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Arsenic in groundwater, Quaternary sediments, and suspended river sediments from the Middle Gangetic Plain, India: distribution, field relations, and geomorphological setting

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Abstract A groundwater arsenic (As) survey in Mirzapur, Varanasi, Ghazipur, Ballia, Buxar, Ara, Patna, and Vaishali districts of UP and Bihar shows that people from these districts are drinking As-contaminated groundwater (max. 1,300 μg/l). About 66 % of tubewells from Buxar to Mirzapur areas and 89 % of tubewells from Patna to Ballia areas have As>10 μg/l (WHO guideline). Moreover, 36 % of tubewells from Buxar to Mirzapur areas and 50 % of tubewells from Patna to Ballia areas have As above 50 μg/l. Most of the As-affected villages are located close to abandoned or present meander channels of the Ganga River. In contrast, tubewells located in Mirzapur, Chunar, Varanasi, Saidpur, Ghazipur, Muhammadabad, Ballia, Buxar, Ara, Chhapra, Patna, and Hazipur towns are As-safe in groundwater because of their positions on the Pleistocene Older Alluvium upland surfaces. The iron (Fe) content in tubewell water samples varies from 0.1 to 12.93 mg/l. About 77 % As-contaminated tubewells are located within the depth of 21 to 40 m in the Holocene Newer Alluvium aquifers. The potential source of As in sediments carried through the rivers from the Himalayas. Maximum As concentrations in the Older and Newer Alluvium sediments are 13.73 and 30.91 mg/kg, respectively. The Himalayas rivers, i.e. Yamuna, Ganga, Gomati, Ghaghara, Gondak, Buri Gandak, and Kosi rivers carrying suspended sediments have high content of As (max. 10.59 mg/kg).

Keywords Groundwater arsenic · Sediment arsenic · Quaternary aquifer · Himalayas river · Geomorphology

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Introduction

The occurrence of high concentrations of arsenic (As) in drinking water has been found in several parts of the world. It is well-established that ingestion of inorganic As may cause liver, lung, kidney, and bladder cancer (Smith et al. 1992). Groundwater As contamination incidence had been reported in different parts of the world including Asia. The mode of occurrence, origin, and mobility of As in sedimentary aquifers in the Bengal Delta and other parts of the world are mainly influenced by local geology, geomorphology, hydrogeology, and geochemistry of sediments and water, as well as anthropogenic activities (Bhattacharya et al. 1997; Nickson et al. 1998; Acharyya et al. 2000; Kinniburgh and Smedley 2001). The upper permissible limit of As in drinking water is 10 μg/l in WHO guideline (WHO 1993) which has been endorsed by Bureau of Indian Standards (BIS 2003). However, in the absence of alternative source, Public Health Engineering Department is supplying drinking water in As-affected areas that have As<50 μg/l. The worst As-affected country in Asia is Bangladesh (Dhar et al. 1997; Nickson et al. 1998; Kinniburgh and Smedley 2001; Ravenscroft et al. 2005; von Brömssen et al. 2007; Uddin et al. 2011). Arsenic contamination in India was first reported from Chandigarh (Datta and Kaul 1976). West Bengal in India is also severely As affected in groundwater (Garai et al. 1984; Mandal et al. 1996; Bhattacharya et al. 1997; Acharyya et al. 2000; Acharyya and Shah 2007; Acharyya and Shah 2010). Groundwater As problem is also reported from Pakistan, People Republic of China, Taiwan, Vietnam, Laos and Cambodia, and Nepal (Tseng et al. 2000; Berg et al. 2001; Sun et al. 2001; Polya et al. 2005; Nickson et al. 2005; Gurung et al. 2005). Moreover, groundwater As is also reported from the Arabian Peninsula in Nile Valley, El Minia district, Egypt

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and Yemen Highland Groundwaters (YHGs), where concentrations of As are fluctuated above and below the WHO guideline (10 μg/l) in drinking water (Al-Mikhlafi 2010; Kashouty et al. 2012). Thus, the As problem is common in several alluvial plains in South, West, and East Asia.

India is the second most populated country in the world, where a large percentage of world's population (17.5 %) is living on limited land area (2.4 %). The Gangetic Plain is one of the vast Quaternary alluvium track in Asia, and groundwater As contamination has been reported in the states of Uttar Pradesh, Bihar, and Jharkhand (Chakraborti et al. 2003; Bhattacharjee et al. 2005; Ahamed et al. 2006; Shah 2008).

The Gangetic Plain of Uttar Pradesh (UP) and Bihar extends from the foothill of the Himalayas in the north to the Peninsular Shield in the south. It consists of fluvial sediments largely derived from the Himalayas and subordinately from the Peninsular India. UP and Bihar are two important states in India and many cities, towns, villages, and hamlets of UP and Bihar are located in the Gangetic Plain. A groundwater As survey in Mirzapur, Varanasi, Ghazipur, Ballia, Buxar, Ara, Patna, and Vaishali districts of UP and Bihar was carried out in tubewells within the Holocene Newer Alluvium aquifers, as well as the Pleistocene Older Alluvium aquifers (Fig. 1). Moreover, the Older and Newer Alluvium sediments, and suspended river sediments were also collected from the Gangetic Plain. The main objective of this study is to investigate the distribution of As in groundwater, Quaternary sediments, and suspended river sediments in the Middle Gangetic Plain under Quaternary geomorphological setting.

Study area

The study areas in the Middle Gangetic Plain are shown in two parts viz., Buxar to Mirzapur areas (Fig. 2) and Patna to Ballia areas (Fig. 3). The geomorphologic and Quaternary morphostratigraphic maps along the Ganga River (85° 21′ 11.80″ E to 82° 27′ 23.56″ E) were prepared based on Survey of India topographic sheets of 1:50,000 scale, with field checks to identify fluvial landforms and soil characters.

The geomorphic features of the Gangetic Plain show differences in their elevations, spatial distribution and nature of sediment, indicating that their formation must have occurred at different times, under different climates, water budget and sediment supply during the Pleistocene–Holocene period. The area is under humid sub-tropical climate, and the annual

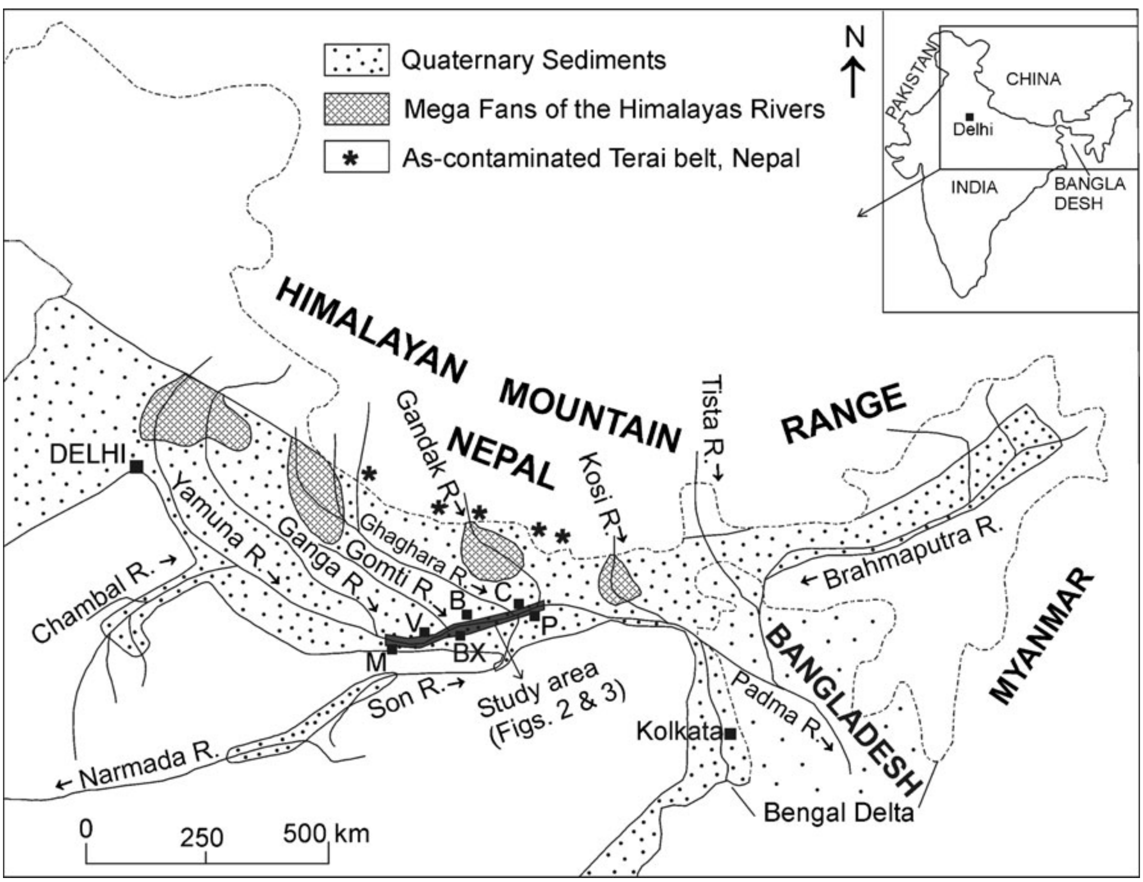

Fig. 1 Quaternary sediments in the Indo-Ganga foredeep and Bengal Basin. The study areas (Figs. 2 and 3) from parts of UP and Bihar are shown in the Middle Gangetic Plain. Abbreviations: *M* Mirzapur, *V* Varanasi, *BX* Buxar, *B* Ballia, *C* Chhapra, *P* Patna

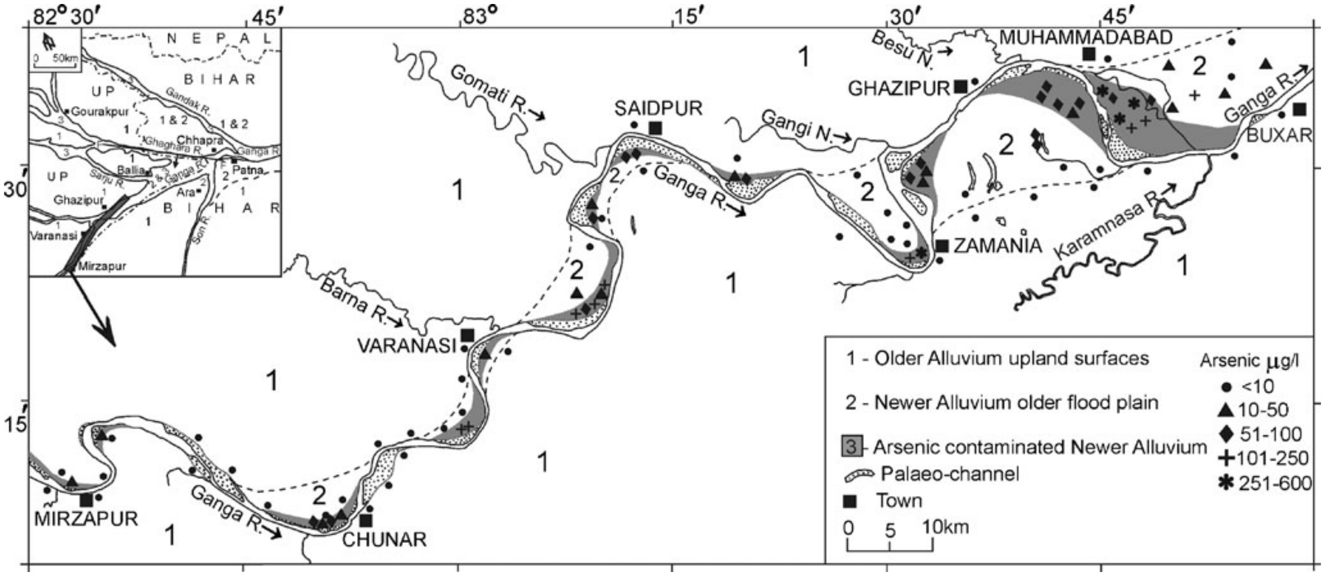

Fig. 2 Groundwater As distribution in entrenched channels and flood plains of the Ganga River from Buxar to Mirzapur towns

average rainfall is 1,041 mm. Temperature variation is ranging from maximum (47 °C) in summer and minimum (2 °C) in winter. The study areas are subdivided into three zones (Figs. 2 and 3), i.e. the Pleistocene Older Alluvium upland Inter-fluve surface (47±12 ka), the Holocene Newer Alluvium river valley terrace surface (3±1 ka), and the Holocene to Recent active channels and flood plains (Kumar et al. 1996; Srivastava et al. 2003).

The major parts of the Gangetic Plain consist of inter-fluve upland surfaces of the Older Alluvium. These Pleistocene Older Alluvium inter-fluve upland surfaces are characterised by yellow-brown coloured sediments with profuse calcareous and ferruginous concretions, and is either exposed or occurs under shallow cover of the Holocene sediments. The Holocene Newer Alluvium surfaces are characterised by organic rich grey to black coloured

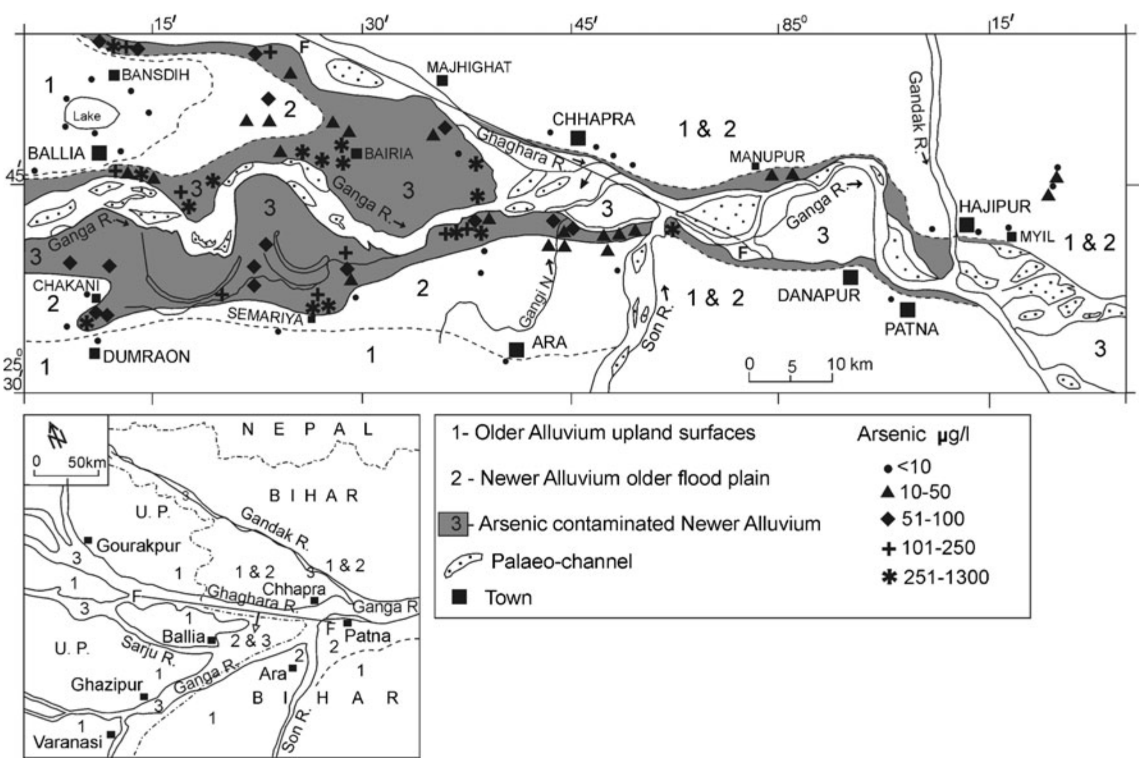

Fig. 3 Groundwater As distribution in entrenched channels and flood plains of the Ganga and Ghaghara rivers system from Patna to Ballia towns

argillaceous sediments in entrenched channels and floodplains of the Gangetic rivers. In this study, approximately 6,500 km^2 have been mapped (Figs. 2 and 3) to delineate groundwater As-contaminated and As-safe areas in the Middle Gangetic Plain.

Geological setting

The Quaternary geology of the Indo-Gangetic Plain has been discussed by Pascoe (1964) who sub-divided the sediments into Older Alluvium and Newer Alluvium. This classification was revised by Pathak et al. (1978) as Upper Siwalik (Upper Pliocene to Lower Pleistocene), Older Alluvium (Middle to Upper Pleistocene), and Newer Alluvium (Upper Pleistocene to Recent) in order of superposition. The Siwalik Supergroup is further subdivided into Lower, Middle, and Upper groups ranging in age from Middle Miocene to Early Pleistocene in northern part of the Siwalik Basin forming the Outer Himalaya. The Banda Group in the southern part corresponds to the Upper Siwalik (Kumar et al. 1996).

The basement and major parts of the Quaternary sediments are concealed in the Gangetic Plain. The deposition of the Quaternary sediments took place in a basin formed during post Siwalik orogeny sometime in Mid Pleistocene. The sedimentation in the basin continued till end of Upper Pleistocene (Dwivedi and Sharma 1992). The first cycle of Quaternary sedimentation in the post-Middle Siwalik basin probably started during Upper Pliocene/Lower Pleistocene with sediment supply in the southern part coming from the Peninsular provenance leading to the deposition of variegated clays followed by clastic sediments overlying Chitrakoot Formation. The Banda Group is the oldest Quaternary lithostratigraphic unit in the southern part of the Indo-Gangetic Plain. The sediments formed Banda Older Alluvium which is characterised by Siwalik boulder conglomerate, sandstone, siltstone, and quartzite embedded with yellowish to brownish coloured matrix (Pathak et al. 1978; Kumar et al. 1996).

A new basin was evolved due to Middle Pleistocene Orogenic movement with upliftment and folding in the northern part of the basin and development of the Himalayan foot hill faults. This basin gradually shifted towards south due to subsidence and received sediments supply from the northern Himalayan provenance. The second cycle of sedimentation during Middle to Early Upper Pleistocene period formed under warm, humid, and dry climate (Dwivedi and Sharma 1992) has refereed here as the Varanasi Alluvium (VA). The Varanasi Older Alluvium constitutes upland surfaces which are occupying the major parts of the Gangetic Plain. The Varanasi Older Alluvium consists dominantly of multiple fill polycyclic sequence of sand, silt, and clay. The Pleistocene Older Alluvium sediments are recognized by yellow-brown coloured sediments with profuse calcareous and ferruginous concretions (Kumar et al. 1996).

The river Ganga draining the area has developed a second geomorphic unit, the low-lying flood plains. The second generation sediments in the Holocene period were deposited within the flood plain domain of each river defined by their palaeobank. These sediments constitute the Newer Alluvium. The main geomorphic features are giant channel bar, natural levee, and flood plains. It is the youngest geomorphic surface present on all other surfaces of the Gangetic Plain. The Holocene Newer Alluvium comprise of grey to black coloured organic-rich argillaceous sediments in entrenched channels and floodplains of the Ganga River. The top thin layer of silt-clay has been deposited in a lacustrine condition towards the close of sedimentation. The Gangetic Plain exhibits variety of landforms, namely incised river valleys, abandoned channels, palaeo-channels, alluvial ridges, ponds, lakes, etc (Singh 2004). The Older Alluvium and Newer Alluvium deposits in the Gangetic Basin have been shown in Table 1.

Materials and methods

In this study, 224 Hand tubewell water samples were collected in acid pre-washed 10 ml plastic bottles. The bottles were kept overnight in dilute laboratory grade nitric acid (1:1) and finally washed with distilled water. Immediately after collection, 1 drop of dilute nitric acid (1:1) GR Grade was added as preservative. At each sampling site, tubewell was purged for approximately 5–10 min to expel any standing water, and the geographic locations of all samples were recorded by a handheld Global Positioning System (Garmin eTrex Vista). The information of tubewell depth was acquired from owner of the tubewell.

The Older and Newer Alluvium sediment samples (approximately 100 gm each) were collected from 8 to 10 m exposed vertical sections of the Ganga River and its tributaries. All samples were collected from 15 to 20 cm inside of vertical sections, and GPS location of all samples was recorded. Each sediment sample of 20 gm was dried in an oven at about 50 °C for approximately 24 h. Sediment samples were ground homogenously and crushed with a mortar and pestle. Analysis of As and iron (Fe) was carried out from the Older and Newer Alluvium sediment samples.

Turbid water of the Yamuna, Ganga, Gomati, Ghaghara, Son, Gondak, Buri Gandak, and Kosi rivers was collected during monsoon period through bucket and poured into a 20 liter plastic bottle. Each sample was collected approximately 50 m away from the riverbank with the help of local people on boat. Plastic bottles were kept in laboratory for a week to settle down the suspended river sediments. After

Table 1 Classification of Quaternary deposits in the Indo-Gangetic Basin (modified after Khan et al. 1996)

Age	Lithounit	Distribution/lithology
Holocene	Channel Alluvium	Active channel deposits of light grey, fine to medium grained micaceous sand and pebbles-cobbles (Himalayan provenance) and red quartzo-feldspathic medium to coarse sand along with drapes of silt and clays (Peninsular provenance).
	Terrace Alluvium	Older flood plain deposits (Terraces) composed of grey fine grained micaceous sand with silt beds in the Himalayan rivers.
	Alluvium Fan Deposits	Occur bordering the Himalayan hill front and composed of sediments ranging from cobbles-pebbles to fine sand and silt.
	Disconformity	
Pleistocene	Varanasi Alluvium (VA)	Polycyclic sequence of clay, silt and micaceous sand of the Himalayan provenance with *Kankar* and ferruginous concretions.
	Unconformity	
	Banda Alluvium (BA)	(b) Chitrakoot Formation: medium to coarse quartzo-feldspathic sand and silt, clay of the Peninsular provenance with minor *Kankar*.
		(a) Variegated clays.
	Unconformity	
Pre-Quaternary	Vindhyan supergroup/Aravalli group	Upper Siwalik Boulder Conglomerate
		Bundelkhand Gneissic Complex in south and (? Marwar Supergroup)
		Lower and Middle Siwalik groups in north

1 week, most of the suspended river sediments were settled down in the bottom of plastic bottle, and top clean water was removed slowly. Thick clayey water in plastic bottle was taken in a beaker and kept in an oven at about 50 °C for dryness. Suspended river sediment samples were used for As and Fe analyses.

Tubewell water and sediment samples were analysed for As and Fe at the School of Environmental Studies, Jadavpur University. Sediment sample weighing 0.5 gm was taken in a Teflon bomb and 2 ml nitric acid was added with it. It was digested at 120 °C temperature for 8 h in hot air oven. Subsequently, the vessel was emptied in a beaker and the sample evaporated near to dryness. After cooling, 5 ml distilled de-ionised water was added in digested sample and it was filtered through 0.45 micron millipore filter and prepared volume 10 ml. During analysis, 5 ml acidified sample was added with 10 ml buffer, 5 ml hydroxylamine hydrochloride, and 2.5 ml phenanthroline and a final volume 25 ml was prepared. The prepared solution was used for As and Fe analyses. Iron in sediment and tubewell water samples was analysed by 1,10 phenanthroline method by the use of UV spectro-photometer.

Arsenic analysis of tubewell water and sediment samples was done at the laboratory through flow injection hydride generation atomic absorption spectrometry (FI-HG-AAS) system. A Perkin-Elmer Model 3100 atomic absorption spectrometer equipped with a Hewlett-Packard Vector Computer with GEM software, Perkins-Elmer EDL system-2, arsenic lamp (lamp current 400 mA) and Varian AAS Model Spectra AA-20 with hollow cathode As lamp (lamp current 10 mA) were used. The minimum detection limit with 95 % confidence level was 3 μg/l As (Chatterjee et al. 1995; Samanta et al. 1999).

A solution of 1.25 % $NaBH_4$ (Merck, Schuchardt, Germany) was prepared in 0.5 % NaOH (Merck, Bombay, India). A 5.0-M solution of HCl (Merck, Bombay, India) was used. All reagents are Analar grade. All these solutions were prepared using distilled de-ionised water. The flow rate for both tetrahydroborate and hydrochloride acid was 1 ml/min. Blank was prepared and measured under the same conditions. Details of the regents and glassware were given elsewhere (Chatterjee et al. 1995; Samanta et al. 1999).

The accuracy of analytical method using FI-HG-AAS was verified by analysing Standard Reference Materials CRM (BND 301) NPL, Indian water (certified value 990±20 μg/l; observed 960±40 μg/l); Water SRM (quality control sample for trace metal analysis) from USEPA Environmental Monitoring and Support Laboratory, Cincinnati, Ohio, USA (certified value 17.6±2.21 μg/l; observed 16±3.5 μg/l). The X-ray Diffractometry (XRD) with Cu target was used (Model Philips APD-15) to determine the constituent minerals in sediment sample.

The results of As and Fe in tubewell water samples, riverbank sediments, and suspended river sediments are shown in Tables 2, 3, 5, and 6. The tubewell As values were plotted in the toposheet to prepare As distribution maps (Figs. 2 and 3) in entrenched channels and flood plains of the Ganga River.

Results and discussion

Distribution of groundwater arsenic from Buxar to Mirzapur towns

Most of the As-contaminated tubewells in between Buxar to Mirzapur towns are located in entrenched channels and floodplains of the Holocene Newer Alluvium surfaces (Fig. 2). A summary of As concentrations of 144 tubewells from Buxar to Mirzapur towns has been shown in Table 2. About 66 % of tubewells have As concentrations above 10 μg/l, the WHO guideline value, and 36 % of tubewells have As above 50 μg/l, the Indian standards for As in drinking water. Maximum concentrations of As and Fe in tubewell are 550 μg/l and 9.3 mg/l, respectively, at Chyan Chappra Village (25° 43.28′: 84° 16.24′), UP.

Table 3 shows that 87 % of tubewell water samples has higher concentrations of Fe beyond its permissible limit of 1 mg/l. The Fe content in tubewell waters varies from 0.2 to as much as 9.3 mg/l. Arsenic-contaminated areas in and around Buxar, Muhammadabad, Ghazipur, Zamania, Varanasi, Chunar, and Mirzapur towns are mainly confined in the meandering belt of the Ganga River. Maximum As concentration at Karkatpur Village is 450 μg/l, which is located in the entrenched channels and flood plains of the Ganga River. However, tubewells located in its opposite side at Zamania town are As-safe in groundwater (<10 μg/l), as the areas belong to the Pleistocene Older Alluvium surfaces. Few As-contaminated areas are also located in the Newer Alluvium older floodplain at the north-east of Zamania and east of Muhammadabad towns. These areas are shallow cover of the Newer Alluvium with As-level ≤100 μg/l. But the Pleistocene Older Alluvium upland surfaces around Zamania and Muhammadabad towns exposing yellow-brown oxidized clay with calcareous and ferruginous concretions are free of As contamination in groundwater. Moreover, tubewells in the Older Alluvium floodplain (As≤50 μg/l) covered with the Holocene sediments on top are locally As contaminated (Fig. 2).

Tubewells located in Buxar, Muhammadabad, Ghazipur, Saidpur, Varanasi, Chunar, and Mirzapur towns are As-safe in groundwater because of their positions on the Older Alluvium upland surfaces (Fig. 2). Depth information (Table 4) of 144 tubewells from Buxar to Mirzapur towns shows that 80 % of tubewells are located within the depth of 10 to 40 m.

Distribution of groundwater arsenic from Patna to Ballia towns

The As-contaminated areas in between Patna to Ballia towns are confined within the Newer Alluvium entrenched channels and floodplains of the Ganga and Ghaghara rivers system (Fig. 3). A summary of As concentrations of 80 tubewells from Patna to Ballia towns is shown in Table 2, where 89 % of tubewells has As concentrations above 10 μg/l and 50 % of tubewells has As above 50 μg/l.

Maximum concentrations of As and Fe in tubewell waters are 1,300 μg/l and 12.93 mg/l at Semariya Ojjha Patti (25° 36.97′: 84° 25.71′) and Pandey Tolla (25° 41.27′: 84° 36.97′) villages, respectively. Table 3 shows that 85 % of tubewell water samples have higher concentrations of Fe beyond its permissible limit of 1 mg/l. The Fe content in tubewell waters varies from 0.1 to as much as 12.9 mg/l. Even As-safe tubewell have higher concentration of Fe (As 6 μg/l and Fe 2.8 mg/l). Depth information (Table 4) of 80 tubewells shows that 74 % of tubewells are located within the depth of 21 to 40 m.

Few As-contaminated areas are also located at the north of Myil, north of Ara, and east of Ballia. All are confined within the Newer Alluvium older flood plain of the Ganga and Ghaghara rivers system. Low level of As concentrations are also observed along the left bank of the Ganga River in the Manupur–Hajipur–Myil areas due to the oxidized Pleistocene Older Alluvium upland surfaces (Fig. 3). A fault is located along the east bank of the Ghaghara River between Chhapra and Majhighat, where down thrown areas on the western bank of the Ghaghara River are As contaminated. Ballia town located on the Older Alluvium or with shallow cover of the Newer Alluvium is free of As contamination. A narrow belt along the southern bank of the Ganga River extending from the confluence point of the Son River to that of Chakani Village is found to be As contaminated (<50 μg/l), as per WHO. Tubewells in Ballia, Ara, Chhapra, Patna, and Hazipur towns are located on the Older Alluvium upland surfaces and are virtually As-safe in groundwater (Fig. 3).

The density of As-contaminated tubewells from Patna to Ballia areas is higher compared to that of Buxar to Mirzapur areas. Table 1 shows 66 % of tubewells have As>10 μg/l from Buxar to Mirzapur areas, whereas 89 % of tubewells have As>10 μg/l from Patna to Ballia areas. Moreover, 36 % of tubewells have As above 50 μg/l from Buxar to Mirzapur areas, whereas 50 % of tubewells have As above 50 μg/l

Table 2 Distribution of tubewell water As (μg/l) in percentage in the Middle Gangetic Plain

Study areas	Samples	As>10	As>50	As>100	As>250	Max. As (μg/l)
Buxar to Mirzapur towns (Fig. 2)	144	66	36	20	8	550
Patna to Ballia towns (Fig. 3)	80	89	50	29	18	1,300

Table 3 Distribution of tubewell water Fe (mg/l) in percentage in the Middle Gangetic Plain

Study areas	Samples	Fe>1	Fe>5	Fe>10	Max. Fe (mg/l)
Buxar to Mirzapur towns (Fig. 2)	144	87	16	–	9.3
Patna to Ballia towns (Fig. 3)	80	85	20	8	12.9

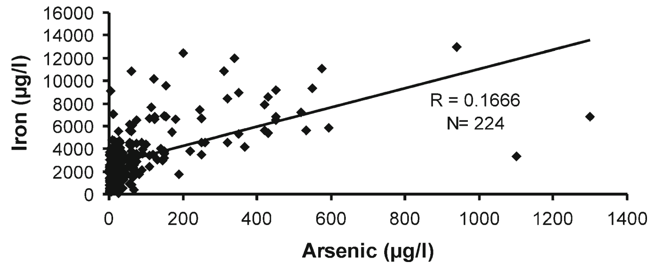

Fig. 4 Correlation between As and Fe concentrations of tubewell water in the Middle Gangetic Plain

from Patna to Ballia areas. So the density of As contaminated tubewells has increased from UP to Bihar states (from west to east) along entrenched channels and flood plains of the Ganga River.

The correlation of As and Fe of 224 tubewells in the Middle Gangatic Plain shows a scatter diagram, where lower value of As corresponds with the higher value of Fe (Fig. 4). Even As-safe tubewell have higher concentration of Fe (As 5 μg/l and Fe 9.1 mg/l).

Depth information of 224 tubewells in the Middle Gangatic Plain (Table 4) shows that 77 % of tubewells are located in shallow depth (21 to 40 m). About 40 % of tubewells has As>50 μg/l within the depth of 17 to 50 m. However, most of the As-contaminated tubewells are located within the depth of 20 to 50 m in the Newer Alluvium aquifers (Fig. 5). About 23 % of tubewells from Patna to Ballia areas are located in very shallow depth (10–20 m), whereas it is only 2 % from Buxar to Mirzapur areas (Table 4). Maximum value of As (1,300 μg/l) corresponds to a depth of 33 m at Semariya Ojjha Patti Village (25° 36.97′: 84° 25.71′).

The age of tubewells (n=224) ranged from one year to 26 years. It is observed that 20 years of old tubewell have As 431 μg/l and 15 years of old tubewell have As 37 μg/l at Garibtola, Ballia district, UP. However, 2 years of old tubewell have As 73 μg/l and 6 years of old tubewell have As 29 μg/l at Gaura market, Ara district, Bihar. Generally, older tubewells have higher concentrations of As, but it is also found that younger tubewells have higher concentrations of As than that of older tubewells. Arsenic concentrations in older and younger age tubewells depend on the lithocharacters of Quaternary sediments and also the As release mechanisms.

Distribution of arsenic and iron in the Older and Newer Alluvium sediments

Chemical analysis of As and Fe in the Older and Newer Alluvium sediments was done from the Middle Gangetic Plain (Table 5), and their distributions in entrenched channels and flood plains of the Ganga River are shown in Fig. 6. Maximum As in the Older Alluvium sediments is 13.73 mg/kg near Saidpur Village, UP (25° 23.20′: 81° 47.58′), whereas maximum As in the Newer Alluvium flood top clay is 30.91 mg/kg on the bank of Ganga River near Loktola Village (25° 18.69′: 82° 04.59′), UP. Maximum Fe in the Older Alluvium sediments at Charri Village (25° 20.20′: 81° 55.69′), and the Newer Alluvium sediments at Pancharukhia Village (25° 45.80′: 84° 21.80′) are 6.15 and 6.31 g/kg, respectively. Iron concentrations in the Older Alluvium and Newer Alluvium sediments are more or less same, but As concentration in the flood sediments is much more higher than that of the Older Alluvium sediments in the Middle Gangetic Plain (Table 5; Fig. 6). However, As and Fe concentrations in the Newer Alluvium sediments in the Bengal Delta are 7.7 mg/kg and 4.26 g/kg, respectively in Manikganj, Bangladesh (Shamsudduha et al. 2008). Arsenic-contaminated tubewells are most occurred in the Newer Alluvium Holocene sediments in the Bengal Delta (Acharyya and Shah 2007; von Brömssen et al. 2007; Shamsudduha et al. 2008).

XRD study of clayey sand was done from the Newer Alluvium sediments near Varanasi area and the mineralogical assemblage is quartz, muscovite, chlorite, montmorillonite, kaolinite, feldspar, and goethite. However, same mineralogical assemblage is also reported from borehole sediments in Jessore district of Bangladesh and Nadia district of West Bengal (Akai et al. 2004; Acharyya and Shah 2007).

Distribution of arsenic and iron in suspended river sediments

Turbid water samples of the Yamuna, Ganga, Gomati, Ghaghara, Son, Gondak, Buri Gandak, and Kosi Rivers in the Gangetic Plain were collected during monsoon period. Seven rivers in the northern part of the Gangetic plains originated from the Himalayas and the Son River flows from

Table 4 Depth (m) of tubewell (%) in As-affected areas in the Middle Gangetic Plain

Study areas	Samples	10–20 m	21–40 m	41–60 m	Max. depth (m)
Buxar to Mirzapur towns (Fig. 2)	144	2	80	17	60
Patna to Ballia towns (Fig. 3)	80	23	74	4	53

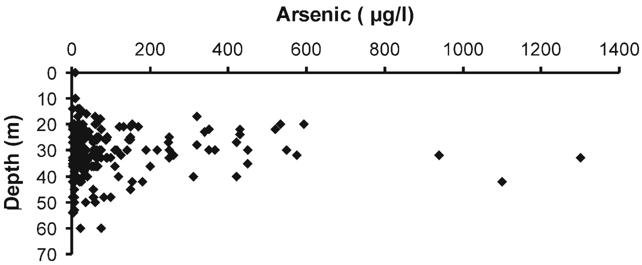

Fig. 5 Relation between As concentration and depth of tubewell in the Middle Gangetic Plain

the Peninsular India. Suspended river sediments of these rivers are very common during monsoon period and sediment samples were analysed for As and Fe (Table 6; Fig. 6). Maximum As in suspended river sediments of the Kosi River is 10.59 mg/kg near Saharsa town (25° 53.07′: 86° 26.21′), and minimum As is 5.61 mg/kg in the Yamuna River near Sihonda town (25° 21.27′: 81° 38.59′). The Ganges is one of the major rivers of the Indian subcontinent and maximum As in suspended river sediments is 9.52 mg/kg near Buxar town (25° 34.58′: 83° 58.25′). The Son River is the largest of the Ganges' southern tributaries, have As 6.60 mg/kg in suspended sediments near Koelwar town (25° 34.11′: 84° 48.31′). Arsenic in suspended river sediments of the Gomati, Ghaghara, Gandak, and Buri Gandak rivers ranges from 7.01 to 9.04 mg/kg. During high water discharge, the contribution to suspended sediments in river most probably originates from erosion and weathering processes of bed sediments and bank scour which results in fluvial transport and sedimentation of As-enriched metal hydroxides especially Fe oxy-hydroxides (Berg et al. 2001). Iron content in suspended river sediments of the Yamuna, Ganga, Gomati, Ghaghara, Son, Gondak, Buri Gandak, and Kosi rivers varies from 5.65 to 1.17 g/kg (Table 6). Maximum Fe in suspended river sediments of the Ganga River is 5.65 g/kg near Buxar town (25° 34.58′: 83° 58.25′). So all the Himalayas rivers have high content of As and Fe in suspended sediments of turbid water and are mostly deposited in the Gangetic Plain. Moreover, the As and Fe bearing suspended river sediments are also associated with organic rich grey to black coloured argillaceous sediments. Arsenic adsorbed on Fe/Mn oxides/hydroxides is released into the groundwater due to a decrease of the redox state in the aquifer (Davis and Kent 1990).

Groundwater arsenic in quaternary geomorphological setting

The Ganga is the axial river in the Himalayan foreland basin. Fluvial sedimentation in the Gangetic Basin was continuing after the depositions of Siwaliks. There have similarities in terms of alluvial architecture, fluvial processes, sedimentation rates, and depositional environments between Siwaliks and post-Siwaliks depositions. A large number of small drainages, dense networks of channel and numerous lakes, and swamps have developed in the Gangetic Basin. The subsurface extension of the Gangetic Basin is limited to the west by the Aravalli–Delhi ridge and to the east by the Monghyr–Saharsa ridge (Singh 2004).

During the Pleistocene time, the mechanical weathering of rocks in source areas (e.g. Himalayas, Indian Shield) was enhanced due to mountain building activities and glaciations. The aquifer sands were largely derived from physical weathering and erosion of the Himalayas. The Ganga Plain has developed a network of small drainages, and entrenchment of major rivers during ~20–13 ka BP. The network became dense during 13–8 ka BP (early Holocene) and moved large amounts of sediment-water to the Ganga Delta. During 8–6 ka BP small drainages changed to large lakes and extensive deposition of sediment took place on the upland interfluve surfaces, and supply of sediment to delta region from the Ganga Plain was reduced (8 ka BP—present). Most of the areas of the Ganga Plain are

Table 5 Distribution of As and Fe concentrations in the Older and Newer Alluvium sediments in the Middle Gangetic Plain

S. no.	Latitude (N)	Longitude (E)	Sample location	Lithology	As (mg/kg)	Fe (g/kg)
S1	25° 23.20′	81° 47.58′	Sahidpur Village, UP	OA	13.73	4.25
S2	25° 28.07′	81° 47.80′	Nima Village, UP	OA	11.57	1.68
S3	25° 20.20′	81° 55.69′	Charri Village(top unit), UP	OA	10.28	6.15
S4	25° 20.20′	81° 55.69′	Charri Village(bottom unit), UP	OA	11.01	3.84
S5	25° 18.69′	82° 04.59′	Flood clay, Loktola Village, UP	NA	30.91	4.00
S6	25° 14.50′	83° 01.73′	Varanasi New bridge, UP	NA	5.26	3.13
S7	25° 24.93′	83° 33.48′	Zamania Village, UP	OA	5.65	3.75
S8	25° 45.80′	84° 21.80′	Pancharukhia Village, UP	NA	8.75	6.31
S9	25° 41.80′	84° 50.36′	Son River bank , Bihar	NA	3.75	2.63
S10	25° 38.44′	85° 16.54′	Myil Village, Bihar	OA	7.28	1.57

OA Older Alluvium, *NA* Newer Alluvium

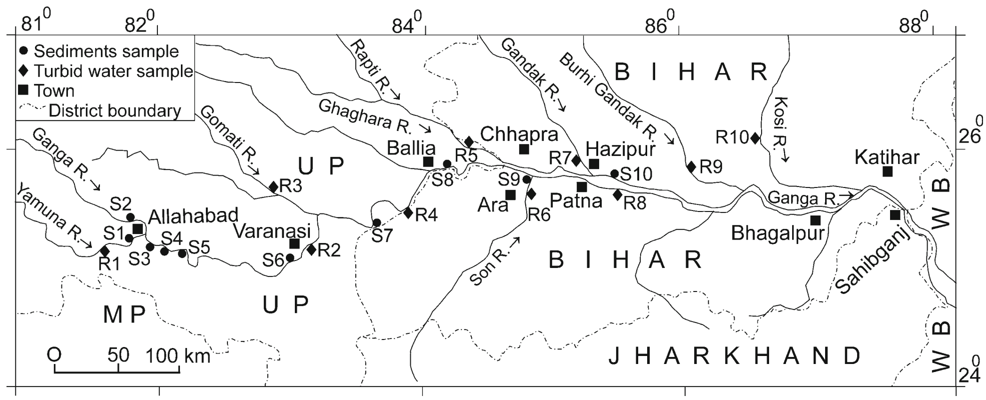

Fig. 6 Distribution of riverbank sediments and suspended rivers sediments in the Middle Gangetic Plain. *Black circle* represents the Older and Newer Alluvium sediments (Table 5), and *black diamond* indicates the suspended river sediments in turbid water (Table 6)

covered by 5–10 m thick muddy sediments, representing deposition during mid to late Holocene (Singh 2004).

The major parts of the Gangetic Plain consist of the Pleistocene Older Alluvium upland surfaces. Groundwater As distribution in entrenched channels and flood plains of the Ganga River is shown in Google satellite imagery (Fig. 7). Geomorphologically, the Gangetic Plain is subdivided into three zones viz., 1—the Pleistocene Older Alluvium upland surfaces, 2—the Older Alluvium flood plains covered with the Holocene sediments on top, and 3—the Holocene Newer Alluvium entrenched channels and flood plains. Tubewells located in the Pleistocene Older Alluvium aquifers are virtually As-safe in groundwater, whereas tubewells located in the Holocene Newer Alluvium aquifers are As contaminated in groundwater. Moreover, the Older Alluvium covered with the Holocene sediments on top are locally As contaminated in groundwater. Arsenic affected villages are located in proximity to meander or abandoned channels (Fig. 7). However, distribution of high content of groundwater As in the Bengal Delta, Mekong River Delta, and Barak Valley is also controlled by the Pleistocene and Holocene aquifers (Polya et al. 2005; Acharyya and Shah 2007; von Brömssen et al. 2007; Shah 2012).

In the Gangetic Plain, the Pleistocene Older Alluvium surfaces were dissected by channels and flood plains and buried under the younger Holocene Newer Alluvium sediments. This yellow-brown coloured oxidizing Pleistocene Older Alluvium surfaces were well flushed by groundwater flow due to high-hydraulic head and devoid of organic matter. The environment of the oxidized Pleistocene Older Alluvium aquifer is not favorable to release sorbed As to groundwater. In the Bengal Delta, high groundwater As concentration is found in the Holocene aquifers, while the Pleistocene aquifers have a low As level (Ravenscroft et al. 2005).

Schematic profile diagram of the Middle Gangetic Plain shows that As-affected aquifers are mostly confined in the Holocene entrenched channels and flood plains. The Holocene Newer Alluvium aquifers are characterised by grey to black coloured organic-rich argillaceous sediments. Tubewells located in the Pleistocene Older Alluvium aquifers are generally As-safe in groundwater (Fig. 8). The Pleistocene Older Alluvium aquifers would be better option for As-safe drinking water. Occurrences of high groundwater As are significantly linked with the Pleistocene and Holocene aquifers, and this model regarding groundwater As can be applied in others fluvial deltaic setting in South and South-East Asia as well. Groundwater As is mostly found in the delta plains, modern floodplains, marshes, and depressed lowlands areas in Bangladesh (Kinniburgh and Smedley 2001). High-As areas are characterised by low

Table 6 Distribution of As and Fe concentrations in the suspended river sediments in the Middle Gangetic Plain

S. no.	Latitude (N)	Longitude (E)	Location of turbid river water	As (mg/kg)	Fe (g/kg)
R1	25° 21.27′	81° 38.59′	Yamuna River, UP	5.61	4.59
R2	25° 18.60′	83° 00.75′	Ganga River, UP	8.07	4.87
R3	25° 47.44′	82° 41.51′	Gomati River, UP	8.01	3.15
R4	25° 34.58′	83° 58.25′	Ganga River, Bihar	9.52	5.65
R5	26° 01.78′	84° 11.29′	Ghaghara River, UP	7.11	3.30
R6	25° 34.11′	84° 48.31′	Son River, Bihar	6.60	1.70
R7	25° 41.48′	85° 11.84′	Gondak River, Bihar	7.01	1.17
R8	25° 29.19′	85° 42.51′	Ganga River, Bihar	6.30	1.90
R9	25° 57.39′	85° 44.46′	Buri Gandak River, Bihar	9.04	3.82
R10	25° 53.07′	86° 26.21′	Kosi River, Bihar	10.59	4.01

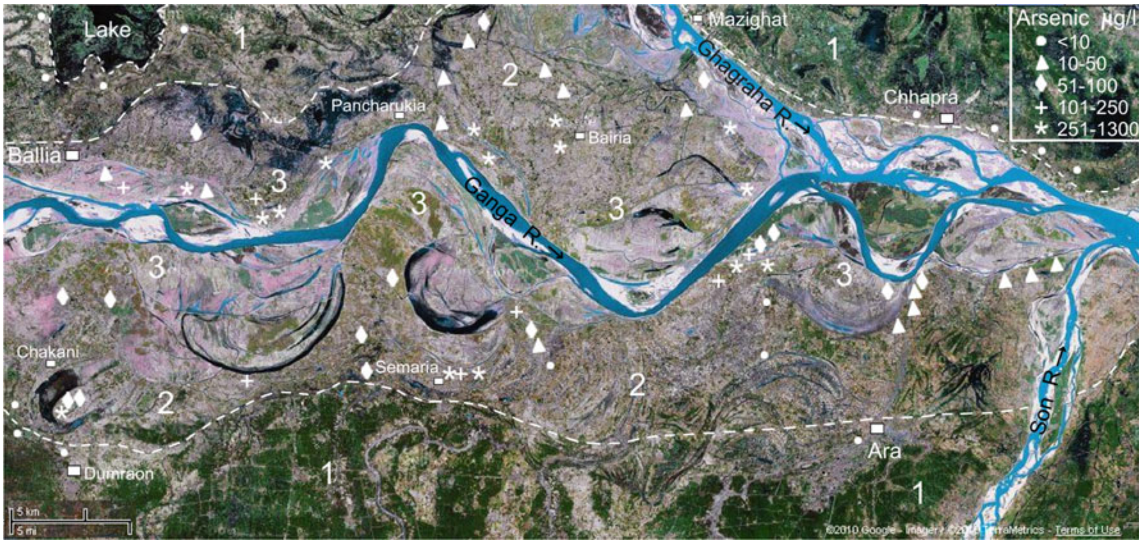

Fig. 7 Google satellite imagery showing spatial distribution of As-contaminated tubewells in entrenched channels and floodplains of the Ganga and Ghaghara rivers system (Fig. 3)

slopes in shallow aquifers where groundwater flow is mainly controlled by elevation and slope variations (Shamsudduha et al. 2008).

Source and release of arsenic in groundwater

No specific sources of As could be identified in the Gangetic Plain but several potential minor sources have been identified both in the Himalayan belt, as well as in the Peninsular India. The Gangetic plains were formed due to accumulation of bulk sediments from the Himalayan hill range, whereas the input of the Peninsular India is minor. There are several As-bearing minerals deposits in the Himalayan hill range including hydrothermal pyrite-chalcopyrite-arsenopyrite-galena mineralization associated with quartz veins in Buniyal, Doda, Almora Garhwal, J and K Hills (Tewari and Gaur 1977). The Indus-Tsangpo suture in north India is marked by ophiolitic rocks, including olivine serpentinites. These ophiolites are composed of serpentinized peridotite, layered mafic to ultramafic rock, volcanic, and oceanic sediments that contain high As (Guillot and Charlet 2007). The Peninsular India is also accounted for source of As where pyrite-bearing shale in Amjhore mine have As 2.6 g/kg (Das 1977). In the gold mineralization belt of Son Valley, As concentrations in bedrock locally reach 28 to 1 g/kg (Mishra et al. 1996).

Groundwater As contamination is also reported from India–Nepal border zone mainly in the Terai belt (Gurung et al. 2005). The Terai sediment has come from the Himalayan hill range. Most of the rivers in the northern part of the Gangetic Plain viz., Yamuna, Ganga, Gomati, Ghaghara, Son, Gondak, Buri Gandak, and Kosi rivers originated from the Himalayas. The possibility of erosion,

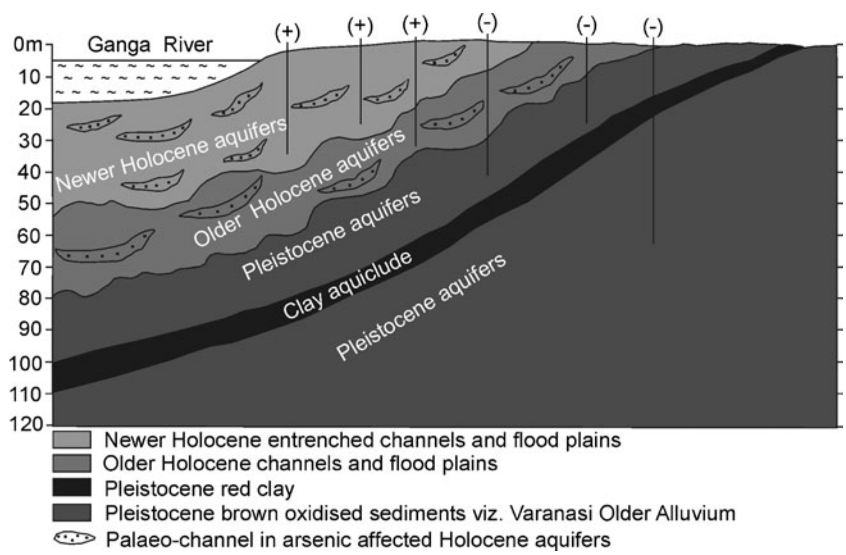

Fig. 8 Schematic profile diagram showing As-contaminated areas in the Middle Gangetic Plain. Palaeo-channels are not shown in the Pleistocene aquifers. Tubewells of As-contaminated and As-safe are shown in symbols (*plus sign*) and (*negative sign*), respectively

oxidation and transportation of As-bearing products in suspension and solution in the Gangetic Plain are high. The southern belt of the Himalayas is subjected to high erosion and intense rainfall during the Holocene time (Williams and Clarke 1984). Most of the As- bearing sediments are enriched with Fe oxides. Oxides of Fe and Mn are potentially the most important source/sink for As in aquifer. Hydrous Fe oxide has a very high specific surface area, and thus a very high adsorption capacity to adsorb heavy metals. Arsenic adsorbed on Fe/Mn oxides/hydroxides is released into the groundwater due to a decrease of the redox state in the aquifer (Davis and Kent 1990). Reduction of hydrated iron oxide (HFO) is common, and high concentration of dissolved Fe (12.9 mg/l) indicates strong reducing condition. Mobilisation of As in the Gangetic Plain is expected to follow a similar course as observed in the Bengal Delta of West Bengal and Bangladesh. Arsenic sorbed on discrete phases of hydrated Fe-Mn oxide coated sediments grains were preferentially entrapped in grey to black coloured organic-rich argillaceous sediments in entrenched channels and floodplains of the Ganga River. Biomediated reductive dissolution of hydrated iron oxide (HFO) by anaerobic heterotopic Fe^{3+} reducing bacteria (IRB) play an important role to release sorbed As to groundwater (Nickson et al. 1998; Kinniburgh and Smedley 2001; Islam et al. 2004).

Conclusions

A groundwater As survey in Mirzapur, Varanasi, Ghazipur, Ballia, Buxar, Ara, Patna, and Vaishali districts of UP and Bihar shows that tubewells located in entrenched channels and floodplains of the Ganga River are prone to As contamination in groundwater. The Holocene Newer Alluvium aquifers are characterised by organic rich grey to black coloured argillaceous sediments and tubewells located in this aquifers are As contaminated. Most of the As-affected villages are preferentially located close to abandoned or present meander channels of the Ganga River. Tubewells in Mirzapur, Chunar, Varanasi, Saidpur, Ghazipur, Muhammadabad, Ballia, Buxar, Ara, Chhapra, Patna, and Hazipur towns are located in the Pleistocene Older Alluvium aquifers and are virtually As-safe in groundwater.

The major parts of the Gangetic Plain in UP and Bihar consist of inter-fluve upland surfaces of the Pleistocene Older Alluvium. These upland oxidizing Pleistocene yellow-brown coloured sediments were well flushed by groundwater flow due to high hydraulic head and devoid of organic matter. The environment of the Pleistocene Older Alluvium aquifers is not favorable to release sorbed As in groundwater and aquifers are generally As-safe. However, in As-contaminated areas, deeper tubewells in the Pleistocene Older Alluvium aquifers would be better option for As-safe drinking water.

The potential sources of As-bearing sediments are mainly from the Himalayas. The Older Alluvium and Newer Alluvium sediments have high content of As. The Yamuna, Ganga, Gomati, Ghaghara, Gondak, Buri Gandak, and Kosi rivers originated from the Himalayas and have high content of As in suspended river sediments, and most of the As-bearing sediments are deposited in the Gangetic Basin.

Distribution of high content of groundwater As in alluvial aquifers in fluvial deltaic setting in South and South-East Asia is controlled by regional geologic–geomorphic units of the Older and Younger Alluvium sediments. Iron in tubewell water is sufficient for reducing condition and biomediated reductive dissolution of hydrated iron oxide by anaerobic heterotopic Fe^{3+} reducing bacteria play an important role to release sorbed As to groundwater.

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