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DOI
10.1016/j.scitotenv.2016.09.074

Publication date
2017

Document Version
Final published version

Published in
Science of the Total Environment

Citation (APA)
Donselaar, M. E., Bhatt, A. G., & Ghosh, A. K. (2017). On the relation between fluvio-deltaic flood basin geomorphology and the wide-spread occurrence of arsenic pollution in shallow aquifers. *Science of the Total Environment*, *574*, 901-913. https://doi.org/10.1016/j.scitotenv.2016.09.074

Science of the Total Environment 574 (2017) 901–913

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

On the relation between fluvio-deltaic flood basin geomorphology and the wide-spread occurrence of arsenic pollution in shallow aquifers

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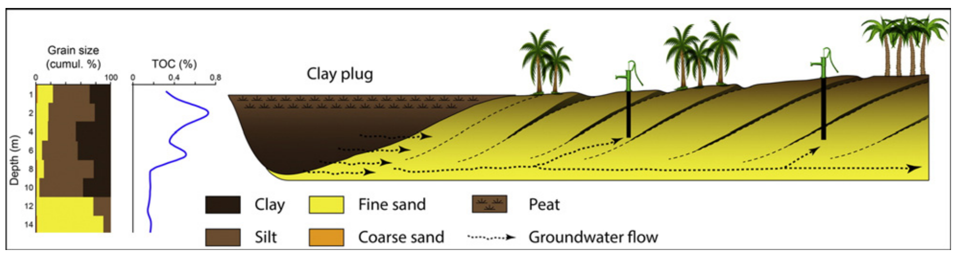
HIGHLIGHTS

- Point-bar and oxbow-lake/clay-plug geomorphological elements are proposed as the coupled source/sink of dissolved arsenic.
- A generic geomorphological model explains the migration and accumulation of dissolved arsenic on entire flood-basin scale.
- Anoxic hypolimnion oxbow-lake water and clay-plug sediments are the loci of reactive organic carbon.
- Released arsenic is trapped in permeable point-bar sands surrounded by low-permeable clay plugs.
- Permeability contrasts in the point-bar geomorphological element cause spatial arsenic concentration differences.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 28 April 2016
Received in revised form 7 September 2016
Accepted 9 September 2016
Available online 14 October 2016

Editor: D. Barcelo

Keywords:
Aquifer flushing efficiency
Arsenic pollution
Holocene flood basin
Microbial action
Permeability heterogeneity
Point-bar sand

ABSTRACT

Pollution of groundwater with natural (geogenic) arsenic occurs on an enormous, world-wide scale, and causes wide-spread, serious health risks for an estimated more than hundred million people who depend on the use of shallow aquifers for drinking and irrigation water. A literature review of key studies on arsenic concentration levels yields that Holocene fluvial and deltaic flood basins are the hotspots of arsenic pollution, and that the dominant geomorphological setting of the arsenic-polluted areas consists of shallow-depth meandering-river deposits with sand-prone fluvial point-bar deposits surrounded by clay-filled (clay plug) abandoned meander bends (oxbow lakes). Analysis of the lithofacies distribution and related permeability contrasts of the geomorphological elements in two cored wells in a point bar and adjacent clay plug along the Ganges River, in combination with data of arsenic concentrations and organic matter content reveals that the low-permeable clay-plug deposits have a high organic matter content and the adjacent permeable point-bar sands show high but spatially very variable arsenic concentrations. On the basis of the geomorphological juxtaposition, the analysis of fluvial depositional processes and lithofacies characteristics, inherent permeability distribution and the omnipresence of the two geomorphological elements in Holocene flood basins around the world, a generic model is presented for the wide-spread arsenic occurrence. The anoxic deeper part (hypolimnion) of the oxbow lake, and the clay plugs are identified as the loci of reactive organic carbon and microbial respiration in an anoxic environment that triggers the reductive dissolution of iron oxy-hydroxides and the release of arsenic on the scale of entire fluvial floodplains and deltaic basins. The

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http://dx.doi.org/10.1016/j.scitotenv.2016.09.074

adjacent permeable point-bar sands are identified as the effective trap for the dissolved arsenic, and the internal permeability heterogeneity is the cause for aquifer compartmentalization, with large arsenic concentration differences between neighboring compartments.

1. Introduction

Natural arsenic pollution of groundwater causes a wide-spread, serious health risk for people who depend on the use of shallow aquifers for drinking and irrigation water. It is estimated that the arsenic pollution affects several hundred million people world-wide (Ravenscroft et al., 2009). Long-term, continued ingestion of arsenic-polluted drinking water, far above the recommended permissible limit of 10 μg/L (World Health Organization — WHO, 1993, 2011) results in the accumulation of arsenic in the human body and a wide array of diseases generally grouped as arsenicosis. The problem of arsenic pollution in drinking water was already described a century ago in Argentina (Litter et al., 2014), but it was not until the early 1980s when the enormous, world-wide scale was discovered with the recognition of arsenic drinking water pollution in the Ganges-Brahmaputra-Meghna Delta in West Bengal, India and Bangladesh (Das et al., 1994; Bhattacharya et al., 1997; Acharyya et al., 2000; BGS and DPHE, 2001; Ahmed et al., 2004; Ravenscroft et al., 2009). Since then, arsenic pollution is recognized globally (Smedley and Kinniburgh, 2002; Ravenscroft et al., 2009), with arsenic hotspots in the Mekong and Red River Deltas in Vietnam (Berg et al., 2001, 2007; Postma et al., 2007; Benner et al., 2008), the Chaco-Pampean Plain in Argentina (Bundschuh et al., 2004; Bhattacharya et al., 2006), the Altiplano Basin in Bolivia (Ramos Ramos, 2014), and Nevada, California and Arizona (SW USA; Ravenscroft et al., 2009), among others.

Pollution of groundwater with naturally-occurring (geogenic) arsenic is concentrated in the shallow aquifer domain of sedimentary basins, and is notably widespread in Holocene fluvial and deltaic flood basins (e.g. Smedley and Kinniburgh, 2002). The arsenic concentration levels in the aquifers are characterized by large lateral variability over distances of 100s of meters (e.g. van Geen et al., 2003; McArthur et al., 2004; Zheng et al., 2004; Harvey et al., 2006; Shah, 2008) and a general strong vertical decrease when the wells penetrate deeper Pleistocene strata (Zheng et al., 2005; Shah, 2008). Arsenic occurs in nature as arsenopyrite, arsenic adsorbed to iron hydroxide, hydrated iron-oxide coatings on quartz and clay minerals, and As–Cu mineralization in granite (BGS and DPHE, 2001; Shah, 2008, 2010). It is generally accepted that the principal process of arsenic release from its solid state to the groundwater occurs in a redox-controlled environment with microbially-mediated reductive dissolution of iron oxy-hydroxides (Nickson et al., 2000; McArthur et al., 2001, 2004; Ravenscroft et al., 2001; Postma et al., 2007; Singh et al., 2010).

Various authors have made the connection between the spatial variability in arsenic concentrations and the geomorphological setting of the shallow aquifers. Ahmed et al. (2004) and McArthur et al. (2004, 2011) related the release of arsenic from iron oxy-hydroxides by the reductive dissolution in the shallow aquifers to the occurrence of organic matter-rich peat layers in the shallow subsurface of the meandering Hugli and Sunti River morphology. Postma et al. (2007, 2012) and Kazmierczak et al. (2016) related the arsenic concentrations to the age and trends of fluvial deposits of Red River deposits in Vietnam. Hoque et al. (2014) concluded that palaeosols act as shields to prevent arsenic to move to shallow palaeo-interfluvial aquifers in the Ganges River floodplain of the Bengal Basin. Sahu and Saha (2015) correlated the variability in arsenic concentrations with the different sediment types in the floodplain of the meandering Ganges River. Nath et al. (2005), Mukhopadhyay et al. (2006), Papacostas et al. (2008), Weinman et al. (2008) and Desbarats et al. (2014) associated the occurrence of high arsenic concentrations to abandoned meander bends (*oxbow lakes*). The oxbow lake water and the fine-grained sediment of filled-in oxbow lakes (*clay plugs*) were proposed as the two main sources for reactive organic matter (Ravenscroft et al., 2001; Harvey et al., 2002, 2006; Islam et al., 2004; McArthur et al., 2004; Meharg et al., 2006; Postma et al., 2007; Neumann et al., 2010; Mailloux et al., 2013). Desbarats et al. (2014) proposed that the organic matter in the clay plugs triggers the reactive dissolution of iron oxy-hydroxides and associated release of arsenic. Ghosh et al. (2015b) and Ghosh (2016) documented TOC values of 0.7% in shallow Holocene clay plug sediments in the Jalangi River floodplain. Desbarats et al. (2014) produced a simplified reactive solute transport model for the migration of arsenic in the clay plug sediments as consequence of irrigation pumping. They concluded that leakage of groundwater through clay-plug sediments (channel-fill sediments in the terminology of Desbarats et al., 2014) is the principal effect of irrigation pumping.

All studies indicate the intricate relationship between the geomorphology, fluvial depositional setting, groundwater migration and intensity of the arsenic pollution. The results of these studies are spot observations that account for local sources and variations in arsenic concentrations in the studied sites, but do not explain the enormous scale and basin-wide extent of arsenic pollution (BGS and DPHE, 2001; Acharyya and Shah, 2007; Ghosh, 2016) which implies a ubiquitous source of microbial respiration to cause the release of arsenic from its solid state.

The aims of this paper are (1) to establish the causal relationship between the alluvial geomorphology in the affected basins and the widespread occurrence and spatial variability in arsenic-pollution concentrations, and (2) to present a generic spatial aquifer architecture model for the release and accumulation of arsenic in fluvial flood basins. The fluvial depositional setting is analyzed for its potential to serve as omnipresent local source of sedimentary organic matter in which microbial communities can thrive and act as agents to release arsenic from its solid state. In addition, the relation is studied between fluvial facies heterogeneity and permeability contrasts in the fluvial sediments, and their impact on aquifer flushing efficiency and related variation in arsenic concentrations. Insight in the spatial variability of arsenic pollution as a function of geomorphological heterogeneity is of paramount importance to design arsenic mitigation strategies and the designation of arsenic-free zones for small-scale piped water supply systems in the affected areas.

2. Data and methods

The study areas of key publications on the spatial variability of arsenic concentrations were analyzed in Google Earth-Pro, and from this an inventory was made of the depositional setting and specific geomorphological conditions (Table 1). In addition, the analysis of time-lapse Google Earth-Pro images provided insight in the depositional processes and velocity of infilling of an abandoned meander bend in the Ganges River floodplain near Suhija, Bhojpur District, Bihar (co-ordinates: 25° 38.934′N, 84° 23.850′E). Two 50-m-deep wells were drilled (percussion drilling) in a large point bar (coordinates Well 1: 25° 40.014′N, 84° 40.913′E) and bordering filled-in oxbow lake (coordinates Well 2: 25° 39.037′N, 84° 40.064′E) along the Ganges River, near Bakhorapur, Bhojpur District in Bihar (Fig. 1). Both wells were fully cored for analysis of the lithofacies heterogeneity. The cores were collected in 60-cm-long PVC core tubes with 4 cm diameter; core recovery was 80%. The core description (grain size, texture, color, sedimentary structures) provided details of the lithofacies succession in the shallow aquifer domain.

Table 1
Literature analysis of geomorphological setting of Holocene aquifers with documented spatial variability of arsenic concentrations.

Authors	Country	Coordinates	Setting	Geo-morphology	Depth (m)	As-concentration range (μg/L)
Berg et al. (2007)	Vietnam (Hanoi)	20° 55.779′N 105° 50.054′E	Holocene floodplain Red River	Point bars — clay plugs	12–45	<10–>300
Bhattacharya et al. (2006)	Argentina (Santiago del Estero)	27° 54.402′S 64° 6.157′W	Holocene floodplain Río Dulce	Point bars — clay plugs, volcanic ash layer	<12	Ave. 743[a]
Bundschuh et al. (2004)	Argentina (Santiago del Estero)	27° 54.402′S 64° 6.157′W	Holocene floodplain Río Dulce	Point bars — clay plugs, volcanic ash layer	?	10–4780[a]
von Brömssen et al. (2007)	Bangladesh	23° 19.746′N 90° 41.616′E	Holocene floodplain Dhonagoda River	Point bars — clay plugs	17–82	5.2–355
Dhar et al. (2014)	Bangladesh	23° 46.937′N 90° 37.013′E	Floodplain Meghan River	Point bars — clay plugs	5-91	<5–860
Enmark and Nordborg (2007)	India (Assam)	26° 17.918′N 90° 43.546′E	Holocene floodplain Manas River	Point bars — clay plugs	12–48.8	0–600
Ghosh et al. (2015a)	India (West Bengal)	23° 55.064′N 88° 33.814′E	Holocene-Pleistocene floodplain Jalangi River	Point bars — clay plugs	50–150	64–131
McArthur et al. (2004)	India (West Bengal)	22° 44.463′N 88° 29.225′E	Holocene floodplain Hugli/Sunti Rivers	Ponded flood basin	<45	<1–1180
Ramos Ramos (2014)	Bolivia (Oruro district)	18° 36.007′S 66° 55.599′W	Holocene Quebrada Pazna River	Point bars	Max. 20	<5.6–142
Shah (2008)	India (UP)	25° 44.360′N 84° 09.510′E	Holocene floodplain Ganges River	Point bars — clay plugs	20–60	3–550
Zheng et al. (2005)	Bangladesh	23° 47.100′N 90° 36.180′E	Holocene floodplain Meghan River	Point bars — clay plugs	<28	<1–800

[a] Highest values: volcanic ash layers within fluvial stratigraphy.

Fresh clay samples were collected in the partly-filled Suhija oxbow lake (coordinates 25° 39.068′N, 84° 23.609′E; see Fig. 1 for location). Clay samples from the boreholes and the fresh clay-plug sediment were analyzed for organic carbon content with the Walkley–Black method (Walkley and Black, 1934). Total arsenic concentrations were measured from soil samples in both wells for analysis of the relation between lithofacies and arsenic content (Tables 2-3). Water samples of hand pump wells were collected from locations around the two boreholes for analysis of the spatial variation of arsenic in the point bar (Table 4). The soil extraction/digestion method was as follows: five (5 g) of soil sample was taken in a 100 mL conical flask and 50 mL of 0.5 M NaHCO3 solution was added. Next, the sample was shaken for 2 h in a "to and fro" horizontal shaker and after completion of shaking, the suspension was filtered through Whatman filter paper no. 42. Analysis of the water and sediment samples was performed with the SDDC (silver diethyldithiocarbamate) method using a UV1 Double Beam UV–vis Spectrophotometer (manufacturer: Thermo Scientific) with a precision level of total arsenic in the sediment of ± 0.9. Processing and analyses of the arsenic concentrations were done at the Dept. of Environment and Water Management (A.N. College, Patna, Bihar, India). Height measurements, grain size and lithology observations of a nearby point bar (measurement date 11 May 2011) in the Ganges River (Fig. 1) served as reference for the analysis of the lithofacies succession in the wells.

3. Results and discussion

3.1. Depositional processes, porosity–permeability distribution and geomorphology

The common denominator of all studies is their location on Holocene floodplains in a meandering river depositional setting (Table 1). The geomorphology consists of extensive flat floodplain areas with scattered curved, ribbon-shaped abandoned river bends that encompass crescent-shaped point bars (Figs. 1–2). The abandoned river bends are in different stages of sediment infill, ranging from (semi-) permanent oxbow lakes to completely sediment-filled clay plugs. Differential compaction resulted in topographical relief: the sand-prone point bars stand out above the compacted clay-prone floodplain and

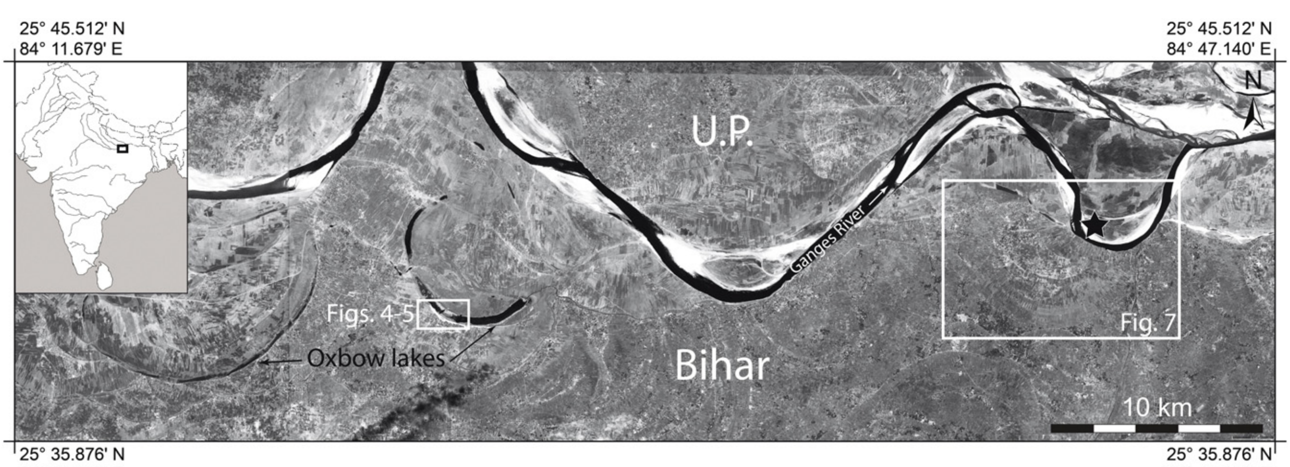

Fig. 1. Google Earth-Pro map of the study areas in Suhija and Bakhorapur (Bhojpur District). Star: location of the present-day point bar used in this study. Location of Figs. 4–5, 7 indicated. Fresh clay samples were taken for organic matter analysis at the location of Figs. 4–5.

clay plugs. Height differences between the highest part of the point bars and the surrounding floodplain in the studied locations are between 3 and 10 m. Villages are concentrated on the topographically high grounds as protection from river floods. The point-bar surfaces show concentric lines with higher and lower topography (or: ridges and swales), which are the expression of alternating sand and clay layers (Sahu and Saha, 2015). Agricultural plots and the dirt roads are aligned along the concentric lines, which facilitates the geomorphological mapping.

3.1.1. Abandoned river bends

Abandonment occurred after lateral expansion and rotation of the meander bends (Fig. 3), associated increase in sinuosity of the active river, and subsequent neck cut-off of the meander loop (Bridge, 2003). After neck cut-off the abandoned river bends converted to permanent standing bodies of water that were inactive in terms of river flow velocity. The oxbow lakes gradually filled with fine-grained sediment (silt and clay) that settled out of suspension in periods of river peak run-off and floodplain inundation. Decayed lacustrine plant and animal life, and organic waste from the hamlets bordering the oxbow lake all contribute to the high organic carbon content in the clay-plug sediment. In the last stage of sediment fill the oxbow lakes are used for rice growth. Analysis of time-lapse Google Earth-Pro imagery from 2003 to 2014, and corroborated by a site visit in January 2013, shows that a present-day oxbow lake (Suhija, Bhojpur District, Bihar) is rapidly filled with a mix of clay that settles from suspension after the yearly monsoonal floods, and water plants that grow from all edges to the lake center and form platforms for new vegetation (Figs. 4–5). When the infill is completed the oxbow lake is converted to a ribbon-shaped clay plug (Fig. 2). Initial porosity of the clay sediment in oxbow lakes is as high as 70–90% and the pore water content ranges from 50 to 80% (Singer and Müller, 1983). Upon continued sedimentation of clay in the oxbow lake, the gravitational loading causes strong porosity decrease by rearrangement of the randomly-oriented clay particles to a preferential orientation perpendicular to the compressional stress direction. Rearrangement takes place in the early stages of compaction, at pressures of only a few kg per cm^2 (Meade, 1966) and results in the formation of horizontally-laminated clay layers in a continuous, low-permeable ribbon-shaped sediment body that encompasses the point bar, and in the expulsion of interstitial water from the clay plug to the adjacent higher-permeable point-bar sand body.

Table 2
Arsenic concentrations in sediment samples of Well 1.

Pipe sample nr.	Sample depth (m)	As-concentration (μg/g)	Depositional unit	Lithofacies association
P1	0.65	2.24		
P2	1.35	3.20		
P4	2.75	50.30		
P6	4.15	1.20	Point bar sand	2
P8	6.25	2.40		
P9	6.95	0.55		
P12	9.05	1.40		
P15	11.15	1.00		
P18	13.25	5.80		
P19	13.95	1.60		
P22	16.05	2.00		
P26	18.85	0.10	Point bar sand	2
P27	19.55	1.60		
P28	20.25	0.30		
P30	21.65	0.10		
P32	23.05	1.26		
P38	27.25	14.40		
P41	29.05	14.40		
P43	30.15	4.70		
P45	31.35	7.80		
P48	33.45	6.20		
P50	34.75	19.80		
P53	36.85	28.80		
P56	38.05	1.42	Braided river	
P57	38.65	0.90	conglomerate and	1
P60	40.45	1.90	gravelly sand	
P65	43.35	0.90		
P68	44.75	0.52		
P70	45.75	13.80		
P71	46.05	0.63		
P74	47.45	9.90		
P75	47.95	2.10		
P77	48.85	16.60		

Table 3
Arsenic concentrations in sediment samples of Well 2.

Pipe sample nr.	Sample depth (m)	As-concentration (μg/g)	Depositional unit	Lithofacies association
P1	0.65	6.97	Clay plug	3
P2	1.35	0.05		
P5	3.45	3.93		
P6	4.15	5.85		
P9	6.25	2.42		
P11	7.65	3.50		
P15	10.65	4.15		
P18	12.75	0.60		
P20	14.15	3.25		
P23	16.25	2.20		
P25	17.65	0.94	Point bar sand	2
P26	18.35	1.17		
P29	20.45	2.79		
P31	21.85	1.27		
P34	23.95	1.14		
P35	24.65	8.35		
P38	26.75	0.92		
P43	30.25	7.41		
P44	30.95	0.14		
P45	31.65	5.52		
P46	32.35	39.10	Braided river	1
P47	33.05	8.86	conglomerate and	
P48	33.75	6.94	gravelly sand	
P50	35.15	4.76		
P52	36.55	2.27		
P53	37.25	8.02		
P54	37.95	0.72		
P58	40.75	8.95		
P61	42.85	1.20		
P63	44.25	1.33		
P64	44.95	1.53		
P66	46.25	4.57		
P69	48.45	10.95		
P70	49.15	7.61		
P72	50.55	2.93		

3.1.2. Point bars

In plan-view, point bars are crescent-shaped geomorphological elements in the convex inner bend of the river (Figs. 2–3). Under normal flow conditions, sediment accumulates onto the point bar by the process of helicoidal flow in the meander bend (Schumm, 1977) and migration of sand dunes obliquely out of the channel floor onto the point-bar surface. Up the point-bar slope the flow energy gradually decreases and the sand dunes come to rest onto the point-bar surface, thereby causing expansion (*lateral accretion*) of the point bar. The up-slope flow-energy decrease results in a vertical fining-upward grain-size succession. In flooding periods the river water expands onto the floodplain, hence the flow energy decreases and fine-grained sediment settles out of suspension and forms a clay drape on the point bar surface. In cross-section the inclined clay and sand layers and a distinct wedge shape define the two-dimensional architecture of point-bar (Fig. 6). In plan-view the heterolithic lithofacies distribution is expressed as a ridge-and-swale topography by the process of differential compaction. The vertical succession and lateral accretion surfaces reflect the expansion in space and time of the inner bend of a meandering river over its channel

Table 4
Arsenic concentrations in water samples of tube wells in the area surrounding the well locations of the present study. See also: Fig. 7.

Sample nr.	Location	Coordinates	Depth (m)	As concentration (μg/L)
01	Bakhorapur	25° 39.870′N 84° 40.993′E	15.2	10.1
02	Bakhorapur	25° 39.941′N 84° 40.697′E	24.4	63.5
03	Bakhorapur	25° 39.941′N 84° 40.702′E	18.3	38.5
04	Bakhorapur	25° 39.756′N 84° 40.613′E	21.3	27.6
05	Sabalpur, Gangali	25° 40.013′N 84° 40.931′E	18.3	2.2
06	Sabalpur, Gangali	25° 39.845′N 84° 40.970′E	15.2	14.6
07	Sabalpur, Gangali	25° 39.835′N 84° 40.972′E	15.2	6.5
08	Sabalpur, Gangali	25° 40.018′N 84° 40.973′E	21.3	223.5
09	Neknam Tola	25° 40.111′N 84° 40.770′E	15.2	48.4
10	Neknam Tola	25° 40.138′N 84° 40.783′E	24.4	232.0
11	Neknam Tola	25° 40.209′N 84° 40.647′E	12.2	14.4
12	Hajipur	25° 40.193′N 84° 40.685′E	13.7	15.3
13	Bakhorapur	25° 39.936′N 84° 40.663′E	12.2	14.3
14	Dubey Chapra	25° 38.776′N 84° 40.436′E	13.7	7.0
15	Dubey Chapra	25° 38.776′N 84° 40.456′E	27.4	70.0
16	Gundi	25° 38.659′N 84° 39.784′E	21.3	11.5
17	Gundi	25° 38.891′N 84° 39.794′E	15.2	22.7
18	Gundi	25° 39.039′N 84° 40.059′E	24.4	338.0
19	Kali Mandir,Raknam Tola	25° 39.447′N 84° 39.918′E	36.6	7.0
20	Raknam Tola	25° 39.477′N 84° 39.872′E	30.5	52.1
21	Gundi	25° 40.291′N 84° 41.726′E	33.5	11.5

floor deposits. Newly deposited and unconsolidated sand is loosely-packed and has an initial porosity of 35–50% (Stone and Siever, 1996). Early-diagenetic gravitational compaction at shallow burial depth involves the mechanical rearrangement to a tighter packing of the grain framework and slight reduction of the porosity to 26% (Graton and Fraser, 1935; Boggs, 2009). The complex spatial arrangement of point-bar grain sizes is reflected in the porosity and permeability heterogeneity of this geomorphological element. The clay plug may be underlain by a thin but continuous layer of bed-load transported clean channel-floor sand with high porosity and permeability (Donselaar and Overeem, 2008). This sand layer laterally continues in the lower part of the point bar. Up the point bar the grain-size decrease from sand to silt and clay (Fig. 6) corresponds with a decrease in porosity and permeability (Xue, 1986; Pranter et al., 2007; Donselaar and Overeem, 2008). The sigmoidal-shaped clay drapes in the upper part of the point bar constitute internal permeability baffles that may compartmentalize the point-bar geomorphological element in terms of permeability. Water injection experiments in hydrocarbon-bearing fluvial reservoir sandstones

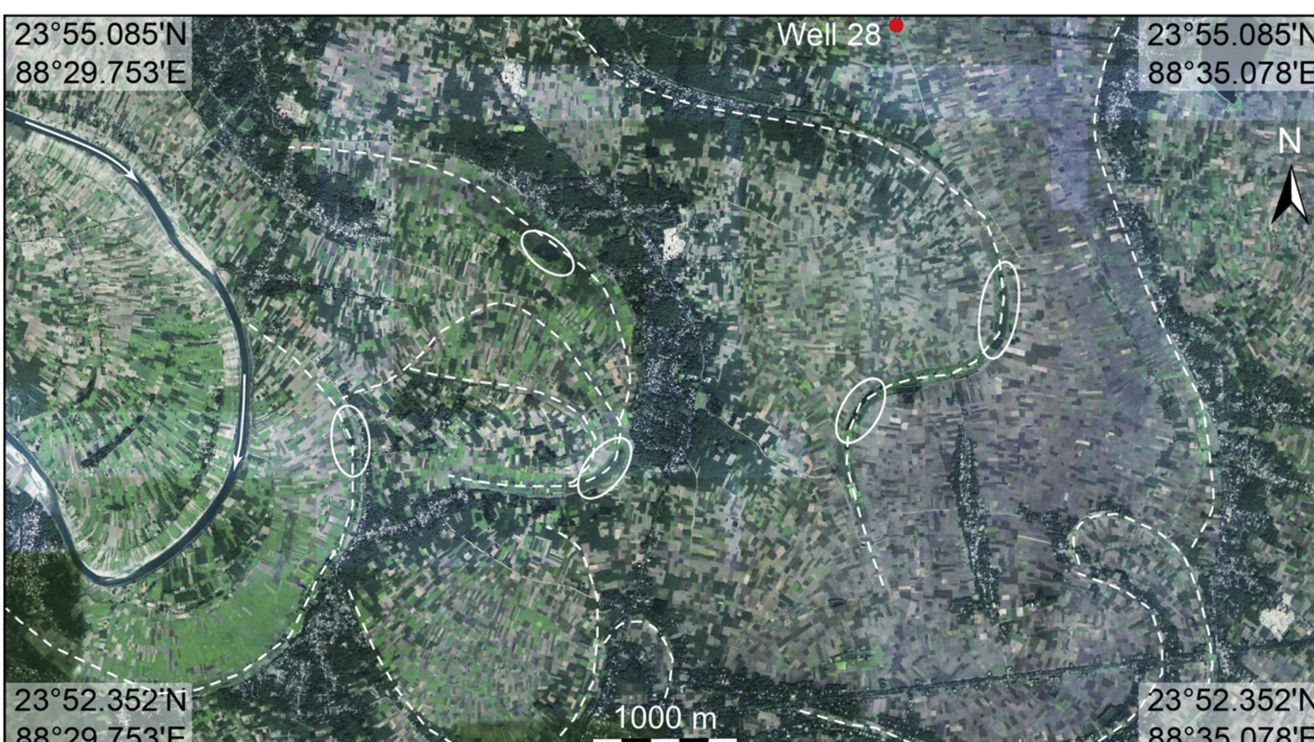

Fig. 2. Google Earth-Pro image of the omnipresent point-bar and oxbow-lake geomorphological elements in the Holocene floodplain of the Jalangi River (Nadia District, West Bengal, India). Study area Ghosh et al. (2015a); their Well 28 indicated. Arrows: flow direction Jalangi River. Dotted lines: ribbon-shaped sediment-filled abandoned river bends with remnant lakes (circled). Note that the agricultural plots, trees, villages and roads align with the geomorphology.

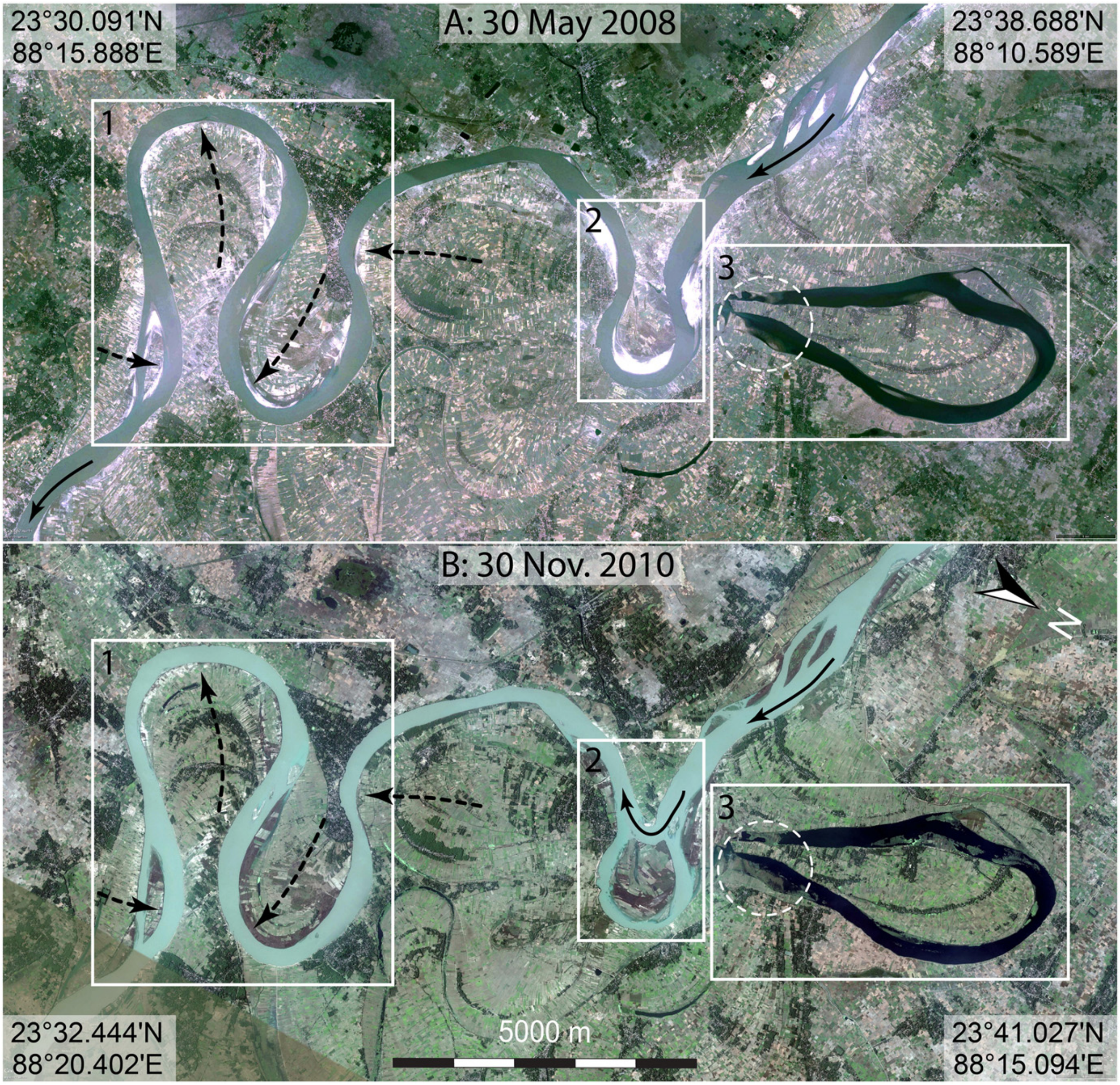

Fig. 3. Successive stages of meander-bend cut-off and abandonment, Hooghly River (West Bengal, India). A: Google Earth-Pro image date 2008-05-30. 1: Highly sinuous river pathway. Point-bar expansion (dashed arrows) and simultaneous erosion of the opposite outer bank will eventually result in meander-bend neck cut-off. 2: Location of the future neck cut-off. 3: Cut-off meander bend is converted to an oxbow lake. Note the partial sediment infill (dashed circle). B: Google Earth-Pro image date 2010-11-30. 2: Recent meander-bend neck cut-off. Water in the bend still in communication with the river. 3: Note the increased sediment infill of the oxbow lake in comparison with image A.

showed that fluid flow is preferentially concentrated in the clean channel sandstone and the lower part of the point bar (Lasseter et al., 1986).

3.1.3. Floodplain

The floodplain is the largest geomorphological element in terms of surface area and volume. Sediment accumulation takes place by settling of clay and silt out of suspension in periods of peak river discharge and subsequent flooding. Local topographic depressions are the locus of shallow ephemeral or permanent ponds. Continued deposition of clay and silt on the floodplain leads to compaction and the strong reduction of initial porosity. In hydrocarbon reservoir modelling studies the compacted floodplain deposits are considered impermeable non-reservoir that hamper connectivity of the fluvial channel and point-bar sandstone (e.g. Larue and Hovadik, 2006).

3.2. Core analysis

The point bars are the most complex geomorphological element in terms of shape, size, and spatial distribution of the lithofacies, and inherently to porosity and permeability heterogeneity which in turn conditions the aquifer flushing efficiency. Population concentrates on the topographically-elevated point bars, and it is here that the arsenic pollution problem is most pronounced.

The studied point bar in Bakhorapur (Bhojpur District) has a surface area of 16 km^2 (Fig. 7). The highest elevation (62 m above sea level) is in the NW corner of the point bar; the surrounding clay plug stands at 55 m above sea level. The stratigraphic succession in both wells consists of three lithofacies associations. The lowermost lithofacies association is separated from the overlying lithofacies associations by a sharp break at

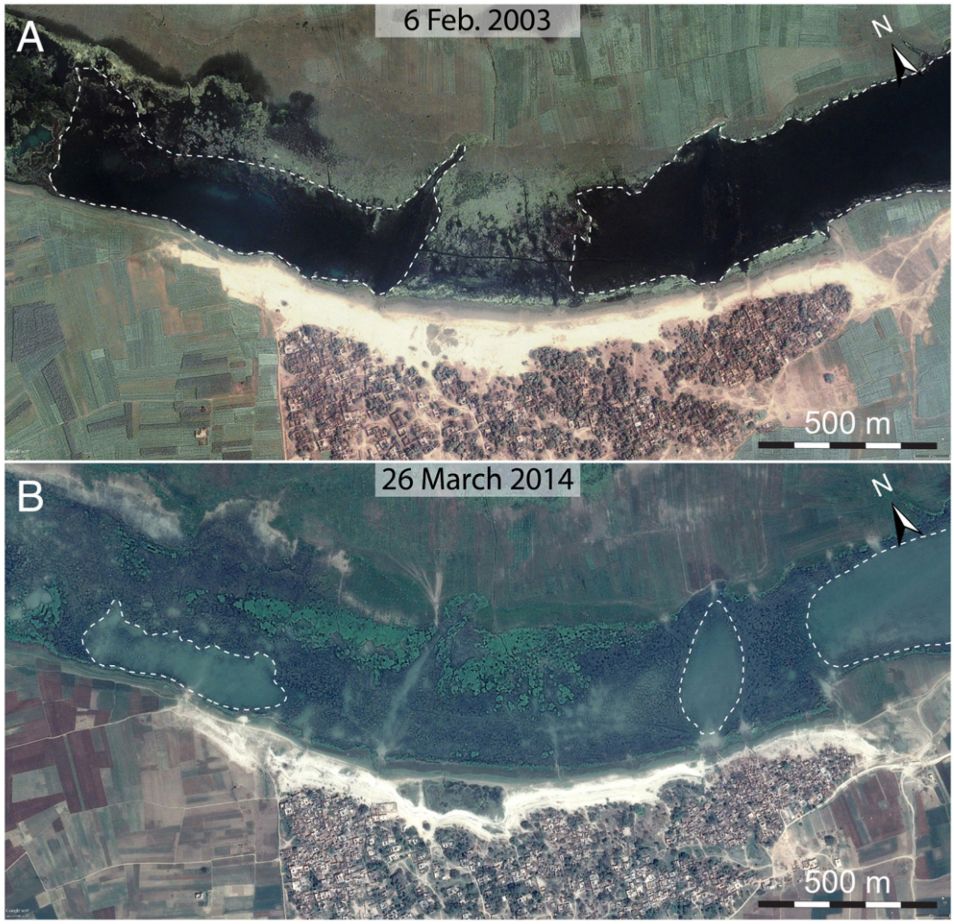

Fig. 4. Google Earth-Pro image sequence of rapid vegetation infill of the Suhija oxbow lake. A: Google Earth-Pro image date 6 Feb. 2003; B: Google Earth-Pro image date 26 March 2014. See Fig. 1 for location. Dotted lines outline the lake surface.

~28 m depth (Fig. 8). The break is interpreted as a sequence boundary which marks the southward shift of the Ganges River belt to this area, with truncation of the upper part of the underlying lithofacies association.

3.2.1. Lithofacies association 1

The lower lithofacies association consists of 0.2–7.0-m-thick conglomerate layers and 0.2–2.5-m-thick, coarse-grained gravelly sand layers (Fig. 9A). Conglomerates are clast-supported, very poorly-sorted, sub-rounded quartz and feldspar granules, pebbles and cobbles (clast diameter up to 30 mm) in a medium- to coarse micaceous sand matrix. The conglomerate layers have a sharp lower surface and a gradually fining top. The gravelly sand layers locally show cross-bedding. Permeability is very high, to the point that the drilling mud (bentonite) has completely invaded the core. This sequence is interpreted as formed by shallow braided rivers. The mineralogy and coarse grain size suggest that the source area of the rivers was in the proximity, and most likely in the south, on the stable Indian Craton.

Fig. 5. Outcrop photo of the vegetation infill. Suhija oxbow lake.

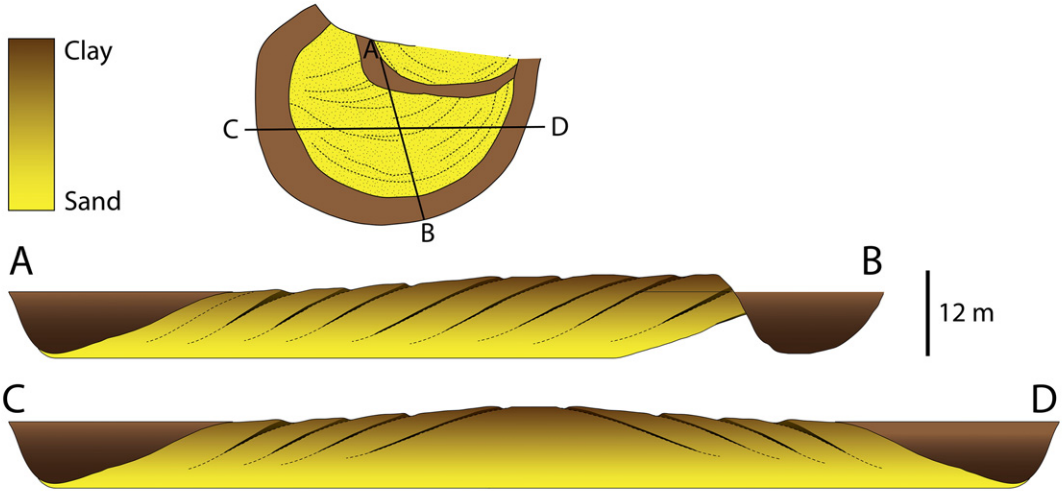

Fig. 6. Transverse and longitudinal cross sections through a point bar. Plan-view drawing corresponds to the studied point bar in Fig. 7.

Arsenic concentrations in sediment of this lithofacies association (Tables 2–3) range from 0.52–39.10 μg/g sediment (average: 7.95 μg/g). The arsenic concentrations in all lithofacies are within the range of 1–13 μg/g as published by Håkanson (1980).

3.2.2. Lithofacies association 2

Lithofacies association 2 consists of a heterolithic succession of medium- to fine-grained, laminated sand, silt and clay, organized in 5–12 m-thick, fining-upward grain-size successions (Fig. 8, Well 01). The sand is light grey, moderately sorted, medium- to very fine-grained and micaceous. Clay pebbles may line the base of the sand layers. The sand is loosely packed, unconsolidated and has a high porosity. Sedimentary structures comprise cross-lamination and horizontal lamination, accentuated by heavy mineral concentrations (Fig. 9B–C). Dark-grey, laminated silt layers characterize the upper part of the fining-upward succession. The frequency and layer thickness increases towards the top of each succession. The silt layers contain organic matter (plant debris), are compact and have a lower porosity than the sand. Lithofacies association 2 represents the vertical stacking of successive generations of Ganges River point-bar sediment. The lithofacies of the present-day

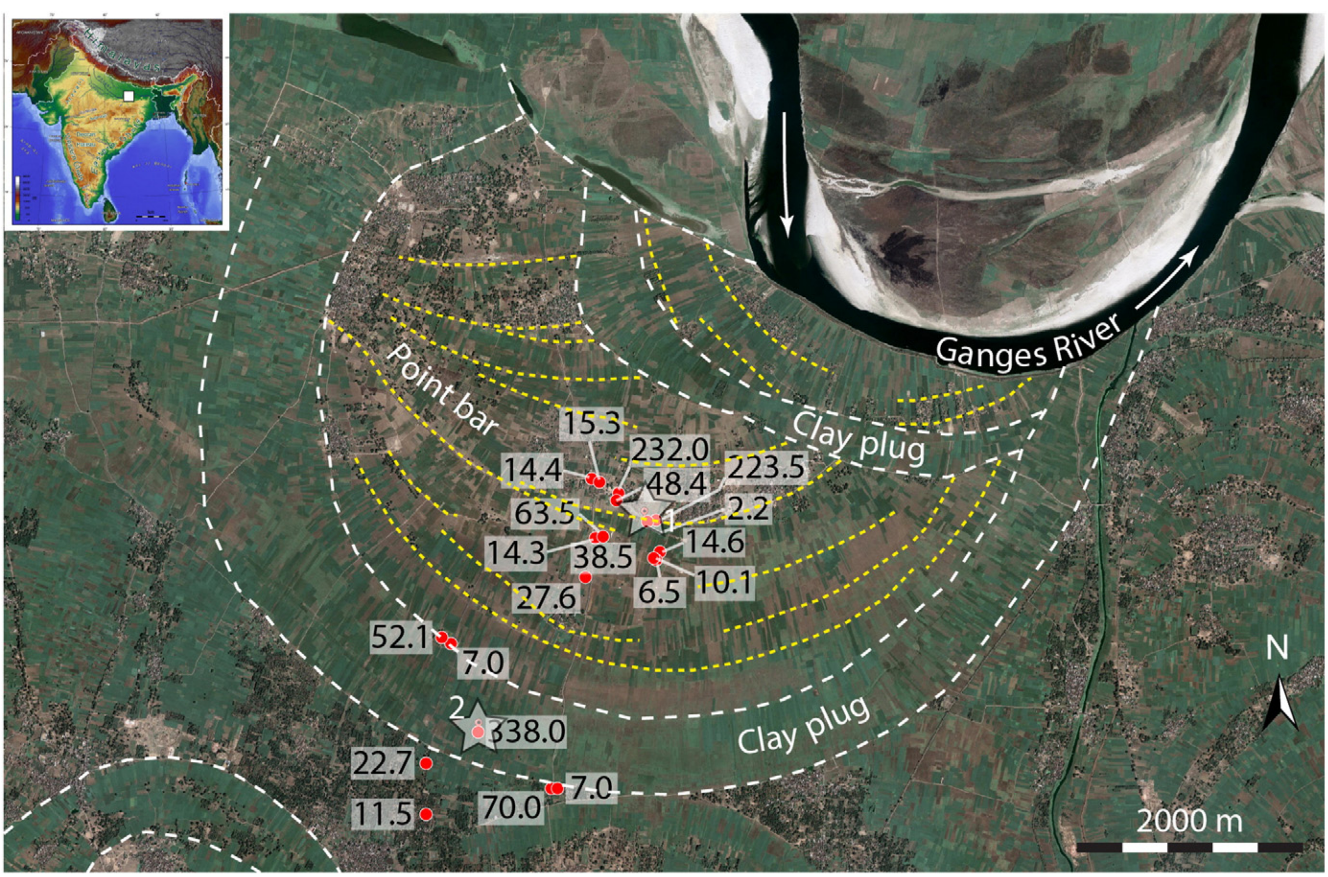

Fig. 7. Interpreted Google Earth-Pro image of the studied point bar and clay plug. Bakhorapur, Bhojpur District. White dashed lines: clay plugs. Yellow dashed lines: ridge-and-swale morphology on the point bar. Stars with numbers 1 and 2: Well locations. Arsenic concentrations from shallow tube wells are indicated (μg/L). Refer to Table 4 for tube-well location names. Note the relation between the geomorphology and the radial orientation of the agricultural plots.

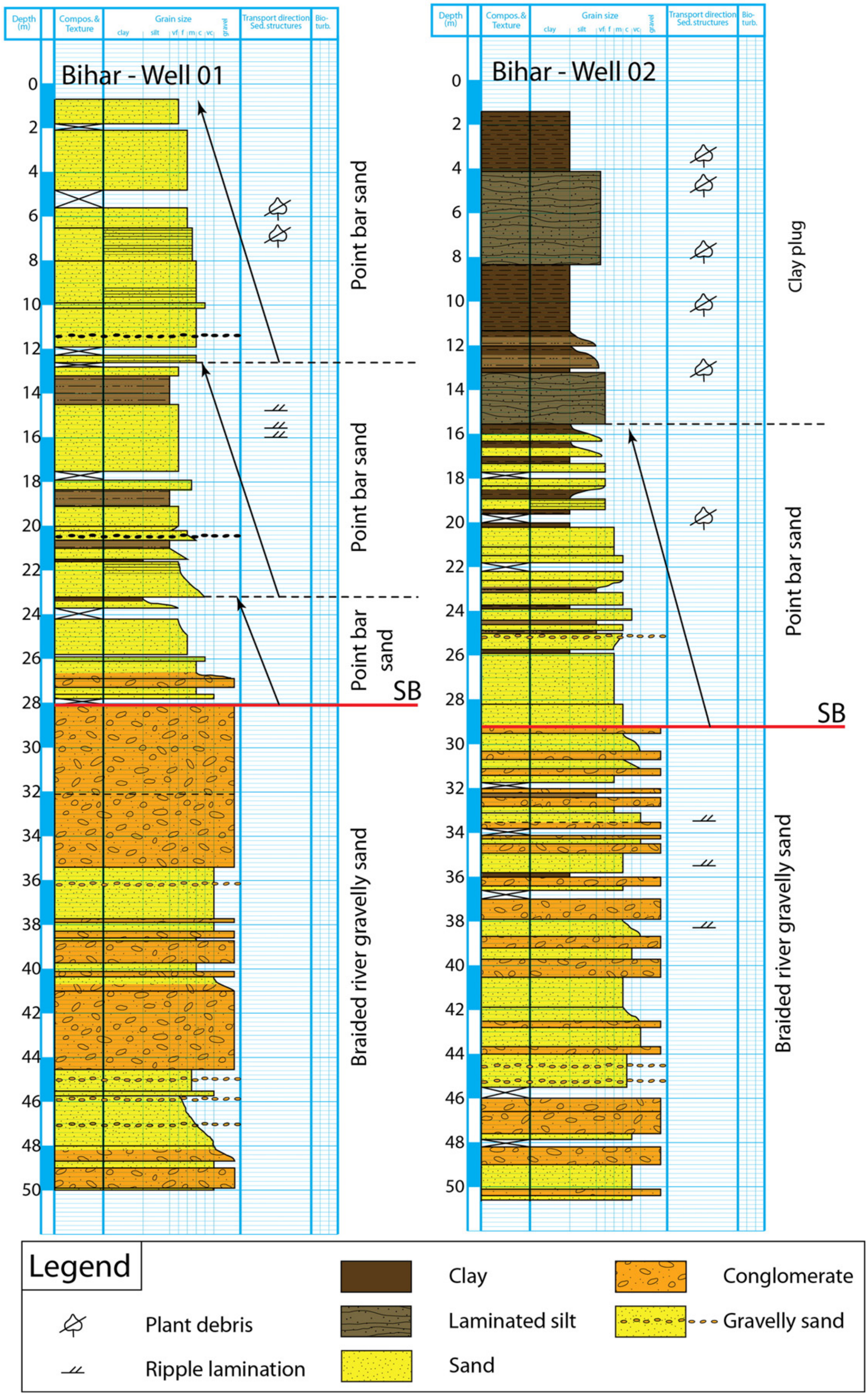

Fig. 8. Core description logs. See Fig. 7 for well locations.

Ganges River point bars in the vicinity of the wells (Fig. 1) is in agreement with the well description, and shows a fining-upward grain-size succession of medium- to fine-grained micaceous light-grey sand at the base to fine-grained at the top. Lateral accretion surfaces with clay drapes and climbing-ripple lamination are the most conspicuous sedimentary structures in the present-day point bar sediment.

Arsenic concentrations in sediment of lithofacies association 2 (Tables 2–3) range from 0.10–8.35 μg/g sediment with one outlier of 50.30 μg/g in Well 1 at 2.75 m depth. The average arsenic concentration is 4.03 μg/g inclusive of the outlier, and 2.18 μg/g without the outlier.

3.2.3. Lithofacies association 3

The uppermost, 12-m-thick point bar sand in Well 1 correlates with the equally-thick lithofacies association 3, which forms the upper part of Well 2 (Fig. 8). Lithofacies association 3 comprises dark-grey to black silt and clay (Fig. 9D). The layers are compact and have a low porosity. The silt layers are horizontally-laminated. The sediment contains organic matter (plant debris) and has a distinctive H_2S smell. The silt and black clay in the upper part of Well 2 are rich in organic carbon; laboratory analysis from clay-plug samples in Well 2 yielded organic matter (OM) content of 2.35% (wet sample) and 2.58% (dry) and organic carbon (OC) content of 1.36% and 1.50%, respectively. Lithofacies association 3 characterizes the clay-plug fill of the oxbow-lake that surrounds the Bhojpur point bar. Thickness measurements at a point bar along the present-day Ganges River just north of the well locations (Fig. 1) showed a total height above the water line of 8 m. Local boatmen calculated the water depth of the Ganges River at the time of the measurements at 4 m. The total 12 m height difference very well matches the thickness of the uppermost point-bar succession in Well 1 and the corresponding clay plug in Well 2.

Arsenic concentrations in sediment of this lithofacies association (Tables 2–3) range from 0.05–6.97 μg/g sediment (average: 3.29 μg/g).

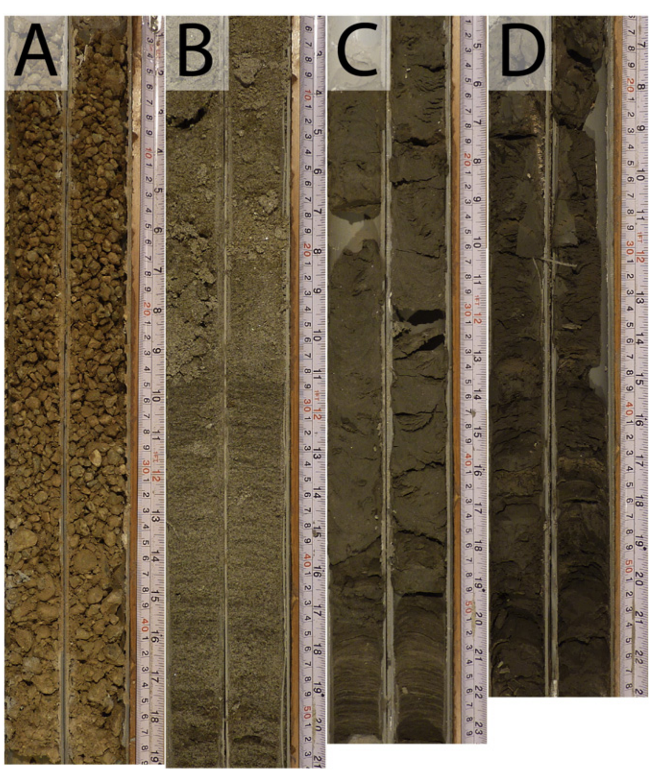

Fig. 9. Core photographs of the lithofacies types in wells 1 and 2. A: Lithofacies association 1: clast-supported conglomerates. Well 1, 38.7–39.1 m. B: Horizontally-laminated, medium-grained sand. Well 1, 6.35–6.85 m. C: Horizontally-laminated very-fine sand fining-upward to silt. Well 1, 14.1–14.6 m. D: dark-grey to black clay. Well 2, 10.16–10.60 m.

3.3. Discussion

Oxbow lakes are standing bodies of water. Water circulation in the lake is limited to the upper part of the water column (epilimnion) and here O_2 diffusion takes place, and plant and animal life thrive. By contrast, the deeper part of the lake (hypolimnion) is poorly oxygenated by the lack of water circulation. The remaining oxygen is consumed by bacterial activity and, as a consequence, depletion of dissolved O_2 occurs which results in an anoxic environment (Nichols, 2009). Plant and animal remains settle out of suspension from the epilimnion on the clay sediment of the lake bottom, and are preserved as organic matter in the absence of oxygen (Fig. 10A). The anoxic conditions in the hypolimnion of the oxbow lake constitute the first source of reactive organic carbon and microbially-mediated reductive dissolution of arsenic from iron oxy-hydroxides (Nickson et al., 2000; McArthur et al., 2001, 2004; Ravenscroft et al., 2001; Postma et al., 2007).

Upon completion of the infill of the oxbow lake, the clay-plug sediment contains co-deposited sedimentary organic carbon. Ghosh et al. (2015b) and Ghosh (2016) reported TOC values of 0.7% in clay deposits at shallow depths (6–12 m) in three boreholes in Nadia district, West Bengal, India (coordinates 22° 56.401′N, 88° 32.389′E; 23° 55.064′N, 88° 33.350′E; 23° 56.352′N, 88° 33.814′E) in a Holocene meandering river geomorphology. The TOC peak is interpreted in the present paper as clay-plug sediment, similar to the one found in the present study. The fresh clay-plug samples from the present-day Suhija oxbow lake (Fig. 5) are black in color, smell of H_2S and have OM and OC concentrations of 1.22–2.64% and 0.71–1.53%, respectively (wet sample). The clay sample from Well 2 (at 24 m depth) is dark-grey to black and has concentrations of 2.35% OM and 1.36% OC (wet sample), and 2.58% OM and 1.50% (dry sample). The clay-plug sediment constitutes the second source of reactive organic carbon (Harvey et al., 2002, 2006; Neumann et al., 2010; Mailloux et al., 2013; Desbarats et al., 2014), where compaction of accumulating clay in the up to 12-m-thick clay-plug successions triggers the expulsion of organic-carbon containing pore fluids to the adjacent permeable point-bar sands (Fig. 10B). Here, the diffusion process of reductive dissolution of iron oxy-hydroxides will release the arsenic from its solid state in the point-bar sands, and the dissolved arsenic enters the aquifer. Arsenic-concentration measurements from tube wells around the well locations (Table 4 and Fig. 7) show high and very variable values (2.2–338 μg/L, with an average of 64.5 μg/L) in the depth range (12.2–27.4 m) of the point-bar successions of lithofacies association 2.

Desbarats et al. (2014) focused their reactive solute transport model on the groundwater flushing in the clay plug (their channel-fill unit) of the geomorphology. In the present study it is proposed that the permeable point-bar sands in contact with the anoxic hypolimnion of the oxbow lake and, after infill of the lake with a clay plug, the permeability contrast between the low-permeable clay-plug sediment and the neighboring high-permeable point-bar sand cause net transfer of organic-carbon containing pore fluids along the permeability gradient. In the case that the permeable point-bar sands are completely surrounded by low-permeable clay-plug sediment (Figs. 2–3, 7) the permeability contrast will effectively isolate the aquifer from the surrounding subsurface, hence the aquifer flushing efficiency is poor and released arsenic will be trapped and accumulates to high concentrations. In the case of an incomplete entrapment the point bar aquifer is in communication with the surrounding subsurface, hence the aquifer flushing efficiency is high and the arsenic will be diluted in the aquifer flow. Extraction of groundwater for consumption and irrigation is highest on the high grounds of the point bars where the villages are concentrated. The extraction creates a pressure gradient which will draw the arsenic-polluted groundwater towards the tube wells and aggravate the health risk. The fluid flow path is conditioned by the permeability distribution within the point-bar sands (Lasseter et al., 1986; Xue, 1986; Pranter et al., 2007; Donselaar and Overeem, 2008). Fluid will preferentially migrate along the high-permeable base of the point-bar sand body and migrate

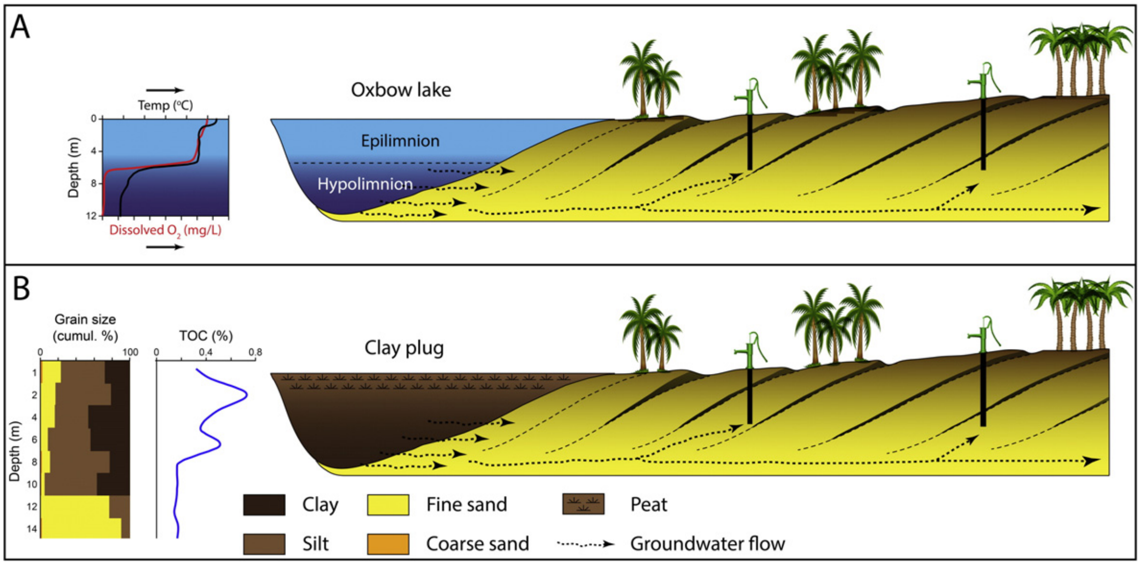

Fig. 10. Generic geological model for the release of arsenic by juxtaposition of clay plug and point bar. A: Temperature and oxygen stratification in oxbow lake. B: Clay infill of the oxbow lake. Arrows indicate expulsion of the pore fluid to the adjacent permeable point bar.
Grain size and TOC data from Ghosh et al. (2015b).

up to the down-hole mouth of the tube well. The upper part of the point bar sediment comprises alternating, inclined layers of permeable sand draped by impermeable clay (Figs. 6, 10). The heterolithic lithofacies distribution causes permeability baffles to fluid flow (Lasseter et al., 1986) which compartmentalize the aquifer. The location of the tube wells determines the fluid flow draw-up from the base of the point bar, whereby large arsenic-concentration differences can occur over short distances in neighboring compartments.

On the basis of the geomorphological juxtaposition of point-bar sand and clay plug, the lithofacies characteristics and inherent permeability contrast between both, and the omnipresence of the geomorphological element pair in the Holocene floodplain environments around the world, a generic geological model is proposed here in which: (1) the anoxic hypolimnion waters and the clay plugs are the loci of microbial respiration that triggers the reductive dissolution of iron oxy-hydroxides and the release of arsenic on the scale of entire fluvial floodplains and deltaic basins such as analyzed in Table 1, and (2) juxtaposed point bars accumulate the arsenic by permeability isolation and poor aquifer-flushing efficiency.

The conglomerates and gravely sands (lithofacies association 1) have an average arsenic concentration of 7.95 μg/g, which is more than twice the value of the other two lithofacies associations. By contrast, the arsenic concentrations from water samples in the deeper tube wells (Table 4) are lower than those of the shallower samples. The lower values are directly related to the depositional setting which consists of coarse-grained, very-permeable braided river sediments which lack clay plugs. Aquifer flushing efficiency is high in such depositional setting and will have flushed out the dissolved arsenic.

Pleistocene aquifers in the Ganges River and Ganges-Brahmaputra-Meghna Delta are generally considered arsenic-free (BGS and DPHE, 2001; Acharyya and Shah, 2007; Shah, 2008, 2010). The so-called orange-brown sand (McArthur et al., 2004) or Older Alluvium (Shah, 2008, 2010) aquifers formed as a prograding clastic wedge during Pleistocene glacio-eustatic lowstand and are characterized by coarser-grained sand, laterite soils and calcretes (Acharyya and Shah, 2007; Ghosh et al., 2015b) deposited as braided rivers in large incised-valley fills separated by interfluve areas (Tandon et al., 2006). It is proposed here that lower coastal plain meandering river deposits are absent in such setting, and that this implies a high flushing efficiency whereby arsenic is effectively removed from the aquifer (Acharyya and Shah, 2007).

Awareness of the relation between geomorphological setting and the variability in arsenic-pollution concentrations in Holocene shallow aquifers is crucial in the predictive spatial modelling of arsenic-safe aquifers, and thus contributes to the development of arsenic mitigation strategies.

4. Conclusions

Arsenic pollution is widespread in shallow aquifers of Holocene fluvial and deltaic flood basins. An inventory of studies dealing with the spatial variability of arsenic pollution in different flood basins around the world shows that in all cases the geomorphological setting consists of meandering river deposits with sand-prone fluvial point-bar deposits surrounded by clay-filled (clay-plug) former meander bends (oxbow lakes). Arsenic concentration levels in the aquifers of these geomorphological elements are characterized by large lateral and vertical variability, which is here interpreted as directly related to the lithofacies heterogeneity and inherent permeability contrasts which condition the fluid flow paths in the aquifer. The high organic matter content and anoxic conditions in the hypolimnion of the oxbow lake, and the clay-plug sediment bordering the fluvial point bars are identified as the loci of reactive organic carbon that triggers the release of arsenic by microbially-mediated reductive dissolution of iron oxy-hydroxides. On the basis of the geomorphological juxtaposition of point-bar sand and clay plug, the lithofacies characteristics and permeability heterogeneity of both, and the omnipresence of the two geomorphological elements in all studied Holocene floodplain settings, a generic geomorphological model is proposed to explain the spatial arsenic concentration variability, and the migration and accumulation paths of dissolved arsenic on entire flood-basin scale.

Acknowledgments

The authors thank their colleagues Nupur Bose (A.N. College, Patna, India) and Hans Bruining (TU Delft) for their discussions on the geology of arsenic pollution, and for the invaluable help with the data acquisition and analysis. Sotiria Lourma and Guido Schaepman are acknowledged for their data acquisition and analysis as part of their TU Delft MSc final thesis work. Damien Boeser is acknowledged for preparing Google Earth interpretation maps as part of his TU Delft BSc thesis. We are grateful for the constructive comments and valuable suggestions of the three anonymous reviewers. Financial support for this study came from the European Union Erasmus Mundus EURINDIA — Lot 13 program. Grant Agreement Number 2009 – 1665/001- 001- ECW.

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