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

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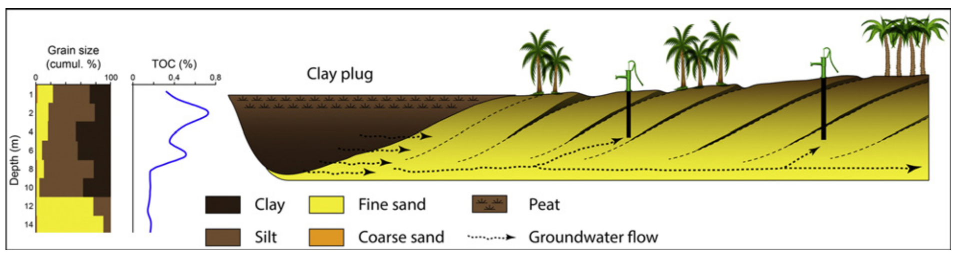
## HIGHLIGHTS

- 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

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## 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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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.