

Arsenic in water and sediments of major rivers in Bangladesh

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Abstract

Arsenic (As) concentrations in river water, suspended sediments and riverbed sediments of three major rivers in Bangladesh – Jamuna, Padma and Meghna - have been assessed. The As concentrations in the river water samples have been found to be very low, mostly below the detection limit of $1\mu\text{g L}^{-1}$. Suspended solids collected from the rivers during post-flood period (September-October) were found to contain significant amount of As, varying from 4.07 to 5.47 mg kg^{-1} and the As concentrations in suspended sediments did not differ significantly among the rivers. Arsenic concentrations of river bed sediments were lower than those of the suspended sediments. Clay fraction of the sediments contained the highest concentration of As, followed by silt and sand fractions. The As load carried annually with suspended sediments of the Jamuna, Padma and Meghna rivers have been estimated to be 3054, 4121 and 4584 tons, respectively. Arsenic concentrations in suspended and riverbed sediments have been found to be comparable to those reported for aquifer sediments in Bangladesh. This study suggests that the recent riverbed and floodplain sediments could be an important source of As, from which As could be mobilized into the groundwater.

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1. Introduction

Bangladesh being located in the downstream of the mighty rivers, the Ganges and the Brahmaputra, works as a retention basin of flood water carried by the river systems for final discharge into the Bay of Bengal. The Ganges-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 (As) and As bearing minerals in the upstream of the rivers carrying water and sediments through Bangladesh (DPHE, BGS and MML, 1999). The river systems in Bangladesh carry 2.4 billion tons of sediments, a part of which is deposited in the floodplains each year (Khan, 1994). The

comparison between deeply flooded areas in Bangladesh and the areas having more than 10 and 50 percent of the tubewells producing water with more than 0.05 mg L^{-1} As (which is the present drinking water standard in Bangladesh) showed that the deeply flooded areas are the main As problem areas in Bangladesh (Ahmed, 2000). The reducing soil environment in the deeply flooded areas appears to be conducive to the release of As from sediment. The tubewells sunk in shallow aquifers in the Ganges and Meghna floodplains, except in the 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 largely free from As contamination (DPHE and BGS, 2001). Arsenic contamination in the flooded areas on both sides of the Jamuna is relatively low, probably because of the dynamic nature of the river at a relatively higher energy level and deposition of sandy soil with low As content on the floodplains. Annual floods usually submerge most of the As contaminated areas, where the soil is characterized by paludal deposit of clay, silt and peat and alluvial deposit of silt and silty clay. The growth of vegetation in nutrient rich floodplains enriches the fine-grained soils with high organic content. The anaerobic condition in soils in the deeply flooded wetlands, produced due to the decomposition of organic matters, 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 biodegradable organic matter. Pore-water devoid of dissolved oxygen produces a reducing environment suitable for reductive dissolution of Fe solids and subsequent releases of adsorbed As. Mok and Wai (1994) reported that under reducing conditions, As mobilization was controlled by dissolution of hydrous iron oxyhydroxides. DPHE, BGS and MML (1999) mentioned that there was a direct relationship between the degree of reduction in groundwater and As concentration. The As associated with the recently deposited sediments could be a major source of As that is mobilized into the groundwater under reducing environment.

In this study, As concentrations in river water, suspended sediments carried by river, and river bed sediments of three major rivers of Bangladesh have been determined in order to estimate the quantity of As coming from upstream sources and to assess its role in As contamination of groundwater in Bangladesh.

2. Materials and methods

2.1 Study area

In this study, As concentrations in water and sediment samples of three major rivers – Padma, Meghna and Jamuna have been assessed. River water, suspended sediment and river bed sediment samples were collected at different locations along these rivers for analysis in the laboratory. Water and sediment samples were collected during the post-flood period (September-October) from 14 locations along these three major rivers of the country. Water and sediment samples were also collected from a limited number of locations along these rivers during the pre-flood period (April). Fig. 1 shows sampling locations along the major rivers. All samplings were carried out during 2001.

2.2 Collection of water and sediment samples

2.2.1 Collection of river water samples

River water samples were collected from each sampling location away from the bank and close to the mid-width of the river. An engine boat was used for traveling in the rivers.

Water samples were collected from a depth of about 30-35 cm below water surface. Water samples were collected in pre-washed 500 ml plastic bottles. For analysis of dissolved As, the water samples were immediately filtered (using 0.45 μm filter) in the field and were subsequently acidified with nitric acid (1 ml concentrated HNO_3 per liter of water). A total of 14 river water samples were collected during September-October 2001, 4 from Jamuna, and 5 each from Padma and Meghna.

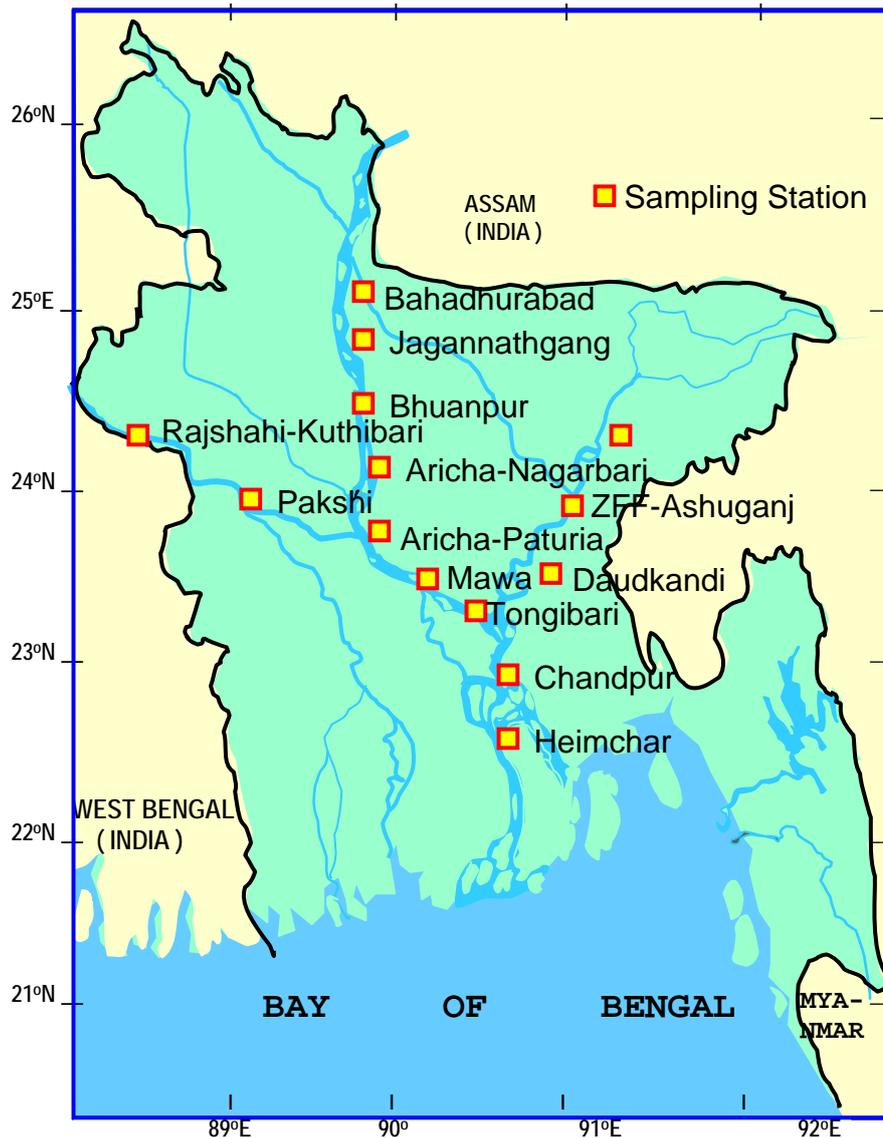


Fig. 1. Sampling locations along the major rivers

2.2.2 Collection of sediment samples

Suspended sediment (SS) samples were collected from 14 locations (see Table 3) during the post-flood period (September-October). Suspended sediments were collected with a suspended sediment sampler. Suspended sediment concentrations in all rivers were very low during pre-flood period (April) and therefore no suspended sediment sample could be collected in April.

During the pre-flood period of April, riverbed sediments were collected from 8 locations (see Table 2) using a grab sampler. During the post-flood period of September-October, riverbed sediments were collected from 14 selected locations along the rivers (see Table 3). The top portion (0-5 cm) of the riverbed sediment was collected using a grab sampler. In addition, riverbed sediment core samples were collected using PVC pipe samplers, which were 5 cm (2 inch) in diameter and 30 to 60 cm in height. One core sample was collected from each sampling location.

2.3 Analysis of samples

2.3.1 Analysis of water samples

The filtered and acidified river water samples were tested for total dissolved arsenic. Arsenic concentrations were determined with an Atomic Absorption Spectrophotometer (Shimadzu, Japan AA6800) attached with a graphite furnace (GF-AAS). The detection limit for this system was $1\mu\text{g L}^{-1}$.

2.3.2 Analysis of sediment samples

The suspended sediment samples were analyzed for total (aqua-regia extractable) As. As noted earlier, two types of riverbed sediment samples were collected: (i) grab samples (0-5 cm) and (ii) core samples. Before analysis, each core sediment sample (about 20 to 25 cm in depth) was divided into two segments, the top segment consisting of the top 5 to 10 cm (referred to as Top riverbed core) and the bottom segment consisting of the rest of the core (referred to as Bottom riverbed core). Each type of riverbed sediment was analyzed for total (aqua-regia extractable) As. Besides, the three types of riverbed sediments were fractionated into sand, silt and clay fractions, following the procedure described in Peck et al. (1998), and analyzed for total (aqua-regia extractable) As.

For determination of aqua-regia extractable As, each type of soil sample was dried in an oven at $110\text{ }^{\circ}\text{C}$ for 24 h. After drying, the sample was ground and sieved before digestion. For digestion, 5 g of sediment sample was weighed into a 500 mL flask, to which 2.5 mL concentrated nitric acid and 7.5 mL concentrated hydrochloric acid were added. The suspension was kept overnight in the flask and then it was refluxed for 2 h, followed by dilution with deionized water to 500 mL, stirring for 5 min and filtering ($0.80\text{ }\mu\text{m}$ filter) after cooling. The filtrate was stored in a plastic bottle for analysis of As using an AAS attached with a graphite furnace (Shimadzu, AA6800).

All acids used in this study were of analytical grade. Standard QA/QC protocol was followed for water and sediment sample analysis, including replicate analysis (1 in every 5 samples) and checking of method blanks (1 in every 10 analysis) and standards (1 in every 10 analysis).

3. Results and discussion

3.1 Arsenic concentration in river water

Table 1 shows As concentrations of river water samples collected during the post-flood period of September-October. It shows that As concentrations of all river water samples are extremely low, mostly below the detection limit of $1\mu\text{g L}^{-1}$. Among the 14 samples,

only 2 had As concentration above $1 \mu\text{g L}^{-1}$. Arsenic concentration of $2.3 \mu\text{g L}^{-1}$ and $1.4 \mu\text{g L}^{-1}$ were detected in the Meghna river water at Heimchar and Chandpur, respectively.

Table 1
Arsenic concentrations in river water samples in post-flood period

River	Location	Arsenic Concentration ($\mu\text{g L}^{-1}$)
Jamuna	Bahadurabad	< 1
	Jagannathgang	< 1
	Bhuanpur	< 1
	Aricha-Nagarbari	< 1
Padma	Rajshahi (Kutibari)	< 1
	Pakshi	< 1
	Aricha-Paturia	< 1
	Mawa	< 1
	Tongibari-Digirpar	< 1
Meghna	Sohagpur	< 1
	Zia Fertilizer Factory	< 1
	Daudkandi	< 1
	Chandpur	1.4
	Heimchar	2.3

3.2 Arsenic concentration in sediment samples

3.2.1 Arsenic concentration in sediments during pre-flood period

Table 2 shows As concentrations of riverbed sediments during the pre-flood period, which shows that As concentration in the sediment samples varied from 2.67 to 3.26 mg kg^{-1} in Jamuna, 1.02 to 3.16 mg kg^{-1} in Padma, and 1.08 to 3.38 mg kg^{-1} in Meghna. It appears that in the Jamuna river at Bhuapur-Sirajganj section, As concentration in riverbed sediments is relatively higher on the Bhuapur side. Similarly, for the Padma river at Bheramara-Pakshi section, As concentration in riverbed sediments is much higher on the Bheramara side compared to the Pakshi side; for the Meghna river at Daudkandi-Gajaria section, As in riverbed sediment is much higher on the Daudkandi side compared to the Gajaria side.

Table 2
Arsenic concentrations of riverbed sediments during pre-flood period

River	Sampling Location	Arsenic Concentration (mg kg^{-1})
Jamuna (Bhuapur-Sirajganj)	Bhuapur	3.53
	Sirajganj (close of middle-river)	2.67
Padma (Bheramara-Pakshi)	Bheramara	3.16
	Pakshi	1.02
	Mid-river	1.11
Meghna (Daudkandi-Gajaria)	Gajaria	1.08
	Daudkandi	3.38
	Mid-river	3.34

3.2.2 Arsenic concentration in sediments during post-flood period

Table 3 shows As concentrations of suspended sediment (SS) and river bed sediments at the sampling locations along the Jamuna, Padma and Meghna rivers during the post-flood period. It shows that As concentrations of SS of the three rivers do not vary significantly. Arsenic concentrations of SS from Jamuna river varied from 4.07 mg/kg (at Jagannathganj) to 5.09 mg kg⁻¹ (at Aricha-Nagarbari); for Padma river As in SS varied from 4.32 mg kg⁻¹ (at Kutibari) to 5.47 mg kg⁻¹ (at Tongibari-Digirpar), and for Meghna river it varied from 4.16 mg kg⁻¹ (at Heimchar) to 4.99 mg kg⁻¹ (at Chandpur). The highest As concentration of 5.47 mg kg⁻¹ was measured in the SS of Padma river at Tongibari- Digirpar, just upstream of the confluence of Padma and Meghna rivers; while the lowest concentration was measured in the SS of Jamuna river at Jagannathganj.

Arsenic concentrations of riverbed sediments (0-5cm) in September-October have been found to be higher than those collected in April (Table 2). At all locations, As concentrations of suspended sediments have been found to be higher than those in riverbed sediments.

Table 3 shows that As concentrations of riverbed sediment (0-5 cm), Top riverbed core and Bottom riverbed core differ to some extent, but do not show any particular trend of variation. The highest As concentration of 4.74 mg kg⁻¹ in the riverbed sediment (0-5 cm) was measured in the sample from Meghna river at Daudkandi, and the lowest concentration of 2.65 mg kg⁻¹ was measured in the sample from Jamuna river at Jagannathganj.

Table 3
Arsenic concentrations in suspended sediment and river bed sediments of Padma, Meghna and Jamuna rivers during post-flood period

River	Location	Arsenic Concentration, mg kg ⁻¹			
		Suspended Sediment (SS)	Riverbed (0-5 cm)	Top riverbed core	Bottom riverbed core
Jamuna	Bahadurabad	4.87	4.41	3.20	3.29
	Jagannathganj	4.07	2.65	2.87	3.54
	Bhuanpur	4.34	4.17	4.19	3.72
	Aricha-Nagarbari	5.09	3.05	2.92	2.68
Padma	Rajshahi (Kutibari)	4.32	3.65	3.78	3.13
	Pakshi	4.32	3.40	2.46	5.81
	Aricha –Paturia	4.34	3.30	3.37	3.20
	Mawa	4.69	3.91	2.71	4.64
	Tongibari-Digirpar	5.47	3.01	2.88	4.68
Meghna	Sohagpur (10 km u/s of Bhairab Bridge)	--	4.31	5.84	3.53
	Zia Fertilizer Factory	--	4.73	5.08	2.04
	Daudkandi	--	4.74	3.31	3.87
	Chandpur	4.99	3.24	3.80	4.23
	Heimchar	4.16	3.29	3.33	4.27

Table 4 shows As concentrations in sand fractions of all riverbed sediment samples. The highest (2.98 mg kg⁻¹) and lowest (1.0 mg kg⁻¹) As concentrations in the sand fractions of riverbed sediment (0-5 cm) were both found in Meghna river at Heimchar and at Zia Fertilizer Factory (ZFF), respectively. Arsenic concentrations in sand fraction of Top

riverbed cores ranged from 1.0 mg kg^{-1} at Kutibari of Padma river to 1.88 mg kg^{-1} at Bhuapur of Jamuna river. Arsenic concentrations of Bottom riverbed cores ranged from 1.1 mg kg^{-1} at Bahadurabad of Jamuna river to 2.47 mg kg^{-1} at Aricha-Paturia of Padma river. In general, As concentrations of sand fractions of riverbed sediments (0-5 cm) were found to be relatively higher than those found in sand fractions of the Top and Bottom riverbed cores.

Table 4
Arsenic contents in sand fractions of riverbed sediments

River	Location	Arsenic Concentration, mg kg^{-1}		
		Riverbed (0-5 cm)	Top riverbed core	Bottom riverbed core
Jamuna	Bahadurabad	2.51	1.85	1.10
	Jagannathgang	2.52	1.83	1.25
	Bhuanpur	Sand absent	1.88	1.25
	Aricha-Nagarbari	1.51	1.08	1.37
Padma	Rajshahi (Kutibari)	Sand absent	1.00	1.41
	Pakshi	Sand absent	1.77	Sand absent
	Aricha –Paturia	2.67	1.81	2.47
	Mawa	2.63	Sand absent	1.60
	Tongibari-Digirpar	2.07	1.72	2.22
Meghna	Sohagpur (10 km u/s of Bhairab Bridge)	2.65	1.61	1.58
	Zia Fertilizer Factory	1.00	1.30	1.54
	Daudkandi	2.45	1.27	2.38
	Chandpur	2.24	1.39	2.43
	Heimchar	2.98	1.13	2.40

Table 5 shows As concentrations in silt fractions of all riverbed sediment samples. Arsenic concentrations of silt fractions varied from 1.86 to 3.78 mg kg^{-1} in riverbed sediments (0-5 cm), from 2.32 to 3.68 mg kg^{-1} in Top riverbed cores and from 2.63 to 3.88 mg kg^{-1} in Bottom riverbed cores. With some exceptions, As concentrations of silt fractions of riverbed sediments (0-5 cm) were found to be relatively higher than those found in silt fractions of riverbed core sediments. A comparison of Table 4 and Table 5 reveals that As concentrations in silt fractions of all the 14 samples are significantly higher than those in the sand fractions.

Table 6 shows that As concentrations in clay fractions varied from 3.87 to 6.28 mg kg^{-1} in riverbed sediments (0-5 cm), from 4.59 to 6.64 mg kg^{-1} in Top riverbed cores and from 4.58 to 5.85 mg kg^{-1} in Bottom riverbed cores. Unlike sand and silt, As concentrations of clay fractions in riverbed core samples (both Top and Bottom) have been found to be higher than those found in clay fractions of the riverbed sediments (0-5 cm). Arsenic concentrations of clay fractions of all the 14 samples have been found to be much higher than those in the sand and silt fractions. For example, As contents of sand, silt and clay fractions of riverbed sediments (0-5 cm) from Jamuna river at Bahadurabad are 2.51 , 3.11 and 4.38 mg kg^{-1} , respectively, and those in the riverbed sediments (0-5 cm) from Padma river at Mawa are 2.63 , 2.84 and 6.28 mg kg^{-1} , respectively. Fine grained fractions such as clay usually contain higher arsenic because of their larger surface area as well as adsorption sites per unit volume/weight. Figure 2 shows As concentrations in sand, silt and clay fractions of riverbed sediments (0-5 cm) from different locations along Jamuna, Padma and Meghna rivers.

Table 5
Arsenic contents in silt fractions of riverbed sediments

River	Location	Arsenic Concentration, mg kg ⁻¹		
		Riverbed (0-5 cm)	Top riverbed core	Bottom riverbed core
Jamuna	Bahadurabad	3.11	2.53	Silt absent
	Jagannathgang	3.14	2.73	3.67
	Bhuanpur	3.31	2.32	3.11
	Aricha-Nagarbari	Silt absent	2.34	3.22
Padma	Rajshahi (Kutibari)	2.86	2.40	3.88
	Pakshi	3.57	2.96	Silt absent
	Aricha –Paturia	3.61	3.27	2.82
	Mawa	2.84	Silt absent	3.08
	Tongibari-Digirpar	2.86	2.56	3.02
Meghna	Sohagpur (10 km u/s of Bhairab Bridge)	3.94	3.68	3.13
	Zia Fertilizer Factory	1.86	2.38	2.63
	Daudkandi	3.78	2.89	3.39
	Chandpur	2.51	3.25	3.48
	Heimchar	3.40	3.59	3.23

Table 6
Arsenic contents in clay fractions of riverbed sediments

River	Location	Arsenic Concentration, mg kg ⁻¹		
		Riverbed (0-5 cm)	Top riverbed core	Bottom riverbed core
Jamuna	Bahadurabad	4.38	Clay absent	Clay absent
	Jagannathgang	3.87	4.97	5.449
	Bhuanpur	4.37	6.46	Clay absent
	Aricha-Nagarbari	4.11	Clay absent	5.26
Padma	Rajshahi (Kutibari)	4.94	Clay absent	5.02
	Pakshi	4.03	5.45	4.86
	Aricha-Paturia	4.83	Clay absent	5.42
	Mawa	6.28	4.93	5.07
	Tongibari-Digirpar	6.18	4.59	4.58
Meghna	Sohagpur (10 km u/s of Bhairab Bridge)	4.58	4.61	5.85
	Zia Fertilizer Factory	Clay absent	Clay absent	4.99
	Daudkandi	Clay absent	5.72	5.15
	Chandpur	Clay absent	6.64	5.28
	Heimchar	Clay absent	Clay absent	Clay absent

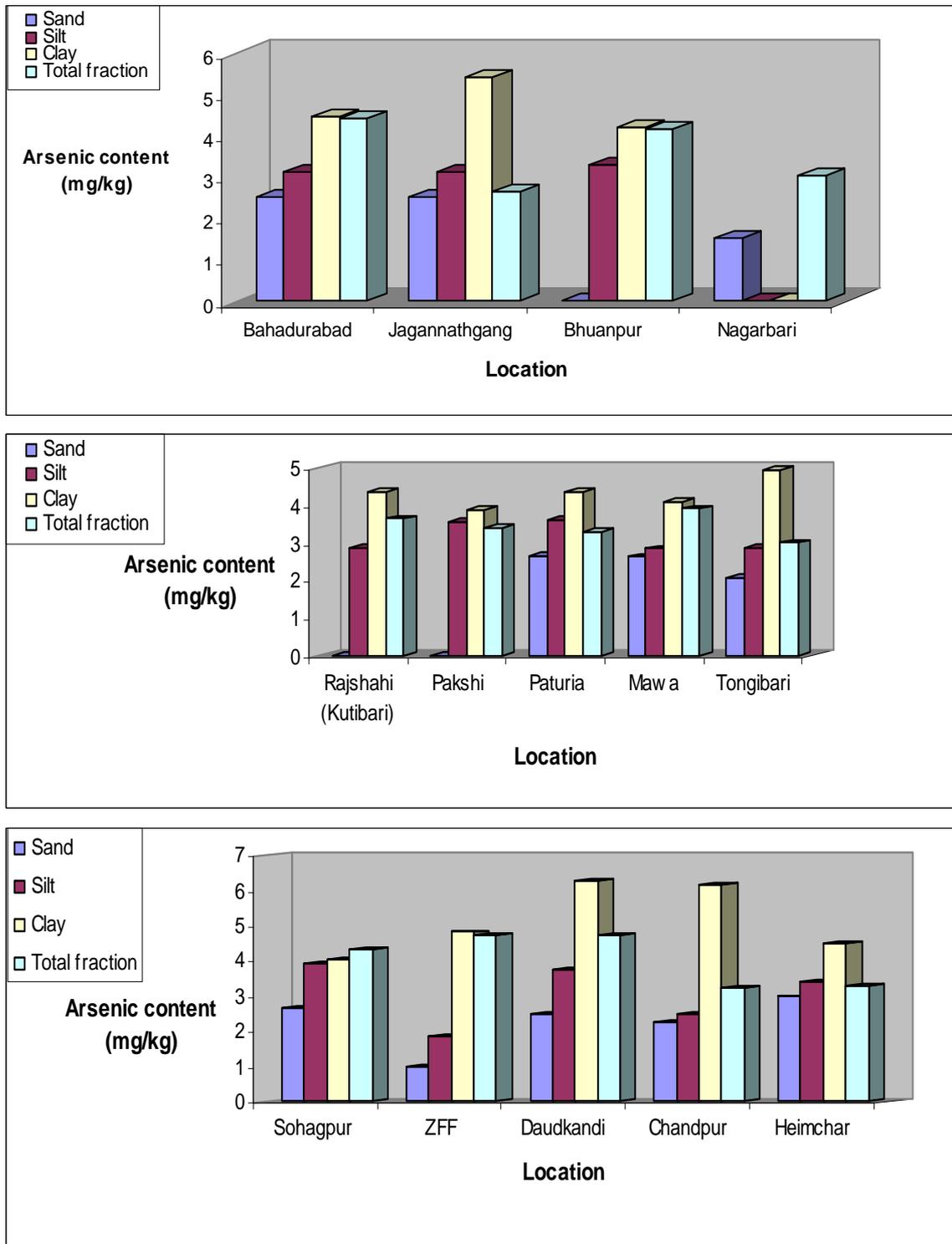


Fig. 2. Arsenic concentrations in sand, silt and clay fractions of the riverbed sediments (0-5 cm) of Jamuna, Padma and Meghna rivers

Arsenic concentrations in suspended and riverbed sediments found in this study are comparable to those reported for aquifer sediments in Bangladesh (e.g., BGS and DPHE, 2001; Smedley and Kinniburgh, 2001; Harvey et al., 2002). For example, BGS and DPHE

(2001) reported average As concentration in alluvial sand and mud/clay in Bangladesh to be 2.9 mg kg^{-1} and 6.5 mg kg^{-1} , respectively. Smedley and Kinniburgh (2001) reported that As concentration in sediments of Chapainawabgang and Lakshmipur varied from 0.4 to 10.3 mg kg^{-1} with an average of 5.9 and 3.2 mg kg^{-1} , respectively.

3.3 Arsenic transportation through rivers

Arsenic is transported through the Jamuna, Padma and Meghna rivers in dissolved form, but mostly in association with suspended sediments. The dissolved As is transported into the Bay of Bengal, but part of the As associated with suspended sediments is deposited in the river bed and the floodplains. The suspended sediment loads transported by the rivers Jamuna, Padma and Meghna integrated over the year were estimated at 499, 890 and 1003 million tons, respectively (FAP 24, 1996). Using average As concentration in suspended sediments of these rivers, the estimated As load carried annually with suspended sediments of the Jamuna, Padma and Meghna rivers are 3054, 4121 and 4584 tons, respectively.

4. Conclusions

Arsenic concentrations of river water and sediment samples along the three major rivers in Bangladesh have been determined. The very low As concentration in river water samples is consistent with the characteristic quality of surface water. Arsenic concentrations in suspended sediments varied from 4.07 to 5.47 mg kg^{-1} and did not differ significantly among the rivers. Arsenic concentrations of river bed sediments were comparatively lower than those of the suspended sediments collected along the rivers. Arsenic concentrations in clay fraction of the sediments were the highest, followed by silt and sand fractions.

The estimated quantity of As carried through Bangladesh by the major rivers with 2.4 billion tons of sediment is about 11,000 tons per year. The *beels* and *haors* receive higher quantity of As as the clayey sediments with higher As content carried through rivers are deposited in these stagnant water bodies. Chandpur, Madaripur, Munshiganj, Gopalganj, Lakshmipur, Noakhali, Bagerhat, Shariatpur, Comilla, Faridpur, Satkhira and Meherpur districts have numerous *beels* and *haors*, and these districts are also severely affected by As contamination of groundwater. This study shows that As concentrations of river sediments and aquifer materials in the contaminated areas are comparable. Therefore, the recent riverbed and floodplain sediments could be an important source of As, from which As could be mobilized into the groundwater.

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