Influence of Upstream Sediment on Arsenic Contamination of Groundwater in Bangladesh

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Abstract

The Ganga- Padma and Brahmaputra-Jamuna river systems in Bangladesh carry 2.4 billion tons of sediments, a part of which is deposited in the flood plains each year. The comparison between deeply flooded areas in Bangladesh and the areas having more than 10 and 50 percent of the tubewells producing arsenic more than 0.05 mg/l shows that the deeply flooded areas are mainly arsenic problem areas of Bangladesh. The reducing soil environment in the deeply flooded areas produced by anaerobic conditions in fine grained sediments rich in organic matters appears to be conducive to the release of arsenic in groundwater. The reducing environment in recently deposited sediments in flood plains appears to be the main cause of arsenic dissolution from soil to groundwater in Bangladesh. The old sediments and oxidized reddish soils in Bangladesh do not release arsenic, rather they adsorb arsenic from water. In this context, presence of significant arsenic in the recent incoming upstream suspended sediments carried by river flow & flood water and sediment deposited in the river flood plain of Bangladesh *significantly influence the arsenic contamination of groundwater in Bangladesh. Arsenic content in surface water of the rivers even after flood is extremely low, almost negligible ranging from 0.11 ppb to 2.25 ppb. The arsenic content of fresh suspended sediment carried by running incoming sediment after post flood period ranges 4.067 to 5.466 mg/kg, which is not significantly higher than the usual arsenic contents of soils in Bangladesh. At every location of each rivers, arsenic in sand fraction is always lower than that in silt and clay fraction. Arsenic content in clay fractions of different type of river sediments ranges from 3.525 to 6.476 mg/kg.*

INTRODUCTION

Bangladesh being located downstream of the mighty rivers, the Ganges and the Brahmaputra, works as a retention basin of excess water carried by the river system for final discharge in the Bay of Bengal. The Ganga-Padma and Brahmaputra-Jamuna river systems drain a huge area of land outside the territory of Bangladesh. There are some direct and indirect references of occurrence of arsenic and arsenic related minerals in the upstream of the rivers carrying water and sediments in Bangladesh (DPHE, BGS and MML, 1999). The Ganga- Padma and Brahmaputra-Jamuna river systems in Bangladesh carry 2.4 billion tons of sediments, a part of which is deposited in the flood plains each year (Ahmed, 2000). The comparison between deeply flooded areas in Bangladesh and the areas having more than 10 and 50 percent of the tubewells producing arsenic more than 0.05 mg/l as demarcated in Figure 1, which shows that the deeply flooded areas are mainly arsenic problem areas of Bangladesh (Ahmed,2000). The reducing soil environment in the deeply flooded areas appears to be conducive to the release of arsenic in groundwater. The tubewells sunk in shallow aquifers in the Ganges and Meghna flood plains except in coastal areas are the worst affected. In the coastal areas, water supply is mainly based on manually operated deep tubewells as the water available in the shallow aquifer is saline. In Bangladesh, deep tubewells at depths greater than 500 ft have been found to be free from arsenic contamination (DPHE and BGS, 2000). Arsenic contamination of flooded areas of both sides of the Jamuna is very low probably because of the dynamic nature of the river and deposition of sandy soil of low arsenic content on the flood plain at a relatively higher energy level. Annual floods usually submerge most of the areas contaminated with arsenic in groundwater as shown in Fig. 1.

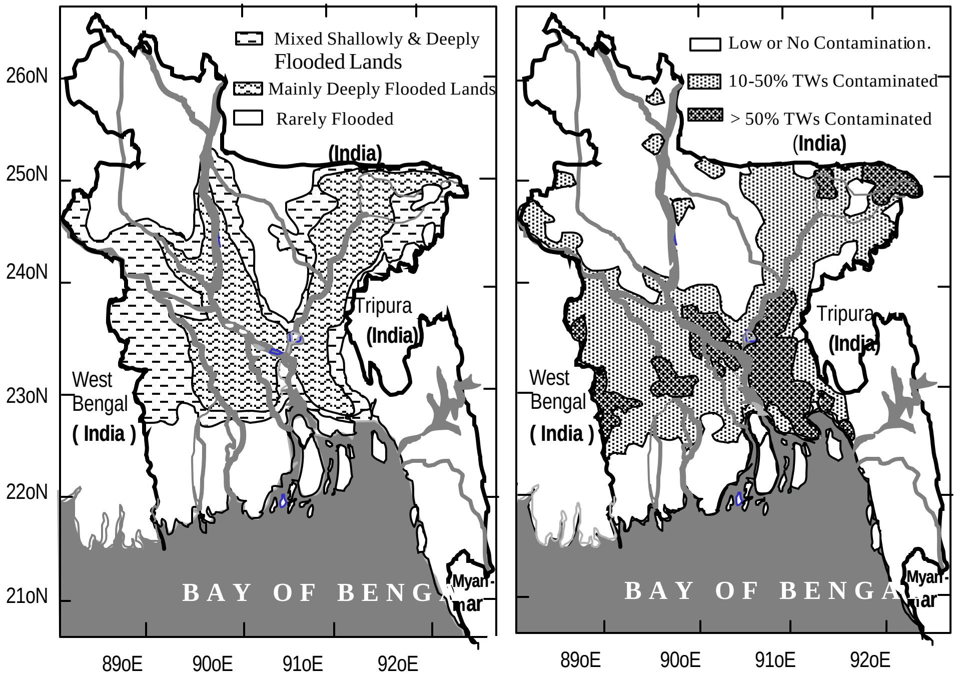

Figure 1: Comparison of Flooded and Arsenic Contaminated Areas of Bangladesh

In these areas, the soil is characterized by paludal deposit of clay, silt & peat and alluvial deposit of silt and silty clay. The growth of vegetation in nutrient rich flood plains enriches the fine-grained soils and inorganic debris with organics, which is used as food for microorganisms. The anaerobic condition in soils in deeply flooded wetlands is characterized by a gray to blackish color and the release of methane gas. The available dissolved oxygen in infiltrated floodwater is exhausted in the topsoil rich in bio-decomposible organic matters. Pore water devoid of dissolved oxygen produces a reducing environment suitable for dissolution of both iron and arsenic. Mok and Wai (1994) reported that under reducing conditions, arsenic mobilization was controlled by dissolution of hydrous iron oxides. DPHE, BGS and MML,(1999) mentioned that there was a direct relationship between the degree of reduction in groundwater and arsenic concentration. The reducing environment in the flood plain is produced by anaerobic conditions in fine grained sediments rich in organic matters. The reducing environment in recently deposited sediments in flood plains appears to be the main cause of arsenic dissolution from soil to groundwater in Bangladesh. The old sediments and oxidised reddish soils in Bangladesh do not release arsenic, rather they adsorb arsenic, if present in water. In this context, determination of presence of arsenic in the recent incoming upstream suspended sediments carried by river flow & flood water and sediment deposited in the river flood plain of Bangladesh was carried out. The main objectives of this study were to identify the source of arsenic in Bangladesh and assess the quantity of incoming arsenic from upstream and its role in contamination of groundwater in Bangladesh.

EXPERIMENT AND STUDY PROCEDURE

Suspended sediment, river bottom sediment and river-bed sediment in flood plain were collected, fractionated into sand, silt and clay constituents and digested by Aqua Regia. After digestion followed by filtration, arsenic content in sediment is determined by AAS (SIMADZU) and the results are analyzed to understand the source, release and transport mechanism of arsenic and its presence in different fractions of materials.

Collection of River Sediment

Suspended sediment (SS) carried by the major rivers Jamuna, Padma and Meghna as well as stirred sediment with surface waters (SSSW) from the bottom of the rivers was collected for analysis. River bottom sediment (BoS) and old river bed sediments (BeS) from flood plains in acute problem areas were also collected. The samples were collected during pre- and post-flood period conditions from selected locations as shown in Figure 2.

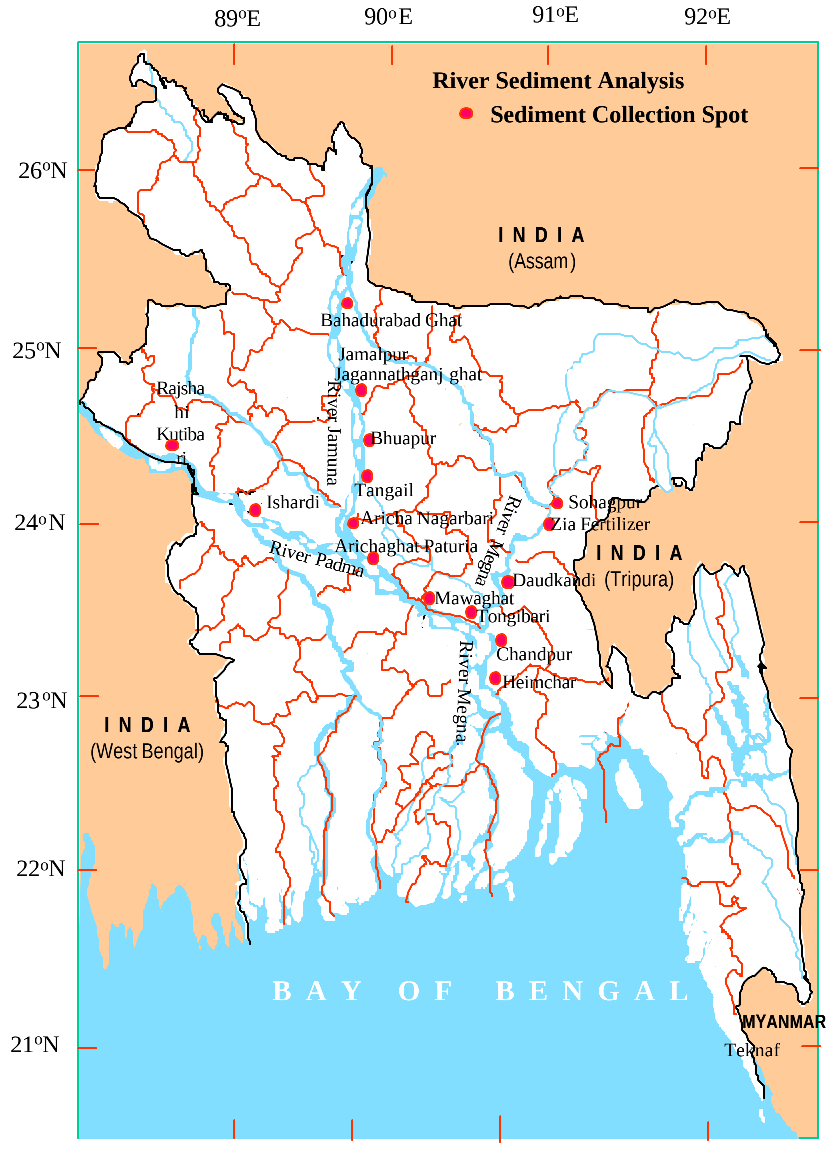

Figure 2: Location of sampling points along the major rivers

Separation and Fractionation of Sediment

Collected sediments were separated and fractionated in different fractions of materials according to the following steps.

- Settling of total sediment was achieved in a large plastic container of 80 litre capacity by allowing the sediment mixed water to settle for at least one week. A significant amount (at least 20 gram) of settled sediment was separated and kept for extracting total arsenic. Remaining sediment with mixed water was used for separating sand, silt and clay fractions of the sediment.
- Settling of sand was achieved in a large cylinder or glass jar or plastic container (of at least 2 litre) by stirring collected river water sample and allowing it for 1 minute to settle. Sand was settled within one minute; the supernatant was transferred into another (second) jar or cylinder.
- Settling of silt was conducted in the second jar by stirring supernatant water and allowing it for one hour to settle. All silt would be settled within one hour and the supernatant containing the clay fractions was taken into the third jar or cylinder.
- The clay was settled by keeping supernatant in the third jar for overnight or several days until the clay particles combine in group or flocculate and settle down.

Also suspended sediment stirred with river water (SSSW), river bottom sediment (BoS) and river bed sediments (BeS) from flood plains in acute problem areas was collected and separated. Rivers bed sediment was collected from the bed of the major rivers Padma, Jamuna and Meghna in their flood plain along the length of the rivers at a few selected locations. Total sediment as well as sand, silt and clay fraction of the rivers bed sediment was separated in the same way.

Test and Analysis of River Sediment Samples

Arsenic, iron, manganese etc. were extracted from the settled sediment, sand, silt and clay constituents by oven drying overnight following digestion by Aqua Regia. After digestion followed by filtration, arsenic was determined by AAS (SIMADZU, Graphite Furnace).

ANALYSIS OF EXPERIMENTAL RESULTS

Experimental results and analysis of the study are detailed in tabular form as follows.

Arsenic Content in Surface Water

For measuring arsenic in surface water of the rivers, water was collected from different selected locations of Padma, Meghna and Jamuna as shown in Figure 2 after flood in September-October of 2001 and settled for about two month. Settled water was tested for arsenic with AAS. Test results shown in Table 1 show that arsenic content in surface water of the rivers is extremely low, almost negligible ranging from 0.11 ppb to 2.25 ppb. Highest amount (2.25 ppb) is found in Meghna water at Heimchar while lowest amount (0.11 ppb) found in Megha water at Shohagpur, 10 km upstream of Bhairab bridge. Arsenic content in Padma water ranges from 0.17 ppb at Mawa to 0.70 ppb at Tongibari while it varies in Jamuna water from 0.31 ppb at Bahadurabad to 0.51 ppb at Jagannathganj ghat.

Table 1: Arsenic in Surface Water (Settled surface water)

River	Location	As Content, ppb	Remarks
Jamuna	Bahadurabad	0.51	Arsenic content is extremely low, almost negligible ranging from 0.11 ppb to 2.25 ppb. Highest amount (2.25 ppb) is found in Meghna river at Heimchar. Lowest amount of arsenic (0.11 ppb) is also found in surface water of Megha at Shohagpur, 10 km upstream of Bhairab bridge.
	Jagannathganj	0.31	
	Bhuanpur	0.40	
	Aricha-Nagarbari	0.47	
Padma	Rajshahi (Kutibari)	0.22	
	Pakshi	0.36	
	Aricha –Paturia	0.21	
	Mawa	0.17	
	Tongibari-Digirpar	0.70	
Meghna	Sohagpur	0.11	
	Zia Fertilizer Factory	0.15	
	Daudkandi	0.61	
	Chandpur	1.35	
	Heimchar	2.25	

Arsenic Content in Suspended Sediment of Rivers During Pre-flood Period

No suspended sediment was found in running free water of rivers during pre-flood period. However, total arsenic in suspended sediment after stirring river bottom with water and settling for 2 month in different locations of Jamuna, Padma and Meghna during pre-flood period was available. Total arsenic content was determined using AAS (Graphite Furnace). Test result is given in Table 2. Test result shows that arsenic content in suspended sediment of river bottom varies from 1.021 mg/kg in Padma river at Pakshi to 3.525 mg/kg in Jamuna river at Bhuanpur.

Table 2: Total Arsenic in suspended sediment stirred with water (Settled for 2 month) during pre-flood period.

River	Location	Arsenic Content, mg/kg
Jamuna (Bhuanpur-Sirajganj)	Bhuanpur Side	3.525
	Middle Jamuna	2.670
	Sirajgang Side	Sediment not Collected
Padma (Veramara-Pakshi)	Veramarah Side	3.16
	Middle Padma	1.112
	Pakshi Side	1.021
Meghna (Dudkandi-Gajaria)	Gajaria Side	1.078
	Middle Meghna	3.343
	Daudkandi	3.376

Arsenic in River Sediment during Post Flood Period (After Flood)

Sediments carried out by the upstream river water to downstream i.e. suspended sediment (SS) from incoming water, suspended sediment after stirring bottom sediment with river water for 2/3 minute (SSSW), fresh sediment deposited at the bottom of the river i.e. fresh bottom sediment (BoS) of the river and old river bed sediment (BeS) from top and bottom portion within 2 feet from the soil surface deposited in the river bed of flood plain for 1 to 5 years was collected during post flood period from September to October 2001. Total arsenic in collected sediment as well as arsenic content in sand, silt and clay fraction of the sediment was determined successively.

Arsenic in Total Sediment

Arsenic contents of suspended sediment (SS) of incoming water, stirred sediment with surface water (SSSW), fresh bottom sediment (BoS) of the river and bed sediment from top portion (BeS - T) and bed sediment from bottom portion (BeS - B) of collected sediment within 2 feet from the soil surface were determined. The results are summarized in Table 3. Arsenic content is higher in most cases in the fresh suspended sediment of running incoming water from upstream as compared to river bottom sediment and old sediment of flood plain. Total arsenic in fresh incoming sediment ranges from low 4.057 mg/kg in the river Jamuna at Jagannathganj ghat to highest 5.466 mg/kg in the river Padma at Digirpar, Tongibari area just before the Junction point of Padma with Meghna. Total arsenic content in incoming suspended sediment ranges from 4.067 to 5.086 mg/kg in Jamuna at Jagannathgang and Aricha-Nagarbari and from 4.317 to 5.466 mg/kg in Padma at Pakshi and Tongibari and from 4.161 to 4.987 mg/kg in Meghna at Heimchar and Chandpur respectively. Unfortunately no fresh sediment in running incoming water was found in three location of river Meghna before the joining point of Meghna with Padma at Chandpur. In case of SSSW, total arsenic content varies from 2.342 to 4.479 mg/kg in Meghna at Chandpur and Daudkandi, from 3.196 mg/kg to 4.459 mg/kg in Jamuna at Jagannathganj and Bhuanpur and from 2.402 to 4.296 mg/kg in Padma at Mawa and Pakshi. Total arsenic content varies from 2.340 to 4.736 mg/kg in Meghna at Chandpur and Daudkandi, from 2.648 to 4.412 mg/kg in Jamuna at Jagannathganj and Bahadurabad and from 3.007 to 3.907 mg/kg in Padma at Tongibari and Mawa in case of fresh bottom sediment. Total arsenic content in upper portion of bed sediment in flood plain ranges from 2.919 to 4.194 mg/kg in Jamuna at Aricha-Nagarbari and Bhuanpur and from 2.462 to 3.784 mg/kg in Padma at Pakshi and Rajshahi and from 3.312 to 5.844 mg/kg in Meghna at Daudkandi and Sohagpur. Total arsenic content ranges from 2.679 to 3.720 mg/kg in Jamuna at Aricha-Nagarbari and Bhuanpur and from 3.129 to 5.810 mg/kg in Padma at Rajshahi and Pakshi and from 2.043 to 4.273 mg/kg in Meghna at Zia Fertilizer Factory, Ashugang and at Heimchar in bottom portion of bed sediment in flood plain.

Table 3: Total arsenic in surface water, suspended, bottom and bed sediment of Padma, Meghna and Jamuna

RIVER	LOCATION	Arsenic Content, mg/kg				
		SS	SSSW	BoS	BeS - T	BeS – B
Jamuna	Bahadurabad	4.873	3.4	4.412	3.195	3.288
	Jagannathgang	4.067	3.196	2.648	2.874	3.543
	Bhuanpur	4.340	4.459	4.167	4.194	3.720
	Aricha-Nagarbari	5.086	3.492	3.051	2.919	2.679
Padma	Rajshahi (Kutibari)	4.317	2.713	3.652	3.784	3.129
	Pakshi	4.319	4.296	3.402	2.462	5.810
	Aricha –Paturia	4.339	3.101	3.303	3.367	3.195
	Mawa	4.693	2.402	3.907	2.711	4.637
	Tongibari-Digirpar	5.466	2.928	3.007	2.881	4.680
Meghna	Sohagpur (10 km u/s of Bhairab Bridge		3.586	4.310	5.844	3.534
	Zia Fertilizer Factory		2.455	4.726	5.083	2.043
	Daudkandi		4.479	4.736	3.312	3.867
	Chandpur	4.987	2.342	3.240	3.803	4.226
	Heimchar	4.161	3.462	3.290	3.327	4.273

Arsenic in sand fraction

Sand fraction from river sediment samples was separated as described above and arsenic content was determined by digestion with aqua-regia. Arsenic contents of sand fraction of different type of sediment are compared in Table 4. Sand fractions were not available in Padma river sediment at Rajshahi and Pakshi in case of SSSW, in Jamuna river sediment at Bhuanpur and in Padma river sediment at Rajshahi and Pakshi in case of BoS, in Padma river sediment at Mawa in case of BeS - T and in Padma river sediment at Pakshi in case of BeS - B.

Arsenic content of sand fraction as shown in Table 4 varied from 1.684 to 3.524 mg/kg in case of SSSW, 1.002 to 2.983 mg/kg in case of BoS, 1.006 to 1.879 mg/kg in case of BeS-T and 1.247 to 2.468 mg/kg in case of BeS-T.

Table 4: Arsenic in sand fraction of suspended, bottom and bed sediments

RIVER	LOCATION	Arsenic Content, mg/kg			
		SSSW	BoS	BeS -T	BeS – B
Jamuna	Bahadurabad	2.233	2.513	1.846	1.097
	Jagannathganj	2.851	2.518	1.825	1.254
	Bhuanpur	2.345	NA	1.879	1.247
	Aricha-Nagarbari	2.568	1.514	1.079	1.372
Padma	Rajshahi (Kutibari)	NA	NA	1.006	1.409
	Pakshi	NA	NA	1.768	NA
	Aricha –Paturia	2.729	2.670	1.813	2.468
	Mawa	2.833	2.634	NA	1.598
	Tongibari-Digirpar	2.629	2.072	1.720	2.215
Meghna	Sohagpur (10 km u/s of Bhairab Bridge	3.524	2.652	1.611	1.583
	Zia Fertilizer Factory	1.684	1.002	1.299	1.541
	Daudkandi	2.619	2.489	1.269	2.382
	Chandpur	2.447	2.242	1.390	2.426
	Heimchar	2.794	2.983	1.127	2.396

NA : Not Available

Arsenic in silt fraction

Settling of silt was conducted in the second jar by stirring supernatant water from sand settling and allowing it for 1 hour to settle. Arsenic in silt fraction of different types of sediment was determined by AAS method following Aqua-regia digestion. Arsenic content of silt fraction of the sediment samples is given in Table 5**.** Silt fractions were not available in Jamuna river sediment at Jagannathgang in case of SSSW, in Jamuna river sediment at Aricha-Nagarbari in case of BoS, in Padma river sediment at Mawa in case of BeS -T and in Jamuna river sediment at Bahadurabad and in Padma river sediment at Pakshi in case of BeS - B.

Arsenic content of silt fraction varied from 2.653 to 3.916 mg/kg in SSSW, 1.858 to 3.784 mg/kg in BoS, 2.379 to 3.681 mg/kg in BeS-T and 2.628 to 3.883 mg/kg in BeS-B. The average arsenic content of silt fraction is higher than that of sand fractions of the sediment samples.

Table 5: Arsenic in silt fraction of sediments

RIVER	LOCATION	Arsenic Content, mg/kg			
		SSSW	BoS	BeS (Top)	BeS (Bottom)
Jamuna	Bahadurabad	2.653	3.109	2.528	NA
	Jagannathganj	NA	3.141	2.732	3.667
	Bhuanpur	3.916	3.312	2.323	3.106
	Aricha-Nagarbari	3.624	NA	2.336	3.223
Padma	Rajshahi (Kutibari)	2.753	2.860	2.397	3.883
	Pakshi	3.489	3.566	2.964	NA
	Aricha –Paturia	2.876	3.605	3.272	2.816
	Mawa	3.601	2.838	NA	3.080
	Tongibari-Digirpar	2.974	2.857	2.563	3.023
Meghna	Sohagpur (10 km u/s of Bhairab Bridge	3.710	3.943	3.681	3.127
	Zia Fertilizer Factory	2.955	1.858	2.379	2.628
	Daudkandi	3.464	3.784	2.890	3.389
	Chandpur	3.733	2.512	3.253	3.481
	Heimchar	3.318	3.401	3.593	3.232

NA : Not Available

Arsenic in Clay Fraction

The clay was be settled by keeping supernatant in the third jar for several days until the supernatant becomes clear. Arsenic in clay fraction of different types of sediment was analyzed using AAS following Aqua-regia digestion. Arsenic content of clay fraction is given in Table 6**.** Clay fractions were not available in different samples for analysis. All the sediment in these samples were composed of silt and sand.

In case of SSSW, arsenic content in clay fraction varies from 4.733 to 5.648 mg/kg in Jamuna at Bahadurabad and Bhuanpur, from 3.525 to 3.816 mg/kg in Padma at Pakshi and Rajshahi and from 3.977 to 5.470 mg/kg in Meghna at Shohagpur and Zia Fertilizer Factory, Ashugang**.** Similarly arsenic content of clay fractionated from BoS, BeS-T and BeS-B samples varied between 3.874 and 6.639. The arsenic content of the clay fraction of the sediment sample was higher than sand and silt fractions.

Table 6: Arsenic in clay fraction of sediments of the river system

RIVER	LOCATION	Arsenic Content, mg/kg			
		SSSW	BoS	BeS - T	BeS – B
Jamuna	Bahadurabad	4.733	4.375	NA	NA
	Jagannathganj	NA	3.874	4.967	5.439
	Bhuanpur	5.648	4.373	6.476	NA
	Aricha-Nagarbari	4.488	4.105	NA	5.258
Padma	Rajshahi (Kutibari)	3.816	4.939	NA	5.018
	Pakshi	3.525	4.029	5.452	4.864
	Aricha –Paturia	NA	4.825	NA	5.417
	Mawa	NA	6.282	4.930	5.068
	Tongibari-Digirpar	NA	6.181	4.585	4.578
Meghna	Sohagpur (10 km u/s of Bhairab Bridge	3.977	4.517	4.611	5.845
	Zia Fertilizer Factory	5.470	NA	NA	4.993
	Daudkandi	NA	NA	5.717	5.153
	Chandpur	4.248		6.639	5.276
	Heimchar	NA	NA	NA	NA

NA : Not Available

DISCUSSION

Arsenic content in surface water of the rivers even after flood is extremely low, almost negligible ranging from 0.11 ppb to 2.25 ppb. Absence of arsenic in water of Jamuna, Padma, and Meghna is the characteristics quality of surface water.

Non-existence of suspended sediment in river water during pre-flood period indicates absence of erosion in the upstream areas of the rivers. Moreover, it can be assumed that no man-made activities like mining and industrial activities in upstream areas is contributiong sediment in downstream areas. However, total arsenic (varying from 1.021 mg/kg to 3.525 mg/kg) in suspended sediment after stirring river bottom with water at different locations of Jamuna, Padma and Meghna during pre-flood period indicates that this deposition of arsenic in bottom sediment occurred after flood.

The higher amount of arsenic in fresh suspended sediment carried by running incoming sediment after post flood period ranging from 4.067 mg/kg to 5.466 mg/kg indicates the transport of arsenic from upstream by the mighty river rivers Padma-Ganges and Jamuna- Bhrammaputra originated from mountainous Himalayan region and the Tibbet Plateue. Obviously high amount of mineral arsenic are geologically eroded by heavy rainfall in rainy season that moves to downstream by downward flood flow and ultimately settles in flood plains of mighty rivers in Bangladesh.

Arsenic content in sand fractions of different type of river sediment varied from 1.002 to 2.983 mg/kg except 3.524 mg/kg at Ashugang in Meghna. The results also showed that at every location of each rivers, arsenic in sand fraction is always lower than that in silt and clay fraction. Arsenic content in silt fractions of different type of river sediment varied from 1.858 to 3.943 mg/kg, while arsenic content in clay fractions of different type of river sediments ranged from 3.525 to 6.476 mg/kg. The arsenic content of fine grained materials is higher because of comparative higher surface area. The coarse grained materials like sand have relatively smaller surface area per unit volume of materials and provides fewer sites for adsorption of arsenic.

CONCLUSIONS

Arsenic content of river water is very low and in most of the cases around minimum detection level of the equipment. It is the characteristic arsenic related quality of surface water. A significant quantity of arsenic is transported in Bangladesh from upstream. All the arsenic carried by the rivers from the upstream is in the solid phase adsorbed on sediment. The arsenic content in deposited bed sediment in the pre-monsoon period is lower that that of suspended sediment. The higher amount of arsenic in fresh suspended sediment during post-flood period ranging from 4.067 to 5.466 mg/kg indicates that mineral arsenic is geologically eroded and moves downstream with downward flood flow and ultimately settles in flood plains of mighty rivers in Bangladesh.

Arsenic content of the sediment is higher in case of fine-grained sediment. The sand fraction of sediment having lower surface area per unit weight of material has the lower capacity to adsorb arsenic than clay fraction. Therefore, the beels and hoars receive higher quantity of arsenic as the clayey materials are deposited in these stagnant water bodies.

REFERENCES

Ahmed M. F. (2000); *Soil environment and arsenic contamination of groundwater in Bangladesh.* Proceeding of International Conference on Geoenvironment 2000, Sultan Qabus University, Muscat, Oman, March 4-7, 2000. Vol. - 1, 256-264.

DPHE, BGS, and MML (1999), *Groundwater Studies for Arsenic Contamination in Bangladesh*, Final Report (Phase-1), January.

DPHE and BGS (2000), *Groundwater Studies for Arsenic Contamination in Bangladesh*, Draft Final Reoprt Summary, March.

Mok W. M. and Wai C. M. (1994); *Mobilization of arsenic in Contaminant River Water in Arsenic in the Environment* (J. O. nriagu Ed.), Jhon Wiley & Sons Inc.