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Refereed article

Tracing Anthropogenic Impact on Arsenic Mobility in the Groundwater of Bangladesh and Southeast Asia — A Review of Methods and Results

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Summary

The effect of anthropogenic impact on groundwater resources in Southeast Asian countries is subject to scientific debate, since it is increasingly considered a contributor to the observed contamination of groundwater with arsenic. It can be diverse, and range from direct pollution, through alteration of hydraulic conditions to indirect effects on geochemical conditions and reactions. While direct pollution can be observed easily, the hydraulic and indirect impact on groundwater quality are more difficult to determine. Nevertheless, considering a generally increasing anthropogenic impact, its complex effects on groundwater chemistry and quality need to be kept in mind, especially if the causes are less obvious. In order to detect this impact, different approaches have been applied over the past few decades. This paper reviews the multiple indicators and approaches used in various studies to trace anthropogenic influences on geochemical conditions leading to high arsenic groundwater content in Bangladesh and South/Southeast Asia. Future research has to address increasingly the spatial complexity and subsurface heterogeneity through clay layers and hydrogeological differences, as well as the temporal development of local contaminant plumes.

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Introduction

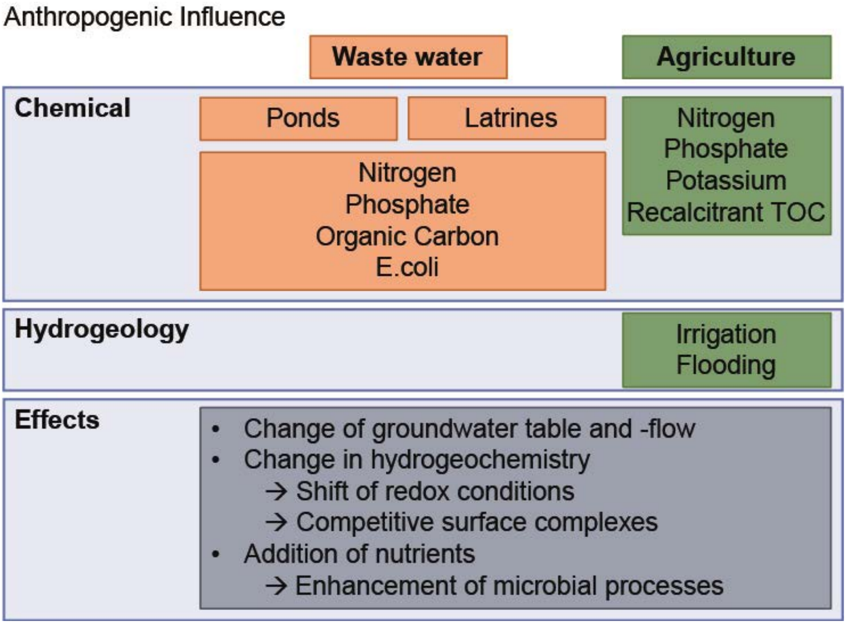
Arsenic in groundwater is a known problem in many countries of the world including Pakistan, India, Cambodia, Taiwan, China and Nepal (Fendorf et al. 2010: 6; Ali et al. 2019: 24). However especially problematic are the high arsenic concentrations in countries like Bangladesh, where groundwater is used directly for drinking water supply, without treatment or permanent monitoring (Chakraborti et al. 2015: 12). The continuous consumption of even trace amounts of arsenic can already cause various negative health effects (Abdul et al. 2015: 19). Therefore, the World Health Organization (WHO) and the United States Environmental Protection Agency recommend a 10µg/l threshold for arsenic in drinking water (WHO 2017: 564).

Since the discovery of these regionally high arsenic concentrations in groundwater, the causes leading to mobilizing conditions have been subject to extensive research (Mandal and Suzuki 2002: 35; Chakraborty et al. 2015: 12; Edmunds et al. 2015: 15; Masuda 2018: 11; Ali et al. 2019: 24). Many influencing aspects have been considered: geological (Mukherjee et al. 2009: 18; Verma et al. 2016: 20), geochemical (Smedley and Kinniburgh 2002: 52; Ahmed et al. 2004: 20; Biswas et al. 2011: 10, 2012: 10, 2014a: 10), mineralogical (Uddin et al. 2011: 14), hydrogeological (Harvey et al. 2006: 25; Stute et al. 2007: 11; Shamsudduha et al. 2009: 30), climatological (Majumder et al. 2016: 11; Kulkarni et al. 2018: 12), geomorphological (Mukherjee et al. 2009: 18) and anthropogenic (Neumann et al. 2010a: 7).

It is generally accepted that arsenic is naturally occurring in the geologically young alluvial sediments of the Ganges-Brahmaputra-Meghna basin (British Geological Survey (BGS) and Department of Public Health Engineering (DPHE) 2001: 289; Seddique et al. 2008: 13; Fendorf et al. 2010: 6). In these regions, reducing conditions are considered the main cause of high arsenic in groundwater. Under water saturation, reducing conditions can develop which lead to the microbially mediated dissolution of arsenic-bearing minerals like iron oxides and carbon minerals. Subsequently the released arsenic is reduced to Arsenic(III), which is more mobile in water than Arsenic(V) (Islam et al. 2004: 4; Itai et al. 2008: 22; Mukherjee et al. 2009: 18; Biswas et al. 2011: 10).

Anyhow, it remains largely unclear until today if, and how much, anthropogenic impacts affect arsenic mobility. This is largely due to the complex effects of anthropogenic influence on hydrogeology, geochemistry and thus geochemical conditions (see Figure 1 below). Therefore this article reviews previous works, aiming to answer the question of in what way and how much of a role these anthropogenic factors play in the arsenic mobilization on-site. Spatial focus will be on Bangladesh and adjoining regions like West Bengal, as geochemical processes are regionally diverse and furthermore strongly dependent on hydrogeological, geological and climatic conditions.

Figure 1. Complexity of Anthropogenic Influence through Waste Water and Agriculture on Geochemistry and Hydrogeology



Source: Author's own compilation.

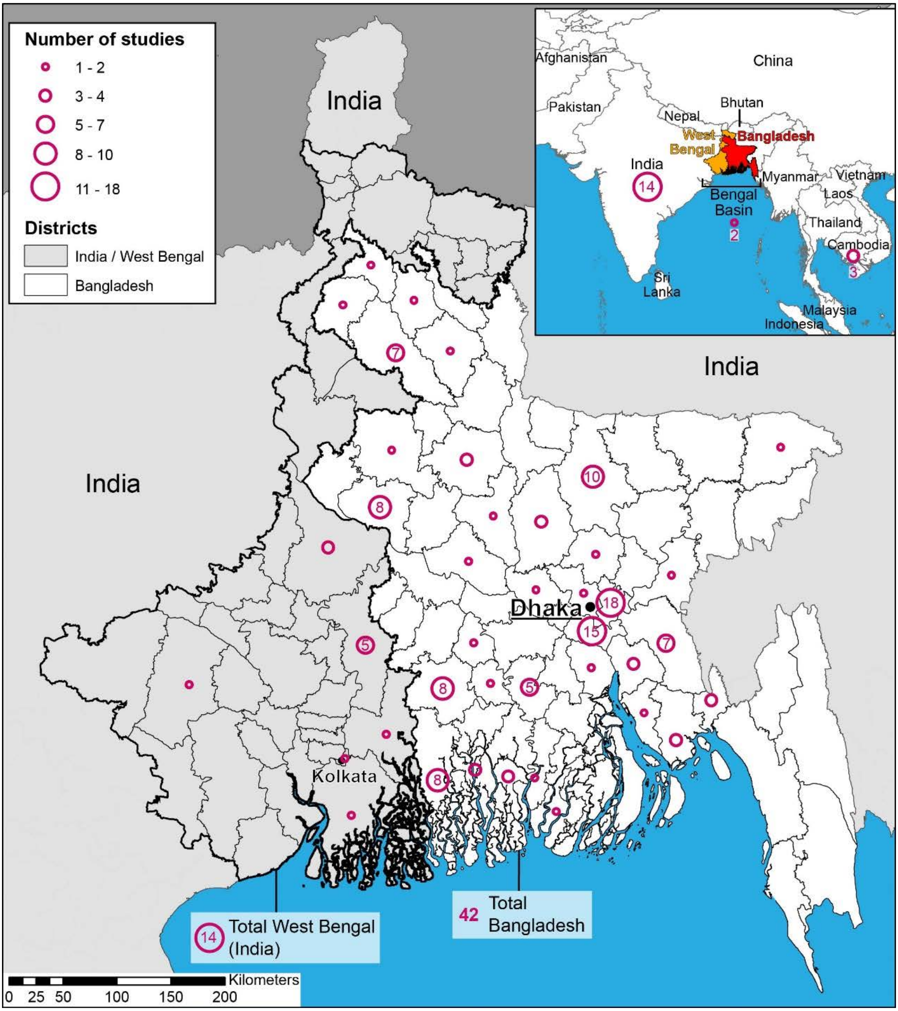
Although difficult to determine and to distinguish from natural impacts, the anthropogenic influence on groundwater in Bangladesh and other arsenic-affected countries has been considered in various studies (Neumann et al. 2010a: 7; McArthur et al. 2012: 13; Whaley-Martin et al. 2017: 9, see also, Table S1 in supplementary information). In the course of this article, different aspects of these approaches are presented and discussed.

Review of methods used to identify anthropogenic influences

Most commonly the chemical characteristics like high concentrations of phosphate, ammonium and chloride, or ratios between certain elements like chloride (Cl^-) and bromide (Br^-), are interpreted as indicators of waste-water or agricultural influence (e.g. Neumann et al. 2010a: 7; Anawar et al. 2011: 27; McArthur et al. 2012: 13). Hydrogeological effects are usually observed in extreme groundwater-table fluctuations and changes of groundwater-flow direction (Harvey et al. 2002: 5, 2006: 25). Waste-water influence is frequently regarded based on indicator bacteria like E.coli or other characteristic substances of waste-water pollution like the sewage contamination index (e.g. Banerjee 2011: 9; Whaley-Martin et al. 2017: 9). Through investigation of the stable isotopes of oxygen, hydrogen, carbon or tritium some studies try to determine the groundwater age and thus estimate at which depth the water is young enough to be impacted by anthropogenic factors (Klump et al. 2006: 8; Stute et al. 2007: 11; Datta et al. 2011: 5; Mihajlov et al. 2016: 26). Nitrogen and carbon isotopes are considered, to gain information about the origins of these elements (Itai et al. 2008: 22; Lawson et al. 2013: 10; Mailloux et al. 2013: 5). Meanwhile, the source and characteristics of dissolved organic matter (DOM) are observed based on spectroscopic signatures, fluorescence and molecular weight (Mladenov et al. 2015: 10). In further studies, batch and column tests are conducted to research the quantifiable effect of anthropogenic substances like waste or fertilizer on arsenic release (Anawar et al. 2006: 13). Other authors take into account the spatial context of anthropogenic influence (Nath et al. 2008: 19; Bhowmick et al. 2013: 9; Chatterjee et al. 2017: 4) or statistically evaluate the impact on arsenic release (Mukherjee et al. 2018: 17; Ahmed et al. 2019: 15; Islam et al. 2019: 21).

In Bangladesh, one of the arsenic hotspots is located downstream from the capital city of Dhaka, which has a large impact on hydrogeological and hydrogeochemical conditions. Two of the most intensively studied areas, Araihazar Upazila (18 studies) and Munshiganj District (15 studies), are located in this region (see Figure 2 below and Figure S1 in the supplementary information).

Figure 2. Spatial Overview of Conducted Studies Considered in This Review based on Tables S2 and S3 in the Supplementary Information



Source: Map data from Awhere-bmgf opendata ArcGIS (2014), ArcGIS Services (2018).

Agricultural influences

Agriculture is considered a main factor in changing hydrogeological (e.g. high pumping rates for irrigation) and hydrogeochemical (e.g. fertilizer use) conditions in Bangladesh. Acharyya et al. (2000: 11), referring to laboratory experiments by Manning and Martens (1997: 7), assume that intensification of agriculture with increased irrigation and phosphate-fertilizer application could enhance arsenic mobility. Whereas the study by Slaughter et al. (2012: 6) indicates that high phosphate concentrations even inhibit microbial processes and thus reduce arsenic mobility, statistical approaches also conclude that agriculture plays the second-most important role in arsenic mobilization after geogenic factors (Islam et al. 2019: 21).

Effects of high pumping rates

Primarily, high pumping rates influence groundwater levels and hydrogeological conditions. How they affect hydrogeochemistry, and thus arsenic mobility, as well as the long-term effects of high irrigation pumping are still subject to debate. Shamsudduha et al. (2009: 30) show that the groundwater levels in Dhaka city are falling at a speed of >1 metre/year due to higher abstraction than recharge rates – but mainly due to household and industrial use, and not to agriculture. Harvey et al. (2006: 25) use a lumped-parameter model to show how high pumping rates during irrigation season change the flow direction and speed of groundwater, as well as recharge rates. They find that during the irrigation season (January through March) groundwater levels drop between 1.2 and 4.9 centimetres a day (Harvey et al. 2002: 5). They also reason that 60 per cent of the aquifer recharge is from monsoon precipitation and 40 per cent from the infiltration of pond water, which is rich in organic carbon and therefore can enhance arsenic mobilization.

With regard to arsenic, some authors assume that the high pumping rates from irrigation wells are likely to decrease groundwater arsenic concentration (Harvey et al. 2005: 12; Stute et al. 2007: 11; Aziz et al. 2008: 15) because of flushing or dilution effects. Harvey et al. (2005: 12) discovered at their study site (Munshiganj, Bangladesh) that the lateral flow vigorously flushes the aquifer of solutes, thus contradicting the common opinion that lateral groundwater flow is very low (2 m/year, following Darcy's law (1856)), causing a relative accumulation of solutes (Smedley and Kinniburgh 2002: 52; Ravenscroft et al. 2005: 25; Anawar et al. 2006: 13).

Based on radiocarbon (^{14}C) measurements, tritium isotopes and hydrologic models, Harvey et al. (2002: 5, 2005: 12) conclude that irrigation strongly enhances downward migration of organic-rich water by intensifying groundwater recharge rates. They found that the water 19m below ground level was less than 50 years old, and reason that organic-rich surface water could reach 30m depth within 6.8 to 29 years. Through helium-tritium isotopes, Stute et al. (2007: 11) show that in the shallow aquifer (< 20m depth) the amount of arsenic increases with groundwater age. They observe that irrigation wells have lower levels of arsenic concentration than household wells, and thus suggest that irrigation pumping could mitigate the problem if the geochemistry is not affected. A similar observation was made by van Geen et al. (2008: 6), who come to the conclusion that the effect of the draw-down of young organic carbon is compensated for by the high flushing rates.

Mailloux et al. (2013: 5) determine the ages of microbial DNA, groundwater and sediment samples through ^{14}C and tritium isotopes. They find microbial DNA and groundwater to be significantly younger by several thousand years than total sediment carbon. Although they notice bomb-labelled[1] ^{14}C in the water up to 19m depth, they could not detect bomb-labelled ^{14}C at 11m depth in the microbial DNA. They thus conclude that anthropogenic organic carbon and perturbation reach depths up to 19m, but do not fuel or influence microbially mediated arsenic release.

Some authors suggest that these hydrogeological modifications and the draw-down of modern, organic-rich water affect chemical and hydrogeochemical processes (Neumann et al. 2010a: 7; Lawson et al. 2013: 10; Biswas et al. 2014b: 13). Other recent studies also consider the risk of arsenic-rich water from shallow aquifers being drawn down into deeper aquifers that have so far been arsenic-free through high pumping rates from deep wells (> 150m) (Mukherjee et al. 2011: 14; Khan et al. 2016: 8; Knappett et al. 2016: 13).

Wet-rice cultivation

Some studies conclude that the cultivation of wet rice enhances the problem of arsenic mobility. Arsenic from irrigation water accumulates in iron-oxide precipitations on the roots of the rice plant. Waterlogging of the fields for wet-rice cultivation alongside the continuous input of organic material lead to the development of strong reducing conditions. The latter cause the remobilization of the arsenic due to dissolution of the iron-oxide precipitate (Farooq et al. 2010: 14; Huang et al. 2012: 9).

Previous studies have shown that irrigation with arsenic-rich water leads to the chemical's accumulation in the surface soil (topmost: 2–75 millimetres) (Dittmar et al. 2007: 6; Bakhat et al. 2017: 17; Chowdhury et al. 2017: 10). Neumann et al. (2009: 15) investigate the groundwater recharge from rice fields, and find that irrigation water contributes to high arsenic concentrations in the uppermost aquifer through the bunds (unploughed rice-field borders). This is also supported by Dittmar et al. (2007: 6) and Saha and Ali (2007: 10), who find that arsenic concentration in the soil decreases during the wet season due to the reductive dissolution of arsenic-bearing iron oxyhydroxides, leached into deeper soil and/or transported away with rainwater.

High levels of arsenic concentration in soil and groundwater can cause increased accumulation of the chemical in crops. How much arsenic is taken up depends on the field crop as well as the means of cultivation (Gillispie et al. 2015: 13; Bakhat et al. 2017: 17). Huhmann et al. (2017: 8) demonstrate that high arsenic concentrations in soil and groundwater cause a lower crop yield.

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[1] Indicating this carbon is disturbed from nuclear bomb explosions between 1961 and 1962 (Stenhouse and Baxter 1977:5).

Changes in hydrogeochemistry through agriculture

Agriculture affects chemical conditions in the underlying aquifer through the addition of DOM and artificial fertilizers. How they influence arsenic mobility is still a topic of debate. Farooq et al. (2010: 14) conduct column tests with sediments to 9m depth taken from a rice field. They show that the DOM derived from the decomposition of the rice plant contributes to arsenic mobility by causing reducing conditions. They also observe a cyclical pattern due to the addition of arsenic to the fields during irrigation season. Neumann et al. (2010a: 7) notice – based on hydrogeological models, isotope signatures and biogeochemical analysis – that organic carbon is mainly added to the aquifer through ponds and the bunds. However, in contrast to other works, they prove that rice fields contribute little DOM to the depths where arsenic is high (30m), and furthermore that this field-derived DOM is more recalcitrant than the more reactive pond-derived DOM. They conclude that rice fields are net sinks of arsenic, because it binds to the soil of the fields and the bunds; ponds and anthropogenic perturbations in the subsoil are the main factors behind arsenic release meanwhile (Neumann et al. 2010b: 89, 2011: 7, 2014: 5).

Different studies have focused on artificial fertilizers, which mainly consist of phosphate and nitrogen compounds like urea (Itai et al. 2008: 22; Weng et al. 2017: 19). It is proven that phosphate strongly contributes to arsenic mobility through surface complexation and by enhancing microbial activity (Neidhardt et al. 2018: 14). Phosphate concentrations in the groundwater of Bangladesh are naturally very high (BGS and DPHE 2001: 289), and the effect of additional phosphate sources thus subject to debate.

Dhar et al. (2008: 15) find, based on time-series data for chemical groundwater composition at different depths, that young (five years old) shallow groundwater contains more phosphorous relative to arsenic content. They conclude that phosphorus is most likely derived from the surface, while arsenic is not. In the older groundwater (> five years old), arsenic and phosphorous have a ratio indicating a similar release mechanism.

Anawar et al. (2006: 13) conduct several incubation studies with different carbon sources and fertilizers. They show that urea and phosphate fertilizer enhance arsenic mobility, both through competition for sorption sites. The phosphate fertilizer used in their study also contained between 2 milligrams/kilograms to 7 mg/kg of arsenic, thus directly contributing to the problem. Lin et al. (2016: 9) perform several microcosm experiments under oxic and anoxic conditions with sediments from Taiwan to observe the effect of different fertilizer amendments. Under both oxic and anoxic conditions arsenic was released from sediment with dipotassium phosphate amendment, under oxic conditions mainly as Arsenic(V) and under anoxic conditions mainly as Arsenic(III). Ammonium sulfate amendments did not result in any arsenic release, which could be explained by the precipitation of the latter with the sulfate. In laboratory experiments Uddin and

Kurosawa (2011: 9) show that the addition of nitrogen fertilizer (urea) strongly triggers arsenic release.

Based on the distinct isotopic composition of nitrogen fertilizer ($\delta^{15}N$ between -4 and +4 ‰), different studies conclude that in Bangladesh it is the main source of nitrogen in groundwater (Itai et al. 2008: 22; Uddin and Kurosawa 2011: 14). Anawar et al. (2006: 13) postulate, though, that low nitrogen-isotope signatures are not an indicator of agricultural contamination. They object to the assumption that nitrate from urea fertilizers enhances microbial activity and thus arsenic release, as nitrate causes oxidizing conditions – consequently leading to iron, and therefore arsenic, precipitation.

The role of artificial ponds

Artificial surface waters are very common in rural Bangladesh, and fulfil different purposes. Household or community ponds are used for hygiene matters like bathing, laundry and waste-water disposal or for watering stock. Some ponds are mainly used for industrial purposes, such as irrigation, dyeing, jute processing or fish farming (Huda et al. 2010: 11; Knappett et al. 2011: 9; Escamilla et al. 2013: 10). Most of these ponds were dug in the last 50 years, and their water levels fluctuate seasonally. In older ponds, an accumulated clay layer at the bottom prevents them from falling dry during the dry season (Harvey et al. 2005: 12, 2006: 25; Huda and Atkins 2005: 8; Huda et al. 2010: 11; Neumann et al. 2010a: 7).

How much these ponds contribute to groundwater recharge is still under debate. Some authors suggest, based on hydrologic models and water-table fluctuations, that these ponds are connected to groundwater (Zheng et al. 2005: 16; Harvey et al. 2006: 25). Other works use indicators like oxygen and hydrogen isotopes or Cl/Br ratios to show that pond water does not contribute to groundwater recharge (Sengupta et al. 2008: 9; McArthur et al. 2012: 13). While Majumder et al. (2016: 11) state that evaporation influences water from surface waters and Cl/Br relations indicate major recharge through vertical mixing at their study site in India, West Bengal, Richards et al. (2017: 14) investigate redox zones based on isotope-mixing models and find that ponds contribute to aquifer recharge and the prevailing redox milieu – especially during rainy season.

Another topic of debate is the organic carbon which is added through these ponds. Neumann et al. (2010a: 7) show – based on numerical simulation of transient three-dimensional groundwater flow and different indicators – that organic carbon from ponds plays a significant role in arsenic mobilization. This is supported by Lawson et al. (2013: 10, 2016: 18) who conclude – from carbon isotopes at sites in Cambodia and West Bengal – that young organic carbon from ponds can be drawn down up to 100m by intensive groundwater abstraction, and thus cause or enhance arsenic release. In contrast, Datta et al. (2011: 5) come to the result that ponds in West Bengal do not contribute to groundwater recharge or organic carbon based on stable isotope (oxygen, hydrogen, carbon) data.

Farooq et al. (2012: 12) investigate the effect of pond-derived organic carbon on a 9m-deep sediment core from a jute-processing pond in West Bengal. They show that the organic carbon from the pond triggered arsenic release, but the groundwater was not affected. The released arsenic was found to be immobilized, and thus enriched in deeper soil regions.

The role of waste and waste water

About 85 per cent of households in Bangladesh own a sanitary latrine, which means faecal matter is disposed of to minimize contact with human beings. Sanitary latrines can be simple pit latrines, septic tanks, or flush toilets with a sewage system. In most parts of the country, the water and waste-water infrastructures are decentralized (Hanchett et al. 2011: 123; Tilley et al. 2014: 180; Gunawan et al. 2015: 58; Hanchett 2016: 22).

Common latrine systems are pit latrines and septic tanks, which need emptying and proper disposal. As studies by the WHO/UNICEF Joint Monitoring Programme show, most latrine systems are never emptied – which implies that they discharge into groundwater or nearby surface waters (Bangladesh Bureau of Statistics and UNICEF 2015: 272). Due to the high population density, wells are usually located close to latrines. Safe distances between latrines and wells are suggested to lie between 3m and 25m, depending on the hydrogeological setting (ARGOSS 2001: 97; Graham and Polizzotto 2013: 10).

Waste-water influence based on E. coli

A commonly used marker for waste-water influence is the faecal indicator bacteria E. coli (faecal coliforms, total coliforms, or thermo-tolerant coliforms), which has been thoroughly researched in Bangladesh. During the rainy season, groundwater tables rise and cause higher groundwater pollution. As some latrines are flooded during the rainy season, the highest microbial contamination with E. coli is observed during this time of the year (Pujari et al. 2012: 13; Escamilla et al. 2013:10; Hanchett 2016: 22; Islam et al. 2016: 10; Dey et al. 2017: 16). Knappett et al. (2011: 9, 2012: 10) show that ponds receiving direct waste water had high E. coli counts but did not contribute faecal contamination to the nearby groundwater, with the exception of newly dug ponds. Banerjee (2011: 9) tests transport distances of faecal indicator bacteria from simple pit latrines under the real hydrogeological conditions found in West Bengal. His work proves that E. coli can be transported up to a maximum of 10m distance from a latrine. The transportable nature of microbes and salt is increased with high groundwater tables and flooded pits during monsoon season, and with artificial pumping.

Several recent works indicate that microbial contaminations from pit latrines are not a sufficient indicator for faecal contamination, as they are conveyed in a very different way from chemical compounds (McArthur et al. 2012: 13; Back et al.

2018: 19). Graham and Polizzotto (2013: 10) show that inside latrine pits a natural biofilm forms that limits the microbial contamination. A recent study by Ravenscroft et al. (2017: 10) even implies that microbial contamination found in groundwater is mainly caused by the poor condition of most wells, like broken surface seals and the use of dirty priming water. Thus they assume that the actual contamination is inside the well pipe and not the aquifer. Leber et al. (2011: 13) find that the geological setting has a major influence on E. coli and arsenic concentrations in groundwater. They describe high arsenic concentrations, but low E. coli counts in areas with a surface clay layer.

Other chemical influences

Besides microbial contaminations like E. coli, waste water from latrine systems contributes chemical components like nitrate, chloride, ammonium, sulfate and organic carbon to subsurface groundwater and ponds (Itai et al. 2008: 22; Graham and Polizzotto 2013: 10). Earlier studies by McArthur et al. (2004: 39) regard the influence of waste water to be a point source of organic matter, causing local reducing conditions with arsenic release. They argue that latrines are evenly distributed across the country, while arsenic is not found everywhere. Furthermore, they consider coliform counts, which were observed to parallel the levels of concentration of ammonium and phosphorous in wells proximate to latrines. However high ammonium also occurred in wells without faecal coliform contamination, and thus a different source like peat was presumed to be the main driver of arsenic release (McArthur et al. 2001: 9, 2004: 39).

In contrast to this observation, Lawrence et al. (2000: 12) describe the complex effects of waste-water input on the geochemical groundwater conditions and arsenic mobility in a rapidly growing city in Thailand. Here the large-scale input of organic carbon from unsewered latrines to ponds, canals and pit latrines led to the development of strong reducing conditions. Consequently, high ammonium, chloride and thus arsenic concentrations were found in the aquifer used for drinking-water supply.

Detailed investigations by van Geen et al. (2011: 7) and McArthur et al. (2012: 13) reveal the nitrate, sulfate and oxygen co-deposited with waste close to latrines, thus causing oxic conditions in direct proximity – along with high microbial contaminations too. Accordingly, reducing, iron- and arsenic- mobilizing conditions evolve with distance away from the latrine – being most likely within 30m downgradient from the waste-water source. Consequently, these authors conclude that faecal bacteria are usually not present in the most contaminated areas of the evolving waste-water plume because they show different behaviour with regard to their transportation further afield. This hypothesis is supported by the bell-shaped solute profile similar to typical contaminant plumes described by Harvey et al. (2006: 25) and as outlined in the geochemical investigations of Mukherjee et al. (2018: 17) too.

In recent studies, additional indicators have been considered to identify wastewater influence on groundwater in Bangladesh. The ratio of Cl to Br is commonly used as an indicator for waste water, as urine contains high amounts of salt. Accordingly, with increasing urbanization chloride concentrations and thus the Cl/Br ratio rises (Alcalá and Custodio 2008: 19; Katz et al. 2011: 16). McArthur et al. (2012: 13) find, based on Cl/Br ratios, that in Bangladesh and West Bengal more than 25 per cent of well water contains more than 10 per cent waste water.

A different indicator is considered by Whaley-Martin et al. (2017: 9), who use the sewage contamination index based on steroid markers found in sedimentary samples in addition to Cl/Br ratios in groundwater. They noticed that the highest arsenic and iron concentrations coincide with high sewage indicators. In contrast, Mladenov et al. (2015: 10) discover, based on investigations of spectroscopic signatures of DOM, that groundwater younger than five years old with low arsenic concentrations showed characteristics of sewage pollution. While highly contaminated with arsenic, old groundwater (> 30 years old) contained primarily DOM from sediments meanwhile.

In the batch experiments conducted by Anawar et al. (2006: 13), the effect of different anthropogenic carbon sources on arsenic mobilization are tested. The results indicate that household waste and cow dung led to a distinctive mobilization of arsenic under anoxic conditions. A different approach includes the consideration of the spatial context, through the statistical evaluation of land-use patterns. In Bangladesh and West Bengal, a strong correlation is described as existing between the use of land for ‘sanitation’ and high arsenic levels (Nath et al. 2008: 19; Bhowmick et al. 2013: 9; Chatterjee et al. 2017: 4).

In Bangladesh and West Bengal, latrine systems are usually seen as a point source of groundwater contamination. In a study by Back et al. (2018: 19) in Malawi (Africa), meanwhile, the septic latrines widely used are regarded as overlapping point sources which impair groundwater quality. The authors do not consider the complex geochemical impact, however.

Estimation of input quantities

Human impact on substance input is definitely the case if considering an estimation of how much nitrate, phosphate and carbon are contributed hereto through waste water and fertilizer application (see Table 1 below). Based on the latest available numbers regarding fertilizer use, Bangladesh applied 1,172,300 tons (or 12.9g/m^2) of nitrogen and 638,480t (or 7g/m^2) of phosphorous fertilizer to its agricultural land in 2016 (Food and Agriculture Organization of the United Nations (FAO) 2020). Even if the crops absorb two-thirds of this nutrient application, 2–4g phosphate or nitrate per square metre might leach into the aquifer (with mostly unknown effects).

It can be calculated that in 2016 the population of Bangladesh produced about 111,793t of phosphorous (0.8g/m^2), 782,550t of nitrogen (5.3g/m^2) from urine, and 977,775t of carbon (6.6g/m^2) through waste water. This is a liberal calculation, which does not consider lower waste production from children and that some of the waste is centrally treated. In contrast to fertilizer, these nutrients are directly discharged into the aquifer through septic tanks, waste-water ponds or latrine pits. These numbers indicate that anthropogenic input of chemical substances through agriculture and latrine systems might become a critical factor in the process of not only arsenic mobilization but also regarding the quality of drinking water in densely populated countries in general.

Table 1: Calculation of Anthropogenic-Substances Input into Groundwater through Fertilizer and Waste Water in Bangladesh

	in tons [t]			in grams per square metre [g/m^2]		
	Phosphorous	**Nitrogen**	**Carbon**	**Phosphorous**	**Nitrogen**	**Carbon**
fertilizer 2016	638,480	1,172,300	n/a	7.0	12.9	
urine (1 year)	82,374	411,868	565,907	0.6	2.8	3.8
faeces (1 year)	29,419	370,681	411,868	0.2	2.5	2.8
waste water (= urine + faeces)	111,793	782,550	977,775	0.8	5.3	6.6

Source: Author's own compilation.
Note: Calculations based on numbers given by Rose et al. (2015: 53) and the United Nations, Department of Economic and Social Affairs, Population Division (2015: 66).

Conclusion

The presented review, featuring diverse approaches by various authors and their subsequent conclusions, covers many years of research in differing but comparable study areas. It leads to the conclusion that although the geological framework plays a major role in arsenic mobilization, the effect of anthropogenic influence on hydrogeology and geochemistry should not be underestimated. Initially, faecal indicator bacteria were interpreted as a sign that anthropogenic input through waste water only contributes locally to arsenic mobilization. The implications of more recent studies based on multiple indicators show that spatial complexity and subsurface heterogeneity through clay layers and hydrogeological differences, as well as the temporal development of local contaminant plumes, need to be addressed in future research.

As of now, the total effect of anthropogenic influences on the hydrogeology and chemical composition of groundwater is not yet completely understood. The sources and roles of substances like phosphate, ammonium or nitrate remain subject to debate. The spatial complexity of the involved processes complicates this understanding, as the released arsenic is transported away from the source. Processes of sorption and desorption also affect its distribution.

Generally, it can be concluded that rising population numbers are likely to have an increasing impact on surface water and groundwater. Rising numbers of sanitary latrine systems should especially be considered, in sum, as more than a local point source of different nutrients. Even if anthropogenic factors do not influence arsenic mobilization yet, as concluded from groundwater dating, the future consequences might be considerable, for example as soon as young organic carbon reaches more critical depths.

Therefore, regular, nationwide testing of tube wells and further studies are advisable in order to understand the manifold pathways of anthropogenic impact on groundwater arising from agriculture, industry and households. So far, it remains unclear which relevant substances are primarily leached into the groundwater and how they possibly affect geochemical processes – especially in tropical and subtropical climatic regions. These components need to be investigated more explicitly, as geochemical and hydrogeological conditions both have a spatial and temporal component. Interdisciplinary research with multilevel groundwater sampling, land-cover and land-use monitoring through remote sensing will be carried out by our work group in the next few years. Still, a nationwide data set of groundwater characteristics is necessary to better understand both past and recent processes.

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