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METALLIC CONCENTRATION IN KARNAPHULY ESTUARY SEDIMENT USING NEUTRON ACTIVATION ANALYSIS TECHNIQUE

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ABSTRACT

Thousands of tonnes of liquid waste, dirt and garbage from different mills and factories from Kaptai to Chittagong are dumped unhindered into the Karnaphuli River. Pollutants enter the river directly from urban sewerage and industrial waste discharges. Ships pollute waterways in many ways. All theses can contribute to the heavy and other metals of the water bodies. To find out concentration of different metallic elements and make a comparison with the world reference values twenty one surface sediment samples were collected from the different locations of Karnaphuly River near Chittagong city drainage outlet. Neutron Activation Analysis (NAA) technique was used for detecting the concentration qualitatively and quantitatively using the 3 MW TRIGA MARK II research reactor of Atomic Energy Research Establishment, Savar, Dhaka. Pearson correlation among the elements obtained using SPSS software. Three transitional metals Fe, Cr and Co, two alkali metals Rb and Na, two lanthanides La and Ce, and two metalloids As and Sb were determined. The concentrations of elements were compared with the world reference values. Some elements are found in elevated level.

Keywords: Environmental pollution, Heavy metal contamination, Karnaphuli River, Neutron Activation Analysis, Nuclear Science application, River sediment.

INTRODUCTION

Heavy metals levels in the environment have increased in the areas where industrial, agricultural and mining activities are widespread (Bryan, G., Langston, W. J., 1992). Heavy metals find their way into aquatic systems as a result of direct input, atmospheric deposition and surface runoffs (Kalay, M. and M. Canli, 2000). Transport, burning of fossil fuels, animal and human excretions and domestic waste contribute to the heavy metals in the water bodies (K. Chandra Sekhar et al., 2003). In the aquatic environment, metals become toxic when accumulation by aquatic organisms reaches a substantially high level (E. E. Obasohan, J. A. O. Oronsaye and O. I. Eguavoen, 2008). Sediments are known to act as a sink for heavy metals from both natural and anthropogenic sources (Pempkowiase J, Sikora A, Biernacka E., 1999). Sediments reflect the current quality of the system (J. Kruopiene, 2007). Karnaphuli River is an important waterway of Bangladesh. The principle seaport of this country is situated at the lower end of this river. It is also a big source of fish and has been playing an essential part in the economic development of the country since 1888. The river seems to be seriously polluted by discharge of industrial effluents, indiscriminate throwing of household, clinical, pathological & commercial wastes, and discharge of fuel and human excreta. In fact, the river has become a dumping ground of all kinds of solid, liquid and chemical waste of bank-side population. Pollutants enter Karnaphuli River directly from Chittagong city sewerage and industrial waste discharges. Once introduced into the water, metals tend to become incorporated into the underlying sediments so that sediments are indicators of metal contamination levels (Wendy Norville, 2005). The objective of the present study is to detect the heavy metals and trace elements concentration present in the Karnaphuli river sediments and establish the baseline concentration data of major, minor and trace heavy metals in the coastal area of the Bay of Bengal, taking Karnaphuli River as an experimental site.

MATERIALS AND METHODS

Many industrial plants and factories (Table - 1) and a densely populated city (Chittagong) are situated at the banks of Karnaphuli. Chittagong sea-port is on its lower end. Samples were collected from three areas of Karnaphuli estuary - Shah Amanat Bridge area, Chaktai Khal area and Bandor area (indicated in the Fig.1). Among these three areas the municipality outlets joined the river at Chaktai Khal and Bandor areas. These two areas are at the downstream of Karnaphuli.

Table 1: Major polluting industries around Karnaphuli River

Polluting Industries	**Quantity**
Tanneries	19
Textile mills	26
Oil refinery	1
TSP plant	1
DDT plant	1
Chemical complexes	2
Fish processing unit	2
Urea fertilizer factory	1
Asphalt bitumen plant	1
Steel mill	1
Paper mill	1
Rayon mill complex	1
Cement factories	2
Pesticide manufacturing plant	2
Paint and Dye manufacturing plant	2
Soap and detergent factories	several

A. H. M. Kamal et. al. (2009) observed that the physico-chemical parameters of the estuarine environment show regular seasonal patterns of variation. The annual surface water temperature (<30ºC) of the estuarine system is closely related to the annual cycle of air temperature. The dissolved oxygen varies from 1.89 mg/l to 5.37mg/l. pH varies from 5.9 to 7.8. BOD varies from 1.02 to 3.51mg/l. The value of TDS varies from 264.3 to 582.9mg/l. And the value of TSS varies from 97.69 to 208.15mg/l during different months of a one year cycle.

The geographical locations of the collected samples are around 22°19.760'N and 91°51.091'E through 2 Km along the Karnaphuli River.

Fig. 1: Bandar, Chaktai Canal and Shah Amanat Bridge in Karnaphuli River

A total of twenty one samples were manually collected using conventional tools. After collection, all the samples were air dried in laboratory temperature. Each sediment sample was sieved to remove the aggregates and organic species, dried at about 80°C in an oven until having constant weight to remove the moisture and crushed into fine powder with an agate mortar and pestle. Weights are taken with the help of a micro balance. The weight of each sample is kept around 80mg. These samples were transferred into ultra clean polyethylene vial individually with identification number. The bullets are then sealed and preserved carefully for irradiation. To apply relative standardization approach, we used three standard / certified reference materials: IAEA-Soil-7, IAEA-SL-1 and NIST 1633b Coal Fly Ash. Each of the standards was prepared the same way as the sample.

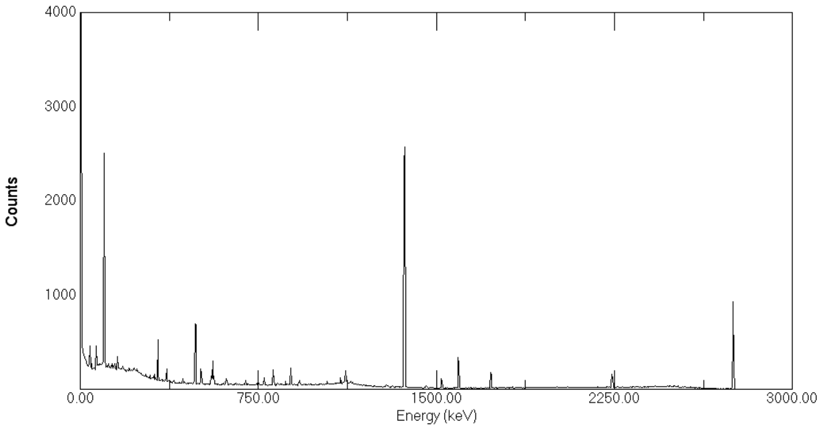

Fig.2: A typical spectrum from soil-7 after 1-day decay of end of irradiation

All the samples and standards, along with some Al-0.1% Fe foils, were taken into a long polyethylene irradiation tube. Irradiations were performed by the thermal neutron flux of 1×10^{13} $n/cm^2/sec$ for 80 minutes. After irradiation, samples were kept in a shielded place for four days to let the short lived radio nuclides decay. The bullet vials were taken out from the big plastic vials and were placed separately onto the HPGe detector. The spectrum was observed on a computer monitor. Care was taken so that dead time does not exceed 10%. The gamma spectrometry of all the irradiated samples and standard / certified reference materials was performed using a pc-based

HPGe detector coupled with a digital gamma spectrometry system. The data acquisition was performed using the software Genie-2000(Canberra) and MAESTRO-32(ORTEC) and the gamma peak analysis was performed using the software Hypermet PC version 5.12. However each of the peaks was checked manually. Fig.2 shows a typical spectrum for Soil-7 after 1 day decay of end of irradiation.

Nuclear data used for the identification and measurement of elements are shown in the Table-2. The peak areas were used to quantify the activities of the individual elements by using Eq. (1). Then using Excel sheet concentrations of elements were calculated.

Table 2: Nuclear data (C. Leclerc and A. Cornu, 1974) used for the measurement of elements

Element	**Target isotope**	**Product**	**Abundance (%)**	**Half life**	**Photopeak (Energy in keV)**	**Gamma Intensity**
Fe	^{58}Fe	^{59}Fe	0.31	45.1d	1099	100
Cr	^{50}Cr	^{51}Cr	4.31	27.8d	320	100
Co	^{59}Co	^{60}Co	100	5.24y	1332	100
Rb	^{85}Rb	^{86}Rb	72.2	18.65d	1076.6	-
Na	^{23}Na	^{24}Na	100	15h	2754	100
La	^{139}La	^{140}La	99.91	1.68d	1595	100
Ce	^{140}Ce	^{141}Ce	88.5	2.07y	145.5	-
As	^{75}As	^{76}As	100	1.09d	559	100
Sb	^{122}Sb	^{122}Sb	57.25	2.75d	564	100

The activation equation for relative NAA is

$$W_{sample} = \frac{A_{sample} e^{\lambda T_{sample}}}{A_{std} e^{\lambda T_{std}}} \times W_s \qquad (1)$$

Here, *A* stands for activity and *W* stands for weight.

Quality control test has been performed to investigate the reliability of the analysis by measuring concentration level in standard / certified reference materials Coal Fly Ash 1633b, SL-1 and Soil -7. It was done by assuming one of these three standards/references as standard for calculation and the rest are unknown. The analytical results for the certified/standard reference material are given in Table3. One may observe from the table that the deviation of the measured values from the certified one (Coal Fly Ash 1633b) is less than or around 10% for most of the elements except Na and Sb, whereas the determination of Na and Sb has less than 5% deviation with Soil-7.

Table 3: Accuracy of the analytical data with reference to the IAEA Certified Reference Materials (CRM) Soil-7 and NIST Standard Reference Material (SRM) Coal fly ash 1633b

Element	**Concentration of element in ($\mu g / g$)**					
	IAEA CRM Soil - 7			NIST SRM Coal fly ash 1633b		
	Measured Value	Certified Value	Deviation	Measured Value	Certified Value	Deviation
As	13.16 ±0.57	13.4	1.79%	122.56 ±4.63	136.2	10.01%
Na	2404.89 ± 73.08	2400	0.20%	2240.74 ±68.64	2010	11.48%
Sb	1.64 ±0.11	1.7	3.53%	5.03 ±0.29	6.0	16.17%
Cr	75.44 ±2.66	60	20.5%	204.42 ±6.86	198.2	3.14%
Co	BDL	8.9	-	48.88	50	2.24%
Fe	26899.84 ±652.92	25700	4.46%	78301.21 ±833.95	77800	0.64%
La	33.17 ±2.31	28	18.46%	105.2 ±7.27	94	11.92%

BDL - below detection limit

The detection limit was calculated using Currie's formula,

$$D_L = 2.71 + 4.65\sqrt{B} \qquad (2)$$

Where, D_L is the detection limit, and B is the background under a γ–ray peak. Detection limit shows that it is well below the values we got.

RESULTS

Total contents of nine elements (metals and metalloids), viz., Fe, Cr, Co, Na, Rb, La, Ce, As and Sb were measured in twenty one sediment samples using neutron activation analysis technique. The metallic concentrations of the Karnaphuli sediment are shown in Table 4. The metal content of the sediment ranged from 0.66 to 7.1% for iron (Fe), 16.85 to 163.03 µg/g for chromium (Cr), 6.44 to 90.82 µg/g for cobalt (Co), 4553.24 to 9279.32 µg/g for sodium (Na), 78.71 to 316.92 µg/g for rubidium (Rb), 24.37 to 53.06 µg/g for lanthanum (La), 12.44 to 148.61 µg/g for cerium (Ce), 3.07 to 43.17 µg/g for arsenic (As) and 0.36 to 3.5 µg/g for antimony (Sb). The average concentration of these elements are 50053.51 (or 5%), 117.68, 63.31, 10154.24, 135.80, 41.72, 105.91, 12.53 and 1.180 µg/g respectively.

Table 4. Some metallic concentration in Karnaphuli estuary sediment

Sample ID	**Fe in μg/g**	**Cr in μg/g**	**Co in μg/g**	**Rb in μg/g**	**Na in μg/g**	**La in μg/g**	**Ce inμg/g**	**As in μg/g**	**Sb in μg/g**
SB-1	11020.36 ±183.95	45.67 ±3.03	17.11 ±0.46	BDL	4553.24 ±130.40	27.93 ±0.67	41.12 ±0.69	3.07 ±0.095	BDL
SB-2	24918.23 ±289.47	72.38 ±4.66	32.33 ±0.69	80.48 ±3.93	5005.08 ±147.76	24.37 ±0.56	59.49 ±0.80	3.48 ±0.108	BDL
SB-3	30374.20 ±339.40	99.27 ±6.36	35.04 ±0.72	78.71 ±4.05	5576.08 ±166.41	37.42 ±0.94	78.82 ±1.00	6.23 ±0.194	0.66 ±0.02
SB-4	70310.72 ±617.93	163.01 ±10.32	87.73 ±1.34	198.84 ±8.25	8420.12 ±326.92	36.60 ±0.92	148.61 ±1.52	8.68 ±0.27	1.02 ±0.03
SB-5	6622.08 ±127.28	16.85 ±1.18	6.44 ±0.28	BDL	9078.31 ±551.77	39.49 ±1.11	12.44 ±0.31	15.48 ±0.48	1.13 ±0.04
SB-6	56028.99 ±521.40	129.77 ±8.26	70.30 ±1.11	144.70 ±6.48	9279.32 ±232.00	43.91 ±1.06	121.57 ±1.34	7.52 ±0.24	1.72 ±0.06
SB-7	45221.09 ±462.51	93.12 ±6.00	54.92 ±0.96	101.28 ±5.21	7885.81 ±459.06	53.06 ±1.61	94.08 ±1.18	14.10 ±0.44	1.40 ±0.05
SB-8	64172.97 ±597.80	150.33 ±9.57	86.01 ±1.31	168.35 ±7.70	6385.67 ±939.84	41.74 ±1.49	125.67 ±1.21	9.34 ±0.30	1.20 ±0.04
SB-9	71548.89 ±651.59	153.83 ±9.79	90.82 ±1.38	162.08 ±7.36	7887.54± 289.76	44.06 ±1.15	148.23 ±1.62	10.27 ±0.36	1.57 ±0.06
SB-10	60167.25 ±574.33	143.25 ±9.14	84.69 ±1.31	161.28 ±7.46	8255.82± 202.74	48.84 ±1.2	132.98 ±1.51	10.26 ±0.32	1.57 ±0.06
SB Average	44038.78	106.45	56.54	136.96	7232.68	39.74	96.30	8.84	1.28
CK-1	57168.98 ±521.71	116.52 ±7.41	69.98 ±1.12	135.9 ±5.94	8149.15 ±206.39	42.54 ±1.03	122.09 ±1.31	11.91 ±0.37	0.36 ±0.03
CK-2	22713.46 ±288.31	110.54 ±7.37	35.61 ±0.7	95.18 ± 4.25	6932.02 ±422.31	28.27 ±0.67	71.18 ±0.97	15.53 ±0.48	1.67 ±0.02
CK-3	69332.21 ±623.21	163.03 ±10.35	84.73 ±1.29	178.58 ±7.83	1404.43 ±38.31	11.08 ±0.25	137.92 ±1.49	43.17 ±1.59	1.44 ±0.05
CK-4	60329.07 ±547.32	121.25 ±8.02	74.39 ±1.16	142.13 ±6.17	2554.67 ±107.45	BDL	126.11 ±1.33	12.67 ±0.36	0.49 ±0.04
CK-5	67118±58 2.67	159.46 ±9.01	79.93 ±1.21	170.09 ±7.1	4843.68 ±127.36	25.35 ±0.34	134.87 ±1.41	9.39 ±0.32	1.38 ±0.05
CK Average	55332.33	134.16	68.93	144.37	4776.79	26.81	118.43	18.53	1.07
B-1	60135.67 ±561.02	138.35 ±8.81	78.59 ±1.22	169.36 ±7.55	53621.66 ±8519.79	84.52 ±3.03	119.10 ±1.35	28.13 ±0.99	3.51 ±0.13
B-2	68175.05 ±620.87	152.20 ±9.68	81.48 ±1.24	316.92 ±13.16	8206.51 ±520.49	45.41 ±1.35	144.52 ±1.58	4.60 ±0.14	1.19 ±0.04
B-3	39635.69 ±440.12	100.11 ±6.38	51.97 ±0.94	120.62 ±6.34	6953.52 ±203.33	39.92 ±1.09	100.28 ±1.23	10.49 ±0.37	1.1 ±0.04
B-4	55605.92 ±536.10	127.64 ±8.16	69.54 ±1.12	140.45 ±6.68	8604.88 ±578.35	44.86 ±1.33	102.03 ±1.24	11.81 ±0.38	0.94 ±0.03
B-5	58149.08 ±523.37	131.54 ±8.62	71.22 ±1.13	147.64 ±6.51	11782.65 ±785.54	BDL	111.28 ±1.29	14.09 ±0.43	1.23 ±0.04
B-6	52375.76 ±527.55	121.39 ±8.01	66.74 ±1.12	139.14 ±5.98	51696.86 ±838.39	76.59 ±2.56	106.26 ±1.24	13.01 ±0.39	1.21 ±0.04
B Average	55679.53 or 5.6%	128.56	69.93	172.36	23477.68	58.26	113.91	13.69	1.53
All Samples Average	50053.51 or 5%	119.50	63.31	150.09	11289.38	41.89	106.60	12.53	1.30

DISCUSSIONS

Concentrations of some elements for the reference sites are tabulated in the Table 5 below. The concentration value of Fe in West river sediment (C. Leclerc and A. Cornu, 1974) ranges from 0.6 to 6.1 with an average value 3.7 in percentage. The concentration value of Cr for Danube sediment (N. Milenkovic et al., 2005) ranges from 30.6 to 112 with an average value 76.2 in µg/g.

Table 5: Element concentrations in the reference sediment

Sediment	Fe (%)	Cr **(µg/g)**	As **(µg/g)**	Sb **(µg/g)**	Ce **(µg/g)**	Rb **(µg/g)**	Co **(µg/g)**	La **(µg/g)**
West River	0.6-6.1 avg. 3.7							
Danube River		30.6-112 avg. 76.2	1.0-44.7 avg. 7.0					
Ohio River	2.1-3.9 avg 3.0%	12-130 avg. 43.3						
Yozgat Turkey		26.7-39.6 avg. 32.0					8.3-19.2 avg. 11.3	
Arno			3-9		35-152	63-150	<3-21	12-76
Dalaio				0.027-1.12 avg. 0.62				

The value for As ranges from 1.0 to 44.7 with an average of 7.0 µg/g for the same sediment. The Fe concentration in Ohio River (John D. Youger and William J. Mitsch, 1989) ranges from 2.1 to 3.9 with an average 3.0 in percentage. The Cr concentration for the same river ranges from 12 to 130 with an average 43.3 in µg/g. Whereas in Yozgat Turkey (M. Soylak et al., 2002) sediment Cr concentration ranges from 26.7 to 39.6 with an average 32.0 in µg/g. The Co concentration for the same sediment ranges from 8.3 to 19.2 with an average 11.3 in µg/g. The Co concentration for Amo river sediment (Enrico Dinelli et al., 2005) ranges from below 3 to 21 µg/g, The Rb concentration ranges from 63 to 150 in µg/g, Concentration for La ranges from 12 to 76 µg/g, Ce concentration range is 35 to 152 µg/g and for As the range is from below 3 to 9 µg/g. The concentration of Sb in Dalaio sediment (Lin, C. Et al.) ranges from 0.027 to 1.12 with an average 0.68 µg/g. Fig. 3 gives a graphical comparison of the metallic concentrations Fe, Cr, Co, Rb, Ce, Na, La, As and Sb with the others. The descending order of the average concentrations of Fe is B>CK>SB>West river sediment > Ohio River sediment.

Lowest Fe concentration of Bandar area sediment is more than the highest value of Fe concentration in Ohio river sediment. The average Fe concentration of Karnaphuli sediment is almost double compared to that of Ohio river. The descending order of the average concentrations of Cr is CK>SB>B>Danube sediment>Ohio river sediment>Yozgat Turkey sediment. All the concentrations (even lower one) for Cr in the sediment of Bandar and Chaktai Khal areas are higher than the highest Cr concentration of Yozgat-Turkey sediment. Average Cr concentration of Karnaphuli sediment is approximately one and half times higher than that of Danube sediment, three times higher than that of Ohio River sediment and four times higher than that of Yozgat-Turkey sediment. The descending order of the average concentrations of Co is B>CK>SB>Yozgat Turkey sediment. All the concentrations (even lower one) for Co in the sediment of Bandar and Chaktai Khal areas are higher than the highest Co concentration of Yozgat-Turkey sediment. Average Co concentration of Karnaphuli sediment is approximately six times higher than that of Yozgat-Turkey sediment.

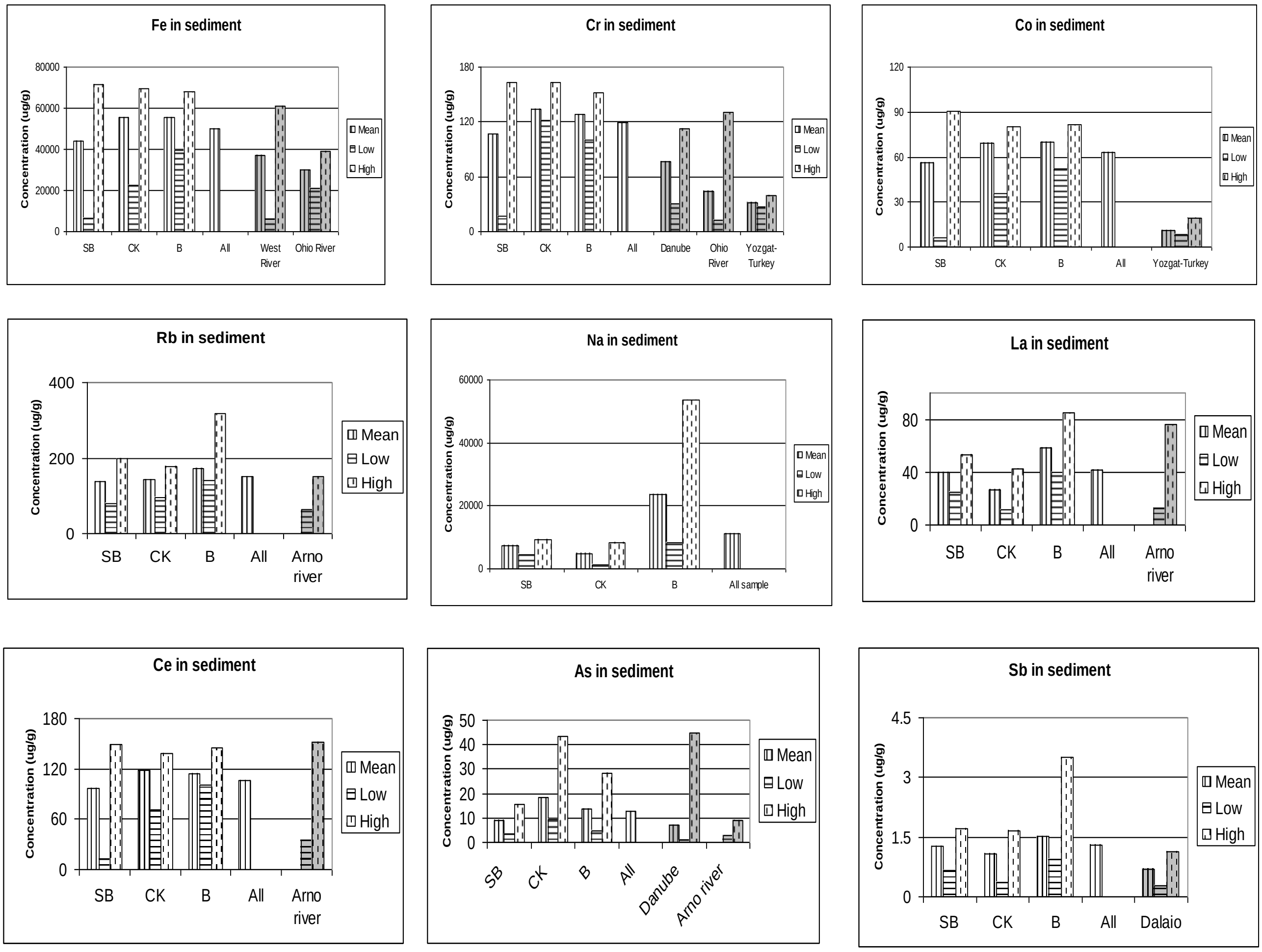

Fig. 3: Concentrations of metallic elements in Karnaphuli sediment

The descending order of the average concentrations of Rb is B>CK>SB. All the highest concentrations for Rb in the sediment of Shah Amanat Bridge, Bandar and Chaktai Khal areas are higher than the highest Rb concentration of Arno river sediment. All the lowest concentrations for Rb in the sediment of Shah Amanat Bridge, Bandar and Chaktai Khal areas are higher than the lowest Rb concentration of Arno river sediment. The Average Rb concentration of Karnaphuli sediment is approximately equal to the highest concentration of Arno river sediment. The descending order of the average concentrations of Na is B>SB>CK. The concentrations for Na in the sediment of Bandar area are significantly higher than that of other areas of Karnaphuli. Even the lowest Na concentration in the sediment of Bandar area is approximately equal to the highest Na concentration of the other areas of Karnaphuli. The descending order of the average concentrations of La is B>SB>CK. Descending order of the highest concentrations of La is B>Arno>SB>CK. Descending order of the lowest concentrations of La is B>SB>CK>Arno. The average value of La concentration in the sediment of Karnaphuli is approximately equal to the mean of highest and lowest concentrations of Arno river sediment. The descending order of the average concentrations of Ce is CK>B>SB. Descending order of the highest concentrations of Ce is B>Arno>SB>CK. Descending order of the lowest concentrations of La is B>SB>CK>Arno. The average value of La concentration in the sediment of Karnaphuli is approximately equal to the mean of highest and lowest concentrations of Arno river sediment. The descending order of the average concentrations of As is CK>B> SB>Danube. All the concentrations of As in the sediment of Chaktai Khal areas are higher than the average As concentration of Danube river sediment. Highest As concentration of Danube River is comparable to the highest As concentration of Chaktai Khal area. Highest As concentration of Arno river sediment and the average As concentration of Danube river sediment are comparable to the average As concentrations of Bandar, Chaktai Khal and Shah Amanat Bridge areas of Karnaphuli river sediment. The descending order of the average concentrations of Sb is B>SB>CK> Dalaio. All the concentrations of Sb in the sediment of Bandar areas are higher than the average Sb concentration of Dalaio sediment. Highest Sb concentration of Dalaio sediment is comparable to the average As concentration of Bandar, Chaktai Khal and Shah Amanat Bridge areas.

Since there is no treatment before waste products are discharged, local environmental degradation has occured. Fish kills and accumulation of toxic substances in fish and shrimp flesh have been recorded in the countries five industrial zone: Dhaka, Chittagong, Narayangaj, Khulna and Ghorashal. Dhaka has a sewage treatment plant, but it can only take care of about a fifth of its population’s wastes. Other big cities have no waste treatement facilities at all. Most freshwater in Bangladesh is, therefore, badly polluted and huge quantities of untreated wastes find their way through open drains, canals and rivers into the Bay of bengal. (Staffan Holmgren, 1994). The continuous increase in heavy metal contamination of estuaries and coastal waters is a cause for concern as these metals have the ability to bioaccumulate in the tissues of various biota, and may also affect the distribution and density of benthic organisms, as well as the composition and diversity of in faunal communities (Geydu-Ababio et al., 1999). The estuarine sediments serve as reservoirs for the heavy metals and their concentrations are controlled by a variety of physical and chemical factors. It is important to determine the source of these heavy metals and to manage their input into the River ecosystem so that their concentrations in the sediment do not reach toxic levels. The heavy metal concentrations in estuarine and coastal sediments also need to be monitored on a more regular basis. (Karen Binning and Dan Baird, 2001).

Table 6: Correlation coefficient (r) values among metallic elements in Karnaphuli sediment

	Cr	**Co**	**AS**	**Rb**	**Ce**	**La**	**Na**	**Fe**	**Sb**
Cr	1								
Co	0.973	1							
As	0.282	0.283	1						
Rb	0.872	0.854	0.137	1					
Ce	0.977	0.978	0.218	0.881	1				
La	0.093	0.184	0.011	0.168	0.097	1			
Na	0.132	0.175	0.326	0.146	0.106	0.835	1		
Fe	0.976	0.992	0.302	0.880	0.984	0.154	0.151	1	
Sb	0.456	0.523	0.554	0.401	0.452	0.722	0.804	0.509	1

Table 6 shows the Pearson correlation obtained using SPSS software. From the above table it is seen that, there lies some strong / positive correlations among Fe, Cr, Co, Rb and Ce. The strength of association between these variables is very high.

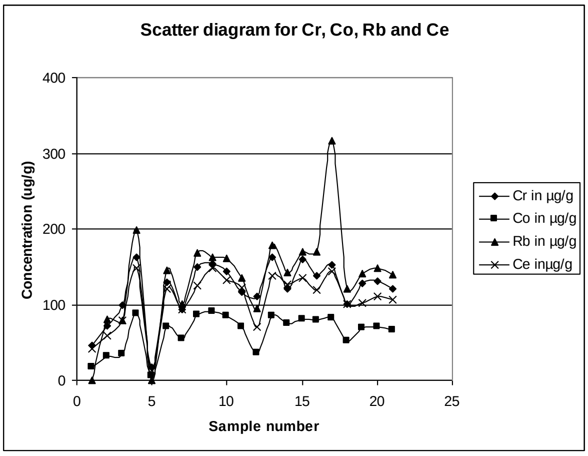

Fig. 4: Scatter diagram of Cr, Co, Rb and Ce

The situation can be observed form the scatter diagram shown in Fig. 4. There are also some positive correlations among Na, La and Sb. Fifty to sixty five percent of variations of these elements are related.

CONCLUSION

In order to evaluate the heavy metal pollution in Karnaphuli river sediment, a number of samples from different locations were collected and analysed using NAA method. The obtained results show that the concentrations are significantly higher than the reference values for almost all heavy metals (Fe, Cr, Co, As, Sb) studied in this research. It is also seen that there are some positive correlations among the concentrations of Fe, Cr, Co, Rb and Ce. It is highly recommended that further investigations and monitoring should be conducted to assess long term effects of anthropogenic inputs into the Karnaphuly River system.

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