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Influence of basin-wide geomorphology on arsenic distribution in Nadia district

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ABSTRACT

The present study depicts the geospatial relation between basinal geomorphology and heterogeneous arsenic (As) distribution in the Bengal Delta Plain (BDP). The distribution pattern largely varies throughout the study area (higher: Karimpur-II As_T average 214.73 μgL^{-1}; lower: Tehatta As_T average 27.84 μgL^{-1}). Both safe (low As) and unsafe (high As) areas are identified within the single shallow aquifer (<50 m), where they are in close vicinity. Statistical analysis shows that Padma river basin is the most contaminated (As_T avg. 214.7 ± 160 μgL^{-1}) and Churni-Ichhamati river basin (As_T avg. 108.54 ± 89.43 μgL^{-1}) is the least contaminated with groundwater As. Moreover, the role of geomorphological features influencing the geospatial distribution of As has been studied and meandering features are found to correlate with high As wells ($r^2 = 0.52$), whereas, natural levees are correlated with safer wells ($r^2 = 0.57$). In the meandering features, the deposition of sedimentary organic matter (SOM) facilitates the reduction of As bearing Fe(III) oxy-hydroxides and subsequent higher As mobilization. In natural levees, surface derived labile organic matter (DOC and FOM, Fresh Organic Matter) from different land-use patterns (Habitation, degraded waterbodies, cattle dwelling, sanitation, etc.) is transported to shallow aquifers (notably protein rich leakage sewage). The fresh organic carbon transported to the shallow aquifers, thereby triggering As release by microbe-mediated reductive dissolution of hydrated Fe(III)-oxides (HFO). Iron reduction (mostly amorphous) is playing an important role in the release of As depending on basin-wise sedimentation pattern, local recharge, accumulation of silt/clay/micas at the top with corresponding reactive oxidation of organic carbon. These are important components and often helping the cyclic water-rock interaction of As causing such heterogeneous geospatial distribution. The delineation of aquifer with regard to safer and unsafe areas would immensely help to supply safe drinking water to the rural community.

1. Introduction

Inorganic arsenic (As) is widely present in groundwater of the shallow aquifers (<50 m) of south Asia [especially the western part of Bengal Delta Plain (BDP), India] (Mukherjee and Bhattacharya, 2001; Bhattacharya et al., 2002a, Bhattacharya et al., 2002b; Bhattacharyya et al., 2003a; Ahmed et al., 2004; Nath et al., 2007, 2008a; Mukherjee et al., 2008; van Geen et al., 2008; Chatterjee et al., 2010a; Fendorf et al., 2010). Usually, the groundwater As concentration in BDP is much higher than the WHO guideline value (As_T <10 μgL^{-1}) (Bhattacharyya et al., 2003b; Ahmed et al., 2004; Rahman et al., 2014). Since the early 80's, many health-related problems have been reported from rural areas of West Bengal, India (Saha, 1984; Guha Majumdar et al., 1988). The drinking water wells (both private and community wells) have been depicted as unsafe raising groundwater quality concerns (PHED, 1993; CGWB, 1999). The extent of the problem is so much threatening that nearly 45 million of rural people of BDP (80% from rural and remote areas) are under high risk from groundwater arsenic exposure

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(RGNDWM, 2001; Chatterjee et al., 2003).

Groundwaters from the alluvial aquifers are usually considered safe from microbial contamination and are frequently used for domestic (cooking, bathing and drinking) and agricultural purposes. Since the last decade, the use of groundwater has been extensively increased and mostly shallow aquifers (<50m) are exploited for these purposes, especially for irrigation started during the 'Green Revolution' (McLellan et al., 2002). During this period (1970-80), a large number of shallow wells (<50m) have been installed.

The mechanism of As release into groundwater has been a matter of serious debate (Stuben et al., 2003; McArthur et al., 2004; Chatterjee et al., 2005, Chatterjee et al., 2010b; Harvey et al., 2006; Mukherjee

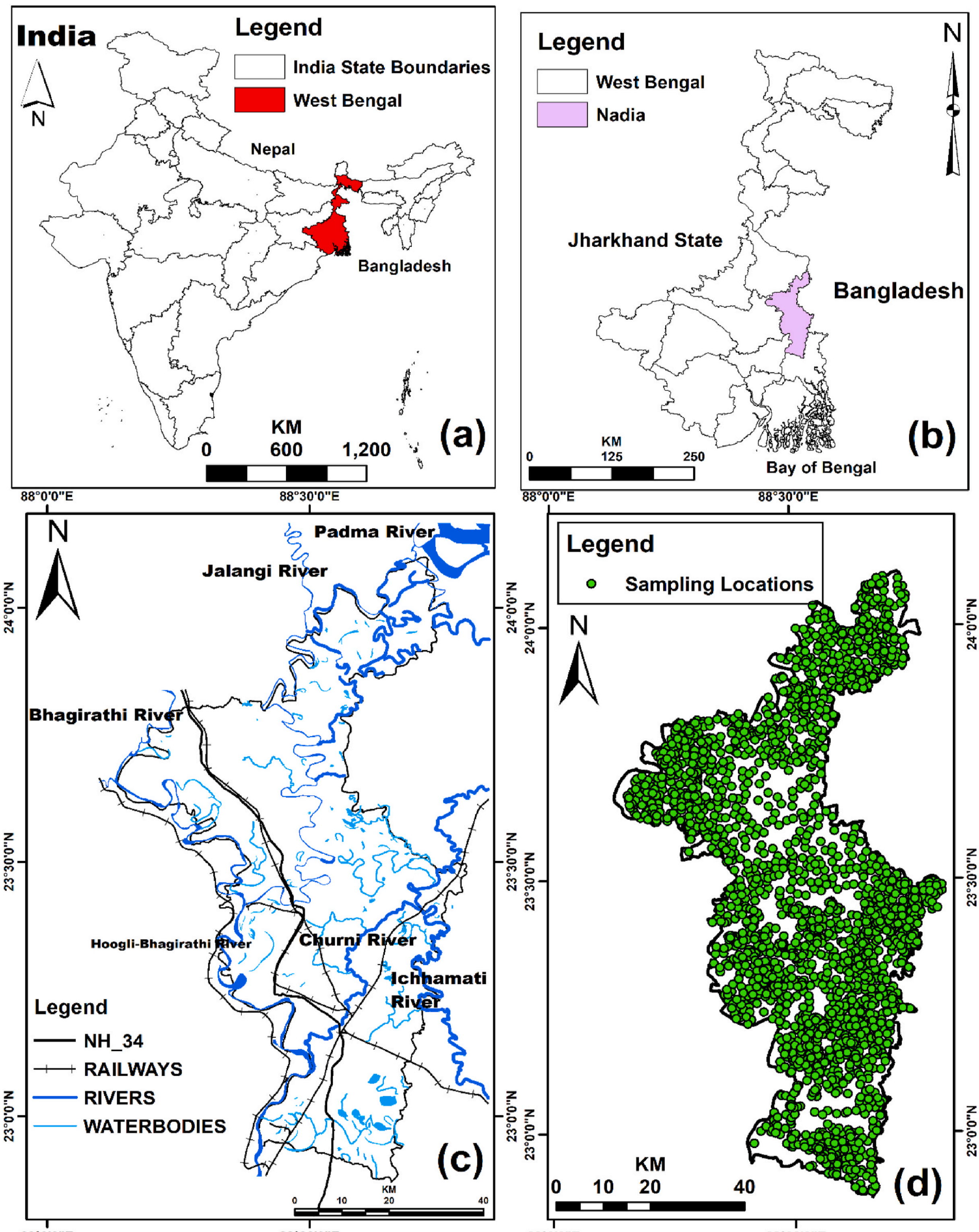


Fig. 1. Study area locations map: (a) India, (b) West Bengal, (c) Nadia district map showing rivers, (d) Sampling locations (green dots, n = 2448 at shallow depth, < 50 m). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

et al., 2008; Neumann et al., 2010). Recent studies raised many questions about prevalent host sedimentary environment including the presence of organic matter with different characteristics, that could in turn play a crucial role for As mobilization in groundwater (Nath et al., 2007; Mukherjee et al., 2008; Lawson et al., 2013; Majumder et al., 2016; Mladenov et al., 2010, 2015; Kulkarni et al., 2017, Kulkarni et al., 2018a; 2018b; Vega et al., 2017; Schittich et al., 2018). In addition, many geomorphologic settings as well as anthropogenic activities like land use pattern, large scale groundwater development for irrigation, and sanitation installment could play a pivotal role to regulate the release mechanism of As into the groundwater based on prevailing redox conditions (Mukherjee et al., 1997; Harvey et al., 2005; Nath et al., 2007; Polizzotto et al., 2008; Neumann et al., 2010; Majumder et al., 2015).

The western part of the BDP (Nadia district, west Bengal) has a special feature with respect to the basinal character, where Hoogli-Bhagirathi, Jalangi, Icchamati, Padma and Churni rivers are contributing to the development of fluvial sedimentation pattern and numerous geomorphological features (RGNDWM, 2001; Chatterjee, 2005; Nath et al., 2007). These geomorphological features (e.g., flood plain, oxbow lakes, meandering belts and spars, abandoned channel, natural levees) are common and often enriched with organic matter of various nature (Papacostas et al., 2008; Nath et al., 2005; Majumder et al., 2016; Bhowmick et al., 2012). The role of organic matter and the importance of river basins as host of arsenic-bearing sediments are already reported from several parts of south Asia (Mukherjee et al., 1997; Harvey et al., 2005; Nath et al., 2007; Polizzotto et al., 2008; Neumann et al., 2010).

The present study deals with the geospatial variability and heterogeneity of As in groundwater across the Nadia district, West Bengal where all the 17 blocks (Karimpur-I, Karimpur-II, Tehatta-I, Tehatta-II, Kaliganj, Nakashipara, Krishnagar-I, Krishnagar-II, Chapra, Krishnaganj, Nabadwip, Santipur, Ranaghat-I, Ranaghat-II, Chakdaha, and Haringhata) are affected with As in groundwater. The study further indicates the understanding of the degree of such geospatial variability in the study area. Attempts have also been made to explain the influence of basinal character (pedo-geomorphic characteristics of the basin), local geomorphology and anthropogenic sources that have some influences on groundwater As distribution pattern. In this context, the role of characteristics of sedimentations and varying nature of organic matter in controlling As mobilization into shallow groundwater has also been discussed.

2. Study area

Nadia district (Fig. 1) is located in the central-east part of the state of West Bengal, India. It is on the eastern wing of the regional river Bhagirathi (22°53′80″- 24°11′78″ N and 88°09′58″- 88°48′30″ E) in the BDP. The entire district lies in the alluvial plain of the Ganges and its distributaries. The district has an area of 3926 km^2 and is predominantly rural. The climate of the study area is typically hot and humid with an average temperature of 10°-40 °C and average humidity of 65–75%. The average annual rainfall of this area has been recorded about 1200–1500 mm. The major river, Bhagirathi generally form the western boundary except for a small strip of land around the city of Nabadwip. Nadia district is mostly abundant with rivers (Bhagirathi, Jalangi, and Churni) courses along with the number of depressions (meander scars and oxbow lakes).

The district is a large alluvial plain spreading southward approximately from the head of the Bengal delta and formed by a succession of river sediments into which the Ganges plays a crucial role. This district is very fertile in connection with agricultural perspectives. There has been a considerable change in the river courses in this area in the last few decades; however, Bhagirathi has been maintaining the same direction towards the south. The general slope of the district is towards south and south-easterly and the whole district is mostly interspersed with jhils (large lakes), lakes, oxbow lakes, marshy lands, old river-beds, river cut-offs and meander channels.

The district of Nadia is the part of the upper delta plain and is characterized by a series of meander belts as a response to the varying hydrodynamic conditions. The basin filled deposits are fluvial and comprise stacks of different cycles of upwards fining sequences (sand-silt-clay) (Bhattacharyya et al., 1997; Ahmed et al., 2004). Such cyclical band is interrupted by the bedding of coarse to medium sand, fine sand, clay and silt respectively. Upward fining cycles of various thicknesses constitute the meander belts of the Bhagirathi and the discontinuities of the cycle of deposition indicate the presence of old meander scars and paleo-channels (Mukherjee et al., 1997; Chatterjee et al., 2010a).

Groundwater usually occurs under unconfined condition particularly in this part of the BDP. The aquifers change gradually from open to semi-confined character towards the south. The closed aquifers are generally interconnected with the upper groups of unconfined aquifers. Fluvial sands and gravels are the major deposits forming most of the aquifers. The sedimentation in the channels raising beds and shifting courses is the natural process of moribund delta (Mukherjee et al., 1997; Chatterjee et al., 2010a).

2.1. Geological settings

The district, Nadia, is an alluvial formation of the rivers belonging to the Ganges-Bhagirathi system. On the top of the surface, it seems to be formed of recent alluvium. However, older alluvium with different materials is found below the plinth. It appears that the underlying older alluvium composed of different materials from the deposits by the Ganges on which the recent upper stratum has been laid down. The BDP is surrounded by the Himalayas and Shillong plateau in north, Indo-Burman uplands in the east, Bihar uplands in the west and Bay of Bengal in the south (Barman, 1992). This recent part of deltaic BDP is mainly the extended portion of the Chota Nagpur plateau, i.e., shield area (Nandy, 2001). The alluvium of BDP is mostly sedimented by the Himalayan rivers passing through the Garo-Rajmahal tract.

A stratum of yellowish clay and sand appears to underlie the upper and younger strata of blue clay and sand (Barman, 1992; Ahmed et al., 2004). It has been found that this delta was constructed by the rivers of the Ganges and its distributaries flowing down towards the south and south-east. The older delta has been suppressed by the newer sediments and now the Ganges seems to have entered upon. The formation on the top of the old sub-stratum was started by the sediments from the neighborhood of Rajmahal tract (Mukherjee et al., 1997).

The deposits of this part of the BDP are fine, medium and coarse sand with gravels. Clayey intercalation is darker in colour which indicates the high contents of the organic matter. Sandy clay and clayey sand, mixed with gravels and coarse to medium to fine sand elucidate the open hydrological system (Bhattacharya et al., 1997). The semi-confined aquifers are the principal deposits of fine white sand with clayey intercalations and coarse sand to gravel towards down. Finer sedimentations have been found at the bottom of most of the aquifers with white sand overlay. Arsenic contamination in the groundwater is characterized with the meander belts comprising Late Quaternary deposits (Chatterjee et al., 2010a). Clayey deposits near the aquifers might act as the source of As in groundwater. It has been noted that the sediments were eroded from the Chota Nagpur-Rajmahal uplands and then deposited in this area by the meandering rivers of the area under very reducing conditions.

In Nadia, the parent sedimentary materials are from Ganges alluvium and on this basis, the different categories of land-forms are distributed all over the district, viz., riverine lands, flatlands, and low lands. The riverine lands are formed of soils on recent alluvial fans, flat plains or other secondary deposits having undeveloped profiles below which lay unconsolidated materials (RGNDWM, 2001). There is no accumulation of clay and lime in the sub-soil. Findings tell us that the top surface is of recent formation and downward deposition of different materials with different textures in different layers in an unsymmetrical manner is also evident (Bhattacharya et al., 1997). Flatlands are formed of younger soils. There is a slight accumulation of clay in the sub-soils underlying unconsolidated materials. In low lands of Ganges delta are composed of claypan soils that are relatively near the surface and are partially impervious to the downward movement of the water. There is a considerable accumulation of calcareous and calco-ferrous materials which do not disintegrate in water (RGNDWM, 2001).

During the early-mid Pleistocene and Holocene, As bearing sediments are deposited by the river meanders and paleo-channels (Chatterjee et al., 2005). The lithology of this area has a succession of sand (channel facies), silt as well as clay (overbank facies) and predominantly a typical upward fining sequence (Bhattacharya et al., 1997; Chatterjee et al., 2010a). The sediment stratum includes both oxidized and unoxidized layers in succession on the partially eroded older platform (Chatterjee et al., 2005). The underlying early Mid-Pleistocene deposits are often oxidized due to long exposure, whereas, Holocene deposits are unoxidized and grey in colour consisting of sand, silt, abundant mica and heavy metals (Goodbred and Kuehl, 2000). The over-bank facies are abundant with finely grained organic substances (Chatterjee et al., 2003).

2.2. Physiographical settings

In BDP, As contamination has been observed notably in the part of interfluves of the Padma-Bhagirathi river which is extended up to the Bay of Bengal. This low-lying area (mainly river banks) of BDP gets flooded by the rivers each year in monsoon (CGWB, 1999). Numerous changes in river courses of river Ganges are due to eustatic sea-level changes and neo-tectonic influences (Bhattacharya et al., 1997; CGWB, 1997). Elevation of the Nadia district varies from 28 to 56 m above the mean sea level. Based on varied pedo-geomorphic characteristics, four river basins, namely – Padma river basin, Bhagirathi-Jalangi river basin, Churni-Ichhamati river basin and Hoogli-Bhagirathi river basin, have been selected to determine the role of geomorphic units controlling the As content in groundwater (Das Majumdar, 1978; Mukherjee and Bhattacharya, 2001; Chatterjee et al., 2005).

The Padma river basin (or its flood plain) has been formed due to huge siltation by river Padma. It lays north-west to southeast direction with a maximum width of 8 km. This tract has experienced various stages of shifting of river Padma since the last 150 years. The swinging nature of Padma, as well as flood, governs the destiny of habitats of this tract (Rob, 2012). The area of the basin is 439 km^2 which includes mainly Karimpur block-I and II. The major land use pattern is agricultural land for cultivation where nearly 80% of the cultivable area that is used for major field and horticulture crops (District Statistical Handbook, 2016). The basic characteristics of the soil are Gangetic alluvium and clayey loam soil.

Bhagirathi-Jalangi river basin formed due to the continuous spilling activity of rivers Bhagirathi and Jalangi and it is the outcome of the rising land along both the river course that is called levee tract of Bhagirathi and Jalangi. Bhagirathi-Jalangi river basin has a prominent history of channel migration and the region is dotted by a large number of marshy lands. Due to over-spilling of the rivers, natural levees have formed along both sides of the rivers. Moreover, for flood protection, embankments have constructed along Bhagirathi in many parts which prevent the spilling activity of Bhagirathi. Elevation of this area varies from 5 to 23 m and the width of tract varies from 3 to 16 kms. There are many lakes and marshes (locally known as bils) as the remnant of the abandoned river courses. These marshy lands are the storehouse of numerous natural resources. This river basin encompasses 923 km^2 of the area which comprises of Nakashipara, Kaliganj, Krishnagar-II (~75%), partly Krishnagar-I (~78%) and partly Nabadwip (~13%). In the Bhagirathi-Jalangi river basin, a large section is covered by low lying depressions, locally known as the "Kalantar area" (Das Majumdar, 1978; CGWB, 1999). The tract is composed of black clayey soil and bears only winter rice. The sediment deposits are clay mixed with fine sand extends down to a depth of about 18 m below ground level (bgl). Below this, one may find fine to medium sand areas up to 78 m bgl with intervening silt horizon (CGWB, 1997; Mukherjee and Bhattacharya, 2001). The average discharge of the river is about 500 cm^3 with an annual silt load of 492 tons km^{-2} (Islam, 1978).

Similarly, the vast flood plain of Churni-Ichhamati is another remarkable river basin which has been created due to overlapping spill channels of several rivers, mainly Churni and Ichhamati. This is a gradational plain of river Bhagirathi, Jalangi and Ichhamati. Elevation of this zone varies from 5 to 22 m and the slope of this region is from north-west to south-east. Physically this plain is almost uniform except some interruptions like bils (Das Majumdar, 1978). The area of the basin (Churni-Ichhamati) is 853 km^2 which includes Chapra, Krishnaganj, a section of Krishnagar-II block (~25%), Ranaghat-II (~53%) and Hanskhali block (~76%). The basinal area is relatively small in comparison to other river basins and usually flat with adequate drainage patterns (Das Majumdar, 1978; Chatterjee et al., 2005). The cultivated crops are mostly paddy (summer, monsoon, and partly winter) along with vegetables of several kinds, cereals, pulses, oil-seed crops, jute, and fiber plants (RGNDWM, 2001). The soil formation is young alluvial fans, old and young flat plains and other secondary deposits with developed profiles underlain by unconsolidated materials. The basin is dominated with clay and silt often loaded with mica (notably fresh) that increases with depth (RGNDWM, 2001; CGWB, 1999). The river discharge, from both Churni and Ichhamati, is fluctuating and increases in monsoon (Mukherjee and Bhattacharya, 2001; Rob, 2012).

Hoogli-Bhagirathi river basin, with new alluvium covered flood plain and prominent imprints of channel shifting, is another prominent river basin of Nadia district. It is formed due to the continuous spilling of river Hoogli-Bhagirathi and flowing towards further south in the Bay of Bengal. In the Gangetic low lands, the association of soil occurs in places where depressions have been created by the swinging movement of Bhagirathi and Hoogli. As a result, soil shows a good accumulation of clay on the surface which is underlain by the unconsolidated materials, more often of riverine profile (Das Majumdar, 1978). The area of the basin (Hoogli-Bhagirathi) encompasses 779 km^2 which includes Chakdah, Haringhata, Santipur, and sections of Ranaghat-I, Ranaghat-II (~47%), Krishnagar-I (~11%) and Nabadwip (~77%). The basin is a Gangetic low land and remarkably noticed in Chakdah (notably south-eastern fringe), Ranaghat (east and north-east portion) and Haringhata (western half) blocks (Das Majumdar, 1978; RGNDWM, 2001; Chatterjee et al., 2005). The paddy (summer, monsoon and winter) is the principal crop of the basin along with vegetables (mostly green and cash crops), jute, oil-seeds (mostly mastered seed) etc. It has been created due to the meandering of different rivers (Das Majumdar, 1978; Mukherjee and Bhattacharya, 2001). Sediment deposit is a lacustrine deposit dominated by clay and silt mixed with clay/fine sand (CGWB, 1999). This basin is mainly the deposit of less compact sediments consisting of fine to very fine grey micaceous sand, silt and dark grey clay of late Holocene age. The elevation of this basin varies from 5 to 18 m and slopes towards south to south-east. This area is mostly found with meander river belts, meanders scars, river cut-offs and oxbow lakes. The discharge of the river is depending on the supply of water from the upper catchment area, particularly Farakka Barrage. The inconsistent river regime controlled through regulated flow from Farakka Barrage has substantial effects on the hydro-geomorphology of the channel, upholding the least stream power throughout the year (Guchhait et al., 2016).

2.3. Site geomorphology

The geomorphological features of this matured deltaic plain can be classified into such classes, viz., Upper Mature Deltaic Plain (UMDP), Lower Mature Deltaic Plain (LMDP) and Flood Plain (FP) (Table 1). The UMDP is abundant by meander scars, abounded channels and oxbow

Table 1
Summary of geological and geomorphic domain along with physiographic features of the Nadia District.

Geomorphological division	Physiography	Geology
Flood Plain (FP)	Channel deposits on the banks of rivers Bhagirathi, Hoogli and Jalangi of lowest topography. The area is prone to flooding and water logging. The relatively younger landforms are still in the process of formation. Channel migration and changing meandering course.	It is the deposit of unoxidized and less compact fine to very fine silver-grey micaceous sand, silt and dark grey clay of late Holocene to recent age. Geologically it is termed as Hoogli formation.
Lower Mature Deltaic Plain (LMDP)	The next higher relief having a narrow stretch of land in the south-western part of the area.	Sporadic occurrences of this Chinsura or Katwa formation of middle to late Holocene age. There are alternate layers of oxidized to unoxidized fine to very fine sand, silt and clay. Sediments of this part are devoid of any ferriginous and calcareous concretions.
Upper Mature Deltaic Plain (UMDP)	It covers a vast area from north to south and occupies high ground above occasional and usual flood levels. A large number of oxbow lakes and meandering scars cover this region as a remnant of the old courses. Prominent wetlands are formed at present over here.	Lithologically it is characterized by grey to yellow very fine sand, silt and silty clay with soft iron nodules and caliches nodules of early to middle Holocene age. Geologically it is termed as Bethuadahari formation.

lakes. The deposits in it indicate the deposition in Holocene ages (Chatterjee et al., 2005). Arsenic contamination in this said region is due to the meander scars and abounded channels (Bhattacharya et al., 1997).

The south-eastern flood plain (FP), i.e., Bhagirathi-Hoogli and Ichhamati basin are As contaminated due to the enrichment of organic matter (Lawson et al., 2013; Kulkarni et al., 2018a).

The LMDP is composed of several tidal creeks, fluvio-estuarine landform, and natural levees along with inter-distributary marshy lands. Fluvio-estuarine lower river plains are contaminated by As and the estuarine-marine depositional regime is generally less contaminated (Chatterjee et al., 2010a). The contaminated groundwater is generally obtained from the young alluvial plain of the LMDP where rivers change their channels frequently during the ages. Moreover, contaminated groundwater is mostly associated with the shifting of paleo-channels and their meander belts (Nath et al., 2005).

3. Materials and methods

3.1. Groundwater sampling

The groundwater samples were collected across all the 17 blocks of Nadia district (Fig. 1) from the private, community and government wells within the shallow range (<50 m) during the period 2013 to 2016. Groundwater samples were collected in prewashed and acid cleaned polyethylene vials (Tarsons, capacity 100/200 mL). Wells were pumped (mostly hand-operated) for few minutes (at least 15 min) to ensure the samples to be the proper representative of the aquifer under investigation. The samples were filtered then acidified on-site by concentrated HNO_3 (1% v/v, Suprapur, Merck). Then the samples were stored at 4 °C. The position of each well was recorded by a handheld GPS device (Garmin). Total 2448 groundwater samples were collected from the entire study area.

3.2. Groundwater analysis

1 mL of each standard (Merck, Germany) and an unknown sample were taken in 50 mL volumetric flasks separately. About 4 mL of concentrated HCl was added to each flask. Then 5 mL of KI and 5 mL of ascorbic acid were added to each of the above-mentioned sets and left for at least 1 h. Both the solutions were made up to 50 mL by volume by adding MiliQ water. Arsenic concentrations of all the solutions (standard and unknown sample) were measured at 193.7 nm wavelength (using HG-AAS mode) by Atomic Absorption Spectrophotometer (Varian, AA-240, Australia). The results were within ±5% for each of the measured standard solutions.

3.3. Geostatistical analysis

DPMS (District Planning Map Series) maps (raster imagery) were procured from the Survey of India, Regional Office, Kolkata and they were georeferenced by the GIS-based software (MapInfo v. 9.2 and ArcGIS v. 10.3) using some fixed-point geographical positions (latitudes and longitudes) that were collected during the sampling. Then the data available after the analysis was pointed on the georeferenced map of Nadia district (DPMS map) along with latitude and longitude of each well. Different thematic maps were produced to delineate the distribution pattern of groundwater As in the study area and the influence of different geomorphological features on the same. The number of different geomorphological features were evaluated by using summary statistical analysis toolbox in ArcGIS v. 10.3. Then percentage of each features (meandering features, water bodies and natural levees) were calculated with respect to total number of features in the study area.

3.4. Statistical analysis

The statistical analyses of the data were done by using SPSS Statistics 23 (IBM Corporation, 2015) using the LSD-Post Hoc package (Kim, 2015; Allen, 2017) to evaluate the mean difference among the As contamination levels within different river basins. The (I-J) values in Table 4 indicate the significant differences between the contamination levels of the river basins. To understand the correlation among the geomorphological features (meandering features, water bodies and natural levees) and arsenic contaminated wells (safe and highest unsafe wells), Pearson correlation matrix (included as Supplementary data Table- 1) has been developed with respective p-values.

4. Results and discussion

4.1. Groundwater As - geospatial distribution

Several studies have been conducted in BDP, notably in Nadia district to understand the spatial distribution of As in shallow aquifers (Mukherjee et al., 1997; Nath et al., 2005; Nath et al., 2007; Majumder et al., 2015). There are distinct geographical features that could be attributed to the spatial distribution pattern. Both longitudinal (88°09′58″- 88°48′30″ E) and latitudinal (22°53′80″- 24°11′78″ N) geographical distribution features are observed, which is patchy in geographical space (Fig. 2). The variable groundwater As concentration areas (based on well concentrations percentage frequency) were categorized, with different colour codes, as: highest unsafe As area (As_T >200 μgL^{-1}, red in colour), high unsafe As area (As_T 120–200 μgL^{-1} orange in colour), moderate unsafe As area (As_T 50–120 μgL^{-1}, yellow in colour), low but unsafe area (As_T 10–50 μgL^{-1}, previous Indian national standard, light green), very low/safer area ($As_T < 10$ μgL^{-1}, deep green) (RGNDWM report 2001; Chatterjee et al., 2018).

Shallow aquifers are mostly exploited for drinking water supply (RGDWM, 2001; Nath et al., 2008a, 2008b; Mukherjee et al., 2011). A

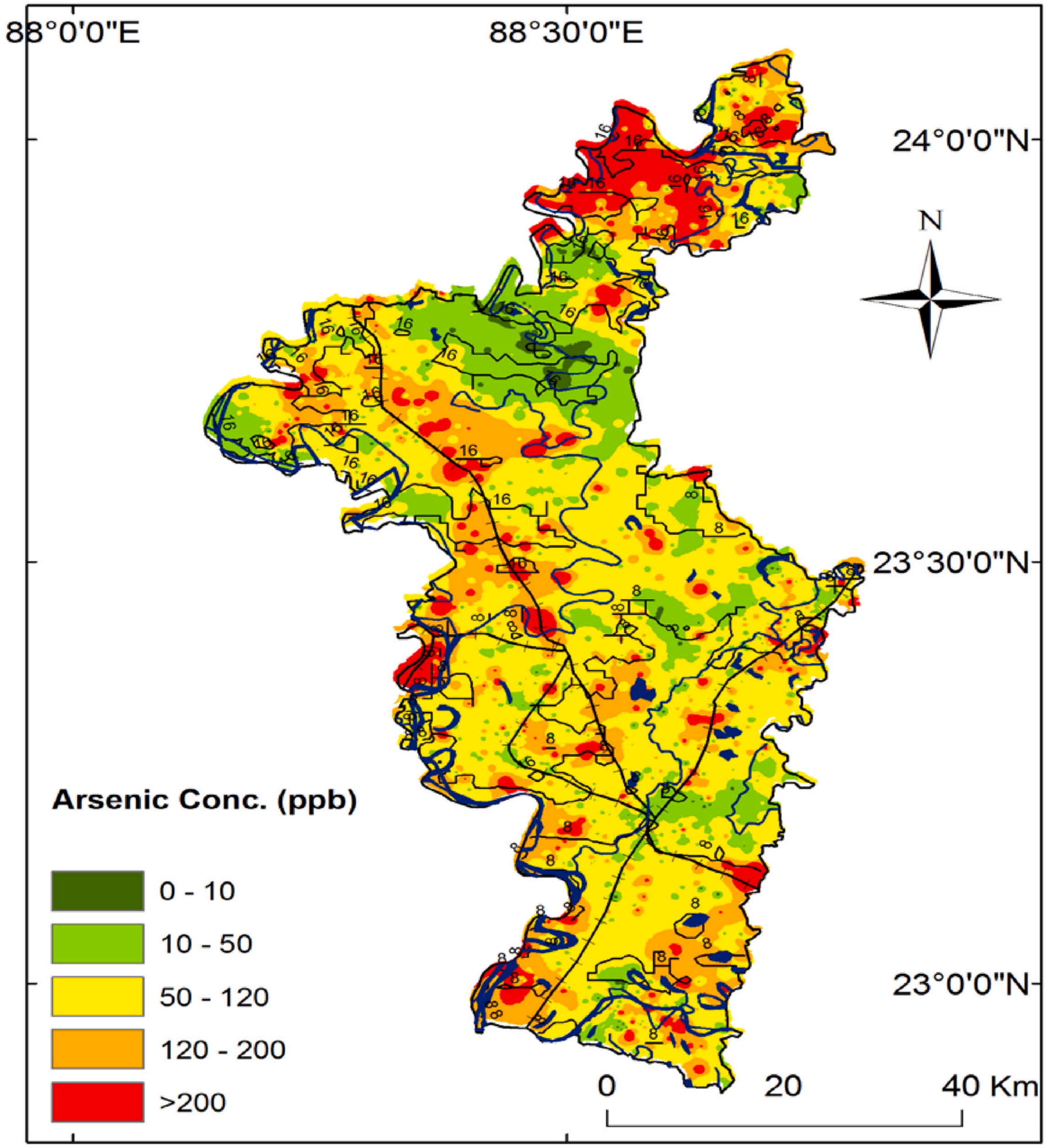


Fig. 2. Map of As distribution pattern of Nadia district, West Bengal (shallow aquifer, <50 m) (modified from Chatterjee et al., 2018). Highest unsafe As area (As_T >200 μgL^{-1}, red in colour), high unsafe As area (As_T 120–200 μgL^{-1} orange in colour), moderate unsafe As area (As_T 50–120 μgL^{-1}, yellow in colour), low but unsafe area (As_T 10–50 μgL^{-1}, light green), very low/safer area (As_T< 10 μgL^{-1}, deep green). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

close observation of the geospatial As distribution pattern (Fig. 2) indicates that the northern part of the district is severely affected with As followed by south-eastern part and south-western part.

It has also been found that all the 17 blocks of the district have been affected with groundwater As occurrence, both high and low (Fig. 2). The highest As concentration areas (As_T >200 μgL^{-1}) are also identified throughout the district. These very high As areas are largely varying concerning geographical areas as well as land-use pattern. The moderate As concentration areas (50–120 μgL^{-1}) have been found notably in the large part of the district (up to 70%) which is mostly covering several blocks in the south-eastern parts of the district. Contrastingly, south-western parts of the district are below the moderate As concentrations.

However, in south-eastern part the groundwater As concentration is also largely varying (Table 2) e.g., Ranaghat-I (As_T average 102.53 μgL^{-1}), Chakdaha (As_T average 132.85 μgL^{-1}) and Haringhata (As_T average 107.75 μgL^{-1}). Relatively higher concentration areas (120–200 μgL^{-1}) have been predominately noticed in south-western part in comparison to south-eastern part. In the south-western part, the highest As concentration areas (Fig. 2) have been found in blocks where several highest As areas have been identified. However, the south-western marginal boundary of the district is another important issue where Hoogli-Bhagirathi is flowing towards the Bay of Bengal. Within this south-western margin the similar pattern of patchy As distribution of both high As concentration areas (As_T range 120–200 μgL^{-1}) and highest As areas (As_T> 200 μgL^{-1}) are identified, unlike the south-eastern margin (farthest eastern part of Chakdaha and Haringhata blocks) where drainage system is practically absent (Dhar et al., 1997; Chowdhury et al., 2000). However, a typical patchy distribution of high As areas (Fig. 2) within high As concentration areas (As_T 120–200 μgL^{-1}) has only been noticed in the furthest south-eastern part of the district

Table 2
In Nadia, block wise (n = 17) groundwater As variation in shallow aquifers (<50 m). [bdl: below detection limit].

Name of the Blocks	No. of tube-wells	Depth range (m)	As distribution				% of High As wells (As> 200 μgL^{-1})
			As_T Avg. (μgL^{-1})	Min. (μgL^{-1})	Max. (μgL^{-1})	Standard deviation	
Karimpur-I	141	9–46	147.8	bdl	786	161.2	27.66
Karimpur-II	127	8–48	214.7	bdl	926	160.0	51.97
Tehatta-I	107	12–46	38.57	bdl	61.97	81.20	07.48
Tehatta-II	85	18–49	27.84	bdl	52.53	55.80	04.71
Kaliganj	235	12–50	101.2	bdl	995	102.2	12.77
Nakashipara	103	6–50	98.39	bdl	586	96.84	21.36
Krishnagar-I	154	15–50	78.34	bdl	610	72.95	09.74
Krishnagar-II	46	14–50	155.7	bdl	1161	184.8	23.91
Chapra	88	12–42	99.68	bdl	514	96.00	10.23
Krishnaganj	192	15–50	96.16	bdl	771	104.9	11.98
Hanskhali	222	12–48	108.54	bdl	518	89.43	07.21
Santipur	121	9–48	97.91	bdl	459	81.84	15.70
Nabadwip	151	12–47	164.17	bdl	712	149.7	11.92
Ranaghat-I	131	12–50	102.53	bdl	1072	112.2	10.69
Ranaghat-II	204	9–49	70.39	bdl	486	69.37	07.35
Chakdah	130	8–50	132.85	bdl	663	120.5	10.77
Haringhata	211	4–49	107.75	bdl	723	123.3	08.53

where Chakdaha and Haringhata blocks are affected. These two blocks have already been identified with several As affected villages such as Ghetugachi, Sahispur, Chakudanga (Chatterjee et al., 2003; Guha Majumdar et al., 2010; Chatterjee et al., 2010a, 2010b; Biswas et al., 2011; Halder et al., 2013; Majumder et al., 2015, 2016). These earlier studies also indicate such spar and inter-spar nature of As distribution pattern in those affected areas (Fendorf et al., 2010; Majumder et al., 2015). The findings of the present study falls in the same line with several earlier studies that have been conducted in Nadia district notably in those two blocks (McArthur et al., 2001; McArthur et al., 2004; Bhowmick et al., 2013a, 2013b; Majumder et al., 2015, 2016).

4.1.1. *Groundwater As - block wise distribution*

The groundwater As concentrations in shallow depth (<50 m) of the study area have been presented in Table 2. The highest average groundwater As concentration (As_T average 214.73 μgL^{-1}) is prevalent in Karimpur-II block, whereas, lowest average As concentration (As_T average 27.84 μgL^{-1}) has been observed in Tehatta-II block. The groundwater As concentration for rest of the blocks, follows the order as: Tehatta-I (As_T average 38.57 μgL^{-1}) < Ranaghat-II (As_T average 70.39 μgL^{-1}) < Krishnagar-I (As_T average 78.34 μgL^{-1}) < Krishnaganj (As_T average 96.16 μgL^{-1}) < Santipur (As_T average 97.91 μgL^{-1}) < Nakashipara (As_T average 98.39 μgL^{-1}) < Chapra (As_T average 99.68 μgL^{-1}) < Kaliganj (As_T average 101.24 μgL^{-1}) < Ranaghat-I (As_T average 102.53 μgL^{-1}) < Haringhata (As_T average 107.75 μgL^{-1}) < Hanskhali (As_T average 108.54 μgL^{-1}) < Chakdah (As_T average 132.85 μgL-1) < Karimpur-I (As_T average 147.82 μgL^{-1}) < Krishnagar-II (As_T average 155.74 μgL^{-1}) < Nabadwip (As_T average 164.17 μgL^{-1}). The interesting observation is the close geospatial association of the highest average As concentration (As_T average 214.73 μgL^{-1}, Karimpur-II block) adjoined by lowest Avg. As concentration (As_T average 27.84 μgL^{-1}, Tehatta-II block). Such a geospatial association of high and low As areas is an essential component of BDP As occurrence (Nickson et al., 1998; BGS and DPHE, 2001; Neumann et al., 2010). These typical associations further indicate the role of the delta building processes. The highly contaminated areas are belonging to mature delta. On the other hand, the lowest contaminated area comes under the levee tract of the Bhagirathi-Jalangi riverine basin. The entire block is under the riverine sedimentation of the river Padma which changes its courses during geological timescale. Tehatta (both I & II) blocks are relatively upland area where the predominant river shifting phenomena is absent. The land-mass is under the influence of active delta and the sedimentation process is different from mature delta building process (Smedley et al., 2002; Neidhardt et al., 2013b).

On the other hand, the overall highest concentration of As in groundwater (As_T 1161 μgL^{-1}, Table 2) has been found in Krishnagar-II block. However, the lowest As concentration is even below the detection limit (bdl). Such high and very low values of As (often bdl) are the characteristic features of Nadia district including the entire BDP (Bhattacharya et al., 1997; Chowdhury et al., 1999; Chowdhury et al., 2000; Nath et al., 2005; Chatterjee et al., 2005; Majumder et al., 2015). The maximum groundwater As concentrations (Table 2) indicate the highest As areas which are more predominant in nature as well as serious public health concern, when those aquifers are used for drinking water purposes (Bhattacharya et al., 1997; McLellan et al., 2002; Kapaj et al., 2006).

Throughout the Nadia district, the highest groundwater As concentration values (Table 2) is largely varying from block to block (even within individual blocks) with a high heterogeneity. The highest groundwater As concentration have been identified nearly in the central part of the study area (Krishnagar; 23°23′19″N and 88°29′26″E, Table 2), close to the bank of the regional river (Hoogli-Bhagirathi), whereas, the entire northern part of the study area (Karimpur-I and Karimpur-II blocks) is highly infested with high As areas. It is pertinent to note that the north-western part (Tehatta-I and Tehatta-II below Karimpur) is usually low in average groundwater As concentration and therefore safer as a drinking water resource. Such As-safe areas (As_T< 10 μgL^{-1}, Fig. 2) have also been noticed in the entire district barring northern part of the district (Karimpur-I & II, Tehatta-I & II).

Another important issue is this patchy high As areas usually occur in the close vicinity of domestic drinking water wells. However, the close examination of groundwater As distribution pattern in Nadia district (Fig. 2) also reveals that such high As areas starting from Kaliganj (23°43′57″N and 88°13′44″E) block are more widely distributed and concentrated in the western part of the district (Kaliganj, Krishnagar, Nabadwip, Santipur; 23°42′43″-23°19′01″N and 88°11′06″-88°27′47″E), where regional river Hoogli-Bhagirathi is flowing with meandering character.

Moreover, number of high As areas (relatively smaller in size) have also been identified in the south-eastern part of the district which has been widely distributed in nine blocks of the district (Chapra, Krishnaganj, Chakdaha, Ranaghat-I, Ranaghat-II, Hanskhali, Haringhata, Karimpur-I, and Karimpur-II).

In this context, the furthest southern part of the district (Chakdaha and Haringhata blocks) where several such high As areas (Fig. 2) have been identified and, widely distributed through both western and eastern margins of the blocks. The formation of high As areas with various size and shapes is a reality, may be, due to several local influencing factors. These factors are local recharge, depth of the wells, basinal character and sedimentation pattern, local geomorphology and land-use pattern, terrain condition and finally well location including surficial features. The variation of these factors and their differential influence will ultimately constitute the size, shape, and nature of such high or low As areas.

4.2. As distribution and basinal geomorphology

Four river basins (namely, Padma river basin, Bhagirathi-Jalangi river basin, Churni-Ichhamati river basin and Hoogli or Hoogli- Bhagirathi river basin) have been studied to investigate the geospatial distribution of As and their association with basinal characters.

The Padma river basin (Fig. 3a) has been noticed to be highly contaminated with As in groundwater (highest As_T average 214.73 μgL^{-1}, Table 2). It has also been noticed that the almost entire northern and north-eastern part of the Padma river basin is highly contaminated with As in groundwater (As_T maximum 926 μgL^{-1}) where the Padma is flowing from north to south-eastern slope towards the Bay of Bengal (Gault et al., 2005; Fryar et al., 2007; Chatterjee et al., 2010a). In this tract, only a very small portion of the south-western part is relatively low in As concentration. In this basin, As contamination has been occurred within the shallow depth zone (8–48 m, Table 2).

Another river basin, Bhagirathi-Jalangi river basin has also been

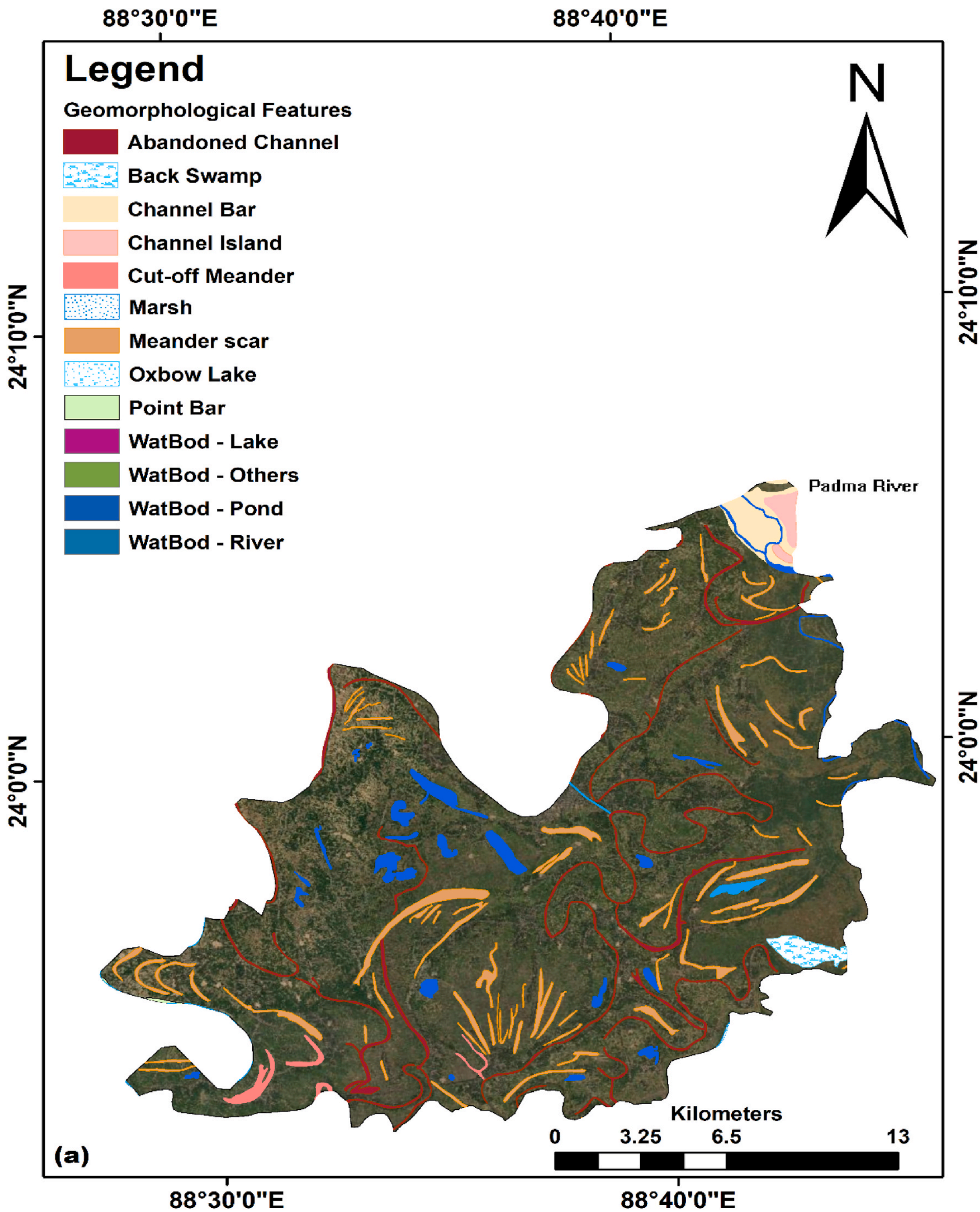


Fig. 3. In Nadia, maps of riverine basinal characters in four different river basins: (a) Padma river basin, (b) Bhagirathi-Jalangi river basin, (c) Churni-Ichhamati river basin and (d) Hoogli-Bhagirathi river basin.

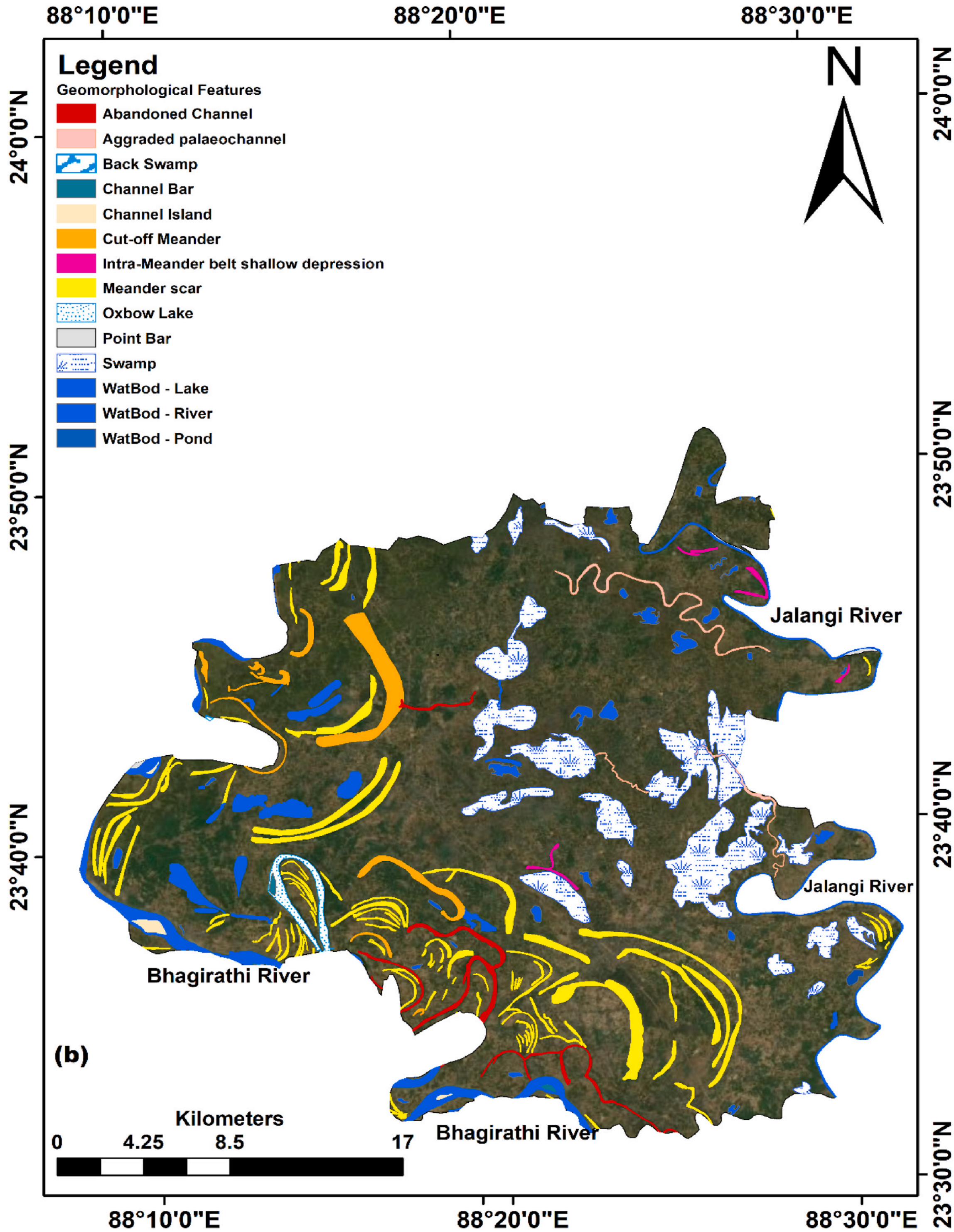
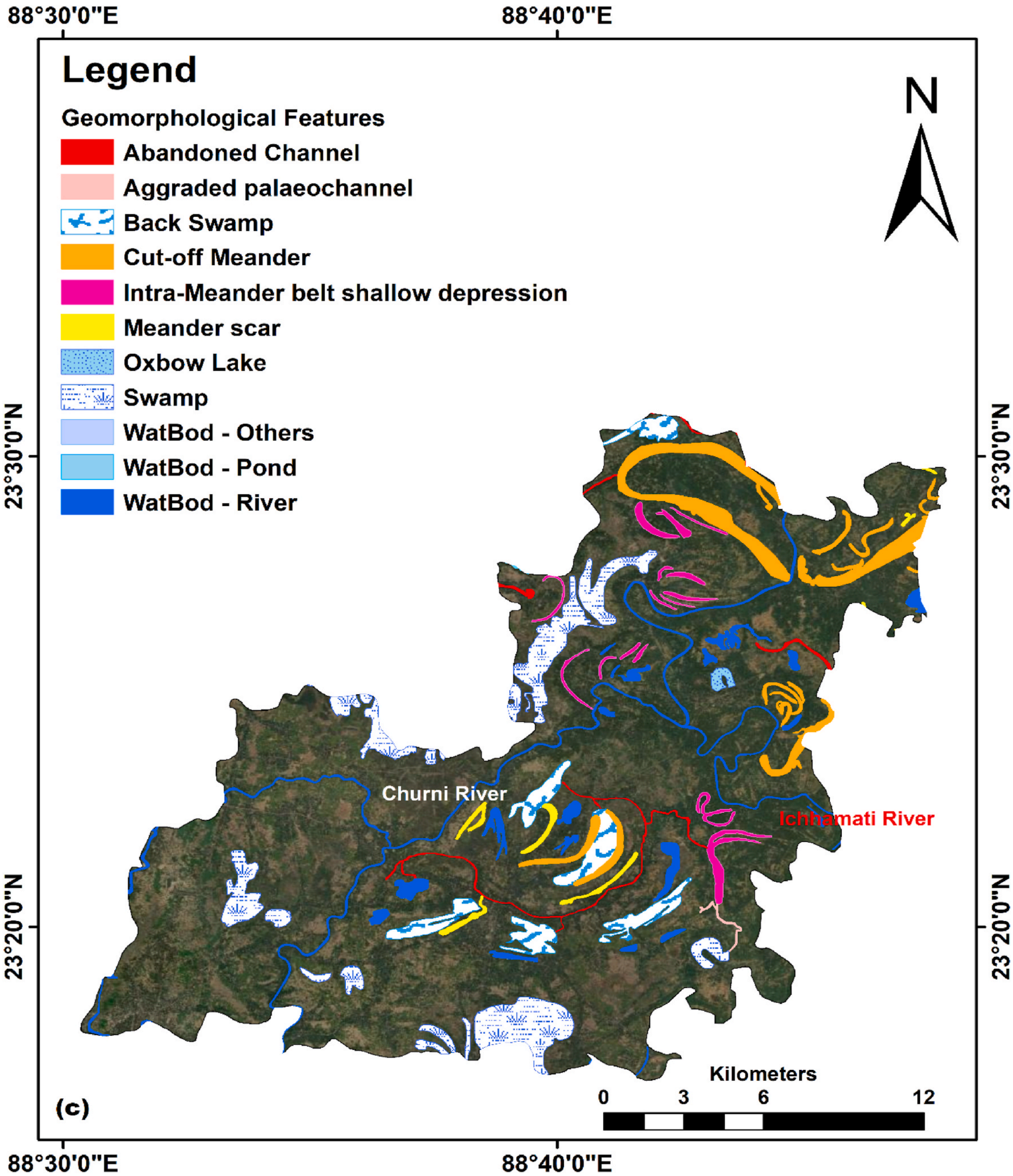


Fig. 3. (*continued*).

identified (Fig. 3b). In this river basin, the As high As areas (hot-spots) are identified mostly in the central part followed by the north-western part, upper northern part as well as the southern part. The said highest As areas ($As_T > 200\ \mu gL^{-1}$, Fig. 3b) are widely distributed throughout the basin with various size and shapes. It has also been found that in north-western part with south-eastward slope, there are safe aquifers which usually contained $As_T < 10\ \mu gL^{-1}$ of As in groundwater. It is important to note that the Bhagirathi river basin is by and large more contaminated with regard to Jalangi river basin (Fig. 3b). The present form of the river Bhagirathi is flowing north-western border of the basin (Nadia district) with a general slope towards the south. The 'Kalantar area' stretches from Beldanga to Kaliganj block in between Bhagirathi and Jalangi river basin. It also consists of large marshes and dead river beds (Neidhardt et al., 2013a). It has no drainage and during monsoon forms shallow lakes. Several such areas are identified in this "Kalantar" stretch generally with north-eastern inclination along the present railway track. It is important to note that, the shape, size and distribution pattern of high As areas are gradually changing from north to south. The large part of Jalangi river basin is relatively less contaminated where several highly contaminated wells have been noticed.



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Another river basin (Fig. 3c) has been identified which is the intervening plane of Churni-Ichhamati river basin, where the Churni was originated from Mathabhanga (Das Majumdar, 1978) and the Ichhamati (an old branch of Bhairab) is flowing through the eastern margin of the basin. Churni was originally a branch of Ichhamati and the flow pattern is influenced by the mixed flow of Ichhamati and Mathabhanga. It is astounding to note that the entire basin is by and large less contaminated with As groundwater in comparison to other basins (Fig. 3b and d). The entire Jalangi river basin is nearly free from groundwater As contamination whereas a part of the Ichhamati basin is contaminated with As in groundwater (Fig. 3c). In addition, only central part of the basin (a small section north to south direction) is largely contaminated with As in groundwater where numerous high As wells (Fig. 3c) have been identified. These areas are often associated with large water bodies (rudiments of river channels) and the presence of railway track (levee part). The spatial distribution pattern of As in groundwater in this basin is less heterogeneous in comparison to other river basins.

Another river basin (Fig. 3d) which is associated with river Hoogli can be designated as Hoogli river basin (or Hoogli-Bhagirathi river basin). The basin is identified in the lower part of the district where Ranaghat, Chakdaha and Haringhata blocks are present (Fig. 3d). River Hoogli with oscillating and highly meandering characters has been flowing through the western margin of the basin with considerable southern slope. Near Chakdaha, the confluence point of Churni and Hoogli-Bhagirathi has occurred. In Haringhata, the river Jamuria (the extension of Mathura bil) has been noticed which is associated with several large water bodies (Solankir bil, Jhakir bil, Mathura bil). The central part of the basin is less contaminated with groundwater As, whereas, both eastern and western margins are contaminated with widespread wells with highest As concentrations (Fig. 3d). Moreover,

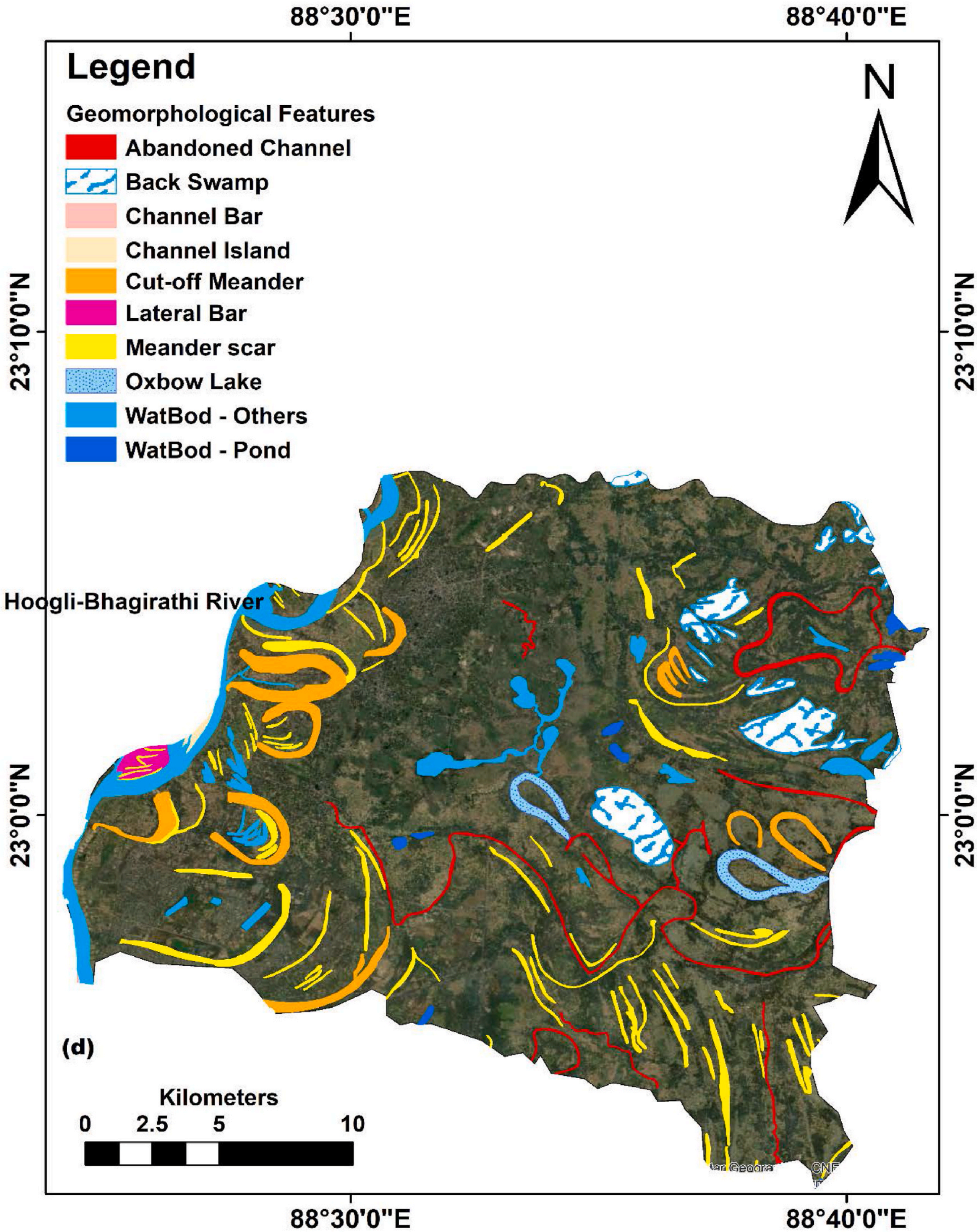

Fig. 3. (*continued*).

relatively lesser highest As areas (Fig. 3d) with high heterogeneity and spatial distribution pattern has been observed in the south-eastern part of the basin.

The highest As areas (As_T> 200 μgL^{-1}) with less heterogeneity have been observed in several parts of Hoogli-Bhagirathi basin (Nakashipara, Krishnagar-II and Krishnagar-I blocks, Fig. 3b), where they are mostly associated with river Bhagirathi. Relatively smaller high As areas with stronger heterogeneity has been observed on both sides of the Jamuria river basin. The multi-aquifer system is mostly predominant near the river basin consisting of sand, silt, and clay layers with upward fining sequences, whereas, thick clayey deposition (Holocene sedimentation) has often been found across the top soil of the river basin (Nath et al., 2008c). Fine to medium sand (unconsolidated) are found under the upper aquitard (silt and/or clay layers), below which, coarse sand of Pleistocene age with high porosity is often noticed (BGS and DPHE, 2001). The present study indicates that the high As areas are in close association with drainage pattern which strengthens the possibility of groundwater recharge through the porous sandy layers, diffusion of atmospheric oxygen, and reducing aquifer geochemistry. The multi-layer aquifers with clayey and/or silty layers near the river channel further suggest that As in the solid phase (aquifer sediment materials of Holocene age) has been released into groundwater (aqueous phase) under reducing conditions. The As distribution pattern in the different river basins from the study area helps to understand the heterogeneous distribution of groundwater As in shallow aquifer system. This is again consistent with river Bhagirathi-Hoogli (confluence point near Nabadwip) that provides evidence for sediment deposition which results in As mobilization into the shallow groundwater. River Bhagirathi and its distributaries form torturous river channels with abundant geomorphological features, which help to supply organic matter that controls the reducing environment of the shallow aquifers. These organic matters are play a key role in mobilization of As into the shallow groundwater. The desorption of As has been promoted by the reductive dissolution of Fe-oxides and/or oxy-hydroxides under the reducing environment of the aquifers, often enriched with fresh organic matter ingested during groundwater recharge. The present study reveals that Bhagirathi-Hoogli river basin is mostly affected than other river basins due to the abundance of geomorphological features and prevailing

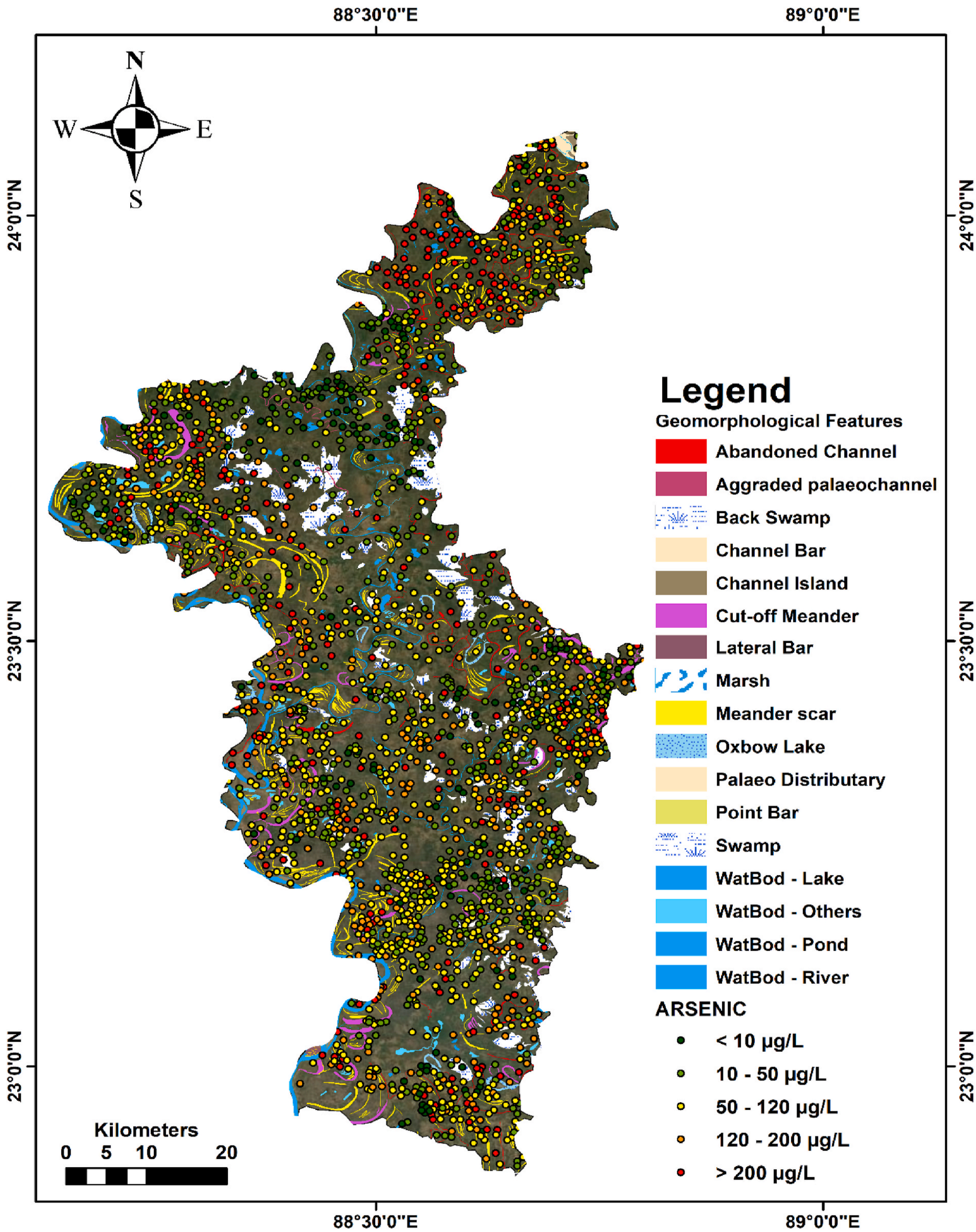

Fig. 4. Geomorphological map of the study area with As wells at different contamination levels.

stronger reducing conditions. Mainly, the depositional environment is controlled by flow velocity of Hoogli-Bhagirathi rivers which results in mobilization of As into groundwater under reducing conditions.

Padma river basin, Bhagirathi-Jalangi river basin and Hoogli-Bhagirathi river basin (Fig. 3a, 3b and 3d) are more affected than Churni-Ichhamati river basin (Fig. 3c). This is possibly due the less abundance of geomorphological features than other three river basins. Among all the river basins, Padma and Hoogli-Bhagirathi river basins are most contaminated with groundwater As.

4.3. As mobilization and geomorphological features

The distribution pattern of groundwater As is connected with various dominant geomorphological features (flood plain, natural levee, inter-distributaries levees, abundant channels, bils, khals, meander scrolls, meander belts, meander scars and oxbow lakes) in Nadia district (Fig. 4). Karimpur (both block I and II) had been identified with several geomorphological meandering features older in origin such as meander channels, meander scrolls, meander belts and meander scars. A large number of high As tube wells are associated with such meandering geomorphological features (Fig. 4).

In Karimpur, several natural water bodies are found (Padma bil, Dhandi bil, Kumari bil, Dighri bil and Dhopagari bil) along with meander belts and meander scrolls are identified. The number of associated high As areas are varying with groundwater As occurrence (n = 105, Table 3). In Tehatta, the number of safe monitoring wells (n = 192, Table 2) are relatively higher in numbers and the high As areas are considerably limited due to lesser number of geomorphological features (Fig. 4). In Kaliganj, the number of water bodies (Boalia bil, Chanduria bil, Bagher bil, Panighata bil) is moderate. The meander belts and meander scars are also relatively low. This reflects on the number of high As areas (n = 30, Table 3) along with safer wells (n = 43, Table 3). In Nakashipara, the safe wells have also been adequately identified and widespread. However, they are lower in number when compared to Tehatta (n = 17, Table 3). On the other hand, the high As wells are identical in number in both the blocks (n = 22, Table 3). In Chapra, the high As areas (As_T>200 μgL^{-1}) are limited (n = 9, Table 3) along with a few safer wells (n = 4, Table 3). In Krishnagar, frequent high As areas have been found while number of safer wells are limited. (n = 3, Table 3). In the said block, several major water bodies (Sujanpur bil, Noapara bil, Bhaluka bil, Hansdanga bil) have been found which are largely distributed throughout the area. In Krishnagar, meander belts, meander scrolls, meander scars are also identified where Jalangi is the principal river. The natural water bodies and other common geomorphological features are possible source of organic matter and rapidly deposited in the sediment (Bhowmick et al., 2013b; Lawson et al., 2013; Kulkarni et al., 2018a). In another block (Krishnaganj), similar prevalence of highest As areas (As_T> 200 μgL^{-1}, n = 23, Table 3) and safer wells have been observed (n = 7, Table 3). Large number of water bodies mostly bils and khals (Damodar bil, Majdia bil, Dharmada bil) are identified in Krishnaganj. The number of safer water wells (n = 7, Table 3) is limited in Krishnaganj. In Nabadwip, lesser number of highest (n = 18, Table 3) as well as safer wells have been observed (n = 3, Table 3). In Santipur, the number of contaminated wells (n = 19, Table 3) is noticed and bit higher number of safer wells (n = 27, Table 3) is recorded. In Santipur, the number of bils and khals (Nutan bil, Pasha bil, Mansadaha bil) are relatively low and smaller in size. In Hanskhali, the situation is in between, where the number of highest As areas (n = 16, Table 3) are relatively low, whereas, the number of safer wells (n = 21, Table 3) is higher. In Ranaghat, the situation is intermediate as the number of highest As areas (n = 29, Table 3) are relatively high. However, it is also interesting to note that there are notable number of safer wells (n = 19, Table 3) present as well. In Chakdaha and Haringhata, there is a similar situation concerning the number of highest As areas (n = 32, Table 3) and the number of safer wells (n = 30, Table 3).

Table 3
In Nadia, the block wise distribution pattern of groundwater As and well occurrence.

Block Names	Safer wells (As_T<10 μgL^{-1})	As_T<50 μgL^{-1}	As_T<120 μgL^{-1}	As_T<200 μgL^{-1}	As_T> 200 μgL^{-1}
Karimpur (I & II)	21	43	30	14	105
Tehatta (I & II)	36	42	8	2	12
Kaliganj	43	4	24	12	30
Nakashipara	17	14	4	7	22
Krishnagar (I & II)	3	29	39	19	26
Chapra	4	14	28	21	9
Krishnaganj	7	31	64	27	23
Santipur	27	47	14	4	19
Nabadwip	3	30	14	17	18
Hanskhali	21	65	65	33	16
Ranaghat (I & II)	19	77	79	19	29
Chakdah	14	29	42	21	14
Haringhata	16	45	66	21	18

A statistical analysis has been carried out to depict the correlation between the occurrences of geomorphic features and As distribution patterns of the groundwater (Fig. 5). The nature of the correlation shows two different scenarios concerning As levels where safer wells (As_T< 10 μgL^{-1}) and wells in high As areas (As_T> 200 μgL^{-1}) are tested. Among the geomorphological elements, natural levees show positive correlation (r^2 = 0.57, Pearson correlation co-efficient = 0.81 at $p < 0.01$, Supplementary data table) with safe wells, whereas, the correlations for natural water bodies (r^2 = 0.30, not significant, Supplementary data table) and meandering features (r^2 = 0.15, not significant, Supplementary data table) are not so pronounced. The relationship is different for unsafe wells where meandering features show some strong evidence of correlation (r^2 = 0.52, Pearson correlation co-efficient = 0.92 at $p < 0.01$, Supplementary data table) as compared to relatively weaker correlation with water bodies (r^2 = 0.33, Pearson correlation co-efficient = 0.56 at $p < 0.05$, Supplementary data table) and natural levees (r^2 = 0.12, not significant, Supplementary data table). A causal linkage between meandering features and groundwater As levels has been already established in BDP across various river basins of south-east Asia (Nath et al., 2005; Van Geen et al., 2006; Berg et al., 2008; Papacostas et al., 2008; Nath et al., 2008a; Bhowmick et al., 2013b).

Another attempt has been made to link the As distribution with various river basins (Table 4). The results substantiate that the basins are significantly ($p < 0.05$) related to the As distribution pattern. The mean difference values (I-J) clearly indicate that the Padma river basin is the most contaminated basin while contamination in Churni-Ichhamati river basin is the least. Significant tests also indicate that the differences (I-J values) observed for As concentrations are in following descending order (Padma > Bhagirathi-Jalangi > Hoogli-Bhagirathi > Churni-Ichhamati).

The statistical analysis reveals that the role of geomorphological elements is significantly contributing to the basin wise distribution pattern of safer and As-contaminated wells. The study demonstrates that the elevated As levels in river basins are often associated with a micro-geomorphological environment, rich in recent organic matter. Thus, they are controlled by fluvial geomorphological processes and local land use pattern. Several paleo-meander belt units are common in the highest contaminated river basin (Padma river basin, Figs. 2 and 4). These units are abundant channels, meander cut-offs, oxbow lakes and flood plains – often covered with rich vegetation. Generally, high As areas are found in recently deposited sediments of the Ganges-Padma river and those deposited in the surrounding areas of those geomorphological units. The rapid sedimentation that takes place in these recent features results in grey fine micaceous sand (reducing in nature), silty clay, soft clay with varying thickness at the top, preserving the sedimentary organic matter

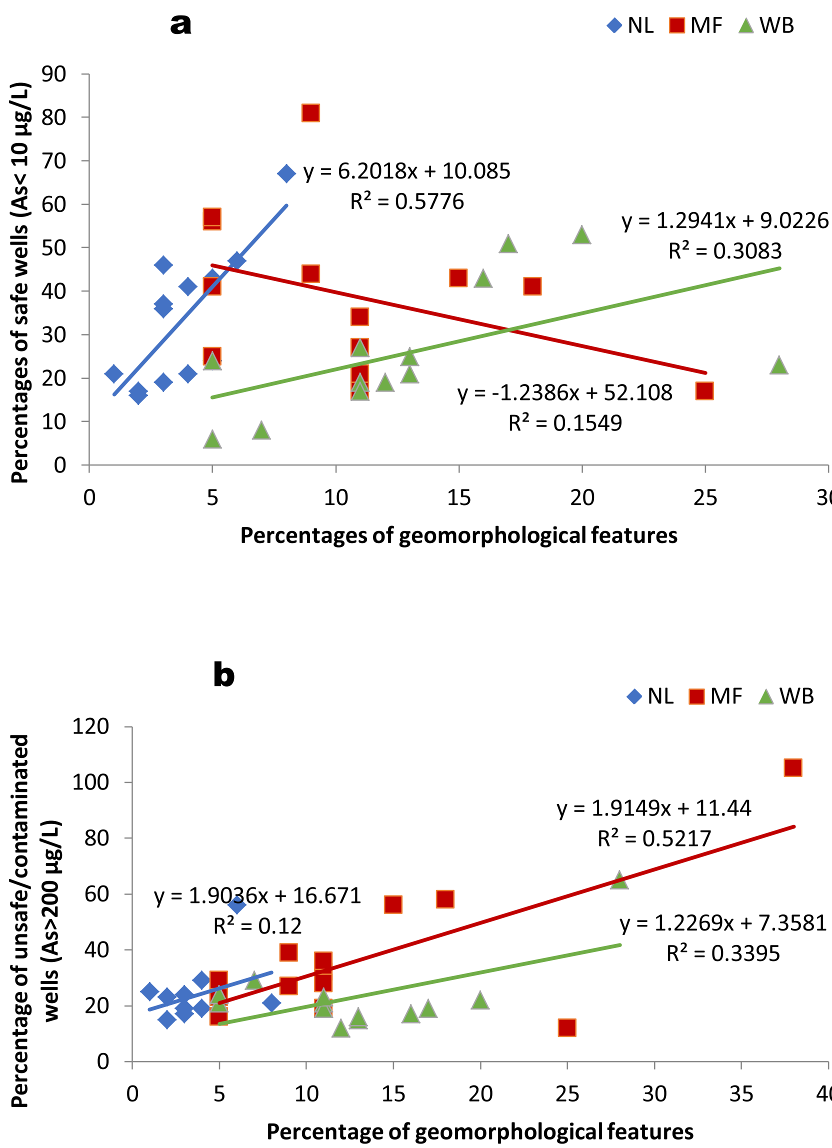

Fig. 5. Correlation of geomorphological features with (a) safer (As< 10 μgL^{-1}) and (b) Highest As areas (As > 200 μgL^{-1}) wells. [The abbreviations NL, MF and WB represent Natural Levee, Meandering Features and Water Bodies respectively].

(SOM) (PHED, 1993; CGWB, 1999; RGNDWM, 2001; DFG-BMZ, 2013; Biswas et al., 2014). A supply of organic carbon (often labile) usually from the shallow aquifer sediment has been reported to be the key player for As mobilization in the entire south-east Asian river basins (BGS and DPHE, 2001; Sengupta et al., 2008; Papacostas et al., 2008; Berg et al., 2008; Datta et al., 2011; Neumann et al., 2014).

The above study indicates that geomorphological features (flood plain, inter-distributaries levees, abundant channels, bils, khals, meander scrolls, meander belts, meander scars, oxbow lakes) do have a spatial connection to the occurrence of As in groundwater. Moreover, the distribution pattern of the geomorphological features with different size, shape, number and proximity are also important factors that often regulate the As concentration in groundwater. This could be the possible reason regarding the variation in the number of wells of highest As areas (As_T > 200 μgL^{-1}) and the number of safer wells (As_T< 10 μgL^{-1}) throughout the entire district. This water bodies are often degraded and centuries old. It is pertinent to note that the water level of these water bodies is fluctuating both in pre-monsoon and post-monsoon (Mukherjee et al., 2007a). As a result, the local recharge pattern is also varying concerning seasonal as well as temporal variation (Mukherjee et al., 2007b). These water bodies can also supply fresh organic matter to the aquifer (Neidhardt et al., 2013a). The nature, characterization, types, concentration, and bio-availability of these fresh organic matters are also important because they can largely contribute towards the reduction of Fe-oxides/oxy-hydroxides and thereby releasing As in groundwater (Biswas et al., 2011). Natural levees are found to be relatively safer from As contamination, which has been statistically supported (correlation analysis, Fig. 5a and b). The more elevated present-day levee is relatively less contaminated when compared with levees of paleo-channels. Around those levees, the As release might be influenced by both geogenic and anthropogenic processes (Nath et al., 2005; Bhowmick et al., 2013b; DFG-BMZ, 2013; Chatterjee et al., 2018). The geogenic framework is sedimentation pattern of fine-grained over-bank deposits (grey sands with silt/clay, Fe-coated colloidal particles, micas and organic-rich clay horizons) combined with natural water bodies (CGWB, 1999; RGNDWM, 2001; Biswas et al., 2014; Chatterjee et al., 2018). In BDP, ponds and wetlands are known to serve as potential sources of surface driven fresh organic matter (FOM) to shallow aquifers (Polizzotto et al., 2008; Lawson et al., 2013; Majumder et al., 2016). The supply of dissolved organic carbon (DOC) from anthropogenic sources could be due to many causes such as habitation (mostly pit latrines), settlements (cattle farming), organic-rich surface run-offs (Nath et al.,

Table 4
Multiple comparisons of mean As concentrations in different basins using LSD-Post Hoc where As concentration (As_T) is an independent variables.

Basin (I)	Basin (J)	Mean Difference (I-J)	Std. Error	Significance level (*p*-value)
Bhagirathi-Jalangi	Churni-Ichhamati	25.0308[a]	8.28893	0.003
	Hoogli-Bhagirathi	16.7195[a]	7.57009	0.027
	Padma	−50.1320[a]	7.48981	0.000
Churni-Ichhamati	Bhagirathi-Jalangi	−25.0308[a]	8.28893	0.003
	Hoogli-Bhagirathi	−8.3114[a]	8.72715	0.001
	Padma	−75.1628[a]	8.65761	0.000
Hoogli-Bhagirathi	Bhagirathi-Jalangi	−16.7195[a]	7.57009	0.027
	Churni-Ichhamati	8.3114[a]	8.72715	0.001
	Padma	−66.8515[a]	7.97208	0.000
Padma	Bhagirathi-Jalangi	50.1320[a]	7.48981	0.000
	Churni-Ichhamati	75.1628[a]	8.65761	0.000
	Hoogli-Bhagirathi	66.8515[a]	7.97208	0.000

[a] The mean difference is significant at the 0.05 level.

2005; Kulkarni et al., 2018a).

As the sediment becomes reduced (grey colour), a series of geochemical processes occur with the locally active organic carbon that leads to As mobilization gradually by reduction of sedimentation rich Fe-oxides/oxy-hydroxides (BGS and DPHE, 2001; Chatterjee et al., 2005; Charlet et al., 2007; Nath et al., 2008a; Sengupta et al., 2008). Arsenic mobilization in shallow aquifers of Nadia is possibly governed by various reactive organic matters in different forms (SOM, DOC and FOM). This is due to geomorphological processes which have formed differential landforms and land-use pattern and coverage. Reactive organic carbon originating from geogenic sources or drawn from anthropogenic resources serves as the energy source for microorganisms to elevate As levels in groundwater. The nature, type, reactivity and amount of the organics are widely varying and thereby producing spatial distribution and heterogeneity of As levels in shallow groundwater.

In this context, the geomorphological features are an important issue because they are the vulnerable sources of organics (mostly fresh organic matter), and thereby, changing the redox condition of the aquifer which leads to the release of As into groundwater.

5. Conclusions

An investigation of basin wise As distribution pattern shows that high As bearing areas are extensive in the north (Padma river basin) where several contaminated wells are found (up to 926 μgL^{-1}). The relatively low As or safer areas are restricted to the south-western part (Churni-Ichhamati river basin) and the range of contamination is varying (96.16–108.54 μgL^{-1}). However, localized high As wells are also noticed in low As areas. The role of geomorphological features and their relationship with groundwater As levels are statistically examined. Among these geomorphological units, natural levees show positive correlation ($r^2 = 0.57$, Pearson correlation co-efficient = 0.81 at $p < 0.01$) with safer wells, whereas, the correlations for natural water bodies ($r^2 = 0.30$, not significant) and meandering features ($r^2 = 0.15$, not significant) are not so pronounced. Contrastingly, for wells with high As concentrations, meandering features show some strong correlation ($r^2 = 0.52$, Pearson correlation co-efficient = 0.92 at $p < 0.01$) as compared to relatively weaker correlation with water bodies ($r^2 = 0.33$, Pearson correlation co-efficient = 0.56 at $p < 0.05$) and natural levees ($r^2 = 0.12$, not significant). The occurrence of higher As occurrence is principally due to the reduction of As-bearing Fe(III)-oxides/oxy-hydroxides under strong reducing conditions, possibly coupled to the mineralization of organic matter. In meandering features, the sediment trapped organic matter (SOM) is providing the necessary energy to dissolute the Fe(III)-oxides/oxy-hydroxides and thereby releasing As into groundwater. In natural levees, the supply of organic matter is surface driven from various land-use patterns and anthropogenic in origin. Under anaerobic condition, Fe(III)-oxides/oxy-hydroxides reduction coupled with microbially mediated oxidation of organic matter is possibly regulating the As release into groundwater. In these shallow reduced sedimentary aquifers, several geomorphological features are common and playing key role for cycling of redox status which is further aggravated by local recharge, sedimentation pattern (notably arsenic scavenging colloids), local groundwater abstraction, geomorphological elements, and land use pattern. Finally, the nature, type, pattern, frequency and distribution of geomorphological elements vary in local and regional scale and thereby controlling this geospatial distribution of As in groundwater of the study area.

Authors' contributions

Ayan Das: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Resources; Software; Writing - original draft Writing - review & editing. Santanu Majumder: Writing - review & editing. Sandipan Barman: Writing - review & editing. Debashis Chatterjee: Conceptualization; Investigation; Methodology; Resources; Supervision. Sutapa Mukhopadhyay: Conceptualization; Investigation; Resources; Supervision. Pinaki Ghosh: Writing - review & editing. Chandra Nath Pal: Methodology; Software; Writing - review & editing. Gopinath Saha: Writing - review & editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

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