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ORIGINAL RESEARCH ARTICLE

Heavy metal concentrations in water from Bakkhali River estuary, Cox's Bazar, Bangladesh

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ABSTRACT

Heavy metals contamination of water is considered as severe global issues for developing countries like Bangladesh. Because heavy metal pollution ruined aquatic ecosystem especially fish diversity which contribute important share on economy of a country. The present study designed to investigate the contamination level of lead (Pb), chromium (Cr), cadmium (Cd), copper (Cu) and zinc (Zn) in surface water of Bakkhali river estuary in Bangladesh. The decreasing order of metals concentration was Cu>Zn >Cr>Pb>Cd with the mean value of 2.6> 0.825 > 0.355 > 0.056 >0.003 mg/L, respectively. The results showed a significant seasonal variation of heavy metals concentration in water. Higher contamination occurred during post monsoon, while lower during the monsoon season. Among the all studied heavy metals Cu concentration was higher (2.6 mg/L) and lower (0.003 mg/L) concentration was Cd. Metal concentrations in water samples exceeded the safe limits of drinking water which indicated that the water from this estuarine river is not fully safe for using/drinking. The study area was not entirely polluted in terms of all metal concentrations. But level of metal concentration in polluted sites supposed risk to ecological health. The findings of present study recommended that continuous monitoring of water should be aimed to evaluate the risk which could help to maintain healthy coastal ecosystem and improve management strategy of this estuarine river.

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INTRODUCTION

The Bakkhali river estuary is located at the south-eastern coast of the Bay of Bengal of Bangladesh. Estuaries provides an important economic function including transportation, tourism, industrial and agricultural activities (Heip & Herman, 1995; Raz-Guzman & Huidobro, 2002), The Bakkhali river estuary server's important economic role of local fishery of Cox's Bazar district (Hena *et al*., 2007), which ensure sustainable livelihood of local community. The substantial environment of this estuarine river serves as feeding, breeding and nursery grounds for a variety of aquatic organisms and animals. This zone is highly productive in terms of nutrient comes from different sources (Kamal & Khan, 2009). The ecosystem of this estuary hampered though environmental disturbance like sedimentation, urban runoff and pollution (Hena *et al*., 2012). Bakkhali River is the main discharge point of the city waste of Cox's bazaar where more than 53 shrimp hatcheries and aqua farms, big fish landing centers, ship repairing, painting and coating; huge hotels for amusement of tourists and industries were located. During the summer, the fish processing, aqua farm operation, hatcheries, crop cultivation and other industrial activities are robustly increased in the sampling sites. The high metal concentrations in this particular period might be attributed to discharge of different salts and chemicals from hatcheries, fish processing industries and aqua farms through their underground pipeline outlets to the sea without treatment. Besides, most of the domestic, municipal sewages and industrial effluents of Cox's Bazar city are incorporated to the Bakkhali River through different uncontrolled canals to the Sea. The waste discharge and chemical spills associated with boat repairing industries represent an additional source of pollutants to the water and sediments in this area.

Heavy metal contamination in water is an increasing worldwide environmental concern. Heavy metals pollution destroys the aquatic ecosystem especially ruined diversified fish community which contributes an important share in the domestic protein demand of the country. Pollution also reduces the fisheries resources. It causes some changes in per capita income, health condition, agricultural change, socio- cultural change, and changes in financial condition of fishermen and boatmen. Heavy metals are very harmful because of their non-biodegradable nature, long biological half-lives and their potential to accumulate in different body parts. Most of the heavy metals are extremely toxic because of their solubility in water. Increasing contamination by heavy metals has a significant adverse effect to humans and aquatic organisms (Islam *et al.*, 2014a; Islam *et al.*, 2015b; Ahmed *et al.*, 2015). However, the level of the risks is tough to accurately measure, due to the difficulty of biologic and chemical interactions that is hugely responsible for the alteration of the bioavailability of metals (Ideriah *et al.*, 2012). Mostly, the soluble forms of the heavy metals maybe found in crustaceans, finfish and shellfish (NNPC & RIP 1986) and ultimately can be transferred to humans via the food chain pathways (Banerjee *et al.*, 2011; Pan & Wang ,2012; Rahman *et al.*, 2013; Islam *et al.*, 2015c).

However, several studies have been conducted on water quality of coastal water along with Bakkhali river estuary (Hasan *et al.*, 2019). But detailed research regarding heavy metal pollution in water of this estuarine river system is still insufficient. As water is most important requirement for all life forms on earth (Bytyçi *et al.*, 2018), the present study was therefore designed to evaluate heavy metal load in water during three different season to evaluate the current pollution status of this estuarine river which could help to maintain healthy coastal ecosystem.

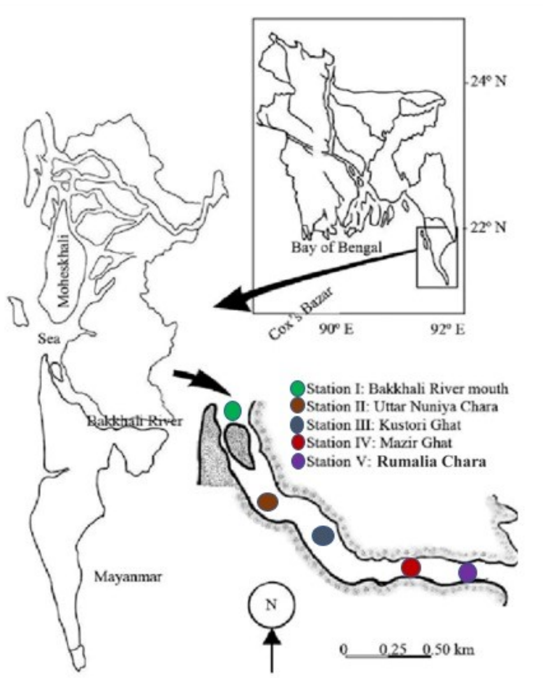

Figure 1. *Map showing the study area and sampling points in Bakkhkali River Estuary, Bangladesh.*

MATERIALS AND METHODS

Study area and site selection

Bakkhali river estuary located between 21°26'01''N latitudes and 91°58'03''E longitudes along the Bay of Bengal and South-Eastern of Bangladesh. The study was conducted from February 2022 to January 2023. Sampling was done on three respective seasons namely pre monsoon (February to May), monsoon (Jume to September) and post monsoon (October to January) in five study sites from the Bakkhali River estuary, Site-1: Bakkhali river mouth, Site-2: Uttar nuniarchara, Site-3: Kosturighat, Site-4: Maizirghat and Site-5: Rumaliachara (Figure 1).

Sampling

For heavy metal analysis, water samples from all five sampling sites were collected at a depth of about 0.3m below water surface into 500 ml plastic bottles. Prior to sampling, the bottles were cleaned with 10% nitric acid and rinsed with distilled water. The bottles were rinsed three times with the river water at the time of sampling. Samples were then collected by direct immersion of the sampling bottle into the river. Immediately after sample collection, 2 ml nitric acid was added to the water samples to reduce adsorption of metals onto the walls of the plastic bottles. Sample bottles were then labeled to indicate date of sampling and the sampling site. Samples were transported in an ice-box to the laboratory and stored at 4°C awaiting analysis.

Determination of physico-chemical parameters

Several physico-chemical parameters including, temperature, pH, salinity and dissolve oxygen (DO) were measured. Water samples were collected for on spot detection of physicochemical parameters. Surface water temperature was measured using a Celsius thermometer. Water pH was measured using an electronic pH meter (Jenway, 3020). Water salinity (ppt)was measured using a hand-held refractometer (TANAKA, New S-100, Adchi-ku, Japan) Dissolved oxygen (DO) was measured by using a portable aquaculture kit (Model FF2, HACH, USA) and DO meter (PDO-519, Taipei, Taiwan).

Digestion of water samples for metal analysis

All standard solution for target element was supplied by Merck Germany with the highest purity level (99.98%). Ultra-pure HNO_3 was used for sample digestion. All other acids and chemicals were either supra pure or ultra-pure received form Merck Germany or Scharlau Spain. Collected water samples were filtered by using Millipore Filtration, using 0.45 mm membrane filter. The filtrated water sampled was then acidified with concentrated HNO_3. 50 mL of well mixed, acidified water sample was taken in a 100 ml beaker, after that heated on a bloc digester to boil until its volume decrease to 20 ml. Another 5ml of concentrated HNO_3 was added and then heated for 10 minutes and allowed to cool. About 5 ml of $HNO_3$was used to rinse the sides of the beaker and the solution filtered using Whatman 0.42µm filter paper into a 100 ml volumetric flask and topped up to the mark with distilled water. A blank solution was similarly prepared. All the digested samples were filtered using Whatman 0.42µm filter paper into a 50 ml volumetric flask and topped up to the mark with distilled water. Digestion of water samples was carried out in Bangladesh Council of Scientific and Industrial Research (BCSIR), Rajshahi.

Metal analysis method

The presence and concentration of heavy metals in the water samples were carried out by using Flame Atomic Absorption Spectrometer (Shimadzu, AA-6800) in central laboratory of University of Rajshahi, Rajshahi. Deionized ultrapure water was used for the experimental procedure. All glassware and containers were cleaned with 20% nitric acid, finally rinsed with deionized ultrapure water several times and oven-dried prior to use.

Statistical analysis

The data analysis and graphs were done by Microsoft Office Excel, version 2010.

RESULTS AND DISCUSSION

Physico-chemical parameters of water

The physico-chemical parameters of different seasons and sites of water measured during the study period are shown in Table 1. The average value of temperature recorded 30.5±0.65 °C during pre-monsoon and 21.3±032°C during post monsoon which was found within the permissible limits set by (WHO, 2004), The findings of our present study quite similar with the findings of Zinat *et al.* (2023) at Passur river 31.39±0.39 °C (Pre-monsoon) and 21.44±0.57 (Post-monsoon), and lower than the findings of Rahman *et al.* (2021) at Meghna river with higher mean temperature (33.7 °C) as this river is highly polluted by hot industrial effluents. The average value of salinity recorded 22.02±0.28ppt during pre-monsoon and 30.4±0.69 during post monsoon. Previous study on seasonal variation of physical-chemical parameters in Bakkhali River revealed salinity level 12 ppt in pre monsoon and 27 ppt in post monsoon (Raknuzzaman *et al.*, 2018). The studied results higher than Masoud *et al.* (2019) revealed the values of salinity in the Reju Khal river estuary ranged from 8.0±3.0 to 29.0±0.6 ppt, and Zinat *et al.* (2023) recorded the values of salinity ranged between 5.35±0.10 to 18.82±0.11 in the Passur river as Bakkhali estuary comprised salt marsh sediment and. meet with Bay of Bengal. The value of Dissolve oxygen (DO) during the study period found 8.3±0.14 mg/L (pre-monsoon), 5.9±0.07(monsoon) and 11.2±0.19 mg/L (post-monsoon), respectively. Zinat *et al.* (2023) at Passur river recorded DO ranged from 4.49 mg/L (pre-monsoon) to 10.12 mg/L (post-monsoon). Previous study on Water quality by Hasan *et al.* (2019) at Bakkhali River revealed DO value 9.95 mg/L during monsoon and the highest 14.436 mg/L during post monsoon. The minimum permissible level for DO set by EQS (1991) for industrial water, fish culture and drinking purpose are 5.0, 4.5 and 6 mg/L. pH value of our present study found highest during post monsoon 8.4±0.09 and found lowest during monsoon 6.36±0.06. Hasan *et al.* (2019) at Bakkhali River recorded pH value 6.5 in monsoon to 7.6 in post monsoon. Zinat *et al.* (2023) at Passur river recorded highest pH value 7.95±0.04 during post-monsoon whereas minimum was recorded 7.19±0.06 during monsoon. The maximum permissible level for pH set by EQS (1991) for Bangladesh water quality standards in industrial discharged water, water for fish culture and drinking purpose are 6.0-9.5, 6.5-8.5 and 6.5-8.5, respectively. The studied results found within the limit.

Table 1. Physico-chemical parameters of Bakkhali River estuary at different seasons and different locations during the study period.

Season	Site	pH	Temperature	Salinity	DO
Pre-monsoon	Site-1	7.66±0.08	32.33±0.91	21.5±0.76	9.12±0.18
	Site-2	7.18±0.07	28.7±0.87	22.5±0.89	8.24±0.17
	Site-3	7.36±0.07	30.3±0.75	23.8±0.50	7.86±0.13
	Site-4	7.7±0.05	31.5±0.56	22.6±0.64	7.45±0.14
	Site-5	7.22±0.08	29.6 ±0.88	19.7±0.43	8.77±0.11
	Mean	7.42±0.07	30.5±0.65	22.02±0.28	8.3±0.14
Monsoon	Site-1	6.67±0.07	25.8±0.45	15.7±0.34	6.21±0.10
	Site-2	6.45±0.05	27.1±0.60	14.22±0.57	6.36±0.12
	Site-3	6.89±0.04	26.8±0.58	13.3±0.40	5.75±0.14
	Site-4	5.8±0.04	26.6±0.68	12.36±0.29	6.12±0.11
	Site-5	6.00±0.07	27.±3 0.77	13.8±0.55	4.87± 0.09
	Mean	6.36±0.06	26.7±0.49	13.9±0.62	5.9±0.07
Post-monsoon	Site-1	9.27±0.12	23.6±0.38	30. 4±89	12.38±0.15
	Site-2	8.43±0.14	18.7±0.52	31.02±0.90	11.24±0.14
	Site-3	8.55±0.11	20.2±0.44	28.6±0.70	10.55±0.13
	Site-4	7.89±0.08	22.6±0.37	32.5±0.62	11.6±0.17
	Site-5	7.75±0.08	21.5±0.29	29.7±0.84	10.25± 0.21
	Mean	8.4±0.09	21.3±032	30.4±0.69	11.2±0.19

Table 2. Heavy metal concentrations of water (mg/L) at different seasons and sites of Bakkhali River estuary during the study period.

Season	Site	Pb	Cd	Cr	Zn	Cu
Pre-monsoon	Site-1	0.055±0.045	0.004±0.002	0.327±0.097	0.747±0.071	2.320±0.097
	Site-2	0.053±0.036	0.003±0.002	0.267±0.075	0.697±0.055	2.693±0.088
	Site-3	0.043±0.030	0.002±0.001	0.260±0.040	0.643±0.052	2.800±0.092
	Site-4	0.056±0.040	0.004±0.003	0.277±0.059	0.733±0.065	2.823±0.080
	Site-5	0.052±0.034	0.004±0.002	0.377±0.035	0.610±0.045	2.523±0.078
Monsoon	Site-1	0.037±0.022	0.002±0.002	0.094±0.007	0.573±0.033	1.833±0.067
	Site-2	0.031±0.013	0.001±0.001	0.077±0.006	0.447±0.040	1.513±0.062
	Site-3	0.025±0.017	0.001±0.001	0.015±0.006	0.400±0.028	1.310±0.092
	Site-4	0.026±0.012	0.002±0.001	0.080±0.003	0.510±0.047	1.430±0.050
	Site-5	0.038±0.020	0.002±0.001	0.116±0.007	0.300±0.0180	1.727±0.056
Post-monsoon	Site-1	0.077±0.055	0.007±0.003	0.587±0.044	1.157±0.125	3.670±0.075
	Site-2	0.061±0.047	0.007±0.004	0.513±0.038	0.883±0.070	3.340±0.117
	Site-3	0.066±0.040	0.005±0.002	0.430±0.022	0.870±0.066	3.213±0.624
	Site-4	0.068±0.049	0.006±0.003	0.517±0.043	1.093±0.143	3.730±0.925
	Site-5	0.064±0.056	0.007±0.005	0.600±0.051	0.86±0.065	3.400±0.965

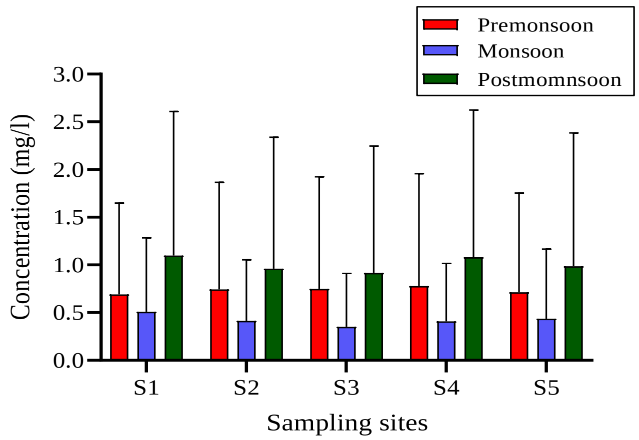

Figure 2. *Metal concentration found in water in different sampling sites in various periods.*

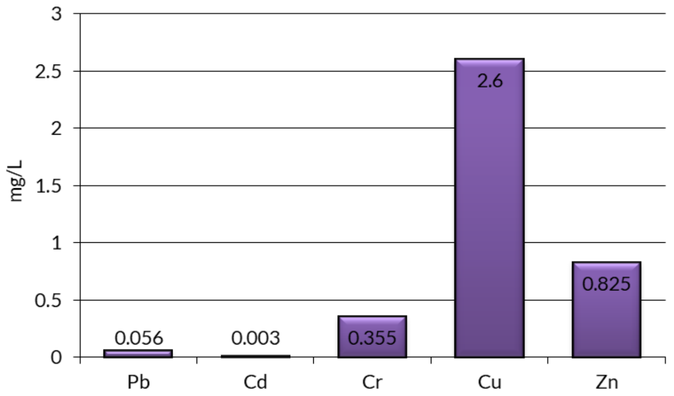

Figure 3. *Mean concentrations of different metals in water.*

Heavy metals in water

Heavy metal concentrations in water of Bakkhali River estuary at different seasons and sites during the study period are shown in Table 2 and Figure 2. Lead (Pb) concentration was highest at Site-1 (0.077±0.055mg/L) during post monsoon which going beyond the WHO (2011) standard level for drinking water, and the lowest value was observed at Site-3 (0.025±0.017mg/L) during monsoon with the average value of 0.051±0.026, 0.031±0.001 and 0.067±0.040mg/L in pre monsoon, monsoon and post monsoon respectively. The concentration of Cadmium (Cd) was highest at Site- 5 (0.007±0.005mg/L) during post monsoon and the lowest value was observed at Site-3 (0.001±0.001mg/L) during monsoon, with the average value of 0.003±0.002, 0.0018±0.001and 0.006±0.004mg/L in pre monsoon, monsoon and post monsoon respectively. The average value within the WHO (2011) standard level for drinking water but during post monsoon the values exceeded the WHO (2011) standard level for drinking water which may be due to effluents from local market and ship breaking area directly discharged into this estuarine river and low water flow during post monsoon precipitated the metal. The concentration of chromium (Cr) was highest at Site-5 (0.600±0.051mg/L) during post monsoon which going beyond the WHO (2011) standard level for drinking water and the lowest value was observed at Site-5 (0.116±0.007mg/L) during monsoon, with the average value of 0.301±0.040, 0.076±0.006 and 0.529±0.043mg/L in pre monsoon, monsoon and post monsoon, respectively.

The concentration of zinc (Zn) was highest at Site-1 (1.157±0.125747mg/L) during post monsoon and the lowest value was observed at Site-5 (0.300±0.0180 mg/L) during post monsoon, with the average value of 0.686±0.052, 0.573±0.028 and 0.927±0.066mg/L in pre monsoon, monsoon and post monsoon respectively. The concentration of cupper (Cu) was highest at Site-4 (3.730±0.925mg/L) during post monsoon which going beyond the WHO (2011) standard level for drinking water and the lowest value was observed at Site-3 (1.310±0.092mg/L) during monsoon, with the average value of 2.32±0.092, 1.513±0.062 and 3.47±0.075mg/L in pre monsoon, monsoon and post monsoon, respectively. The table and graph show that heavy metal concentration seasonally varied, higher concentration occurred during post monsoon as water flow lower than monsoon and pre monsoon causes precipitation of metal and lower metal concentration occurred during monsoon may be due to dilution effect of water through rainfall and runoff from local market and aquaculture farms as drainage system directly connected with this estuarine river. The mean value of Pb concentration in water found 0.056 mg/L (Figure 3). The concentration of present study was higher than the findings of Zinat *et al.* (2023) at Pasur River Estuary (0.024 mg/L) and Rahman *et al.*, (2021) at Meghna River estuary (0.009 mg/L). This higher concentration of present study may be due to the combined effect of sewages effluents discharged into water and chemical and oil from the fishing vehicles also discharged directly into the river.

The mean value of Cd found 0.003mg/L (Figure 3) lower than the findings recorded by Zinat *et al.* (2023) at Passur River Estuary (0.006 mg/L) and Ali *et al.* (2016) at Karnafuli River (0.01 mg/L). Untreated pollutant from local market, ship breaking area cause precipitation of this metal in Bakkhali River estuary. The Mean value of Cr found 0.355 mg/L (Figure 3) of present study was higher than the findings of Zinat *et al.* (2023) at Pasur River Estuary (0.050 mg/L) and Rahman *et al.* (2021) at Meghna River estuary (0.054 mg/L). Reason for high Chromium concentration is likely untreated sewage, ship breaking activities, agricultural and industrial effluents discharged into the river. According to Hossain *et al.*, (2021**)** Chromium widely used as anti-corrosive, plating agent in ship braking and boat repairing industry therefore cause high concentration in surrounding area. The Mean value of Zn found 0.825 mg/L (Figure 3) of present study was higher than the findings of Zinat *et al.* (2023) at Pasur River Estuary (0.014 mg/L), and lower than the concentration revealed by Hossain *et al.* (2021) at Turag River (3.2 mg/L). In present study increased concentration of Zinc may be due to raw pollutant from shrimp and fish industry (aquaculture farm, hatchery), effluents from house hold local market, battery and pharmaceuticals industry directly discharged into the river. Lubricating oil which containing Zn also used as source of Zinc in this estuarine river. The mean value of Cu found 2.6 mg/L (Figure 3) present study was higher than the recorded value of Zinat *et al.* (2023) at Pasur River (0.021 mg/L) and Uddin *et al.* (2019) at Karnaphuli River (0.0189 mg/L). This metal entered into coastal water body through $CuSO_4$ used in metal plating and fishing operations used as algaecide. Copper also used as fertilizer and fungicidal spray. Domestic and aquaculture discharges, sedimentation also used as source of copper in this estuarine river.

Conclusion

Bakhkhali river estuary was not entirely polluted in terms of all metal concentrations. But during level of metal concentrations in polluted sites exceeded the safe limit of WHO which indicated that water of this estuarine river is not safe for drinking and supposed high risk to ecosystem. In Bakkhali river heavy metals originated mainly from domestic and industrial discharges, gas production plant, agriculture and shrimp farming. The growing threat from resistant organisms, caused by increased heavy metals pollution into aquatic environment like the Bakkhali river estuary, should alert scientists and political authorities of the need for concerted action in order to prevent the emergence of new resistant strains. Therefore, the present study recommends that point sources of heavy metals in the surface water of the Bakkhali River water should be strictly monitored to reduce metal contamination of water and improved the health of the riverine ecosystem.

Competing interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Authors contribution

Conceptualization, M.A.S.J. and S.J..; Methodology, S.J. and M.A.S.J.; Software, S.J..; Validation, J.A. and S.J..; Formal analysis, S.J. and J.A..; Investigation, M.A.S.J..; Data curation, S.J..; Writing—original draft preparation, S.J. and M.A.S.J.; Writing—review and editing, S.J. and S.A.; Supervision, M.A.S.J. All authors have read and agreed to the published version of the manuscript.

Conflicts of interest: The authors declare no conflict of interest.

Ethical approval: Not applicable.

Data availability: The data that support the findings of this study are available on request from the corresponding author.

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