Arsenic Geochemistry and its Impact in Public Health: The Bangladesh Case (Geoquímica del Arsénico y su Impacto sobre la Salud Pública: El Caso de Bangladesh)

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Abstract

Considered the king of poisons, arsenic occurs naturally in the environment being present in air, soil, water and food. Its presence in drinking water is of global concern. Initial chronic exposure is manifested by skin lesions. Additionally, arsenic consumption impairs certain visceral organs: bladder, liver, prostate, etc. More over, arsenic is a recognized carcinogenic substance.

When in Bangladesh started the program to lead safe drinking water in the 60's, they never imagined the catastrophic consequences. Water wells were drilled in the whole country. The arsenic problem was recognized recently in the 90's.

In order to understand the nature of arsenic in the environment and how it could possibly reach groundwater in Bangladesh, this work explains some chemical characteristics of arsenic, the geological formation of the basin, and its mobility.

The origin of arsenic contamination in the Bangladesh Delta is due to the geologic nature of the basin rather than the possibility of an arsenic rich mineral. The profile of sediments shows that the Delta is not homogeneous, but rather heterogeneous even in closer areas. The driving process for arsenic mobility is mainly the reduction by iron oxyhydroxides coupled with organic matter, including other factors such as particle size, depth, morphology, metal content, as well.

Keywords: Arsenic, Bangladesh, arsenic geochemistry, arsenic mobility, arsenic diseases

1. Introduction

Considered the king of poisons, arsenic occurs is found naturally in the environment being present in air, soil, water and food. (Nriagu et al. 2007) But human intervention has modified its natural occurrence making its presence quite dangerous. In many parts of the world (i.e.

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Argentina, China, Taiwan, Chile, etc.) it has been reported cases of chronic toxicity due to water with arsenic consumption. (Smedley and Kinninburgh 2005, Rahman 1999) It is not clear what the lowest safe limit for arsenic presence in drinking water is, but it is recognized that at high levels it has negative consequences in human health. (Rahman 1999) Hyperkeratosis and hyperpigmentation of skin are the initial signs of chronic poisoning and this is used to identify arsenicosis and as a biomarker in humans. (Kinninburgh 2003, Mazumder 2003).

It is the case of Bangladesh, one of the most densely populated countries in the world, where the presence of arsenic in groundwater and its consumption has caused the greatest environmental disaster and impact in public health in the world.

Aided by international agencies, starting in the 1960's, the Bangladesh government started a program with the aim to provide safe drinking water to about 97% of the population. For this, more than 4.5 million shallow and deep tube wells were drilled, without analyzing the quality of its water. The low cost was very attractive, US\$ 0.30 per meter. In 1980 were detected the first cases or arsenic chronic poisoning, but in the '90s this was recognized as a problem, and so was its real magnitude by beginnning of the century. (Mukherjee & Bhattacharya 2001)

This work will attempt to present a geological perspective of the problem related with health, showing the sources of arsenic and its mobilization.

2. Arsenic Chemistry and Geochemistry

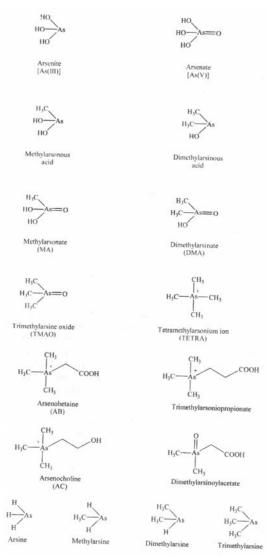
Presence of Arsenic in the Environment

Arsenic (MW = 74.921 g-As·mol⁻¹, AN = 33) is an ubiquitous element with metalloid properties. In its elemental form, arsenic is a grayish metal, and is found next to selenium in the periodic table (group V-A), in the same group as phosphorous. In our context it will be referred to either as arsenite or arsenate, at least when further precision is not required.

Arsenic, the king of poisons, is a natural constituent in the Earth crust and ranks 20th in abundance, with an average of 2-3 mg-As·kg⁻¹. (Nriagu et al. 2007) Arsenic is present in the environment in two forms: organic and inorganic. The organic form is quite diverse, produced mainly by microbial activity, or found in marine and terrestrial living organisms: microorganisms, algae, terrestrial fungi and lichens, plants, and terrestrial and marine animals. Some important compounds are arsine, arsenobetaine, tetramethylarsonium ion, arsenocholine, methylarsonate, dimethylarsinate, trimethylarsine oxide, methylarsines, arsenosugars, etc. (Figure 1). (Kumaresan & Riyazuddin 2001, Francesconi & Kuehnelt 2002, Bissen & Frimmel 2003)

The sources of arsenic in the environment can be natural or anthropogenic. The natural sources are much larger than the anthropogenic ones, mainly from volcanic activity and low temperature volatilization (biological methylation). It is also found in drinking water from wells. On the other hand, anthropogenic sources of arsenic originate from the use of pesticides,

copper smelters, mining activity, landfill from dumping slag, wastewater from smelters and refineries, which contribute indirectly to land and water contamination. (Hutchinson & Meena 1987, Bissen & Frimmel 2003)



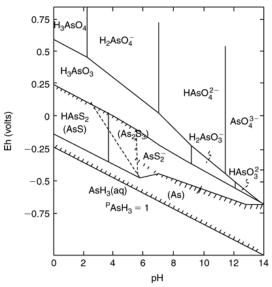


Figure 2. Arsenic speciation in water at different pHs. Source: Francesconi & Kuehnelt 2002

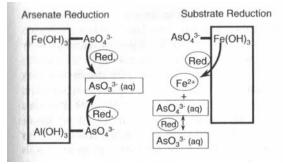


Figure 1. Some inorganic and organic arsenic compounds and their respective chemical structures. Source: Francesconi & Kuehnelt 2002

Figure 3. Posible reduction mechanism on clay surfaces. Source: Inskeep et al. 2002

Arsenic is also found in mineral forms, being arsenopyrite (**FeAsS**) the most abundant arsenic-containing mineral from more than 200 minerals. Other important minerals include realgar (As_4S_4) and orpiment (As_2S_3), which are reduced forms of arsenic. Arsenolite (As_2O_3) is an oxidized form. Other minerals consist on olivenite (**CuOHAsO**₄), cobaltite (**CoAsS**), prousite ($Ag_3 AsS_3$), nicolite (**NiAs**), etc. (Francesconi & Kuehnelt 2002, Bissen & Frimmel 2003, Nriagu et al. 2007) Table 1 shows the percentage of natural abundance of As in crustal material.

Rock type	Range of As
	concentration (mg⋅kg ⁻¹)
Igneous rocks	
Ultrabasics	0.3-16
Basalts	0.06-113
Andesites	0.5-5.8
Granites/silicic volcanic	0.2-13.8
Sedimentary rocks	
Shales and clays	0.3-490
Phosphorites	0.4-188
Sandstones	0.6-120
Limestones	0.1-20
Coal	
Bituminous	9.0±0.8
Lignites	7.4±1.4
Peat	16-340

Table 1. Natural abundance of As in crustal materials. (Source: Nriagu et al. 2007)

Aqueous Chemistry of Arsenic

In freshwater systems arsenic concentrations vary according to the geological composition and the input of anthropogenic sources, being reported 0.1-80 μ g-As·L⁻¹ as a typical range, but in seawater it is typically 1-3 μ g-As·L⁻¹. Arsenic in water takes mainly the form of pentavalent arsenate, **As(V)**, which predominates under oxidant conditions; on the other hand the trivalent arsenite, **As(III)**, is what prevails in reducing environments. Both forms (protonated oxyanions in aqueous solutions) and the degree of protonation are pH dependant. Figure 2 shows the different species of arsenic under different redox conditions. (Kumaresan & Riyazuddin 2001, Le 2002, Francesconi & Kuehnelt 2002, Bissen & Frimmel 2003, Stollenwerk 2003)

The controlling factor on environmental fate and transformation of arsenic in the subsurface is driven by natural and biological processes. Although arsenite is thermodynamically unstable in aerobic environments, its conversion to **As(V)** occurs very slowly. However, the conversion rates increase when sensitive redox species are present. Diverse researches have connected the presence of clay minerals, manganese oxides and hydroxides (goethite) directly to the oxidation process (Kumaresan & Riyazuddin 2001, Le 2002, Stollenwerk 2003)

Arsenic Sorption

It is believed that the sorption of arsenate and arsenite on Fe(III)-oxide and aluminum solid phases is perhaps the most important sink for As in aquatic and terrestrial environment. Additionally, it is necessary to take into account that some microorganisms are capable to use

arsenic for metabolic purposes. It means that arsenic mobility depends on pH, anaerobic/aerobic conditions, sorption on Fe(III)–oxide phases, and microbial activity. (Inskepp et al. 2002, Stollenwerk 2003) Figure 3 shows a possible mechanism on Fe(III)– oxide surface, which is largely being controlled by pH, especially at lower values for arsenate in contrast to arsenite with a maximum adsorption at about pH 8-9. Figure 4 shows the bonding nature of arsenite and arsenate on it.

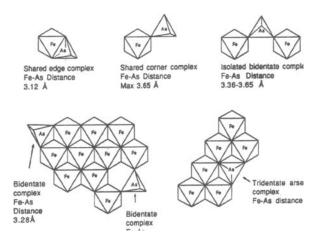


Figure 4. Possible bonding sites of As(V) on hydrous ferric oxide phase such as goethite. Source: Inskeep et al. 2002

3. Arsenic and Health

Basically, humans are exposed to arsenic in three ways: via air, food and water. The present work is focused on arsenic exposure by drinking water.

Related Arsenic Diseases by Drinking Water Consumption

Arsenic exposure is better evaluated taking into account not only the concentration in water but also the daily water consumption. When arsenic is consumed it accumulates in different parts of the body or excreted by it. Arsenic can be found in blood, nails and hair, and excreted by urine and feces. (Yamauchi & Fowler 1994, Yoshida 2004) Bladder, kidney, lung liver and prostate are target organs impaired by arsenic in drinking water consumption, being cancer the malignant effect of chronic exposure. (Rhaman 1999, Yoshida 2004)

Studies carried out concluded that chronic arsenic poisoning resulted from chronic exposition of as little as $0.15-3.3 \text{ mg-As} \cdot \text{day}^{-1}$ in drinking water and in food prepared with this water. (Pontius et al. 1994, Rahman et al. 1999) The effects by the consumption of drinking water contaminated by arsenic are delayed, appearing in the form of skin lesions including hyperpigmentation, hyperkeratosis (Figure 5 A, B), and various cancers. The first manifestations appear after 5 years of low exposure. (Pontius et al. 1994, Chen & Lin 1994, Yoshida 2004)

Arsenic is recognized as a human carcinogen. There is a strong correlation between the consumption of arsenic in drinking water and skin and visceral cancer, depending on age and

time exposure. The major incidence of cancer is during middle age, which reflects long latent period until the expression of this disease. (Chen & Lin 1994, Yoshida 2004)

Changes in the skin such as melanosis, dermatitis and hyperkeratosis are reported as well. According to Rahman studies in Bangladesh (1999), there is a substantial epidemiological evidence of association between well water containing inorganic arsenic (used as drinking water) and the occurrence of black foot disease. This disease is a vascular disorder resulting in a dry gangrene and spontaneous amputation of affected limbs (Figure 5 C-D). This disease was named after its most striking clinical feature, i.e. blackish discoloration of feet or hands.









Figure 5. (A-B) Hyperkeratosis and hyperpigmentation. (C-D) Loss of limbs by Blackfoot disease.

Source: http://www.sos-arsenic.net/english/latest.html; http://sitemaker.umich.edu/section7group1/effects_of_arsenic_contamination

Studies carried out by Tondel et al. (1999) show that hypertension and neuropathy are associated to low levels of consumption (less than $0.2 \text{ mg} \cdot \text{L}^{-1}$) of arsenic in drinking water.

It is significant to highlight that dietary (Rahman 1999, Kile et al. 2007) and socio-economic condition (Rahman 1999, Argos et al. 2007) also plays an important role in the entailed sensitivity to arsenic toxicity. Also the gender difference is another important factor, being males more susceptible to this toxicity than women. (Rahman et al. 2006)

Most studies related to Bangladesh case and other associated are focused on the above mention arsenic diseases, especially on skin. However, other aspects of chronic exposure arsenic less studied are related with miscarriages (Rahman et al. 2007), intellectual impairment in children (Wasserman et al. 2007), or diabetes (Navas-Acien et al. 2006). In those studies it was shown a significant link between dose and the response to arsenic exposure.

Because most studies are mainly focused on arsenic, other substances of public health interest present in Bangladesh groundwater are not considered or are studied apart, such as manganese. More over, there are very little studies which try to correlate arsenic and other substances, such as the role of humic substances (Wang et al. 2003, Warwick 2005); evidently it will complicate the studies, but it could complete the whole view of the problem.

4. The Bangladesh Case

Mukherjee and Bhattacharya (2001) strongly suggest that the most acceptable hypothesis for the widespread contamination in Bangladesh is a geologic cause. When high-As groundwater sources were first discovered, it was thought that the cause was a possible As-rich source mineral, because it is a common problem specially coming from mining activities. However, a more extensive research realized that high-As groundwaters are possible, though following other mechanism for its extension. (Kinniburgh et al. 2003) In this section it will be presented the geologic nature of the Bengal Delta Basin and some explanations about the arsenic mobilization in the basin.

Bengal Basin Geology and Morphology

The Indo-Burman range at the east, an uplifted block of Precambrian Shield (Shillong Plateau) in the north, and Precambrian basement complex (Indian Shield) in the west surrounds The Bengal Delta Basin (Figure 6). (Ahmed et al. 2004)

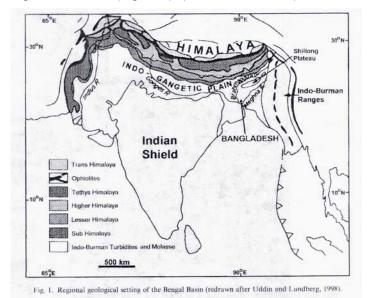


Figure 6. Regional geological setting of the Bengal Basin. Source Ahmed et al. 2004

The Bengal Delta Basin (BDP) lies on mainly Holocene, shallow aquifers (<150 m deep), composed of *alluvial* and *deltaic sediments*, which are deposited by the flow of Ganges, Brahmaputra, and Meghna river systems (GBM). (Smedley & Kinninburgh 2005) It is necessary to indicate that more than 16 km thick synorogenic Cenozoic sediments are deposited in the basin derived from the Himalaya. (Ahmed et al. 2004) In addition, a 15km sequence of Cretaceous to Recent sediments occupies some 100,000 km² of lowland floodplain and delta. (Kinninburgh et al. 2003)

Coarse alluvial fan deposits with dominant sand and gravel prevails in the upper reaches of basin; meander belts consisting on levee, blackswamp, ox-bow lakes, and abandoned channel deposits are common in the middle parts; and fine silts and clay but with some sand horizons can be found in the lower reaches of delta, marsh and tidal flat deposits. (Smedley & Kinninburgh 2005) Sediments can be characterized by its origin as: Tertiary sediments represented by sandstone and shale sequences; Pleistocene sediments mainly by clay, overlain by Holocene alluvium. In the Holocene plain lands includes piedmont plains, flood plains, delta plains and coastal plains. (Ahmed et al. 2004)

The geomorphology of the GBM system has developed in response to a series of glacioeustatic⁴ sea-level cycles and long term tectonic activity. (Kinniburgh et al. 2003) The next geomorphological units in the Holocen landmasses can be defined: fan deltas of the Tista and Brahmaputra; fluvial flood plains of the Ganges, Brahmaputra, Tista and Meghna rivers; delta plain of the lower GBM system south of the Ganges and Meghna valleys, including the moribund Ganges delta and the Chandina plain; Pleistocene Terraces and the subsiding basins in the eastern Ganges tidal delta and the Sylhet basin adjacent to the Dauki Fault (Figure 7). (Ahmed et al. 2004)

During the late Quaternary or the Holocene age were formed the sediments deposits pool of arsenic-contaminated groundwater, being the parent rock source from the Himalayas and the highlands closer to the area. The sediments form a succession series of layer, its lithology includes sand, representing the channel facies⁵, and the over bank facies, comprising predominantly silt and clay. The fined-grained overbank facies are rich inorganic matter.

⁴ Series of retreats of ice sheets with increase of sea level.

⁵ A part of a rock or group of rocks that differs from the whole formation (as in composition, age, or fossil content)

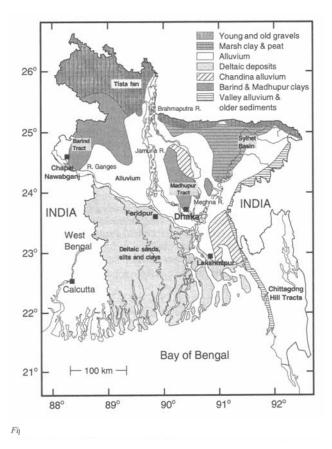
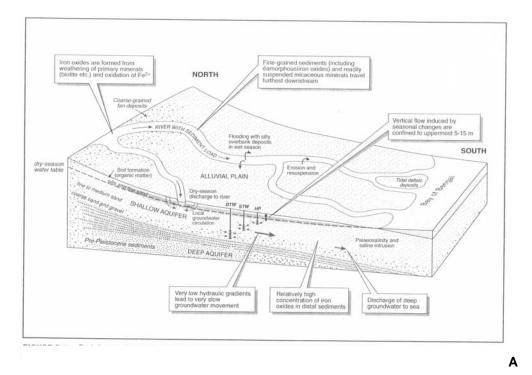


Figure 7. Bengal Basin sediment distribution. Source Ahmed et al. 2004

Figure 8 shows the process of in the mobilization and speciation of arsenic in the delta. Figure 9 represents a typical sedimentary succession with stacks of fining upward sequences in Tala, Satkhira district in southwestern Bangladesh. (Mukherjee & Bhattacharya 2001)

In addition, several isolated geological formations have been found as source of arsenic: Samthan Hill near Darjeeling, West Bengal, India; Singbhum in Bihar India, arsenic-bearing nickel ores, Chottonagpur and Shillong Hills, India; iron ore deposits in eastern India from the upper to the lower part Ganges Bengal Basin; Gondwana coal; volcanic terrain.



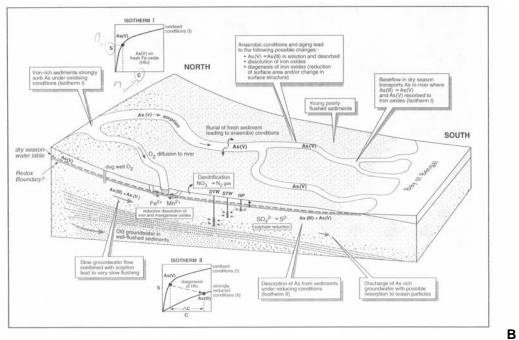


Figure 8. Mechanism of iron and arsenic mobilization in the basin. Source Mukherjee & Bhattacharya 2001

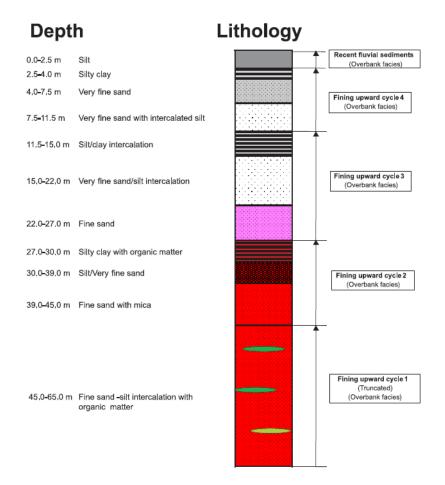


Figure 9. Example of sediment profile in the studies basin. Source Mukherjee & Bhattacharya 2001

Arsenic Mobilization

Ahmed et al. (2004) concluded that arsenic is mainly released in groundwaters of GBM Delta due to the reductive dissolution of iron oxyhydroxides. In addition, organic matter degradation is also coupled to that process. However, the extension of this mechanism depends on many factors. Among those factors it can be pointed out: biogeochemical process, Fe, Mn and Al content in the sediments, size of sediments, and morphology of terrain (Tareq et al. 2003, Ahmed et al. 2004)

5. Conclusions

The origin of arsenic contamination in the Bangladesh Delta is due to geologic nature of the basin rather than the possibility of an arsenic rich mineral. It is shown that the profile of sediments is not homogeneous in the Delta, but it is rather heterogeneous even in closer areas. The reduction by iron oxyhydroxides coupled to organic matter acts as the driving force of arsenic mobility. Its extension would have to include factors such as particle size, depth morphology, metal content, etc.

It is clear that the associated diseases will continue appearing, especially after a long term exposure to low content of arsenic.

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