

MAGNITUDE OF INORGANIC ARSENIC CONTAMINATION IN GROUNDWATERS OF MIDDLE GANGETIC BASIN OF UTTAR PRADESH AND BIHAR, INDIA

**RAMANATHAN, AL. and
KUMAR, Manoj**



Contact No: 91-11-26704314
alr0400@mail.jnu.ac.in

**School of Environmental Sciences
Jawaharlal Nehru University
New Delhi-110067**



Introduction

- Geogenic or non-point source arsenic (As) contamination of groundwater is widespread in South-East Asian countries.
- It threatens to human health and poses a challenge to water supply in the affected areas.
- The level of the As contamination in Bengal basin (the lower region of Ganga-Brahmapura-Meghna) has been well documented and known as the most severely affected region of the world (Nickson et al. 1998; Burgess et al. 2010).
- A survey was done in central Gangetic basin (CGB) and found ~ 59 % of the hand pumps of the villages (20–40 m depth) were reported with (As > 50 µg/L) (Chakraborti et al., 2003).

Cont...

- It is noteworthy that mobility, (phyto)toxicity, potential risk, retention and bioavailability of As in the environment depend on the form in which they enter and the final form in which they are present (Sanz et al., 2007)
- Determining the total As concentration alone is insufficient for many environmental exposure scenarios
- Nonetheless, As speciation analysis is a powerful tool that can generate accurate assessments of environmental impact and risk to human health (Sanz et al., 2007; Rahman et al., 2009; Vassileva et al., 2001)
- Arsenic remedial action strongly depends on arsenic speciation, the presence of other possible contaminants, and on the general water composition (Katsoyiannis et al., 2007)
- The presence of As in the central Gangetic Basin is now well documented by various studies that have examined water, agricultural soil and subsurface sediments (Srivastava et al., 2013; Kumar et al., 2016; Saha et al., 2013; Kumar et al., 2012). But all these studies are limited to total estimations, except very few studies and those are limited to groundwater only.

Health problem due to consumption of elevated Arsenic through drinking and food

(a) Diffuse melanosis,

(b) Spotted melanosis,

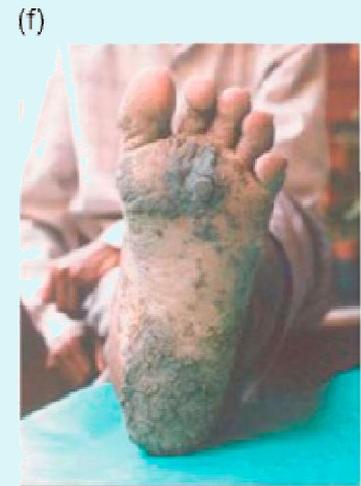
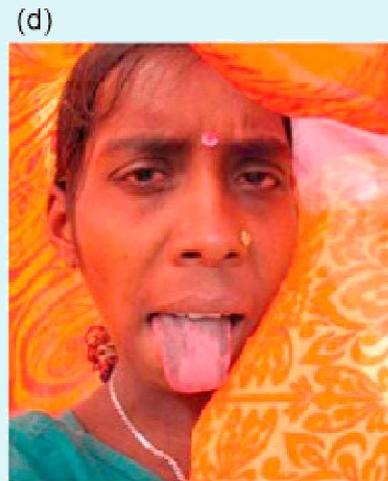
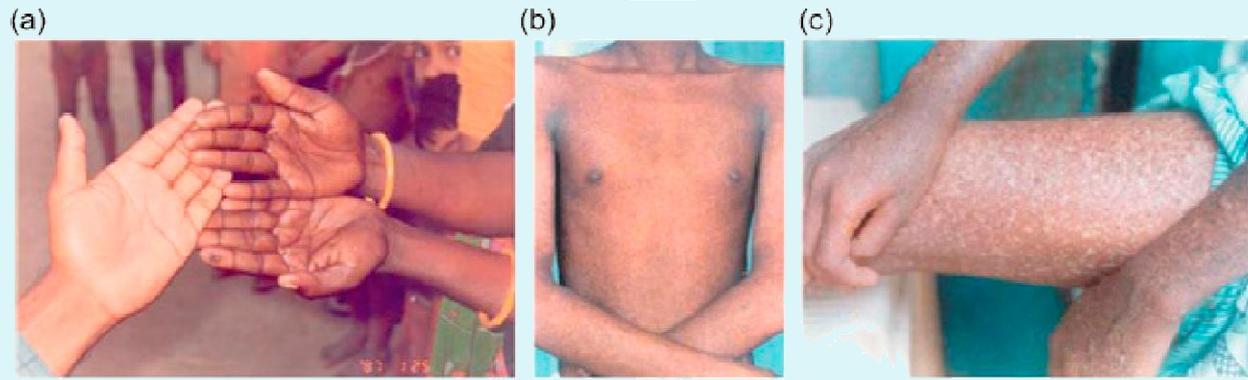
(c) Leucomelanosis,

(d) Tongue melanosis,

(e) Keratosis on palm,

(f) Spotted keratosis on sole

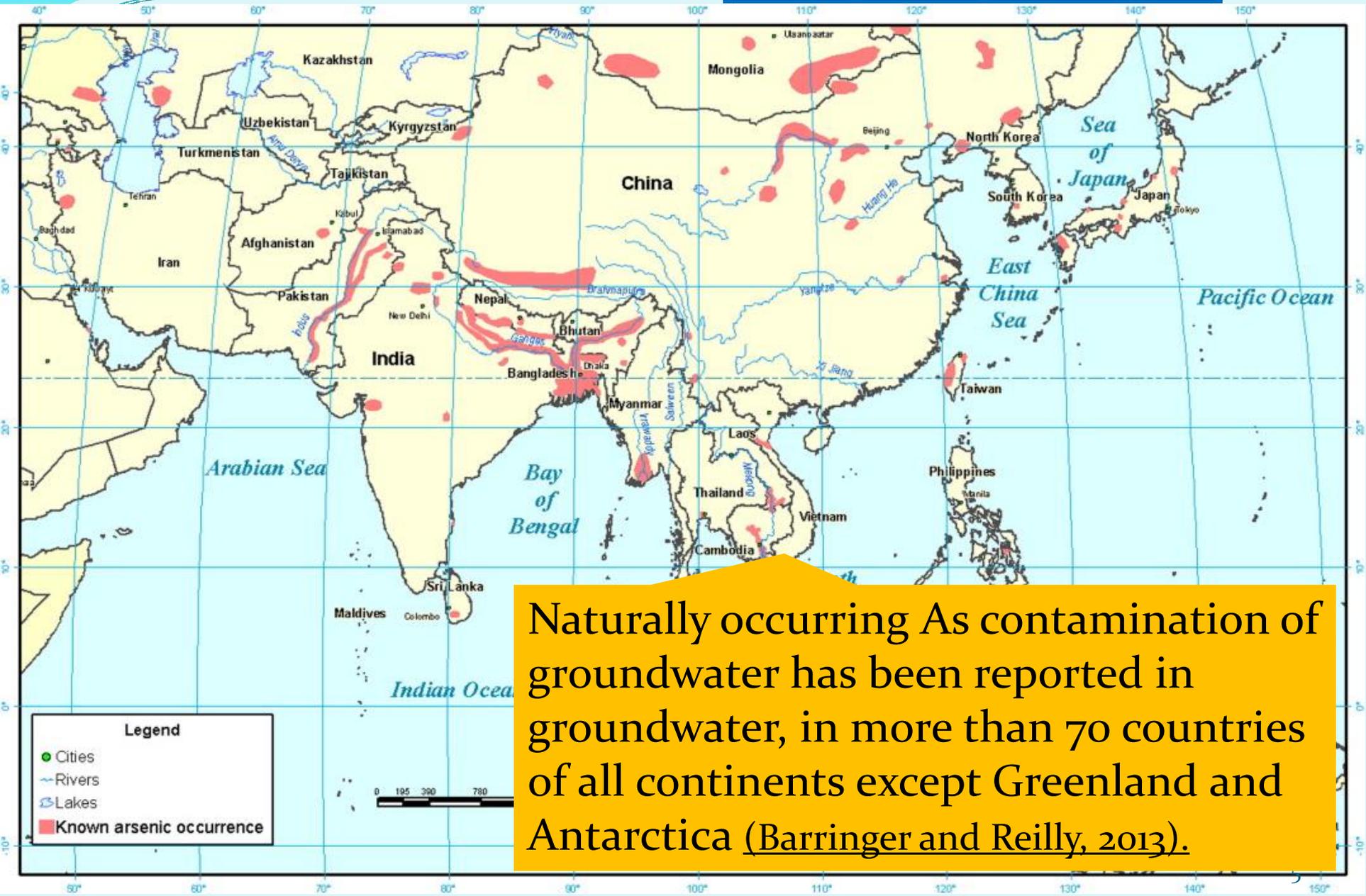
(g) Dorsal keratosis.



Images: Different skin symptoms due to arsenic toxicity

Chakraborti et al., 2016

Distribution of arsenic contamination of groundwater in South and South-East Asia (Brammer and Ravenscroft, 2009)



Naturally occurring As contamination of groundwater has been reported in more than 70 countries of all continents except Greenland and Antarctica (Barringer and Reilly, 2013).

List of arsenic endemic states in India

Area of endemic states = 529674 sq km (approx)

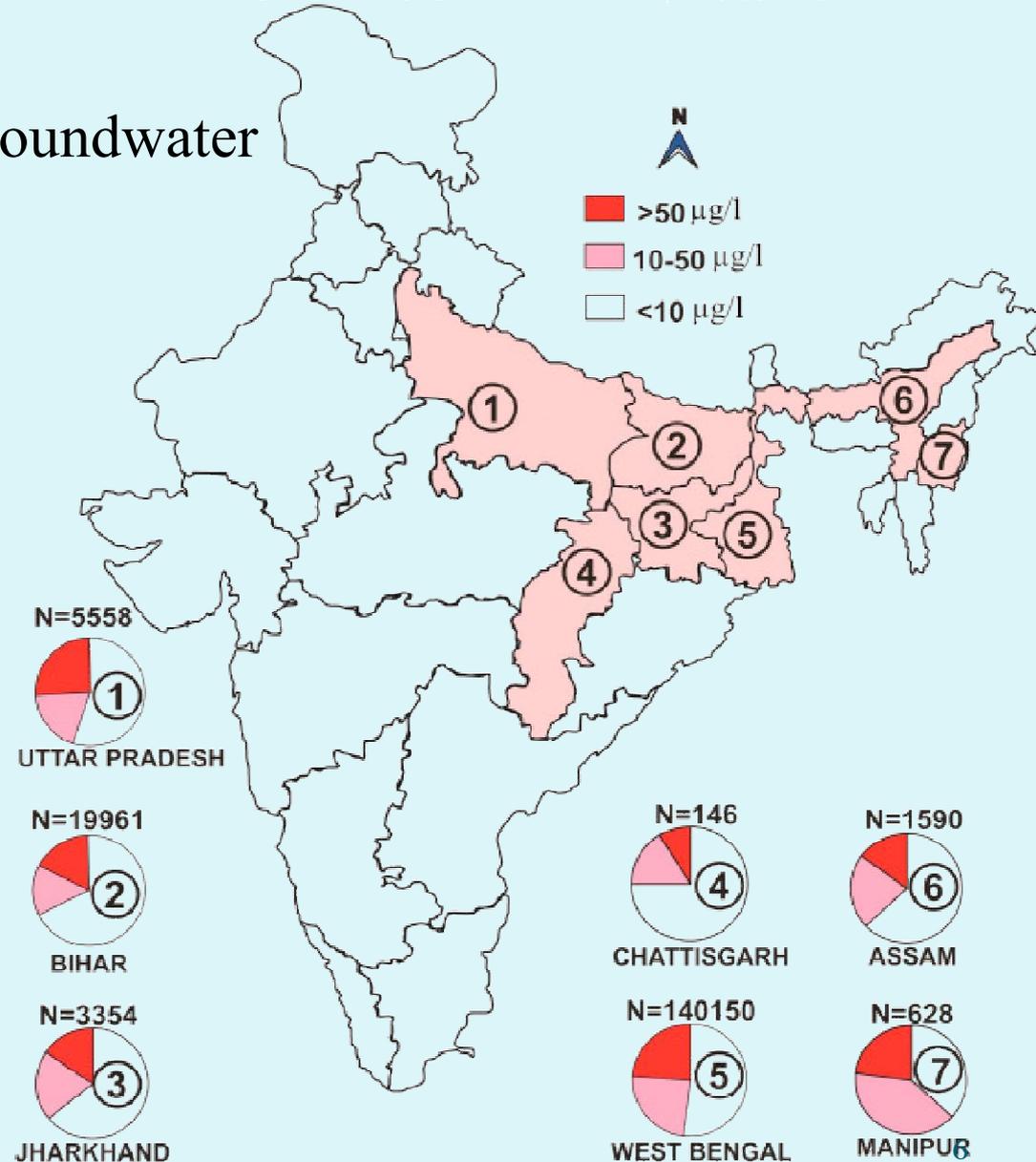
Population of endemic states = 359 million (approx)

Potentially exposed population = 50 million (approx)

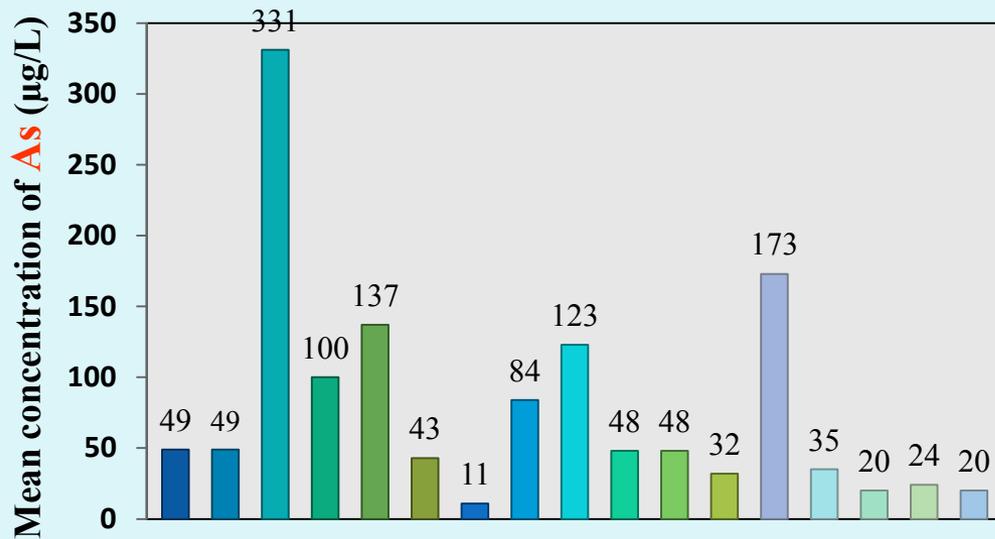
India is one of the worst As Groundwater affected country in the world
(Chakraborty et al., 2016)

In 6 states of the (GB-Plain),
70.4 million people are potentially
at risk from groundwater As toxicity
($>10 \mu\text{g/L}$)

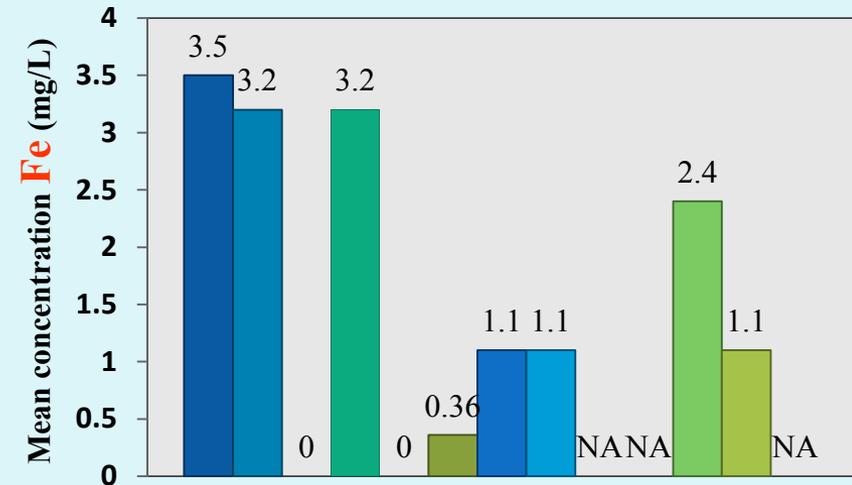
- West Bengal
- Bihar
- Uttar Pradesh
- Assam
- Manipur
- Jharkhand
- Chhattisgarh



As and Fe in central Gangetic basin



- Ballia, Uttar Pradesh (Chandrasekharam et al., 2007)
- Ballia, Uttar Pradesh (Chauhan et al., 2009)
- Ballia and Ghazipur, Uttar Pradesh (Srivastava and Sharma 2013)
- Ballia Uttar Pradesh Shah (2008)
- Ballia, Uttar Pradesh and Patna, Bihar (Chandra et al., 2010)
- Ghazipur, Uttar Pradesh (Kumar et al., 2010b)
- Varanasi, Uttar Pradesh Raju (2012)
- Bhojpur, Bihar (Saha et al., 2009)
- Bhojpur, Bihar (Saha et al., 2011)
- Bhagalpur, Bihar (Kumar et al., 2010a)
- Samastipur, Bihar (Saha and Shukla, 2014)
- Samastipur, Bihar (Kumar et al., 2016)
- Bhojpur, Bihar (Chakraborti et al., 2016)



- Ballia, U.P. (Chandrasekharam et al., 2007)
- Ballia, U.P. (Chauhan et al., 2009)
- Ballia & Ghazipur (Srivastava and Sharma 2013)
- Ballia U.P. (Shah 2008)
- Ballia & Patna, Bihar (Chandra et al., 2010)
- Ghazipur, U.P. (Kumar et al., 2010 (b))
- Varanasi, U.P. (Raju, 2012)
- Bhojpur, Bihar (Saha et al., 2009)
- Bhojpur, Bihar (Saha et al., 2011)
- Bhojpur, Bihar (Saha et al., 2011)
- Bhagalpur, Bihar (Kumar et al., 2010 (a))
- Samastipur, Bihar (Saha and Shukla, 2014)
- Samastipur, Bihar (Kumar et al., 2016)

Most acceptable mechanism for As mobilization in CGB

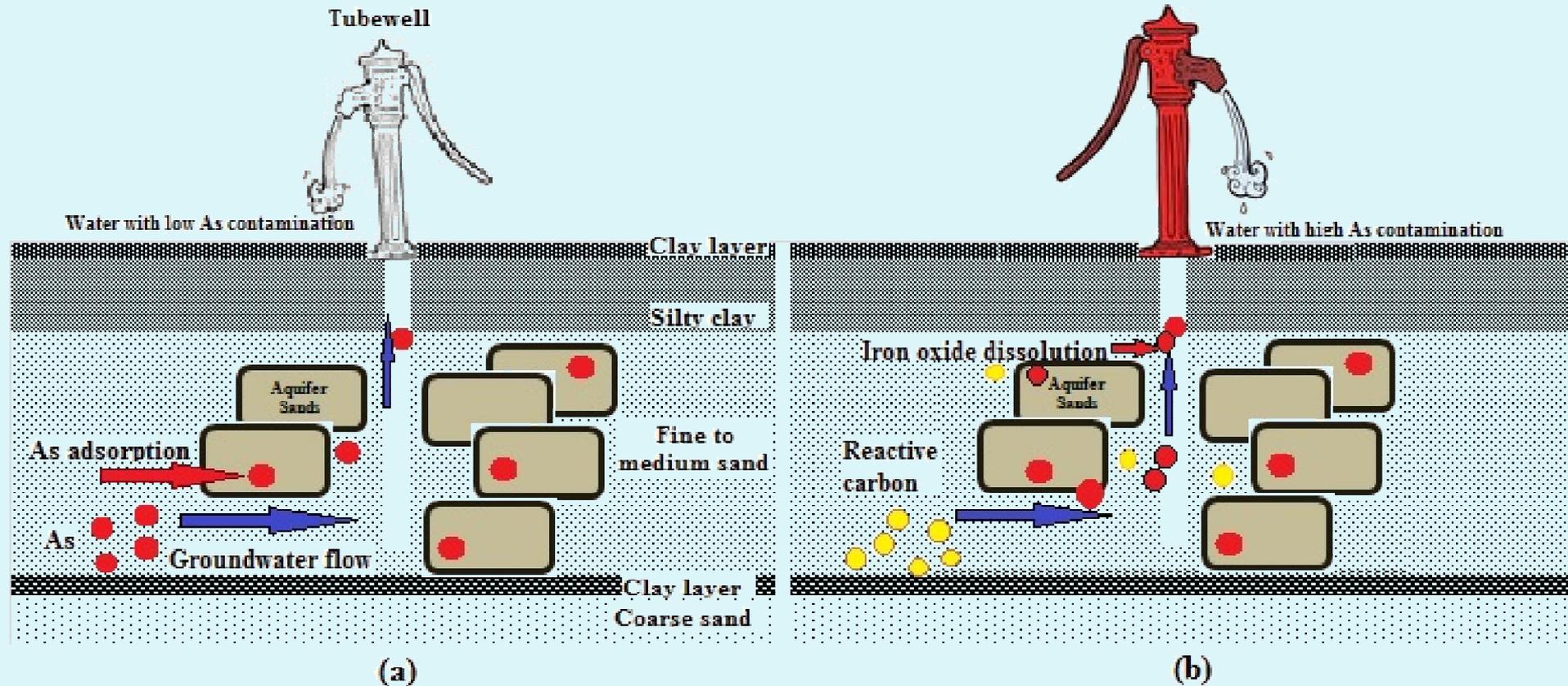
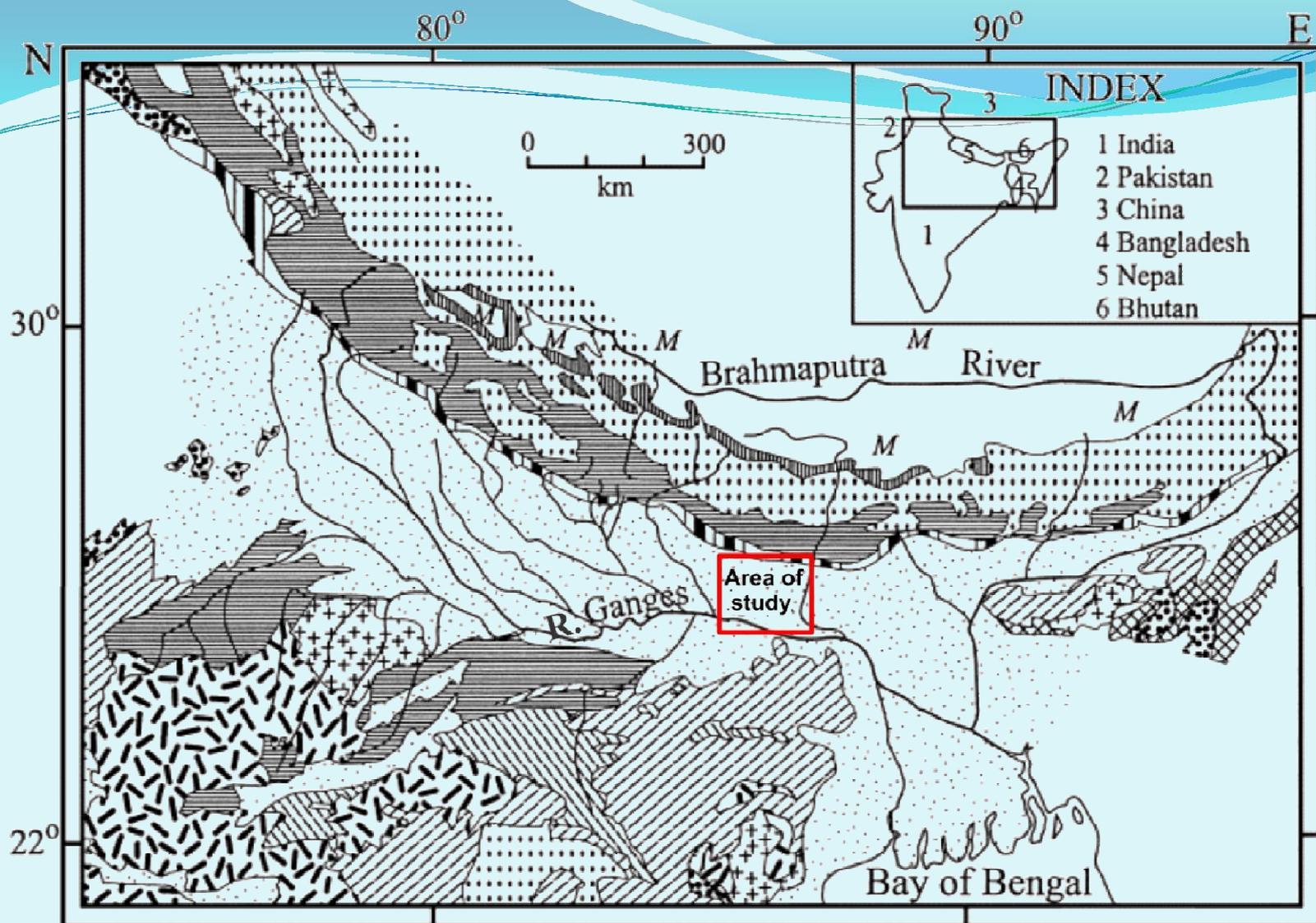


Fig. Groundwater flow could play a role by supplying As directly or indirectly by supplying reactive organic carbon that triggers reductive dissolution of iron oxides (modified from van Geen, 2011)

Study Area

- The area undertaken for the study encompasses parts of the flood plain deposits of the Ganges and its tributaries in the states of Bihar.
- The area extends from Bank of the River Ganges near district Patna in the south to the Terai plains in the north, located in the foothills or piedmont of the Himalayas near the Nepal border.
- North of the Ganges (study area), the alluvial deposits are much more pronounced.
- Tectonically, the study area may be regarded as a part of the Himalayan foreland basin.



- | | | |
|---|---|--|
| Quaternary alluvium | Mesozoic/L. Tertiary mafic effusives - Deccan, Rajmahal Traps | Precambrian metamorphics - schists, gneisses, quartzites, limestones |
| Miocene sandstones - Siwaliks | Mesozoic sandstone, shale, limestone | Precambrian metamorphics and younger felsic intrusions |
| Oligocene sandstones, shales - Barisals | Paleozoic granites, gneisses, charnockites | Cambrian sandstones, phyllites, dolomites |
| Eocene sandstones, limestones, shales | Paleozoic-Mesozoic sandstone, shale, limestone | Archean granites, gneisses |

Sample locations

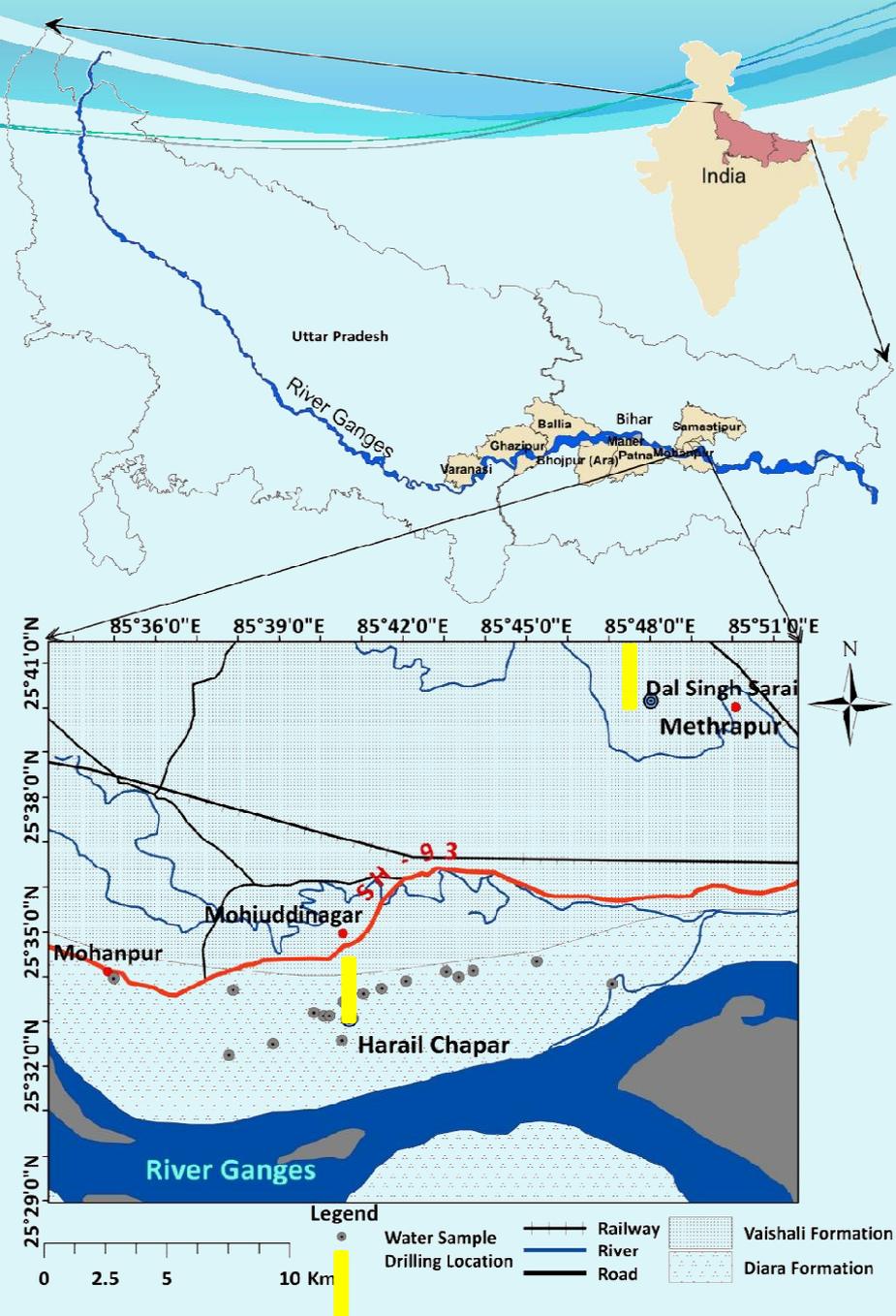


Figure. Study area map: India, Uttar Pradesh and Bihar and water sample and drilling locations.

Material and methods

Sample collection

- Two sites, specifically a new alluvium region (also known as the Diara formation) and older alluvium deposition (also known as the Vaishali formation) were selected to fulfill this objective
- Water samples were collected from tubewells, aquifer sediment was collected using hand-flapper method and agricultural soil samples (0–15 cm depth) were collected from 11 different locations

Digestion procedure for the total elemental extraction

- A microwave digester (CEM, MARS 6) consisting of 42 digester vessels was employed to digest all soil and sediment samples
- **Extraction procedure of soil and sediment samples**
- Microwave-assisted extraction technique along with 1M Ortho-phosphoric acid were used to extract the As species from agricultural soil and subsurface sediment samples.
- **Analysis:** ICP-MS for As and other elements, (IC) coupled with ICP-MS for speciated As, Scanning electron microscopy (SEM) and X-Ray diffraction (XRD analysis

Table

Arsenic species present in groundwater (results expressed in $\mu\text{g/L}$ and are based on peak area calculation)

S. No.	Depth (m)	pH	pe	As ^{III} ($\mu\text{g/L}$)	As ^{III} (%)	As ^V ($\mu\text{g/L}$)	As ^V (%)	As (t) ($\mu\text{g/L}$)	As ^{III} /As ^V
1.	30.5	7.4	-2.4	17.0	57.8	12.4	42.2	29.4	1.4
2.	24.4	7.8	-4.9	78.0	95.8	3.4	4.2	81.4	23.0
3.	15.2	7.2	-2.5	15.8	44.9	19.4	55.1	35.1	0.8
4.	21.3	7.6	-3.0	5.1	84.6	0.9	15.4	60.0	5.5
5.	18.3	7.7	-1.9	11.6	91.8	1.0	8.2	12.6	11.2
6.	30.5	7.5	-1.6	1.1	76.7	0.3	23.3	1.5	3.3
7.	27.4	7.4	-1.4	2.3	88.1	0.3	11.9	2.6	7.4
8.	54.9	7.4	-1.5	0.5	8.1	5.1	91.9	5.6	0.1
9.	36.6	7.4	-2.7	10.0	67.6	4.8	32.4	14.8	2.1
10.	18.3	7.4	-2.0	13.6	70.9	5.6	29.1	19.2	2.4
11.	19.8	7.4	-1.9	0.8	57.7	0.6	42.3	1.4	1.4
12.	36.6	7.4	-0.5	2.5	87.5	0.4	12.5	2.8	7.0
13.	24.4	7.1	-2.5	7.2	42.9	9.5	57.1	16.7	0.8
14.	36.6	7.2	-4.2	4.1	58.9	2.9	41.1	7.0	1.4
15.	21.3	7.8	-3.4	1.3	94.6	0.1	5.4	1.3	17.4
16.	19.8	7.3	-3.7	0.4	14.8	2.3	85.2	2.7	0.2
17.	27.4	7.6	-0.0	2.7	11.6	20.8	88.4	23.5	0.1
18.	21.3	7.5	-0.3	83.4	79.6	21.3	20.4	104.7	3.9
19.	30.5	7.5	-0.2	10.9	77.8	3.1	22.2	14.0	3.5
Mean	27.1	7.5	-2.1	14.1	63.8	6.0	36.2	20.1	1.8

Figure Inorganic As species in groundwater samples.

Approximately 73% (14 out of 19) of the samples shows As^{III} as the dominant species while 27% (5 out of 19) reveals As^V is the dominant species

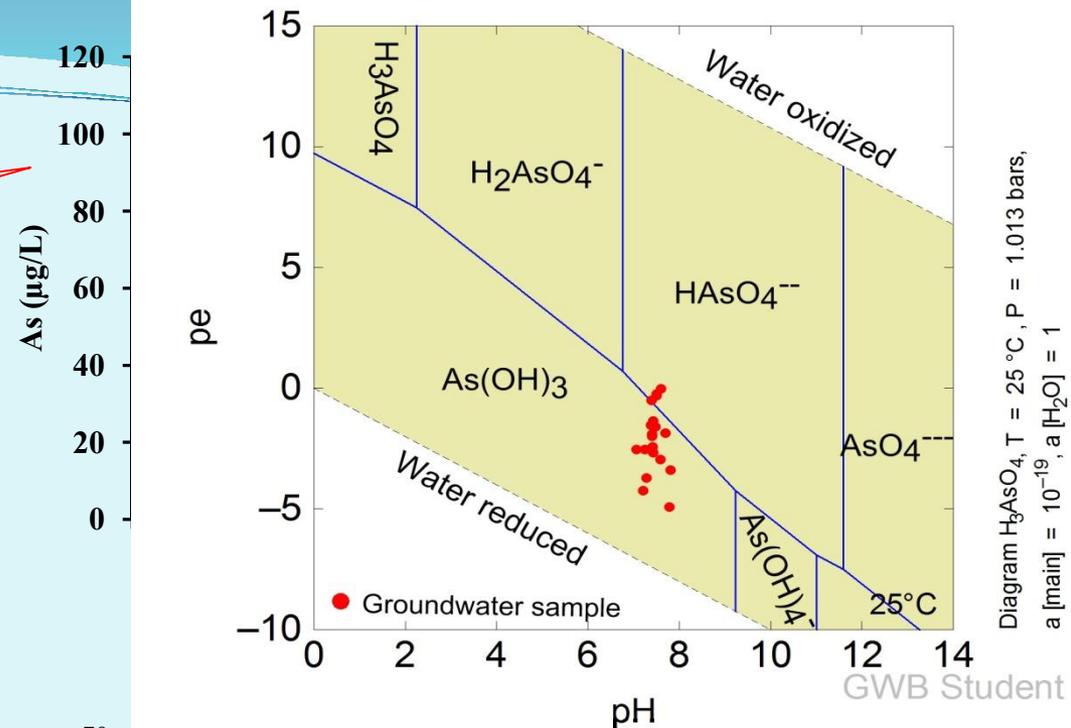
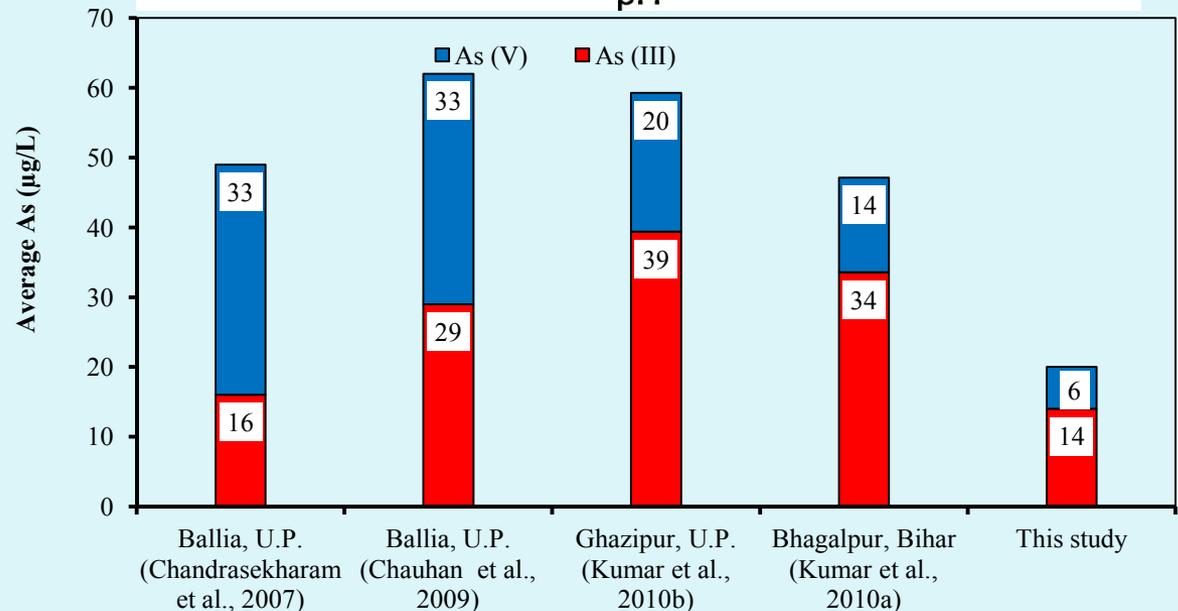


Figure Comparison of mean concentration of inorganic As species in groundwater from the middle Gangetic plain of Uttar Pradesh and Bihar of India.



Arsenic species present in groundwater

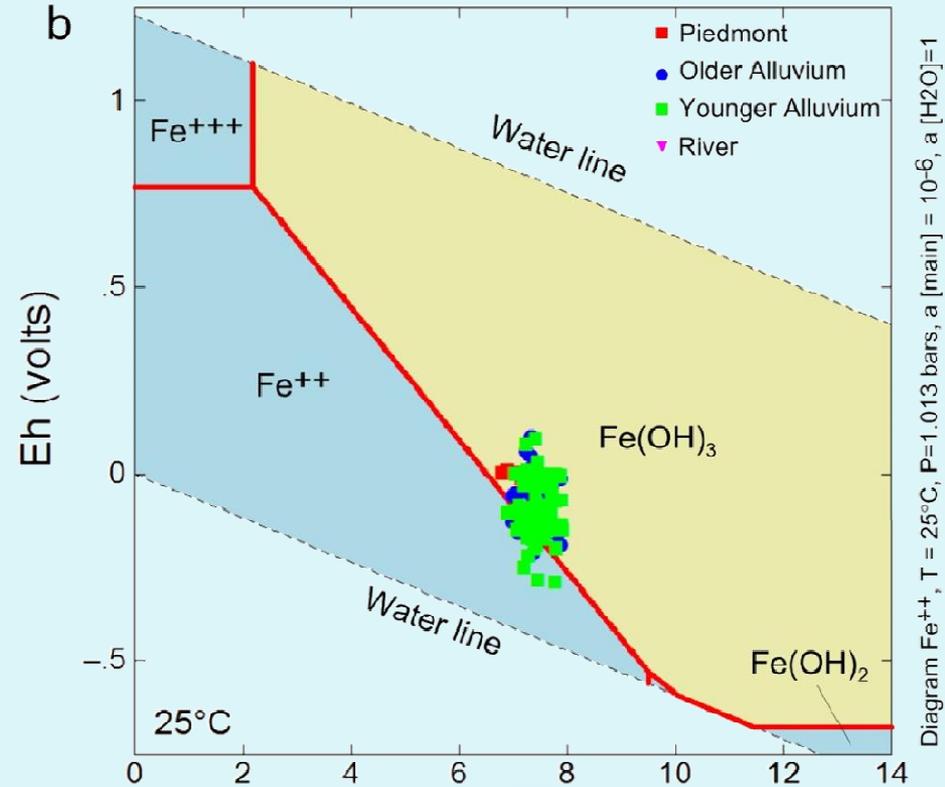
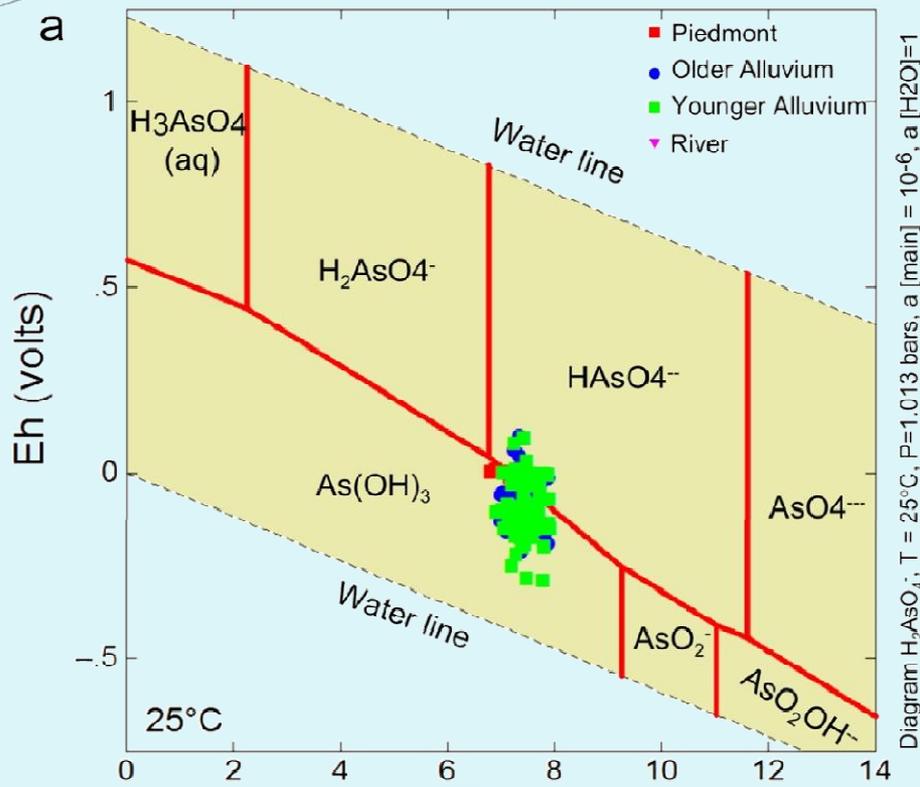


Figure 10.10 Eh-pH diagrams for As and Fe species in groundwater

More than 73% of the samples fall under the $As(OH)_3$ section (AsIII) while the remaining falls under $HAsO_4^-$ (i.e. As V).

More than 30% of the samples fall under the $Fe(OH)_3$ section Fe(III) while the remaining falls under Fe (II)

Total and inorganic As in agricultural soils

Table

Concentration of inorganic As species with their respective extractable amount (%) in agricultural soil ($n = 11$) (results expressed in $\mu\text{g}/\text{kg}$ and are based on peak area calculation)

S.ID.	Latitude	Longitude	As ^{III}	As ^V	As (ext)	As (tot)	% extractable
1	25.572	85.668	Nd	1050	1050	3528	29.8
2	25.570	85.638	Nd	2413	2413	10600	22.8
3	25.554	85.669	39	6835	6874	12940	53.1
4	25.561	85.718	20	5913	5933	10830	54.8
5	25.563	85.723	Nd	3420	3420	9527	35.9
6	25.563	85.713	31	4257	4289	9525	45.0
7	25.539	85.642	40	5404	5444	9742	55.9
8	25.579	85.677	Nd	4403	4403	12410	35.5
9	25.545	85.662	0.1	4513	4513	11630	38.8
10	25.608	85.666	12	3766	3779	13490	28.0
11	25.549	85.670	Nd	6152	6152	14690	41.9

As (ext) = total sum of arsenic species extracted, As (tot) = concentration of arsenic by using aqua regia, Nd = not detected.

The concentration of As^{III} and As^V varied from not detectable – 40.1 and 1050–6835 $\mu\text{g}/\text{kg}$, respectively.

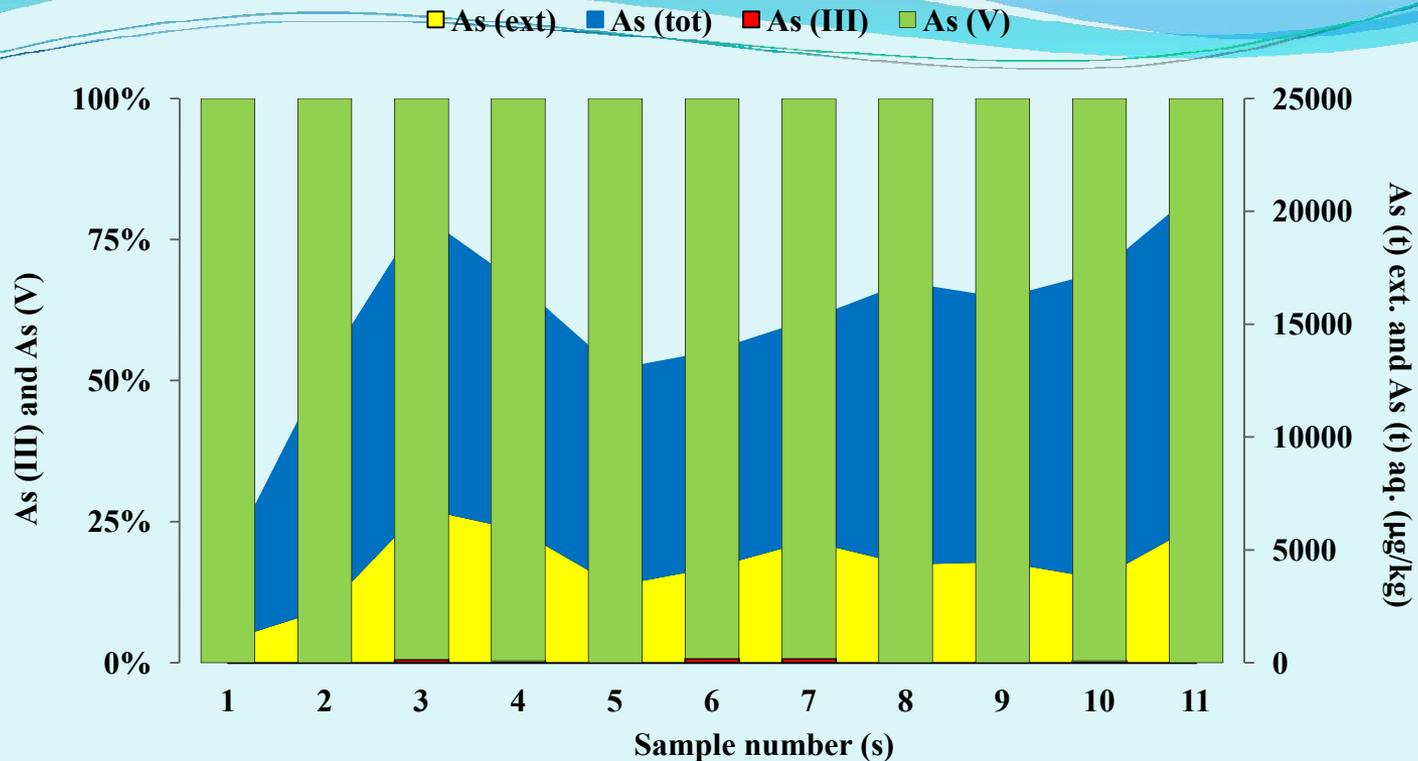


Figure Inorganic As species in agricultural soil samples. Values at secondary axis are the sum of As (ext) and As (tot) ($\mu\text{g}/\text{kg}$)

The total concentration of As observed in agricultural soils ranged from 3527–14690 $\mu\text{g}/\text{kg}$

A high discrepancy was observed in the concentration of As in agricultural soils in the study area.

Sediment color and As severity in the sediment

A modified Munsell Color Chart which was reduced into a four color tool (*see* Figure) on the basis of comparative analysis of 2240 sediment samples was served to categorize the samples into four colors

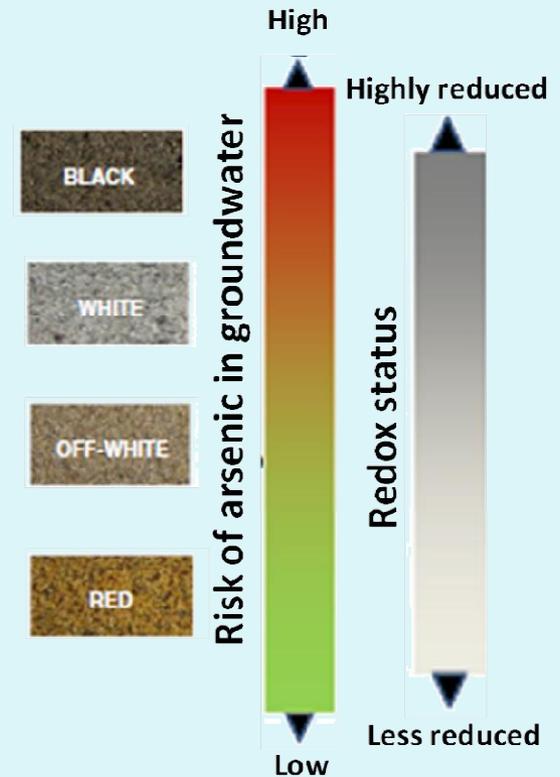


Figure Four color sands with corresponding risks of As concentration in groundwater under varying redox status adopted from (Hossain et al., 2014).

Table

Concentration of inorganic As species in subsurface sediments

Depth (m)	pH	As ^{III}	As ^V	As (ext)	As (tot)	(%) ext.	Sand (%)	Silt (%)	Clay (%)	EOC (%)	OM (%)
Methrapur											
1.5	8.3	Nd	4328.0	4328.0	16007.1	27.0	27.7	63.8	8.6	0.8	2.2
4.5	8.2	Nd	3480.0	3480.0	16450.1	21.2	29.7	66.8	3.6	0.6	1.8
9.1	8.3	Nd	3203.7	3203.7	9876	32.4	26.5	63.9	9.6	0.5	1.6
21.3	8.4	264.2	6288.6	6552.8	20056.7	32.7	89.8	9.4	0.7	0.5	1.3
36.5	8.3	117.8	2328	2445.8	11291	21.7	39.0	57.4	3.7	0.5	1.6
42.6	8.2	Nd	3332.5	3332.5	9119	36.5	56.2	41.1	2.7	0.6	1.8
Mean	8.3	191.0	3826.8	3890.5	13800.0	28.6	44.8	50.4	4.8	0.6	1.7
Min	8.2	Nd	2328.0	2445.8	9119.0	21.2	26.5	9.4	0.7	0.5	1.3
Max	8.4	264.2	6288.6	6552.8	20056.7	36.5	89.8	66.8	9.6	0.8	2.2
Harail Chapar											
1.5	8.3	Nd	8820.6	8820.6	15663.4	56.3	31.4	59.2	9.4	0.5	1.3
9.1	7.8	368.5	6921.8	7290.2	19681.1	37.0	21.7	66.3	12	0.9	2.7
18.3	8.2	22.8	7224.8	7247.6	13654.7	53.1	35.1	56.5	8.5	0.5	1.3
24.4	8.3	Nd	2211.3	2211.3	4788.9	46.2	47.7	51.8	8.1	0.8	2.2
33.5	8.0	Nd	1134.9	1157.7	5515.6	21.0	82.5	15.3	2.1	0.5	1.6
Mean	8.1	195.7	5267.2	5345.5	11860.7	42.7	43.7	49.8	8.0	0.6	1.8
Min	7.8	Nd	1134.9	1157.7	4788.9	21.0	21.7	15.3	2.1	0.5	1.3
Max	8.3	368.5	8820.6	8820.6	19681.1	56.3	82.5	66.3	12.0	0.9	2.7

Figure Litholog prepared from core samples along with their corresponding photographic view of sediment (a), Graphs shows inorganic As species in subsurface sediment samples of Methrapur, Dal Singh Sarai (b).

Larger amounts of As(t) along with As^V and As^{III} in Methrapur, Dal Sing Sarai were reported at a depth of 21.3 m bgl with concentrations of 20057 and 6289 and 264 µg/kg, respectively

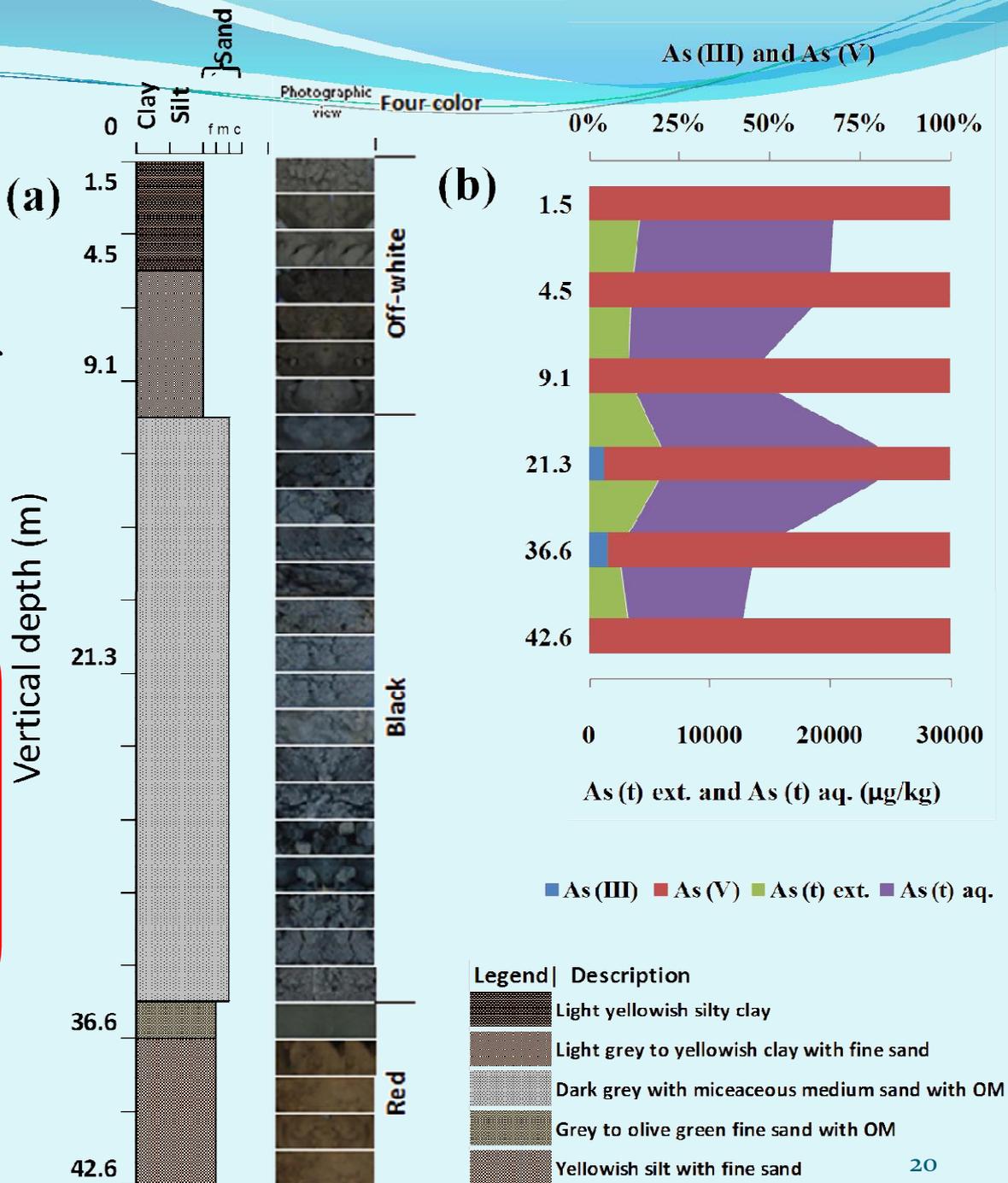
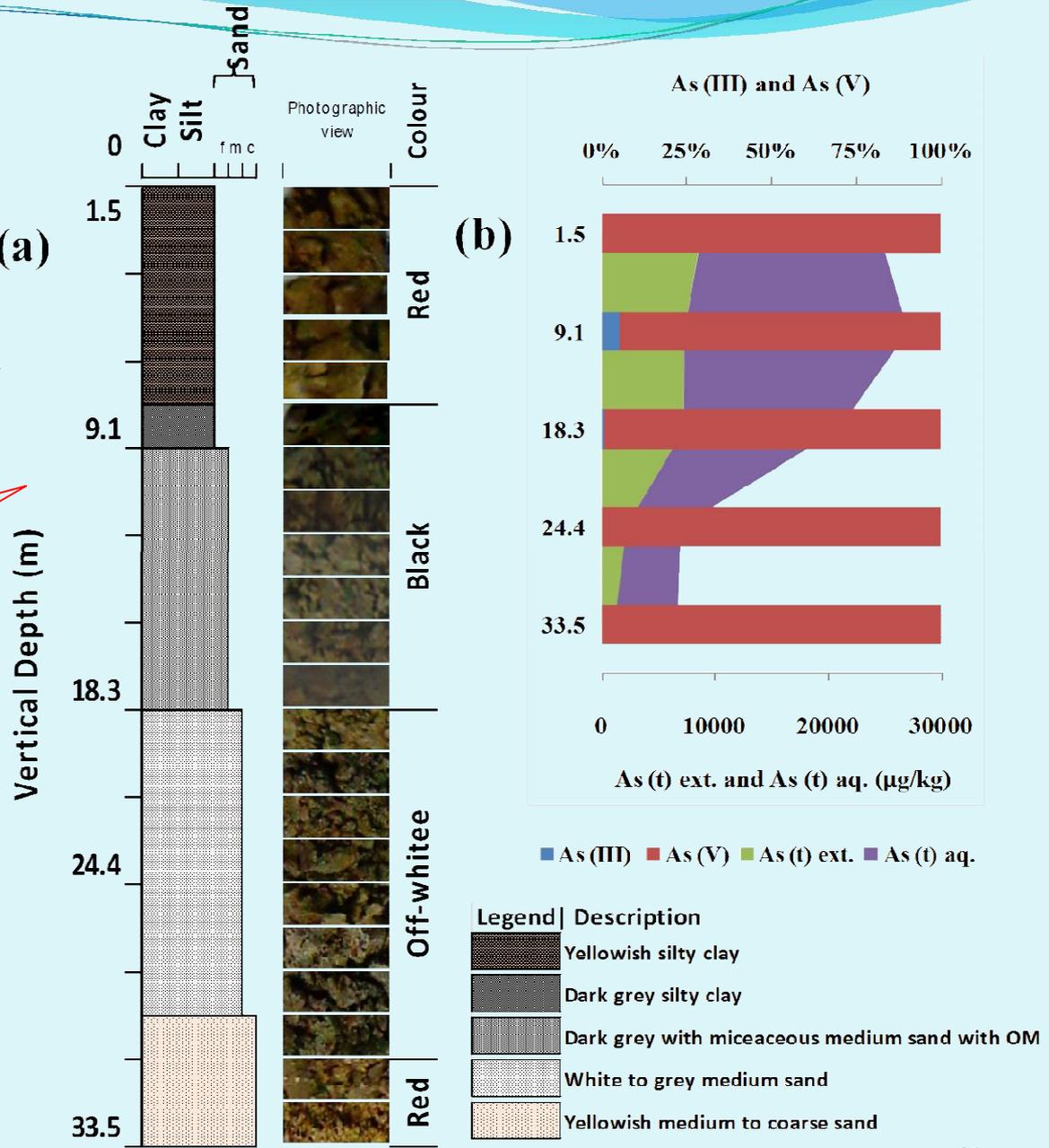
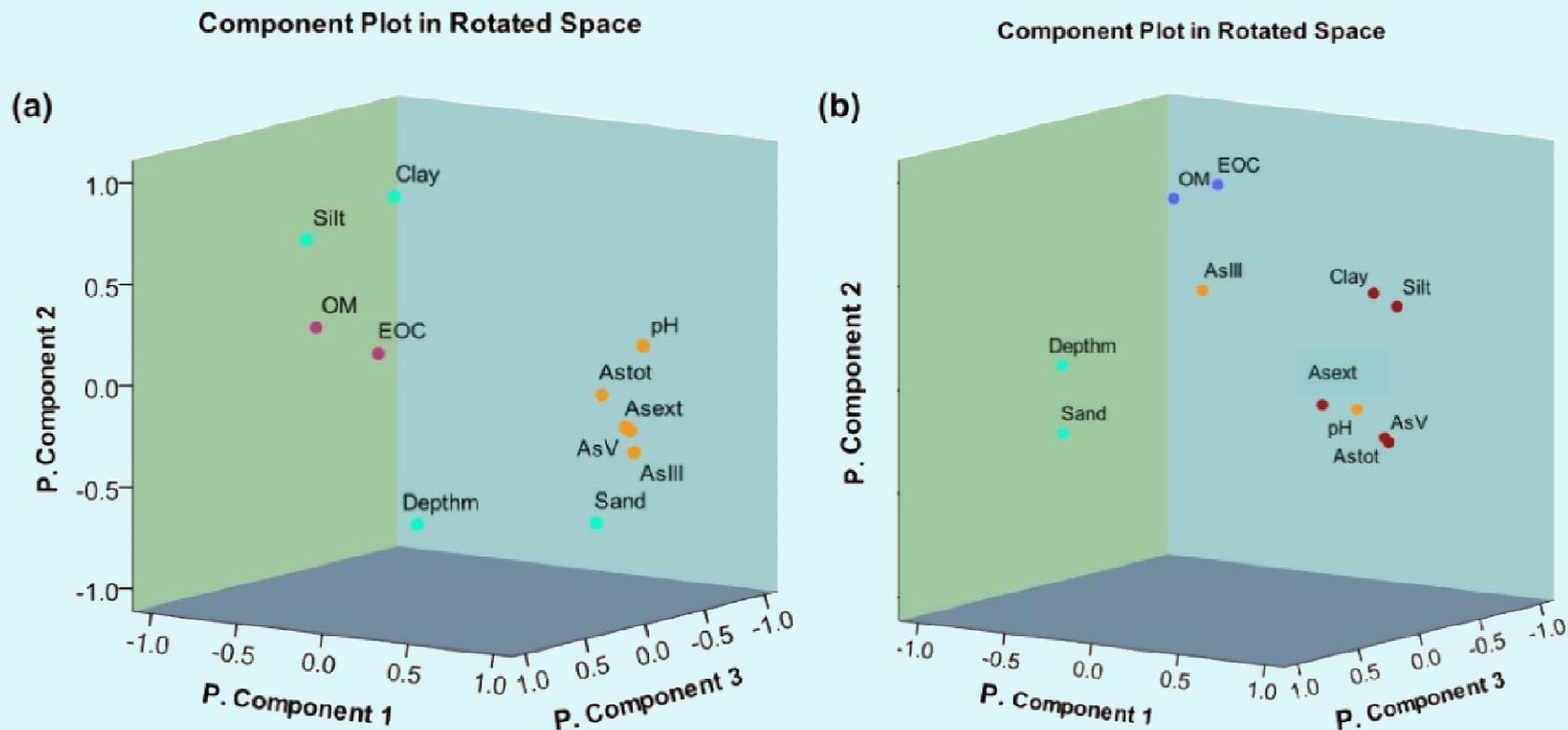


Fig. 9. Litholog prepared from core Samples along with their corresponding photographic view of sediment (a). Graph shows inorganic As species in subsurface sediment samples of Harail Chapar, Mohiuddin Nagar (b).

In Harail Chapar, larger amounts of As(t) and As^{III} were observed at a depth of 9.1 mbgl having a range of 19681 and 368 µg/kg, respectively. Meanwhile the highest As^V concentration was observed at a depth of 1.5 mbgl and having a value of 8821 µg/kg



Statistical analysis for the subsurface arsenic mobilization



Multivariate analysis, principal component in rotated space (a) Methrapur, (b) Harail Chapar

Mineralogy of the aquifer sediment

Methrapur, Dalsingh Sarai

Figure Comparative XRD pattern of three different depths

- (a) upper light yellowish silty clay (off white) at 1.5m vertical depth
- (b) dark grey with micaceous medium sand with OM (black) at 21.3 m vertical depth and
- (c) lower yellow silt with fine sand (red) sediments at 42.6 m vertical

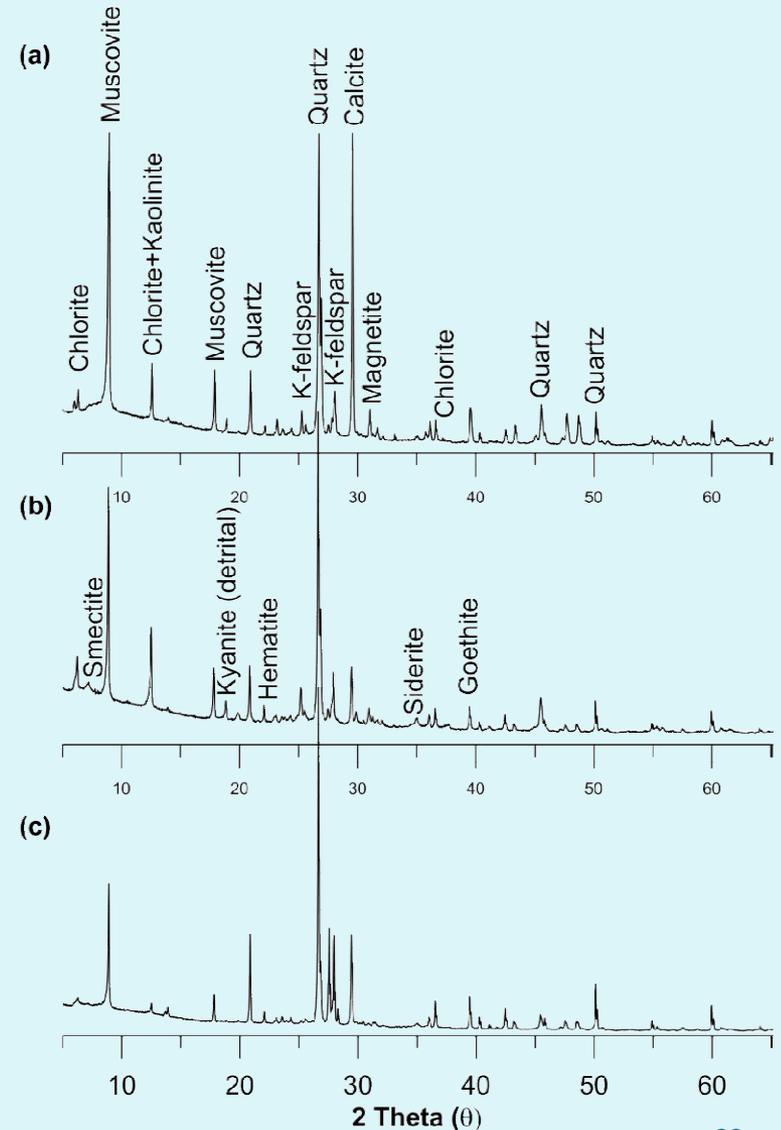


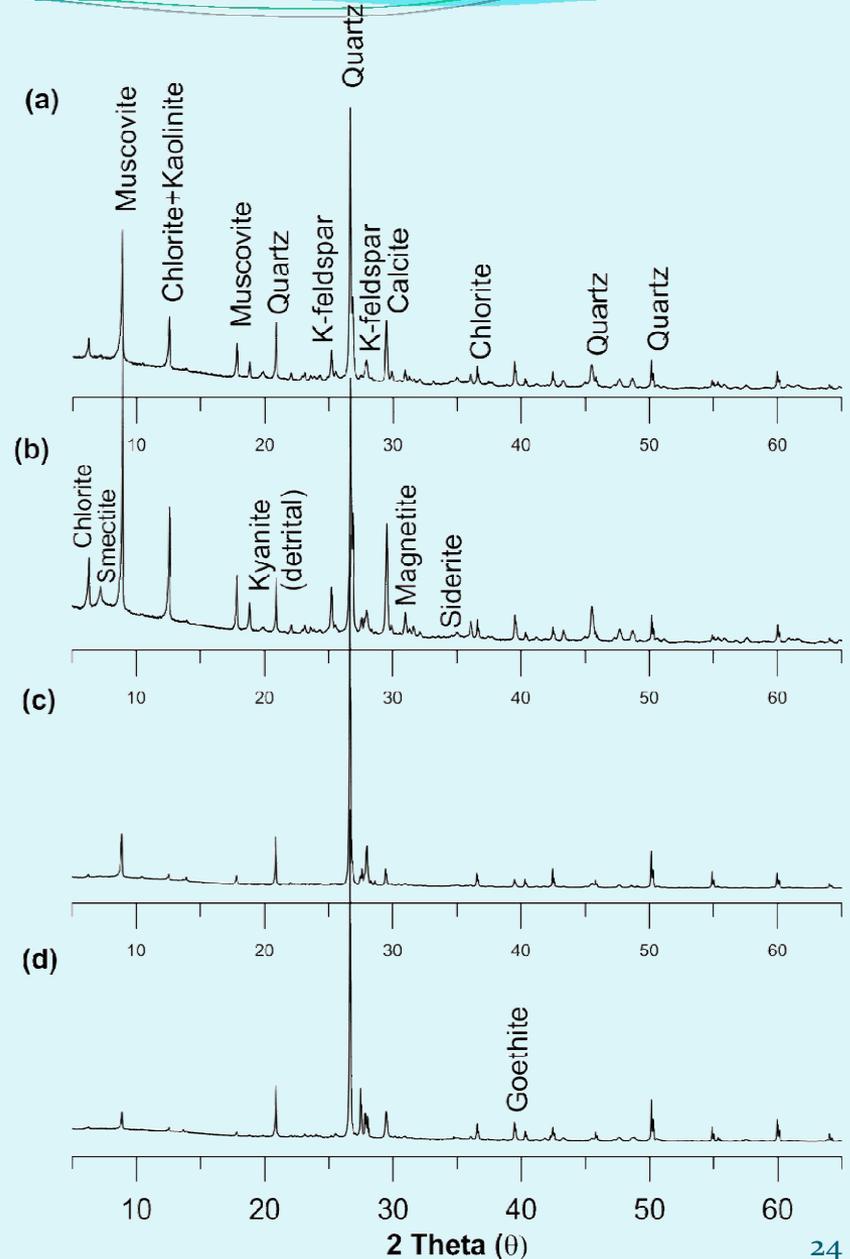
Figure Comparative XRD pattern of four different depth

(a) upper light yellowish silty clay (red) at 1.5m vertical depth

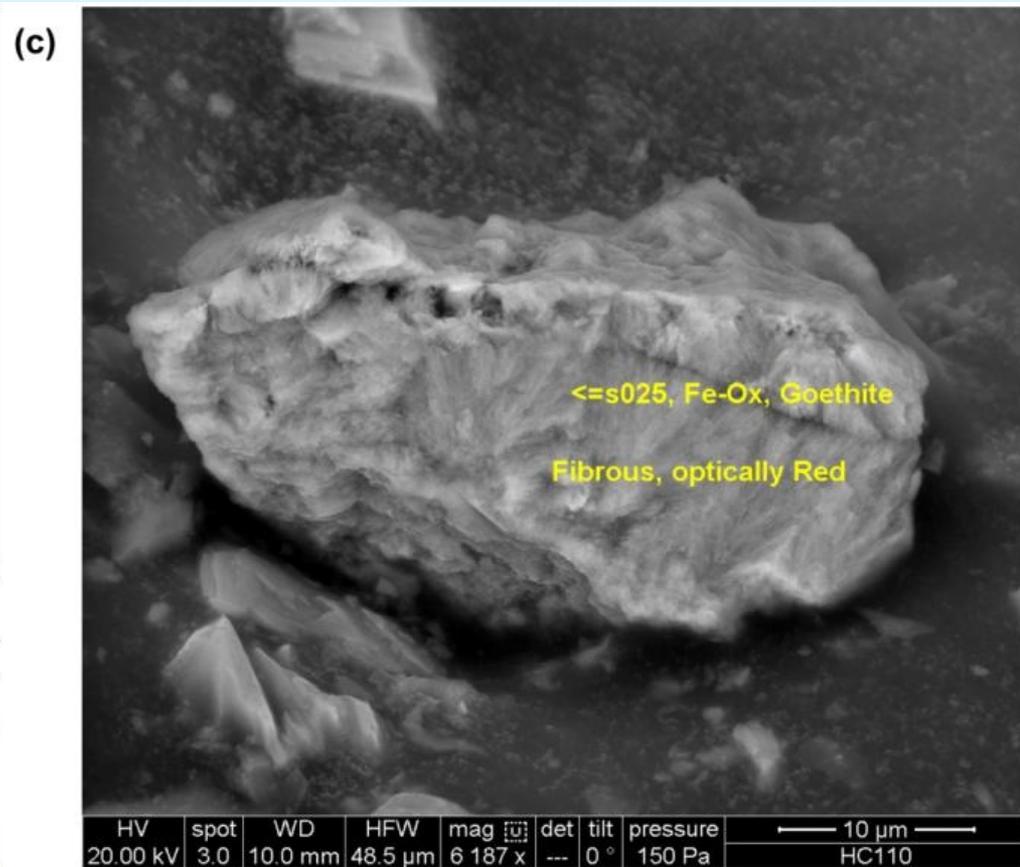
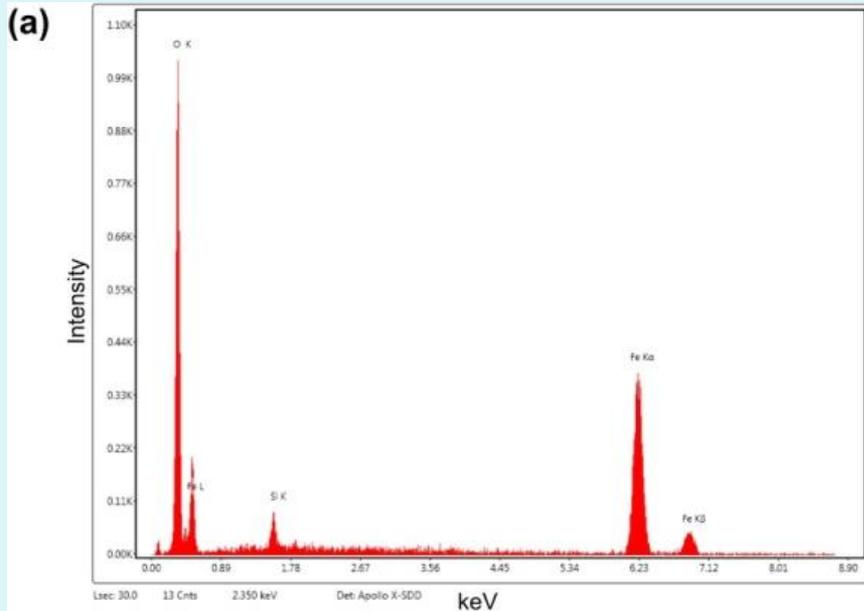
(b) Middle dark grey silty clay (black) at 9.1m vertical depth

(c) middle white to grey sand (off-white) at 24.4 m vertical depth and

(d) lower yellowish sand (red) sediments at 33.5m vertical depth



Scanning electron microscopy of subsurface sediment



(b)

Element	Weight (%)	Atomic (%)	Error (%)	Net Int.	K Ratio
C K	2.1	5.6	29.0	1.9	0.0061
O K	25.6	51.8	8.4	101.7	0.1343
Si K	0.7	0.8	33.6	5.3	0.0039
Ti K	2.4	1.6	16.6	17.0	0.0267
Fe K	69.2	40.2	2.9	227.2	0.6379

Tool used for identification of SEM images (a) intensity vs. keV plot (b) table of elements identified (c) SEM image of FeOx.

Scanning electron microscopy of subsurface sediment

Methrapur, Dalsingh Sarai

Scanning electron microscopic images of the minerals support the XRD results in Methrapur as shown in **Figure**

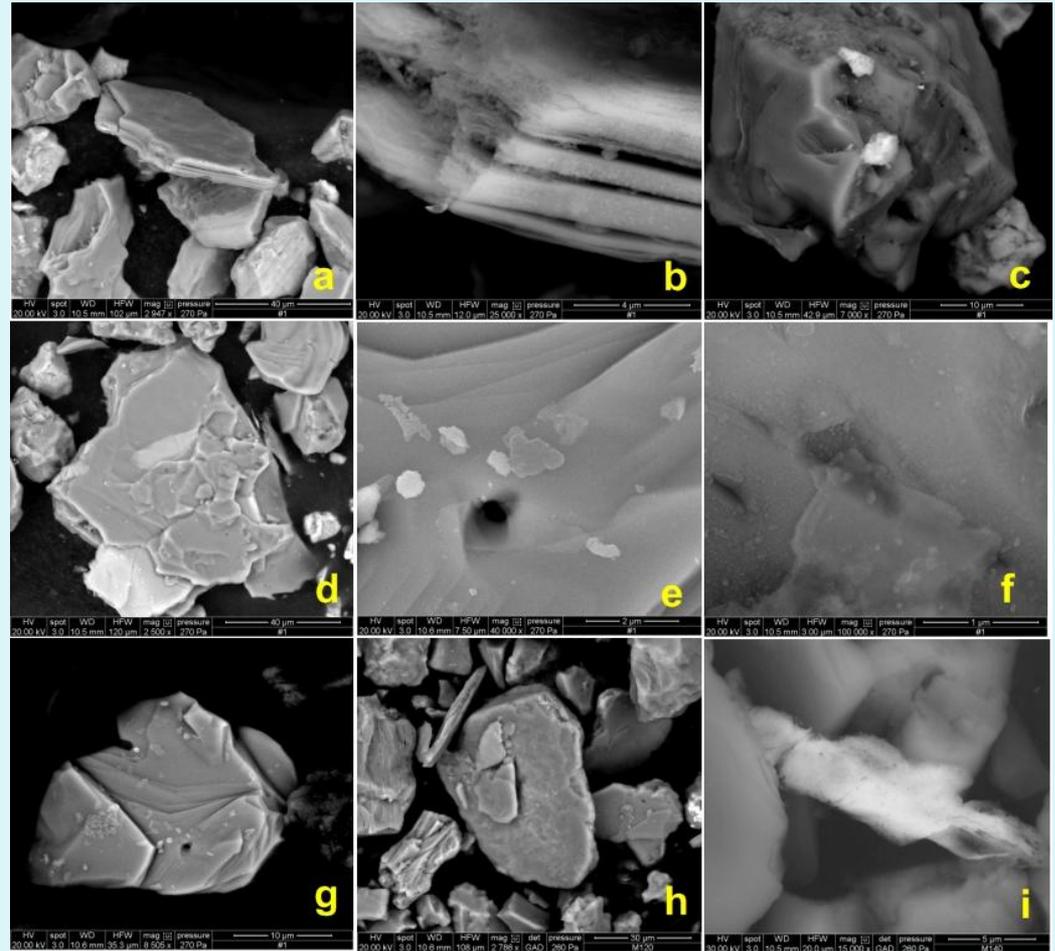


Figure Backscattered close view obtained by Environmental SEM of sediment of different depths: **(a)** layered mica **(b)** calcite **(c)** Fe-oxide on Al-silicate **(d)** weathered pyrite **(e)** Ferric-oxide **(f)** Show intense chemical weathering **(g)** hematite **(h)** muscovite on biotite (large piece) **(i)** Fe-oxide in precipitated form (bright object).

Harail Chapar, Mohiuddin Nagar

Weathered pyrite framboid was noted in lower dark grey micaceous sand at a depth of **9.1m**, indicating signature of sulphate reduction in some extent and prevalence of highly reducing condition in the aquifer.

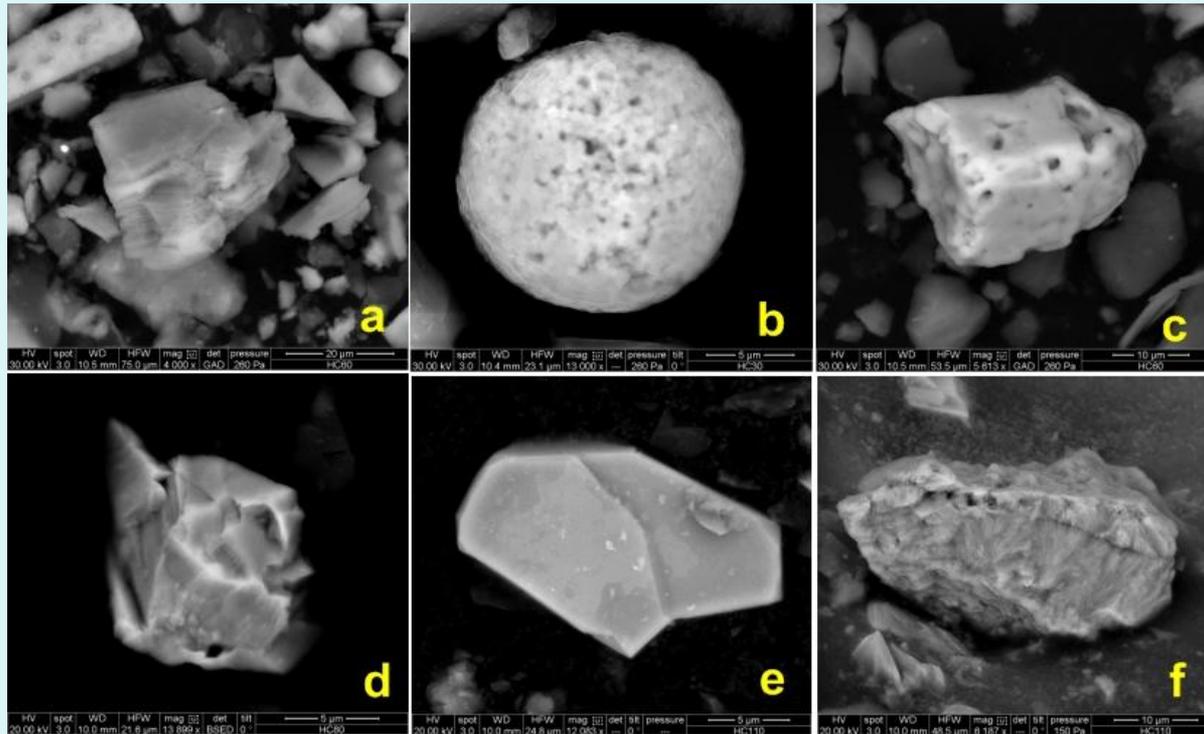
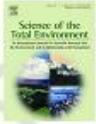


Figure Backscattered Environmental SEM images of sediment of different depths: (a) Na-Mg-Fe-Al silicate (b) weathered pyrite framboid (c) highly weathered zircon (d) Fe-oxide (e) Fe-oxide (surface alteration) (f) fibrous goethite.

Conclusions

- The present study finds that As(III) is dominant in groundwater while As(V) is dominant in agricultural field soil and subsurface sediment.
- Concentrations of As(III) were higher in subsurface sediment than the agricultural field soil.
- The As in extracted or in totally digested samples indicates that subsurface sediments are necessarily the source to soils.
- Variations inorganic As forms were associated with: firstly, the redox conditions of the aquifer depicted in sediment's color; and secondly, with grain size, dark sediment with fine grain size contains more As(III) as well as As(V).
- Siderite appears in the reduced portion of the sediments, and it is a known sink for As.
- The alteration of Fe-oxides and presence of fibrous form of goethite indicating partial solubilization of oxyhydroxides in this area.
- This current study did not include any microbiological component to determine whether microbes are involved in mobilization of As in the middle Gangetic plain.
- Future studies should include the involvement of microbes to discuss the biogeochemical process of As mobilization from the study area. Further studies with larger core sediment samples are required to understand the As mobilization process in the middle Gangetic plain.



Concentrations of inorganic arsenic in groundwater, agricultural soils and subsurface sediments from the middle Gangetic plain of Bihar, India

Manoj Kumar^{a, b, c}, A.L. Ramanathan^c, Mohammad Mahmudur Rahman^{a, b}, Ravi Naidu^{a, b, *}

^a Global Centre for Environmental Remediation (GCER), Faculty of Science and Information Technology, University of Newcastle, University Drive, Callaghan, NSW 2308, Australia

^b Cooperative Research Centre for Contamination Assessment and Remediation of the Environment (CRC-CARE), University of Newcastle, ATC Building, University Drive, Callaghan, NSW 2308, Australia

^c School of Environmental Sciences, Jawaharlal Nehru University, New Delhi 110067, India

ARTICLE INFO

Article history:
Received 26 May 2016
Received in revised form 21 July 2016
Accepted 16 August 2016
Available online xxx
Editor: F.M. Tack

Keywords:
Inorganic arsenic
Agricultural soil
Subsurface sediments
Extractable arsenic
Middle Gangetic plain

ABSTRACT

Concentrations of inorganic forms [arsenite, As(III) and arsenate, As(V)] of arsenic (As) present in groundwater, agricultural soils and subsurface sediment located in the middle Gangetic plain of Bihar, India were determined. Approximately 73% of the groundwater samples ($n = 19$) shows As(III) as the dominant species while 27% reveals As(V) was the dominant species. The concentration of As(III) in agricultural soil samples varies from not detectable to 40 $\mu\text{g}/\text{kg}$ and As(V) was observed as the major species (ranging from 1050 to 6835 $\mu\text{g}/\text{kg}$) while the total As concentration varied from 3528 to 14,690 $\mu\text{g}/\text{kg}$. Total extracted concentration of As was higher in the subsurface soil (range 9119–20,056 $\mu\text{g}/\text{kg}$ in Methrapur and 4788–19,681 $\mu\text{g}/\text{kg}$ in Harail Chapar) than the agricultural soil, indicating the subsurface sediment as a source of As. Results of X-ray diffraction (XRD) and environmental scanning electron microscope (ESEM) revealed the presence of hematite and goethite throughout the vertical section below while magnetite was observed only in the upper oxidized layer at Methrapur and Harail Chapar. Alteration of Fe-oxides and presence of fibrous goethite indicating presence of diagenetic sediment. Siderite plays a crucial role as sinks to the As in subsurface sediments. The study also concluded that decomposition of organic matter present in dark and grey sections promote the redox conditions and trigger mobilization of As into groundwater.

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1. Introduction

For the past few decades, arsenic (As) contamination of groundwater and soil has become an important environmental problem globally. Many As compounds exist in the environment and biological systems as well. Naturally occurring As contamination of groundwater has been reported in groundwater, in > 105 countries of all continents except Greenland and Antarctica (Barringer and Reilly, 2013; Naujokas et al., 2013). The major natural source of As entering the soil is weathering of rocks containing As, while the anthropogenic sources include combustion of coal, activities like smelting of base metal ores, application of As-based pesticides, chromated copper arsenate (CCA) for wood preservation and mining activities (Smedley and Kinniburgh, 2002). Since these activities, and very low depletion are mainly characterized by plant uptake, methylation, leaching or erosion, As may accumulate rapidly in the soil environment and also act as a sink in As-contaminated belts of alluvial plains (Sanyal and Nasar, 2002). Another reason may be the tendency of As to mobilize in the environment strongly, a phenomenon influenced by its association with the solid phase in sediments and soils (Brammer, 2009; Pandey et al., 2004; Winkel et al., 2008). Association of an elevated

As in agricultural soils and the tubewell water used for irrigation for a long time have been confirmed by various studies from Bangladesh (Ahsan et al., 2009; Brammer and Ravenscroft, 2009; Karim et al., 2008; Meharg and Rahman, 2003), Nepal (Dahal et al., 2008), West Bengal (Roychowdhury et al., 2005) and middle Gangetic plain (Srivastava and Sharma, 2013). Higher As concentration (46,000 $\mu\text{g}/\text{kg}$ dry wt.) was reported in agricultural soil in most affected zone compared to a low level of As (< 10,000 $\mu\text{g}/\text{kg}$) in As safe groundwater zone (Meharg and Rahman, 2003). The level of As uptake and accumulation in crops is strongly influenced by the concentrations of As in soils and irrigated water (Smith et al., 2006). A study reported that 6.4 t of As was falling on the soil as a result of using water from 3200 contaminated tube-wells for agricultural irrigation in 201 km^2 area of Deganga block, North 24 Parganas, West Bengal (Chowdhury et al., 2001). Another study reported that total As withdrawn by the four shallow tubewells, used for agricultural irrigation in As-affected areas of Murshidabad district, West Bengal was 6.8 kg per year (Roychowdhury et al., 2005). Continuous irrigation with As-contaminated groundwater may lead to a build-up of this toxic element in soils, which in turn may trigger its entry into the food chain in harmful concentrations. This process further depends on soil type and plant species.

Arsenic added by irrigation water is adsorbed to ferric oxyhydroxides in the topsoil (Roberts et al., 2007) where it gradually accumulates over time (Meharg and Rahman, 2003; Roychowdhury et al., 2005). This accumulation is more prevalent in soils used for paddy

* Corresponding author at: Global Centre for Environmental Remediation (GCER), Faculty of Science and Information Technology, University of Newcastle, University Drive, Callaghan, NSW 2308, Australia.

Email address: ravi.naidu@crccare.com (R. Naidu)

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