chemical analysis of drinking water and health and vulnerability issues due to the consumption of arsenic-contaminated drinking water has been studied in Bihar (Singh et al. 2014, 2016a; Singh and Choudhary 2011).

Geogenic contamination of arsenic is a threat to the population and hinders sustainable development. Mitigating arsenic health impacts through community level has been studied by Bhatia et al. (2016), while Bose et al. (2016) suggested the need for sustainable development through mitigation. Singh et al. (2017, 2018) developed sustainable arsenic mitigation technologies and prediction algorithms for arsenic mitigation in the middle Ganga plains of India. There is a need for more study about arsenic in drinking water and the associated health issues in Bihar, and in the current paper, we report a study of five arsenic contaminated villages from Shahpur block in the Bhojpur district from Bihar state.

3 Methodology

3.1 Study Area Description

The state of Bihar consists of 38 districts. Each district is further divided into subdivisions and several blocks, and blocks are further divided into Gram Panchayats (GPs) and GPs have several villages (Census 2011). GPs are the smallest elected administrative representation in the state of Bihar. Shahpur was chosen for our study area for two reasons: it is among the most arsenic concentrated blocks, and the arsenic was first identified from this block. Shahpur block contains 10 GPs and 86 villages with a total population of 2, 12,170 (Census 2011). From the 38 districts, the drinking water of 15 districts, 65 blocks and around 900 habitations contain greater than 0.05 mg/L arsenic (Thakur and Gupta 2016). Figure 11.1 shows the districts in Bihar with more than 0.05 mg/L concentration of arsenic in drinking water.

3.2 Water Sample Collection and Instrumentation

We collected water samples from 173 surveyed households from their hand tube wells. Arsenic concentration in the water samples was tested using a field test kit at the time of the survey. The kits were manufactured by Prerana Laboratories which

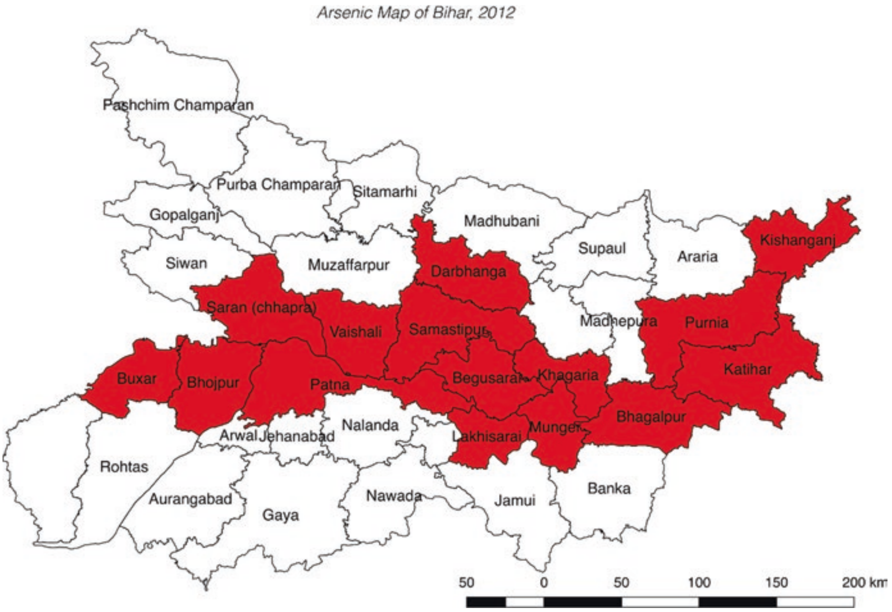

Fig. 11.1 Districts in the Bihar state of India where arsenic concentration in drinking water is more than 0.05 mg/L

Table 11.1 Arsenic concentration in water samples from the surveyed villages from Shahpur block in the Bhojpur district of Bihar state in India

SI	GPs	Village	Habitation (Tola)	Water samples tested	Arsenic concentration level (in mg/L)	
					Maximum	Minimum
1	Semariya	Gosaipur	Gosaipur	21	0.3	0
2		Dudhghat	Dudhghat	19	0.5	0
3		Bhaisaha	Bhaisaha	9	0.05	0.01
4		Semariya Ojha Patti	Semariya Ojha Patti	61	0.5	0
5	Bariswan	Bariswan	Bariswan	63	0.3	0
6	Total household survey			173	0.5	0

Source: Field Survey 2014–15

is recognized by both BIS and WHO. The field test kit provides results from 0 to 0.5 mg/L, and arsenic levels higher than 0.5 mg/L cannot be accurately determined using the test kit. The results were confirmed using a few samples (10%), which were tested by the State Arsenic Test Laboratory and the results were similar to those published by earlier work (Rahman et al. 2005; Thakur and Gupta 2014, 2016). Our results were used to identify the concentration of arsenic in the water samples and the affected population. The surveyed study area is given in Table 11.1.

The present study was also conducted to collect primary data through a questionnaire-based survey from February to April 2014. The questionnaire was divided into three sections – the first section aimed at collating demographic information and details of the households. The second section was based on information related to various health issues including health cost and other medical expenditure. In this section, we collected the cost of visits to doctors, diagnostic and medication cost, and the cost of hospitalization. The third section was dedicated to various socio-economic issues. It was difficult to identify patients with arsenicosis without the aid of trained medical practitioners and to obtain that was beyond the scope of this study. Therefore, the present study relied on the symptoms of primary, secondary and tertiary diseases induced due to arsenic in drinking water, as described by the WHO (2012), Thakur and Gupta (2016), and Chakraborti et al. (2003, 2016a, b).

4 Results and Discussion

4.1 Households in the Study Area

The primary survey of 173 households comprises 1472 individuals out of that 814 were males (55.29%), and 658 (44.71%) females. The mean age of the respondent is 44 years and the average years of schooling are 5.46 years. Around 31.1% of the surveyed population was illiterate. Majority of the households were male-headed. The average household's size was 8.5 which is higher than the national and state

Table 11.2 Summary of the surveyed household

Variables	Mean	Standard deviation
Arsenic in drinking water (in μg/L)	64.87	106.16
Age of the respondent (in years)	54.29	13.66
Household size (in number)	8.56	4.1
Respondent education (in years of schooling)	5.51	5.11
Agriculture as a primary source (in binary)	0.43	0.49
Household income (in INR monthly)	23,277.46	14,843.89
Household expenditure (in INR monthly)	10,947.25	8081.97
Household health expenditure (in INR monthly)	1748.19	9009.8

Source: Field Survey 2014–15

average. Sex ratio among low-income households is higher than higher income households. Nearly half of the households are involved in the farming activity, and the rest of the households have more than one occupation. The main source of drinking water is through hand pump well, and more than 90% of the households have access to drinking water. Majority of households are Hindu while more than half of them have a joint family. Arsenic and iron problem is prevalent among households drinking sources, and few of the households are facing various water-borne health issues. Table 11.2 provides information on the summary of the surveyed household.

4.2 *Water Sample Test Results*

The results of the water sample test revealed Dudhghat village of Semariya GPs have the highest arsenic concentration than the other villages (Table 11.3). In Dudhghat, 10.53% of the water samples contain more than 0.3 mg/L arsenic and more than 57% of the water samples contain higher than 0.1 mg/L arsenic in drinking water. In the Gosaipur village, around 71% of the water samples tested for arsenic concentration were found to be safe for drinking, and 9.52% of the samples contain higher than 0.1 mg/L arsenic. Bariswan and Bhaisaha villages have an arsenic concentration in the range of less than 0.3 mg/L. Despite of so many programmes available by the government, the arsenic concentration level in Semariya Ojha Patti village[1] is still poor and more than 50% of the households still drink water that contains more than 0.05 mg/L of arsenic. If we see the concentration level of all the villages together, results revealed around 36% of the water samples do not contain arsenic, around 10% of the water samples contain between 0.051 and 0.105 mg/L arsenic, 16% have between 0.101 and 0.3 mg/L and 2.89% of the tested water samples contain more than 0.3 mg/L arsenic. The results of the water samples test

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[1] The first incidence of arsenic in drinking water in Bihar was found in Semariya Ojha Patti village of Shahpur block from Bhojpur district (Chakraborti et al. 2003).

Table 11.3 Arsenic concentration levels in drinking water in the study villages. (Values shown are in percentage)

Study villages	Arsenic concentration levels (in mg/L)						Total number of samples
	Below detectable limit (BDL)	0.001 to ≤0.01	0.011 to ≤0.05	0.051 to ≤0.1	0.101 to ≤0.3	0.301 to ≤0.5	
Gosaipur	71.43	4.76	4.76	9.52	9.52	0.00	21
Dudhghat	21.05	5.26	10.53	5.26	47.36	10.53	19
Bhaisaha	0.00	33.33	66.67	0.00	0.00	0.00	9
Semariya Ojha patti	31.15	11.48	6.56	21.31	26.23	3.28	61
Bariswan	38.1	33.34	22.21	3.17	3.17	0.00	63
All villages	35.84	19.07	15.60	10.40	16.20	2.89	173
Total (samples)	62	33	27	18	28	5	173

Source: Field Survey 2014–15

support earlier reported results by others (Chakraborti et al. 2016a; Thakur and Gupta 2014, 2016). More details on the levels of arsenic in household drinking water are shown in Table 11.3.

4.3 Health Issues

From the survey, 60 (out of 173) households have either primary, secondary or tertiary health problems. Primary health problems include black spots on the body, conjunctivitis, gastroenteritis, and inflammation of the respiratory tract, while secondary health problems include white black spots on the body, hyper-keratosis, non-pitting edema, peripheral neuropathy, and liver and kidney disorders. Some of the tertiary problems we found during our survey include gangrene and urinary bladder cancer. Figure 11.2 presents arsenic-induced health issues in the study area.

4.4 Patients with Skin Lesions

From the survey, we found that many inhabitants were suffering from skin lesions due to excess concentration of arsenic in their drinking water. Around 2.19% of cases of surveyed male, 2.9% of surveyed female, and 3.04% of surveyed children are suffering from skin lesions. The incidence of skin lesions found to be higher among children than adult male and female. In Dudhghat village of Semariya GPs, a group of children who consume highly arsenic contaminated drinking water (higher than 0.1 mg/L) have skin lesions. From the survey, we found that people were heard about arsenic but were not aware of the health-related effects due to excess arsenic in drinking water. From our survey, we found that only 13.29% of the

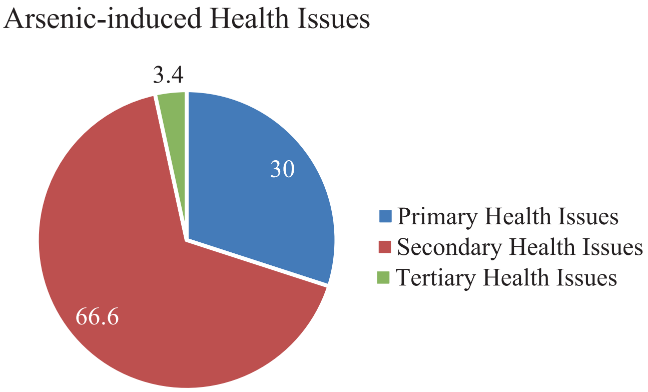

Fig. 11.2 Arsenic-induced health issues in the study area (in percentages)

respondent were aware of the health effects due to arsenic while around 49% of the respondent heard about arsenic.

4.5 Other Issues

Survey reveals that households are suffering not only due to health issues but various other social and economic issues such as unable to get married, discrimination, and divorce or separation and social exclusion are the prominent issues amongst the affected individuals and households. Although from the survey, it was found that only 1% among the surveyed population in Shahpur block facing problem-related to the skin which leads to the issues in their marriages. An unmarried female poorly reported skin problems, and thus the number of respondents with marriage issues related to their skin problems could be more. Poor households suffer socially due to the lack of adequate information on the arsenic caused health issues. From the field observations, it was found that cases with the suicidal tendency and depression due to skin lesion problems are prevalent among the affected individuals.

5 Conclusion

Arsenic contamination of groundwater in India and particularly in Bihar state is reportedly increasing at an alarming rate after each new survey has been done. Arsenic is a human carcinogen, and excess dosage of arsenic in drinking water is considered as a human health hazard and can be fatal. From the analysis of the water sample from the study area, it was revealed that the arsenic concentration in drinking water is more severe in the Semariya Ojha Patti GPs than the Bariswan GPs from Shahpur block. Around 46% of the tested samples of the study area contain excess arsenic over 0.01 mg/L. In the surveyed villages, Dudhghat and Semariya have an excessive amount of arsenic in drinking water. Around 10.53% of the samples contain an excess of more than 0.3 mg/L concentration levels, and more than 57% of the water samples contain an excess of 0.1 mg/L arsenic in Dudhghat. Although Bariswan and Bhaisaha have excess contamination levels in drinking water but the extent of contamination levels found to be lesser than other surveyed villages. In Gosaipur, around 29% of the samples were found excess concentration in the water.

As discussed earlier, excess concentration leads to health effects and various other issues. Majority of inhabitants who suffer health issues are in the form of skin lesions. The incidence rate of skin lesions among children found to be higher than the adults. Many children have skin lesions in Dudhghat where arsenic concentration level is higher than the other areas. We also found from the survey that, households have other problems besides health issues such as social problems, discrimination, suicidal tendency, depression, and social exclusion besides economic problems.

Lack of awareness and an alternative source of drinking water is common in the study area. Half of the respondents (51%) were not aware of the arsenic menace although they heard about arsenic. The increasing incidence of skin lesions among children make the group more vulnerable, and it leads to a disturbance on inter and intra-generation (Thakur and Gupta 2016). Drinking water is essential for a human to survive it needs urgent attention from the Government to work on sustainable management of water through various channels. The state of Bihar has adequate water resources, and proper management of water resources may help to reduce the problem. Otherwise, in the future problem will be more severe.

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Chapter 12
Arsenic Contamination of Drinking Water and Mitigation in Pakistan: A Case of Indus River Basin

Unaib Rabbani and Zafar Fatmi

Abstract Most of the arsenic-exposed population of the globe reside in South Asia. Over 100 million people living around the basin of the Ganges River in Bangladesh and West Bengal in India alone are exposed to arsenic through underground drinking water, and have received much attention. Millions in Pakistan also are exposed to arsenic through underground drinking water along the basin of Indus River. However, it has not raised eyebrows for health workers and policymakers in Pakistan. This chapter reviews the available evidence on arsenic exposure to the population in Pakistan through the Indus River, its severity and association with disease, and prediction of long-term consequences. It also dwells on the inadequate measures so far undertaken for the mitigation and control of the impending disaster. With the growing population in Pakistan, water is becoming scarce; and consequently, an increasing number of people are resorting to underground water. The future health consequences of arsenic exposure through drinking water could be enormous and could have detrimental impacts on the overall development of the population. This chapter begins with the situation analyses of arsenic exposure and toxicity by reviewing the published and unpublished evidence for the quality of water and its health consequences. It takes into account of the available literature, and the example of Bangladesh when discussing the health consequences as the Indus River shares the same Himalayas origin as the rivers in Bangladesh. It then outlines the arsenic mitigation efforts so far undertaken in Pakistan. It further discusses the policies and strategies for arsenic affected areas (mainly along the bank of river) of Pakistan where the mitigation efforts would be most effective in terms of population benefits. In discussing this, it structures a framework for policies and strategies for action for low resource countries for arsenic mitigation.

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1 Arsenic Contamination of the Indus Basin in Pakistan

1.1 Indus Basin

The Indus river is one of the longest rivers in the world with a length of about 2000 miles (3200 km) (Inam et al. 2007). The basin of the Indus River covers a large population and area. It has the total drainage area of 1,165,000 km^2, out of which 712,000 km^2 is in Pakistan (Nasir and Akbar 2012). The Indus river arises from Tibetan Plateau in western China then passes through Himalayan valleys. After crossing Kashmir region it enters and traverses through Pakistan and finally meets the Arabian Sea. (Inam et al. 2007). There is seasonal and spatial variation in the flow of principal rivers of Indus system as they are snow-fed (Ahmad 1993). Like other large river in South Asia, i.e., Ganges and Brahmaputra, Indus also has its origin in the Himalayas (Fig. 12.1).

Pakistan, with its total population of 208.7 million, is situated in the north-western part of the South Asian subcontinent (Pakistan Bureau of Statistics 2017). Total land area of Pakistan is about 796,096 km^2, which also features a diversified terrain and topography. Agriculture is the largest working sector in Pakistan, employing about 42% of the total workforce and contributing about 20% of the total gross domestic product (GDP) (Farooq and Wasti 2017). Underground water is the major source of drinking water, providing for about 63% of the population for household use (Pakistan Bureau of Statistics 2016). Additionally, groundwater is

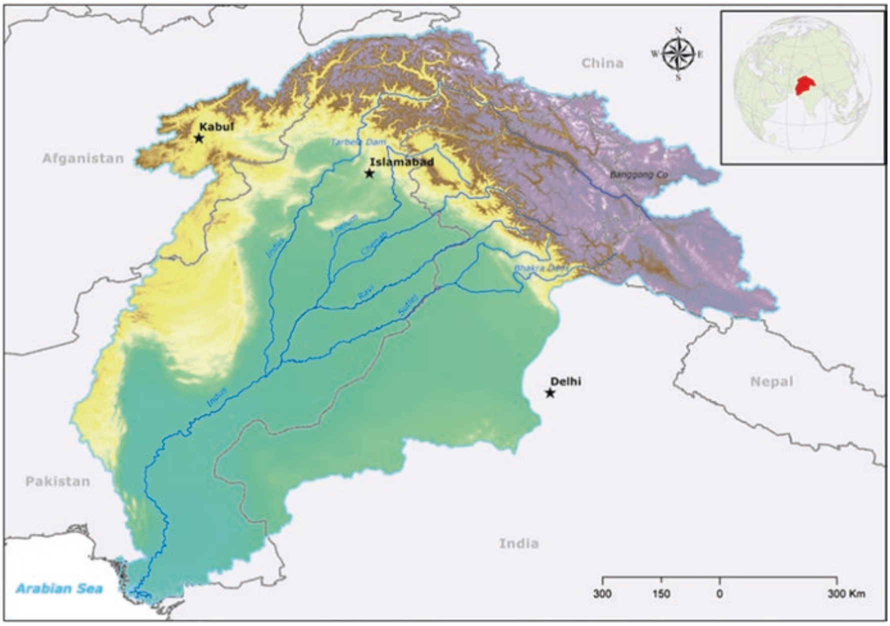

Fig. 12.1 Map of Indus Basin. (Reproduced with permission from Johnston and Smakhtin (2014))

also used for irrigation purposes, which is more common in Sindh and Punjab provinces (Bhutta and Alam 2006).

After reports from Bangladesh and India about arsenic (As) contamination of groundwater resources, preliminary and national survey for arsenic contamination of water sources showed unsafe levels of As in the groundwater sources (Ahmed et al. 2004). A large proportion of the population in Pakistan who depends on groundwater for drinking and domestic use may have been thus potentially exposed to unsafe levels of arsenic. It is therefore necessary to assess the problem and develop strategies for mitigation of arsenic in the country to identify a way forward for protecting the health of the population.

1.2 Arsenic in Underground Water in Pakistan

The issue of arsenic contamination in Pakistan brought to surface after reports of widespread arsenic (As) contamination in neighboring countries such as India and Bangladesh. Several organizations have done an assessment about water quality for arsenic contamination including; Pakistan Council for Research in Water Resources (PCRWR), United Nations Children's Emergency Fund (UNICEF), and Aga Khan University. In addition, Sindh Health and Educational Development Society and Human Resource Development Society carried out awareness-raising and dissemination of information about arsenic in the communities.

The initial assessment of the arsenic contamination of water was carried out in six districts of Punjab by PCRWR with assistance from UNICEF in 1999. It identified that about 14% of the drinking water samples had arsenic levels above the World Health Organization's (WHO) guideline of 10 ppb (Ahmed et al. 2004). A national survey for arsenic in drinking water was carried out in 2001. A total of 35 of 104 districts from all the four provinces viz. Punjab, Sindh, Khayber Pakhtunkhwa (KPK), and Balochistan, were included in the survey. Overall 9% of the 8712 samples had arsenic concentration more than 10 ppb. Punjab and Sindh were the most affected provinces with 12.2%, and 11.0% of the drinking water samples had arsenic concentrations more than 10 ppb, respectively (Ahmed et al. 2004). The national survey was followed by blanket testing (all water sources in selected sub-districts) of arsenic in water sources in high-risk districts of Punjab and Sindh provinces. The districts of Dadu, Khairpur, Nawabshah and Tharparkar in Sindh province and Multan, Rahim Yar Khan, and Bahawalpur in Punjab province were included in the survey. About 21% in Sindh (n = 20,158) and 14% of drinking water samples in Punjab (n = 11,975) were contaminated with arsenic at a level above 10 ppb (Ahmed et al. 2004).

Studies from districts of Sindh province estimated 2–64% prevalence in Khairpur district, 30% in Matiari and 57% in Thatta district (Arain et al. 2007; Fatmi et al. 2009; Rubab et al. 2014). The studies conducted in districts of Punjab showed a prevalence range between 32% and 100% (Ali et al. 2015; Arshad and Imran 2017; Malana and Khosa 2010; Qurat ul et al. 2017; Rasool et al. 2017). Only one study was conducted in the province of KPK in the district of Peshawar where 11 out of 13 samples reported arsenic concentration in drinking water to exceed 10 ppb (Ishaq et al. 2013). All these studies were small-scale with small sample size and confined to specific geographical areas. Given these limitations, these studies cannot be used to draw a comprehensive picture of arsenic contamination in Pakistan. However, these are useful for the identification of high-risk areas in the country. A recent study with a larger sample (n = 1184) size from all over Pakistan, showed that 66.3% of the tested samples were contaminated with arsenic and that the Indus plain was a high-risk area (Podgorski et al. 2017). They estimated about 50–60 million people in Pakistan are at risk of arsenic exposure. The study raised concern among authorities. However, there was disagreement regarding its findings due to its methodological limitations (Niazi 2017; Rabbani et al. 2017). Table 12.1 summarises the studies conducted to assess the arsenic contamination of water sources.

Various studies across the world have shown a relationship between arsenic concentration in groundwater and the distance from the river (Berg et al. 2007; Hoang et al. 2010). Areas located near the bank of rivers are the ones that are highly affected by the arsenic contamination. Similar pattern of arsenic distribution exists in Pakistan. Results from the national survey showed that districts located near the river Indus had higher arsenic concentrations in Punjab and Sindh provinces. Another study from Khairpur also reported the similar distribution of arsenic contamination (Fatmi et al. 2009). Further analysis by Rabbani et al. found that wells located within 18 km from the bank of river Indus in district Khairpur had a higher concentration of arsenic than distant wells (Rabbani et al. 2016).

2 Evidence of Arsenic Exposure and Its Health Impact Studies

2.1 Arsenic Exposure Related Studies in Pakistan

Initial surveys conducted in Pakistan showed widespread arsenic contamination. There was a need to assess exposure and health effects in the population. In this regard, UNICEF invited department of Community Health Sciences, Aga Khan University Karachi to design and conduct a study to assess the health burden of arsenic contamination of underground water in one of the heavily affected districts of Sindh, Pakistan. Therefore, a cross-sectional multi-stage stratified cluster random sampling survey was conducted in district Khairpur in the province of Sindh in 2006 by Fatmi et al. (2009), which aimed to determine the prevalence of arsenicosis, relation between arsenic levels in underground drinking water and frequency of manifestation on skin (arsenicosis), and the role of nutrition on arsenicosis prevalence. In this study, a total of 3874 individuals were examined and interviewed; and a total of 2517 water samples and 505 urine samples were tested on Atomic Absorption Spectrophotometer (AAS) for arsenic levels.

Table 12.1 Summary of studies conducted to assess the arsenic contamination of water sources in Pakistan

Author (year)	Site (province)	N	Prevalence[a]	Comments
Shakoor et al. (2018)	Punjab	123	75	Small sample size
Podgorski et al. (2017)	Pakistan	1184	66.3	Large sample size covering large geographical area. Overestimation of exposed population due to methodological limitation.
Qurat ul et al. (2017)	Rahim Yar Khan Punjab (Punjab)	51	32.5	Small sample size
Arshad and Imran (2017)	Kasur,	35	100	Small sample size Selected villages only
	Islamabad	18	0	
	Rawalpindi	15	0	
	Bhawalpur	18	72	
Rasool et al. (2017)	Mailsi (Punjab)	44	100	Small sample size
Rasool et al. (2015)	Mailsi (Punjab)	52	86	Small sample size
Ali et al. (2015)	Thar (Sindh)	3	59.5 ± 1.88	Only three aquifers were sampled during different quarters throughout the year.
Brahman et al. (2014)	Thar (Sindh)	~150	BDL in fresh samples 194–683 μg/L in stored water	Fresh and stored rainwater samples during two consecutive monsoon seasons and at different periods were tested for As concentrations. As levels increased over time in the stored water.
Sultana et al. (2014)	Lahore (Punjab)	30	87	Small sample size
Rubab et al. (2014)	Thatta (Sindh)	37	37	Small sample size
Ishaq et al. (2013)	Peshawar (KPK)	13	85	Small sample size
Malana and Khosa (2010)	DG Khan (Punjab)	32	18	Small sample size
Fatmi et al. (2009)	Khairpur (Sindh)	2517	12	An epidemiological study from Sindh to assess the burden of arsenicosis in the community. Wide variations in arsenic levels with distance from the bank of the river.
Arain et al. (2007)	Matiari and Khairpur (Sindh)	94	47	Small sample size

[a] Percentage of wells with arsenic concentrations more than 10 ppb

Approximately 12% of water samples showed arsenic level above 10 ppb, and 2.2% above 50 ppb. Rabbani et al. (2016) in their study used geographical information system (GIS) data and techniques, in which they developed risk maps of district Khairpur Sindh and estimated the population at risk of arsenic exposure. In the study, authors used village-level maps provided by the revenue department of district Khairpur, population data from district census report and Global Positioning System (GPS) coordinates of water sources. All this information was used for spatial analysis. Results showed that the majority of the wells located near the bank of the river Indus were contaminated with higher arsenic levels while those away from riverbank were generally safe. It was identified that a band of about 18 km along the river Indus was the high-risk area where the majority of the wells were arsenic contaminated. Based on this information, authors extrapolated result to the whole length of river Indus and estimated that about 13–16 million people in Pakistan were exposed to arsenic-contaminated water, including 6 million women and 5.6 million children under 15 years of age (Rabbani et al. 2016).

Some studies also assessed the intake of arsenic through water and food sources. Average daily dose was reported in some studies from Punjab (0.036–12 μg/kg/day) (Rasool et al. 2017; Shakoor et al. 2015), Sindh (5.7 μg/kg/day) (Ahmed et al. 2014; Arain et al. 2009; Shah et al. 2011) and KPK (0–0.0056 μg/kg/day) provinces (Muhammad et al. 2010). The exposure dose in Punjab and Sindh were higher than the US environmental protection agency reference dose (RfD), i.e., 0.3 μg/kg/day (U.S. Environmental Protection Agency 1991). The level of arsenic in drinking water was positively correlated with urinary arsenic concentration, and this is a more sensitive indicator of higher arsenic exposure. The median urinary concentration in Punjab has been reported to be as high as 118 ppb (Bibi et al. 2015; Sughis et al. 2014) while in Sindh this value is 28.5 ppb (range: 0.1–848) (Ahmed et al. 2014). Another indicator of arsenic exposure is scalp hair levels which also has a strong positive relation with arsenic in drinking water (Kazi et al. 2011). Higher levels of arsenic in scalp hair samples have been reported in various studies in Pakistan (Baig et al. 2011, 2016; Kazi et al. 2011; Shah et al. 2011). Few studies have also assessed the arsenic levels in nail and blood in Pakistan. All of these studies are limited by the scope and scale as most of them were done in selected areas and a particular group of the population. Therefore, these values should be considered as an indicator of exposure, but may not represent the exposure of the population. Presence of arsenic in the water and soil can also lead to bioaccumulation in the food chain. Thus, studies have shown higher levels of arsenic in vegetables and fishes as well in Pakistan (Nawaz et al. 2010; Rehman et al. 2016; Shah et al. 2011; Waheed et al. 2013). Although few studies were conducted but these suggest that millions of people in Pakistan are exposed to arsenic. Major source of exposure is through the contaminated groundwater as well as food. These people have the potential risk of developing adverse health effects due to arsenic exposure.

2.2 Health Effect Studies

The science about the adverse health impacts of arsenic is still unfolding, and it seems to involve every organ system of the body. Arsenic exposure leads to skin and non-skin health effects. Arsenicosis is the disorder of skin—characterized by hyperpigmentation and hardening of the skin of palms and soles on both sides (symmetrical) (Kadono et al. 2002). These skin lesions may culminate in cancers of the skin. Besides skin, arsenic exposure has been associated with cardiovascular diseases, hypertension, diabetes mellitus, decreased lung functions, gastrointestinal disturbances, liver disorders including enlargement, tenderness, jaundice, ascites and elevated liver enzymes, and internal cancers including liver and bladder (Ahsan et al. 2006; Brown and Ross 2002; Chakrabarti et al. 2018; Khan et al. 2003; Nafees et al. 2011; Smith et al. 1992). The spectrum of these disorders depends on the exposure dose, i.e., concentration in the source and exposure duration (Nafees et al. 2011; Yoshida et al. 2004). Literature on assessment of health impact due to arsenic exposure is scarce in Pakistan.

Initial epidemiological investigations of arsenicosis carried out by the Institute of Public Health Punjab, reported a lower prevalence of definitive and borderline arsenicosis 11 and 130 per 100,000 respectively (Ahmed et al. 2004). This study, however, did not find any association between groundwater arsenic concentrations and dermatological lesions. A later study was also conducted in northern Punjab where the prevalence of clinical arsenicosis was found to be 92 per 100,000 while that of borderline cases was 242 per 100,000 individuals of the population (Ahmed et al. 2004). Fatmi et al. (2009) in Sindh conducted a more detailed study in which 3874 individuals were examined and interviewed, 2517 water samples and 505 urine samples were tested on Atomic Absorption Spectrophotometer (AAS) for arsenic levels besides testing of water samples for arsenic contamination. The burden of arsenicosis was reported to be higher in Sindh. The study disclosed that the numbers of definitive cases of arsenicosis and suspected cases were 3.4 per 1000, and 13 per 1000 population (among ≥15 years of age) in district Khairpur, respectively. The mean arsenic level of water (17.2 ppb) and urine (56.4 ppb) were significantly high in arsenicosis cases compared to mean arsenic level of water (5.5 ppb) and urine (38.5 ppb) in normal individuals. Arsenicosis was significantly higher in malnourished, BMI < 18.5 kg/m^2 (25.3/1000) than adequately nourished, BMI > 18.5 kg/m^2 (10.5/1000) individuals, indicating higher susceptibility of malnourished people to health effects of arsenic (Fatmi et al. 2009). This prevalence was higher 13.5% among households exposed to a higher arsenic concentration >50 ppb (Fatmi et al. 2013). A study also reported a decrement of lung functions with exposure to arsenic (Nafees et al. 2011). Changes in the lung functions start even before skin manifestations. Arsenic exposure has been linked to the reduced activity of antioxidative enzymes (Bibi et al. 2015). Some case-control studies have reported higher levels of arsenic in biological samples of cancer and hypertension patients compared to controls (Afridi et al. 2014; Arain et al. 2015b; Wadhwa et al. 2011, 2013). Another study reported higher levels of N-acetyle β glucosaminidase

Fig. 12.2 Skin manifestations of arsenic. (Reproduced with permission from Fatmi et al. (2009))

(NAG), a biomarker of proximal tubular damage in kidney, among people exposed to high levels of arsenic (Arain et al. 2015a).

Although the literature on health effects due to arsenic exposure in Pakistan is scanty but the available studies suggest that people are suffering from adverse health effects resulting from arsenic exposure. No epidemiological studies were conducted to attribute arsenic exposure to non-communicable diseases; therefore, this side of the picture is still veiled. The scenario will be worse in the future if this exposure is continued and may result in a rise in cancerous and other chronic disorders (Fig. 12.2).

3 Arsenic Mitigation in Pakistan

Since 1999 when a first preliminary survey for arsenic contamination of groundwater sources was conducted, a number of activities were carried out by different stakeholder for arsenic mitigation in Pakistan. These included; assessment of water quality, advocacy and social mobilization, development of mitigation plan and prevention of exposure (Ahmed et al. 2004). However, these activities were not concerted and lacked sustainability. Therefore no tangible outcomes were observed (Islam-ul-Haque and Nasir 2015). Next few paragraphs will present the status of arsenic-related intervention so far conducted in Pakistan.

Formal efforts of arsenic mitigation started in 1999 with the first arsenic survey conducted by PCRWR with support from UNICEF (Ahmed et al. 2004). This was later up-scaled to a national arsenic survey in 2001. This time the Public Health Engineering Department (PHED) and Local Government and Rural Development were involved and they surveyed one third (34 of 104) of the districts in Pakistan (Ahmed et al. 2004). These surveys were later followed by capacity building for mitigation of arsenic. UNICEF team visited Bangladesh to learn from their experiences, and this experience was applied in Punjab by local government and Non-Governmental Organizations (NGOs) in Sindh. Local governments in Sindh and

Punjab organized provincial level seminars to publically disseminate the information with government officials, NGOs, and media about arsenic contamination status in Pakistan and developed provincial mitigation frameworks. Capacity building activities included training of officials, NGOs and local representatives regarding arsenic exposure, its effects on human health, water sampling, use of arsenic field testing kits, collection and analysis of data and identification of alternate safer water source for consumption. This training, however, did not include the training of health care providers regarding the identification and diagnosis of arsenicosis. Social mobilization and advocacy started after the surveys. However these activities were mostly one-time effort and not sustained to change community knowledge and behaviors (Islam-ul-Haque 2015). At the community level, various NGOs in Sindh (SAFWCO and SHED) and Punjab (HDRS) started disseminating information about the arsenic exposure and its hazards and marking safe and contaminated wells for identification and use. Later in 2004, blanket testing of all water sources was carried out in high-risk districts of Sindh and Punjab (Findings described earlier in the text).

Epidemiological studies were conducted to assess the burden of health effects due to arsenic exposure. In this respect, the first epidemiological study was conducted in seven districts of Punjab in 2002–03 by Institute of Public health, Lahore and sponsored by UNICEF. In Sindh, UNICEF invited department of Community Health Sciences, Aga Khan University Karachi to design and conduct a study to assess the health burden of arsenic contamination of underground water in one of the heavily affected districts of Sindh, Pakistan. Therefore, a more precise cross-sectional multi-stage stratified cluster random sampling survey was conducted by Dr Fatmi and his team in 2006 in district Khairpur of Sindh province, which had the objectives to determine the prevalence of arsenicosis, relation between arsenic levels in underground drinking water and frequency of manifestation on skin (arsenicosis), and role of nutrition on arsenicosis prevalence (Fatmi et al. 2009).

Dr. Fatmi and his team from the Department of Community Health Sciences, Aga Khan University with the support of UNICEF developed capacity building project. Main objectives of the project were at a primary level, to train master trainers on arsenic and utilize these master trainers to train health professionals at the secondary level and also to develop training/awareness material on arsenic. In this regard, training material (training/evaluation modules) for primary and secondary level training on arsenic mitigation was developed (Fatmi et al. 2008). In the development of manual, help was taken from the field guide for detection, management and surveillance of Arsenicosis prepared by South East Asian Regional Office, WHO and the Chinese manual for diagnosis of Arsenicosis. An extensive review of the literature and expert consultations with dermatologists including Chinese research experts were undertaken. This was followed up by a visit to China where meetings/interviews with arsenicosis patients were held in order to examine clinical variation in the presentation of the disease, which may be geographically and/or genetically influenced (Fatmi et al. 2008). This material was utilized in arsenic workshops to train master trainers at primary training workshop and health professionals at secondary level training workshops. Twenty master trainers and 100 health professionals (60 doctors and 40 paramedics) from seven highly affected districts of Sindh were trained on arsenic mitigation in the primary and secondary level training workshops. For the awareness of affected communities and local health care providers, team translated English version of training manual of "diagnosis and management of arsenicosis in Pakistan" into local languages (Urdu/Sindhi) and also developed arsenic awareness brochure in English/Urdu/Sindhi languages. Provincial framework for arsenic mitigation was developed in arsenic workshops. This project resulted in the development of training and evaluation modules for master trainers, 20 master trainers, 100 arsenic trained health care providers, training material, health education material for communities and a provincial framework for arsenic mitigation in Sindh.

Arsenic removal technology across the world ranges from municipal levels plants to home-based units. Arsenic removal methods are based on chemical processes such as oxidation-reduction, precipitation, adsorption and ion exchange, solid-liquid separation, physical exclusion, and coagulation (Nicomel et al. 2015). Available technologies are based on these processes and include; air oxidation, chemical oxidation, alum coagulation, ion coagulation, sorption techniques using activated alumna, iron coated sand or ion exchange resins, membrane technologies such as nano-filtration, reverse osmosis and electrodialysis (Nicomel et al. 2015). These methods are effective in the removal of arsenic from the drinking water but require technical expertise, high operational costs, and energy. Given the socio-economic status of the population affected by arsenic in Pakistan, these methods are not feasible. PCRWR in collaboration with UNICEF started research to locally developed technology for arsenic filtration and treatment. In this regard clay-pitcher, plastic gravity flow, and ceramic cartridge arsenic removal filters were developed (Fig. 12.3). These low-cost arsenic removal technologies were developed keeping in view the socio-economic profile of the rural and urban population in Pakistan (Government of Pakistan 2007). These technologies were evaluated for a period of 6 months. The evaluation was done on the parameters like the composition of pre and post-filtered water, the life of arsenic removal

Fig. 12.3 Arsenic removal filters developed by PCRWR. (Government of Pakistan 2007)

media, flow rates, trace element distribution, microbiological effectiveness, and estimated cost. Evaluation results found that clay pitcher was most effective among the three technologies. On the ground, however, there was a limited production of clay pitchers, and no promotion in the communities was carried to implement in the communities while gravity flow arsenic removal cartridge filter was not available for community use (Islam-ul-Haque 2015). Community-based arsenic removal tanks were installed with UNICEF support, but out of 25 units, only two were found functional after 1 year in Rahim Yar Khan district of Punjab (Islam-ul-Haque 2015). Reasons for failure were; non-availability of filter media, lack of community capacity to test water and non-willingness, no mechanism for monitoring and testing from program office and no monitoring of project and promotion (Islam-ul-Haque 2015). UNICEF in Rajanpur Punjab provided a sachet containing chemical coagulants and disinfectants. However, no follow-up was done to assess its community acceptance and feasibility.

First formal arsenic mitigation plan, the National Action Plan for Arsenic Mitigation (NAPAM) 2007–2011 (Government of Pakistan 2007), was developed to protect the population from hazardous health effects of arsenic exposure. An inquiry of NAPAM 2007–2011 is given in the following paragraphs.

Given the widespread arsenic contamination of water sources, Pakistan needed a comprehensive mitigation plan to prevent arsenic exposure and avoid health consequences resulting from its long-term exposure. The central body that led to the development was the Ministry of Science and Technology, Government of Pakistan. UNICEF was the leading partner in arsenic mitigation efforts and policy development in Pakistan. Other stakeholders included; PCRWR, the governments of Sindh and Punjab, Public Health Engineering department, local government, Ministry of the environment, departments of health and NGOs. Development of NAPAM involved a series of seminar-cum workshops at the provincial and national level. Components suggested to be included in the policy in these workshops were; provision of alternate water supply, water testing and treatment technologies, advocacy, developing an institutional framework, monitoring, and surveillance of water sources, capacity building, and research and development. A number of objectives were set in NAPAM. Major components of NAPAM included; screening of water sources, health effect assessment, and development of diagnosis and management protocols, provision of safe drinking water, arsenic removal technologies, institutional framework, and social mobilization.

According to plan screening of the whole Indus basin was to be completed within 2 years of plan development; however, no progress was made in this regard. Health effects assessment remained confined to a few studies only. Diagnosis and management protocols were developed and translated into local languages but could not be implemented on a large scale. Similarly, no progress was made in the provision of alternate water supply. In highly affected areas surface water was to be treated as alternate source, but no treatment plants were established to serve the population. Deep wells as alternate source were part of the plan. However, no systematic attempts were made to dig deep wells according to local geography. Arsenic removal technologies developed locally were not produced at a mass scale to bring costs down. Maintenance facilities for arsenic removal technologies and filter media were not made easily available in the areas worse hit by arsenic contamination. Behavior change communication strategies were not sustained to change community behaviors and improve the acceptability of the technologies. Provision of potable water through tankers was also kept as a strategy, which was not feasible economically as well as road infrastructure is not available in rural areas. Monitoring and surveillance of water sources could not be carried out, however plan was to establish lab facilities at tehsil levels. This should have been done in a stepwise manner by capacity building of locals and provision of basic equipment for water monitoring in high-risk areas and then subsequently into other areas. Involvement of the private sector and NGOs was not clearly defined. It is important to effectively involve the private sector to reach maximum population affected by arsenic contamination. No mechanism of coordination among various stakeholders was defined clearly to make the process smooth. Monitoring and evaluation framework for the action plan was given. However, no indicators were developed and included to track the progress of plan. Similarly, no protocol was given for the evaluation of the plan.

3.1 Complacency Regarding Mitigation Efforts

Despite nearly two decades since UNICEF led the investigation of water sources and identified arsenic as a public health problem in Pakistan, and millions of people are at risk of chronic effects of arsenic exposure, no systematic effort has been done to mitigate the adverse effects. This chronic exposure to arsenic will result in future losses to the nation in the form of disability and premature deaths and direct costs of health care. There is a number of reasons that Pakistan failed to control the problem of arsenic contamination and protect people from exposure.

The first and most important among the causes of failure is the lack of ownership of arsenic mitigation by the governments. It was UNICEF, which started mitigation activities and provided financial and technical support. But later governments did not fulfill their commitments to finance and continue mitigation efforts. Activities suggested in NAPAM were not made part of the annual development plans (ADP) (Islam-ul-Haque and Nasir 2015). Provincial governments also did not make any tangible efforts for the implementation of NAPAM. Arsenic mitigation activities remained confined to papers and small-scale projects by NGOs. Provinces did not functionalize steering committees, and no actual mobilization of resources was done at the provincial level. Various factors play behind this lack of political interest in developing countries. These include; lack of tangible outcome, extra work, and competition with other problems and differing motivation of various stakeholders (Summerill et al. 2010a, b). Historically governments in Pakistan had a tendency to spend more on structures such as roads, which are more visible compared to health and other social domains, which often take longer to give visible outcomes. Poor human resource management in terms of an adequate number of personnel and skills lead to overburdening of existing staff. Budgetary allocation for health is low in most of the developing countries due to which additional programs in the health sector are not given propriety. Additionally, in Pakistan, this is also due to political instability in the country and discontinuation of old policies by new government regimens. Local government system had provided infrastructure to reach the communities effectively and engage them in arsenic mitigation efforts. However, this opportunity was never utilized for arsenic mitigation in Pakistan. This lack of political ownership led to stagnant progress in arsenic mitigation.

Financial support has been identified as one factor affecting the sustainability of the arsenic mitigation program (Rahman and Paul 2013). Since this program was donor-driven, after the withdrawal of donor funding, activities came to a halt. NAPAM was not implemented in its true spirit as suggested in the implementation plan, and national steering committee did not continue its function.

Screening of water sources was not continued and expanded therefore a large proportion of the population remained unidentified as risk exposed group (Islam-ul-Haque and Nasir 2015). Similarly provision of alternate water supply to the identified exposed population was not done. There was a lack of coordination between various governmental departments, which hindered the arrangements of alternative water sources. Behavior change communication activities initiated earlier were one-time activity and therefore could not bring changes in the population regarding arsenic.

Arsenic-based removal technologies (ART) installed at community levels were not maintained, and people returned to the untreated water. Lack of financial resources and political interest led to the discontinuation of research at PCRWR. On the other hand inability of the affected population to pay for the removal media and maintenance of the units also contributed to the abandoning of ARTs. Additionally, spare parts and removal media used in ARTs were also not available in the local markets rendering it difficult for users to continue use of units (Islam-ul-Haque 2015).

Although Pakistan made rapid progress in arsenic mitigation initially during which assessment of water sources, capacity building and research and development were carried out. However, this progress lasted only a few years after which things became stagnant. Lack of political commitment, dependence on foreign aid and lack of financial resources lead to the discontinuation of arsenic mitigation efforts in Pakistan (Authors' observations).

3.2 Future Prospects for Pakistan

In order to protect the health of the population from hazardous health consequences of arsenic exposure, Pakistan needs to take immediate actions. Successes and failures of Bangladesh in arsenic mitigation provide lessons for other low-income countries like Pakistan.

The first and most important requirement for addressing any issue is political commitment. Government and policymakers need to show and practice commitment to the protection of the population. In Pakistan, rapid progress was made for arsenic mitigation after initial surveys, and NAPAM was also developed. However, this lasted only for 5–6 years after which progress came to a halt. Donor aid is important for low resource countries like Pakistan, but over-reliance on donors for financial and other resources results in non-sustainability (Alymkulova and Seipulnik 2005). For example, Sono filters were introduced in Bangladesh with the help of donors. However, the project lacked networking among stakeholders, and adequate need assessment was not done before the launch of the project (Kundu et al. 2016). UNICEF led the arsenic mitigation in Pakistan by providing financial and technical support to the government and private institutions. But as soon as it withdrew support, the work on arsenic mitigation stopped. There is a need for commitment from the governments of the affected countries to allocate resources for arsenic mitigation with technical support from international organizations and minimal dependence on foreign aid. This will not only result in sustainability but also a local capacity building, which will further complement sustainability. A study reported that institutional weakness and lack of stakeholder accountability were hindering factors in the sustainability of arsenic mitigation in Bangladesh (Khan and Yang 2014). An institutional framework should be developed for progressing arsenic mitigation, and accountability should be ensured such as suggested by Islam-ul-Haque and Nasir (2015).

Human and cultural factors affecting sustainability and community buy-in of interventions need to be considered while developing an arsenic mitigation plan. Human and cultural factors challenging sustainability of mitigation program include; community readiness, engagement, support, knowledge, resources, acceptance, and participation (Kot et al. 2014; Rahman and Paul 2013). Further sustainability of any intervention for arsenic mitigation depends on cost, trust, geographical access and knowledge about the health effects of arsenic (Etmannski and Darton 2014; Singh et al. 2018). Advocacy and social mobilization for arsenic mitigation through various media is another issue that needs to be considered by policymakers and researchers in Pakistan. Effectively communicating evidence on arsenic contamination and health effects in the population to the political leaders is important to bring problem on policy agenda. Similarly, behavior change communication is important for disseminating knowledge and changing the behavior of the population regarding arsenic and gaining the trust of the communities regarding interventions (Singh et al. 2018). Electronic media can be used to bring a problem to the surface so that it gains population attention and also improve the knowledge of the population regarding arsenic exposure and health effects. This will create demand-side call for action. Studies from Bangladesh showed that using educators from or outside of the community is equally effective in improving knowledge of community regarding arsenic (George et al. 2013). However, this may not translate into behavior change and needs parallel actions such as well testing and support for alternate water source by the government to facilitate change (Madajewicz et al. 2007). This is necessary to sustain such efforts as knowledge fades with time and people tend to return to contaminated wells (Balasubramanya et al. 2014; Bennear et al. 2013). While designing any health education campaign, it is important to consider local context because areas may vary in socio-demographics, water contamination levels, and available water sources. Identifying and involving local activists and leaders is important for social mobilization and implementation of arsenic mitigation. Such mobilization will result in local ownership of mitigation efforts and sustainability. Lack of strong social network around and intervention may result in failure of intervention at the community level (Kundu et al. 2016). It is important to generate local resources so that dependence on government and NGOs is minimized because complete dependence will result in an inability to maintain technology use or maintenance of new wells (Bhattacharya et al. 2017).

Arsenic mitigation cannot be carried out in isolation, as this requires the involvement of various departments from within the government such as water boards, municipal corporations, irrigation department, local governments, health department, and research institutions. In order to carry out activities effectively, it is necessary to have proper collaboration and coordination among the various stakeholders. This will help to streamline activities towards a common goal and prevent waste of efforts and resources. Role of the private sector and local NGOs are also important in resource-constrained countries like Pakistan where the government cannot reach all areas. Partnerships with the private sector and NGOs should be designed in such a way that there is a clear demarcation of roles and responsibilities of each of the party. Proper mechanisms of monitoring and evaluation of the private sector should be developed to ensure accountability. It is important to streamline the activities of all agencies working on arsenic mitigation to avoid duplication of efforts. In many parts of Bangladesh failure resulted from a lack of coordination among different stakeholders (Milton et al. 2012). There was unplanned installation of new wells by different agencies where sustainability was not considered (Bhattacharya et al. 2017).

It is important for the sustainability of a program that it should be led by local leaders. Bangladesh was able to sustain its arsenic mitigation program partly because of leadership by local people (Jakariya et al. 2003). In Pakistan, UNICEF initiated local capacity building. A team was sent to Bangladesh to learn their experiences of arsenic mitigation and then implement these in Pakistan. In order to effectively utilize such experience, there should be a mechanism through which those who have experience in arsenic mitigation should transfer this knowledge. Second order learning is also important to run programs and bring innovations to sustain activities (Kundu et al. 2016). Experts from within and outside the country should carry out training of the local people in arsenic mitigation. Similarly, identification and training of people from communities for water sampling, testing, and health education should be implemented. This will provide a network of local people who can monitor water quality and provide continued health education to the community regarding arsenic. A study from Bangladesh showed improved knowledge of people after the implementation of health educators from the community (George et al. 2013). However, these workers should be monitored, and periodical refresher training should be provided to enhance knowledge and skills. The health department needs to develop and implement guidelines for the identification, diagnosis, and management of arsenicosis. Health care workers should be trained on these guidelines. Arsenic sources, exposures and health effects should be included in medical curricula.

In NAPAM 2007–11, a budget was proposed to carry out planned activities. This also described the proportion of budget from federal, provincial and donor agencies. However, this commitment of financial resource allocations was not fulfilled, and no implementation of NAPAM was done. In order to run the arsenic mitigation program, the government needs to prioritize the issue and allocate proper resource for implementation. In addition to money allocation, there is a need to place a governance mechanism to protect the leakage of resources and improve financial efficiency.

3.3 Intervention Options for Pakistan

In order to protect the population from arsenic exposure and its health effects, there is a need to work on the provision of safe water and addressing the health effects of exposed population. There are various interventions to protect from arsenic exposure of population through drinking water. Some interventions are short-term and required for worst arsenic hit areas immediately while other interventions are sustainable and useable in the long run.

3.3.1 Short Term

Short-term interventions should target populations, which are at the highest risks of arsenic toxicity. For these populations immediate action would be to provide with acceptable and affordable arsenic removal technologies at community and household levels, switching to deep wells and treatment of surface water.

Arsenic removal technologies While selecting any technology, it is necessary to consider long-term sustainability, operation/maintenance requirement, engagement of stakeholder and socio-economic status of the community (Bhattacharya et al. 2017). In addition to arsenic removal technologies developed by PCRWR in Pakistan, there are other similar technologies developed in India and Bangladesh and found effective. These can be modified and adopted in Pakistan. Ion exchange and membrane purification methods are hi-tech, require more costs and well-trained personnel to operate therefore these methods are not suitable for Pakistan (Luqman et al. 2016). Oxidation and precipitation methods are better options for Pakistan (Luqman et al. 2013).

Coagulation and flocculation This involves co-precipitation and subsequent coagulation. This process uses a locally available material such as alum, iron sulfate and iron chloride. This system has relatively low cost and simple in operation. However needs pre-oxidation and has low arsenic removal capability (Ahmed et al. 2005).

Jerry can system In this system, there is a adsorption followed by precipitation and sedimentation. It has a capacity of 50 L. Lifetime cost is 35,000 PKR and yields

73,000 L of water (Luqman et al. 2016). This system can be adopted in rural areas because of its cost-effectiveness.

Arsenic removal using bottom ash This uses coal ash, which is easily available from coal power plants and combustion units. This ash is combined with $Fe(OH)_3$. Life time cost is 8400 PKR and yields 73,000 L (Berkeley Arsenic Alleviation Group 2007). Initial and total costs of this system are low which make this system affordable for poor rural communities.

Solar distillation This method uses sunlight for evaporation of water and subsequent condensation. This is eco-friendly technology, and it can provide arsenic-free water at a very low cost (Pearce and Denkenberger 2006).

Gravity flows arsenic removal technology In this system, two plastic pitchers are placed over one another. There is a layer of silver coated sand on which different arsenic removal media can be used. Compared to other methods, it is more costly (156,000 PKR for 73,000 L), but mass production will bring the costs downs (Tahir 2004).

Kanchan arsenic filters This system uses iron nails, brick chips, fine and coarse sand, and gravel filters. It can filter water at a rate of 15–20 L/h (Ngai et al. 2007). The cost for 80,000 L filtered water is approximately 4500 PKR. This is also a cheaper option for arsenic removal at the household level.

Successful implementation of these technologies depends upon strong advocacy, sustained behavior change communication strategy, ownership by communities and government and availability of materials required for operations in the local markets. Most of the arsenic-exposed people are often rural poor, and may not be able to pay for the installation of such units. In this regard, the government can provide arsenic removal units at subsidized costs. Further production of these units and materials locally at a mass level should be encouraged to bring costs further down. Another important consideration for arsenic removal technologies is the disposal of exhausted/concentrated media. Improper disposal may lead to contamination of nearby water sources. Regeneration of adsorption media is a feasible option even in distant localities, where local trained persons can regenrate used adsorption media (Jiang et al. 2012).

Well switching Well switching is another strategy for the provision of arsenic-free water in short terms (Van Geen et al. 2002). The first option is to switch to safer wells. In this regard, the first step is to mark safe and unsafe wells with green and red colors respectively in a locality so that the community knows which well to use for drinking purpose. The second step is to promote well sharing within communities. An alternative option for areas where no safe wells are available is to dig deep wells. Studies have shown that deeper wells have low arsenic concentrations and are safer for use. However, while implementing well switching it is important to consider the following point;

- Considering local geological and hydrogeological features.
- Site approval by authorities.
- Regulated withdrawal as massive withdrawal may lead to movement of shallow water with high arsenic concentrations to the deep water and contaminate it.
- Continuous monitoring of new wells for arsenic and other parameters.

This needs active involvement of communities in the process of technical support from the government and/or NGOs. Effective behavior change communication strategy is essential to raise the awareness of communities about arsenic and changing behaviors for well sharing and switching (Balasubramanya et al. 2014; Bennear et al. 2013).

Water treatment options In areas where deep wells cannot be used as an alternate source of water, surface water such as river, ponds and harvested rain water may be treated to make fit for human use. In this regard, establishing water treatment plants with rapid sand filtration and chlorination facilities is needed in priority areas that are the worst arsenic hit. The public health engineering departments, local governments, and municipal corporations should work in coordination for the provision of treated surface water. For domestic use, the package -type slow sand filters are low-cost technology for medium size settlements in arsenic affected areas. They are efficient in the removal of turbidity and microorganisms; however, if the bacterial contamination is very high, there may be a need for chlorination (Ahmed et al. 2005).

3.3.2 Long Term

In areas of arsenic contamination long-term options to protect harmful exposure of population include; piped water supply, monitoring of water sources and data management and social mobilization and advocacy. The section that follows provides an account of these options.

Piped water supply There is a need to provide a piped water supply from arsenic free sources. In this regard, the government needs to allocate resources and develop infrastructure to ensure a continuous supply in the long run to the communities affected by arsenic contamination. This supply could be in the form of house connections, yard connections or standpost depending on the availability of the resources and affordability of the communities. Standposts have been found to be the most cost-effective options compared to new hand pumps and arsenic treatment units (Singh 2017a).

Micro-watershed management system Through this system, rainwater can be stored and treated to be provided to the communities to meet their household requirements. For this purpose, surveys should be conducted to identify appropriate locations for storage, based on rain patterns. Unequal distribution of rain throughout the year requires larger storage capacities to provide continuous water supply.

Soil and sub-surface soil investigation There is a need to continuously assess the quality parameters of soil as this affects the arsenic content in the water. This will provide data to be used in the further planning of well digging and agriculture.

Institutional arrangements A clear policy guideline and framework should be available to carry out arsenic mitigation activities. This should be supported with the capacity building of personnel at all the levels such as field-testing, geographical data analysis and interpretation and diagnosis and management of arsenicosis. There should be clear guidelines on a partnership with NGOs and the private sector to maximize the benefits of arsenic mitigation and reach the population most vulnerable to the health effects of arsenic.

Research and information There is a need to research to develop new effective and affordable technologies for arsenic removal and assess the feasibility in the communities. Further research is needed in the area of epidemiological studies to assess the burden of health effects due to arsenic exposure. There should be a system of regular data on surveillance of water sources and physical and chemical features of the soil. This data should be used for the planning and management of water sources.

Social mobilization and advocacy Active involvement of communities is central to the success of any intervention. There is a need to mobilize communities through effective and sustained behavior change communication strategies. Heath education regarding arsenic exposure and prevention through various media should be provided to the communities. Although these interventions are effective in preventing arsenic exposure in the short and long run but these need to be supplemented with continuous political commitment, adequate financial resources and institutional framework for arsenic mitigation. These interventions can be supplemented with the use of information and computer technology such as the development of arsenic-related database available to the researchers, policy makers and the general population (Singh 2017b).

4 Conclusion

Pakistan is an agrarian country with almost two-third of its population using groundwater for drinking and irrigation purposes. Various large and small-scale studies have reported widespread contamination of groundwater sources across its length. Millions of people are potentially exposed to unsafe levels of arsenic through drinking water and are at risk of developing adverse health effects due to continued exposure. Pakistan made some progress initially with the support of local and international partners. Nationwide surveys were carried out to assess the extent of contamination, which was followed by the development of the National Action Plan for Arsenic Mitigation. Arsenic mitigation efforts failed in Pakistan, and a number of factors played in the failure such as lack of political commitment and ownership by local leadership, dependence on foreign aid, lack of sustainability of mitigation efforts. Continued exposure of the population to unsafe levels of arsenic may cost Pakistan huge economic and health burden in the future. Low-cost arsenic removal technologies with the availability of raw material in the local markets, well switching and treatment of surface water can be considered in short-term. While piped water supply, micro-water shade management system, soil investigations research and information systems are an option in long-term planning for arsenic mitigation. These interventions should be paralleled with continuous social mobilization and advocacy and political commitment.

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