RESEARCH ARTICLE

Hydrological distribution of physicochemical parameters and heavy metals in surface water and their ecotoxicological implications in the Bay of Bengal coast of Bangladesh

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Abstract

Accumulation of heavy metals in the coastal ecosystem has become a prodigious problem in any developing countries like Bangladesh. The impact of human activities on some physicochemical parameters and heavy metals was studied in surface water of the Bengal coast, Bangladesh. For ease of description, the fourteen study stations were categorized into four regions of the coastal sites of Bangladesh to determine physicochemical parameters and nine heavy metals like chromium (Cr), nickel (Ni), copper (Cu), arsenic (As), cadmium (Cd), lead (Pb), iron (Fe), manganese (Mn), and zinc (Zn) in water samples. The mean concentrations for water physico-chemistry are temperature (27.7±1.2 °C), pH (7.4±0.27), electrical conductivity (EC) (41.8±6.6 mS/cm), dissolved oxygen (DO) (6.7 \pm 0.69 mg/L), turbidity (58.5 \pm 12.0 NTU), fluoride (1.4 \pm 1.2 mg/L), chloride (126 \pm 66.3 mg/ L), sulfate (120±90.5 mg/L), nitrate (4.7±2.5 mg/L), and phosphate (4.7±2.5 mg/L). While the mean concentrations of Cr, Ni, Cu, As, Cd, Pb, Fe, Mn, and Zn were 150±58.3, 40.2±10.1, 186±114, 77.3±31.3, 32.7±20.7, 66.7±32.5, 871±268, 178±41.4, and 222 ±100 μg/L, respectively. As a whole, average concentration of studied metals in surface water followed the decreasing order of $Fe > Zn > Cu > Mn > Cr > Pb > As > Ni > Cd$. Heavy metals in water samples were much higher than the water quality guidelines for freshwater quality criteria for protection of aquatic life and drinking, indicated that the water of the study areas may create health hazard. The outcomes of the contamination factor (CF), pollution load index (PLI), Nemerow's pollution index (NPI), degree of contamination (C_d) , and modified degree of contamination (mC_d) varied spatially and most of the water samples were moderately to heavily polluted.

Keywords Water quality; Heavy metals . Bengal coast . Contamination factor . Pollution index

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Introduction

Waters are the essential requisite for all life forms on the earth (Bytyci et al. [2018\)](#page-12-0) and are also known as the most important irreplaceable natural resource on which the socio-economic growth and sustainable development of a country depends to a large extent (Pobi et al. [2019](#page-13-0); Proshad et al. [2020\)](#page-13-0). However, despite its importance to the human being, water is the most poorly managed resource in the world especially in developing countries and is facing a serious threat as a result of the wide array of anthropogenic activities (Islam et al. [2014;](#page-12-0) Ustaoğlu and Islam [2020](#page-13-0)). Deterioration of the world marine ecosystems due to the introduction of different types of pollutants like heavy metals and nutrient enrichments from excessive anthropogenic activities such as marine dumping, dredging, industrial effluents and incessant municipal, agricultural waste, domestic discharges, and atmospheric depositions

has been a worldwide concern in recent years (Raknuzzaman et al. [2016;](#page-13-0) Zaher et al. [2017](#page-14-0); Bristy et al. 2021). These activities and wastes with toxic contaminants into surface waters have negative effect on human health. In addition, bacteria, viral, and parasitic diseases like typhoid, cholera, encephalitis, hepatitis, skin infection, hair loss, liver cirrhosis, renal failure, and neural disorder are spreading through dermal and oral ingestion of metals contaminated water (Chowdhury et al. [2015;](#page-12-0) Islam [2021\)](#page-12-0). According to World Health Organization (WHO) about 80% of disease are water borne, hence, surface water is a major source of infection to marine organisms and human.

Among the pollutants, heavy metals are considered as one of the most serious environmental pollutants because of their high toxicity, abundance, and ease of accumulation by various organisms (Swarnalatha et al. [2013;](#page-13-0) Islam [2021](#page-12-0)). The study of heavy metals is crucial due to their nonbiodegradable nature, accumulative properties, and long biological half-lives. Once they enter into the water body, it is difficult to remove them entirely. Therefore, heavy metals persist in the aquatic environment; gradually move through the lower food chain to upper trophic levels and bioaccumulate in the higher organisms (Aderinola et al. [2009](#page-12-0)), which can eventually lead to adverse health effects on humans due to the consumption of contaminated seafood (Wang [2002](#page-14-0)). Heavy metal contamination can affect the physical, chemical, and biological characteristics of water and its ecosystem. Therefore, physicochemical water analysis is valuable in assessing water quality and composition (Nubi et al. [2019\)](#page-13-0). With the increasing use of wide variety of metals in electronics and petrochemicals industries in Bangladesh way of disposing waste indiscriminately, there is little awareness of detrimental health effect of heavy metal pollution in the coastal environment. Researchers therefore have started focusing in assessing the hydro geochemical distribution of heavy metals in the surface water with their human and ecological risks (Wang et al. [2018](#page-14-0); Qu et al. [2018;](#page-13-0) Saleem et al. [2019](#page-13-0); Proshad et al. [2020\)](#page-13-0).

Bangladesh is a riverine country. According to Bangladesh Water development board about 230 rivers currently flow in Bangladesh with 580 km coastline of the Bay of Bengal. The Ganges is a trans-boundary river of Asia which flows through India and Bangladesh. The 2525 km length of different river rises in the western Himalayas of Uttarakhand, India, and flows south and east through the Gangetic Plain of North India into the Bay of Bengal of Bangladesh (Al-Mamun et al. [2019;](#page-12-0) Bristi et al. [2021](#page-12-0)). The Yamuna River is the second-largest tributary river of the Ganga and the longest tributary in India and flows into Bangladesh. These two rivers have been identified as the major recipients of industrial effluents through Padma and Jamuna Rivers forming the Meghna estuary (Raknuzzaman et al. [2016](#page-13-0)). In Bangladesh, Meghna estuary is the most biogeochemically active zones and is more easily affected by anthropogenic nutrient loading

and pollutants from urban expansion, untreated effluents from widespread ship breaking and port activities, excessive operation of mechanized boats, deforestation, and intensive agriculture, and aquaculture activities (Smith et al. [2012;](#page-13-0) Islam [2021;](#page-12-0) Bristi et al. [2021\)](#page-12-0). The waste materials coming through these connecting tributary rivers with the Bay of Bengal are also accelerating the coastal water pollution that might contaminate the fish, crustaceans and others aquatic biota. The concerned authorities, local government, and general people of Bangladesh have not been aware about the present worse situation and no detailed study has been carried out so far in the coastal area of Bangladesh. Therefore, the objectives of the present study are to investigate the impact of human activities on some physicochemical parameters (e.g., temperature, pH, dissolve oxygen, conductivity, and turbidity), nutrient (e.g., nitrate, phosphate, sulfate, fluoride, and chloride), and heavy metals (Cr, Ni, Cu, As, Cd, Pb, Fe, Mn, and Zn) distributions in the surface water of the coastal sea and estuarine areas using water pollution index such as contamination factor (CF), pollution load index (PLI), Nemerow's pollution index (NPI), degree of contamination (C_d) , and modified degree of contamination (mC_d) in order to evaluate ecotoxicological effect on marine organisms and teeming population in the coastal areas of Bangladesh.

Materials and method

Description of the study area

In order to explore the influence of the potential sources of heavy metals, we have investigated fourteen sampling sites from four coastal areas in the southeast and southwest part of the Bay of Bengal coast of Bangladesh. These sampling sites located in Cox's Bazar, Chittagong, Meghna Estuary, and Sundarbans are shown in Fig. [1.](#page-2-0) The first site, Cox's Bazaar (Site 1), is a seaside tourist town with an unbroken 125 km world's longest natural sandy sea beach. Considering two diverse ecological aspects, it was divided into two sub sites: hatchery area (CX1–CX2) and Bakkhali estuary (CX3–CX4). More than 53 shrimp hatcheries and aqua farms, fish landing centers, huge hotels for the amusement of tourists, and some industries are located in the hatchery site. Likewise, the Bakkhali estuary, regarded as an important economic part of Cox's Bazar district with a harbor and imperative local fishery, is about 0.5 km wide and >10 m deep at its mid-point and directly influenced by semi-diurnal tides. Moreover, Maheshkhali Island is one of the important tourists attracting economic zone incorporating to Bakkhali estuary through 9 to 11 kilometer long Maheshkhali channel to Cox's Bazaar central island. Consequently, these sampling areas are mostly influenced by the municipal sewage and

Fig. 1. Study area map of different locations of coastal sea and estuarine area in Bangladesh

industrial wastage discharged from these unplanned industries, hatcheries, and aquaculture farms (Al-Mamun et al. [2019\)](#page-12-0).

The second site, Chittagong (Site 2), is located near the Chittagong port (CT1–CT2) and ship breaking area (CT3– CT4). This is the southeastern principal seaport region of the country, where a significant ecological change is pronounced due to huge discharges of untreated or semi treated domestic and municipal sewage as well as effluents from multifarious industries (namely, paper and pulp, tanneries, textile, chemicals, pharmaceuticals, rubber and plastic, oil refinery, steal rerolling, leather, jute, tobacco, fish processing plants, paint, coal-based thermal power plants, fertilizer, rechargeable batteries, jewelry, plating, automobile engine, and electronics industries, etc.,) as well as contaminated mud disposal from harbor dredging. Besides, Chittagong ship breaking yard is world's second largest ship breaking area confined to 18 km² areas along the coast of Sitakunda Upazilla, particularly

Bhatiary to Kumira in Chittagong division. While dismantling the ships, the industry generates huge organic and inorganic wastes which is discharged into the nearby area and thus polluting the coastal water and sediment.

The third site (ME1–ME3), Meghna Estuary (Site 3), is an estuarine area where the main rivers mix together to the Bay of Bengal. This site is influenced mainly by the domestic and industrial effluents carried by the inland rivers from the country and trans–boundary countries. The fourth site, Sundarbans (Site 4), is located near the southwest part of the coastal area which is regarded as a large mangrove ecosystem in Bangladesh. This sampling area (SN1–SN3) is also mostly influenced by different anthropogenic and industrial activities like cement factories, export processing zone, sea port, paper industries, oil refinery industries, steal rerolling, fertilizer industry, hatcheries and aqua farms, fish processing industries, leather industries, dyeing industries, paint industries in Khulna and Mongla area. An elaborative description of the study area

is given in the Supplementary information (Table S1). All the industrial activities were observed during the sampling campaign of the present study.

Sample collection, preparation, and quality assurance

A total of 42 composite surface water samples were collected during the transition of summer and rainy season starting from early September to end of November in 2020. The reasonable and adequate ground for the selection of seasons were to estimate the fluctuation of heavy metals in surface water in relation to the variation of the physicochemical properties, as the industrial and agricultural activities like fish processing, hatcheries, crop cultivation, and ship dismantling activities are vigorously increased in the sampling sites during this particular period. The tide in the coastal and estuarine area was semidiurnal and the samples were collected during the period of low tide. Unfiltered surface water samples were collected from fourteen coastal sampling stations. Immediately after collection, the samples were transferred into 100 mL polypropylene bottles which were previously washed with dilute nitric acid and deionized water. Then the samples were stored in refrigerator in the Department of Soil Science, Patuakhali Science and Technology University, Dumki, Patuakhali, Bangladesh.

Water samples were filtered (0.45 μm filters, cellulose nitrate, Millipore) into poly propylene tubes using a plastic syringe (BD Plastipak, 50 mL) for dissolved heavy metals concentrations. Samples were acidified to 0.24 M with $HNO₃$ (65% supra pure, Merck) and kept at 4 °C in the dark until analysis. The concentrations of As, Cd, Pb, Cr, Ni, Cu, Fe, Mn, and Zn in surface water samples were measured using inductively coupled plasma-mass spectrometry (ICP-MS, Agilent, 7700). The detection limits for Cr, Ni, Cu, As, Cd, Pb, Fe, Mn, and Zn by ICP-MS analysis were 0.2, 0.05, 0.1, 0.05, 0.02, 0.02, 0.1, 0.02, and 0.3 ng/mL, respectively. The calibration curves with $R^2 > 0.999$ were accepted for concentration calculation. Multielement solution (Agilent Technologies, USA) 1.0 μg/L was used as tuning solution covering a wide range of masses of elements. All test batches were evaluated using an internal quality approach and validated if they satisfied the defined Internal Quality Controls (IQCs). Quality assurance and quality control for water samples were carried out using triplicates, method blanks, and standard reference material (STD TMDA-70.2). The percent recovery for all heavy metals ranged from 99.1 to 101.9% (Table S2). Comparison is made with the certified values, which in both cases confirmed that the sample preparation and instrumentation conditions provided good levels of accuracy and precision.

Physicochemical parameters analysis

Analysis of common inorganic anions like fluoride, chloride, and sulfate was done using Ion Chromatography (IC). The physicochemical parameters such as pH, conductivity, dissolved oxygen (DO), turbidity, and temperature of water samples were measured immediately after sample collection by multi water quality checker (U-50, Horiba, Ltd., Kyoto, Japan). Water samples collected for dissolved nutrients were analyzed for Nitrate and Phosphate. Nitrates were determined using ammonium chloride. Phosphates were determined using the molybdenum-blue method. All the dissolved inorganic nutrient analyzes followed the methods of Parsons et al. [\(1984\)](#page-13-0) using UV/VIS spectrophotometer at 410 and 820 nm for nitrate and phosphate, respectively. The common laboratory apparatus which were used during the study include; different sized beakers flasks, funnels, volumetric flasks, block digester, fume hood, droppers, glass pipettes, spatula, measuring cylinders, vinyl gloves, analytical balance, and conical flasks.

Water pollution index

Most studies about the water quality assessment use a water quality index (Popoola et al. [2015;](#page-13-0) Yingran et al. [2017;](#page-14-0) Tanjung et al. [2019\)](#page-13-0). The water pollution index is often used to understand and evaluate the level of water pollution in the coastal areas. The pollution index results can give an advice to the decision makers in order to know the quality of waters and make policy that would prohibit the deterioration of water quality if there is a quality reduction due to the existence of pollutants (Tanjung et al. [2019\)](#page-13-0). A coupling of specific and combined approaches was used to appraise of heavy metal pollution level. Contamination factor (CF: Hakanson [1980\)](#page-12-0), and Nemerow's pollution index (NPI) were adopted for the specific appraisal of heavy metal pollution (Dan et al. [2014\)](#page-12-0), whereas the degree of contamination $(C_d:$ Hakanson [1980\)](#page-12-0), modified degree of contamination (mC $_d$: Abrahim and Parker [2008](#page-12-0)) index, pollution load index (PLI: Tomlinson et al. [1980\)](#page-13-0). All The mathematical equations, brief description and physical significances of these indices are shown in Table S3.

Statistical analyzes

The data produced from this study were statistically analyzed using the statistical package, SPSS 16.0 (SPSS, USA). The means and standard deviations and inferential statistics (oneway ANOVA and correlation) of heavy metal concentrations in surface water samples were calculated. The data were compared by one- way ANOVA to determine if the difference was significant using 95% confidence interval ($P-value < 0.05$). Correlation matrix analysis was used to identify possible significant relationships among physicochemistry, nutrients, and heavy metals in surface water at both 0.01 and 0.05 significant levels. Multivariate post hoc Tukey test was performed to detect significant differences in concentration of physicochemical parameters and risk index for different sampling sites. Other calculations were performed by Microsoft Excel 2010. A multivariate method in terms of principal component analysis (PCA) was used to obtain the detailed information of the data-set and gain insight into the distribution of heavy metals by detecting similarities or differences in samples. The PCA was performed using Varimax normalized rotation on the data-set.

Results and discussions

Physicochemical parameters distribution within water column

The physicochemical parameters of surface water samples from coastal sea and estuaries in Bangladesh are shown in Table 1. A wide range of physicochemical properties in water samples were observed among the sampling sites. Factors such as salinity, DO, pH, Temp., geomorphological setup, and terrestrial runoff might have played a role in the variation of physicochemical parameters. Water temperature is important because it increases/decreases the rate of chemical reactions as well as solubility of different gasses most especially oxygen (Ali et al. [2013\)](#page-12-0). Water temperature for Cox's Bazar Coast ranged between 26.3 and 29.9 °C with a mean of 28.0 ±1.1 °C, for Chittagong Coast ranged between 26.9 and 29.6

°C with a mean of 28.4±0.75 °C, for Meghna Estuary ranged between 25.1 and 28.7 °C with a mean of 27.0±1.2 °C and for Sundarbans ranged between 25.5 and 29.71 °C with a mean of 27.1 ± 1.3 °C, respectively. Temperature values for surface were below 35 °C standard given by FEPA ([1991](#page-12-0)). The minimal temperature values recorded across the stations could be attributed to the cloudy sky that was observed during sampling. The pH is a measure of the concentration of hydrogen ion and a very critical environmental indicator which can affect the composition of the organisms in the aquatic environment and the availability of nutrients as well as the relative toxicity of certain heavy metals. The mean \pm SD value of pH in surface water was 7.6±0.39, 7.2±0.07, 7.4±0.05, and 7.5 ±0.05 at Cox's Bazar Coast, Chittagong Coast, Meghna Estuary, and Sundarbans, respectively which is slightly alkaline. The pH values obtained are typical of the brackish water tidal climate (Fabunmi et al. [2015\)](#page-12-0). Generally the alkaline nature of the study area could be due to the presence of carbonates and bicarbonates in the water bodies (Murhekar [2011\)](#page-13-0). Higher value of alkalinity recorded at harbor area could be attributed to the buffering effect of the sea and it is an indicator of environmental condition closeness to the sea in agreement with the study of Onyema and Popoola [\(2013\)](#page-13-0). The pH values recorded across the sample stations were within the permissible limit (6.5–8.5) set by WHO ([1993](#page-14-0)). The pH values were well within the susceptible range for optimal aquatic productivity (6.5–9.0) and livable range of 5.5–10.0 (Wetzel [2001\)](#page-14-0).

Electrical conductivity (EC) is a measure of water capacity to convey electric current through the water and it signifies the

Table 1 Physicochemical parameters distribution within water column at different locations of the coastal sea and estuarine area in Bangladesh and comparison with the standard limit $(n=3)$

Sampling sites		Temperature $(^{\circ}C)$	pH	EC (mS/cm)	Dissolved oxygen (DO) (mg/ L)	Turbidity (NTU)	FI^{-} (mg/L)	Cl^2 (mg/L)	SO_4^2 (mg/L)	NO ₃ (mg/L)	PO ₄ ³ (mg/L)
Cox's Bazar Coast	$Mean \pm SD$	28.0 ± 1.1^a	7.6 $\pm 0.39^{\mathrm{a}}$		44.2 ± 5.6^a 6.8 $\pm 0.71^a$	56.8 ± 10.9^a 1.6 ± 1.2^a		$168 \pm 63.5^{\circ}$	$176 \pm 108^{\rm a}$	$5.1 \pm 2.4^{\text{a}}$	3.4 ± 0.96^a
	Range	26.3-29.9	6.9-7.9	30.8-50.7	$5.2 - 7.7$	$45.1 - 76.2$	$0.18 - 3.4$	50.5-255	46.3-351	$1.9 - 8.9$	$1.9 - 5.0$
Chittagong Coast	$Mean \pm SD$	$28.4 \pm 0.75^{\text{a}}$	7.2 $\pm 0.07^{\rm a}$	$42.0 \pm 5.5^{\circ}$	6.8 ± 0.60^a	$57.6 \pm 13.8^{\rm a}$	$1.3 \pm 1.3^{\rm a}$	$98.9 \pm 52.7^{\rm b}$	$77.2 \pm 43.1^{\rm b}$	$6.5 \pm 2.4^{\text{a}}$	$3.9 \pm 1.3^{\rm a}$
	Range	26.9-29.6	$7.0 - 7.3$	34.7-48.9	5.5-7.8	40.1-72.5	$0.11 - 3.3$	39.9-195	38.5-138	$2.5 - 9.2$	$2.0 - 5.5$
Meghna Estuary	Mean±SD	$27.0 \pm 1.2^{\rm a}$	7.4 $\pm 0.05^{\mathrm{a}}$		$43.6 \pm 5.8^{\text{a}}$ 6.4 $\pm 0.84^{\text{a}}$	54.1 \pm 9.9 ^a	1.8 ± 1.1^a	$154 \pm 54.9^{\rm a}$	$161 \pm 83.7^{\rm a}$	2.9 ± 1.3^{b}	2.6 ± 1.0^{b}
	Range	25.1-28.7	$7.3 - 7.4$	33.1-51.8 5.2-7.6		42.9-71.6	$0.22 - 3.5$	66.8-216	42.3-285	$1.2 - 4.8$	$1.2 - 4.1$
Sundarbans	Mean±SD	$27.1 \pm 1.3^{\text{a}}$	7.5 $\pm 0.05^{\mathrm{a}}$	36.4 ± 7.7^b	$6.6 \pm 0.63^{\text{a}}$	$66.2 \pm 10.8^{\rm b}$	0.6 ± 0.9^b	79.3 ± 55.2^b	$56.9 \pm 42.0^{\rm b}$	3.7 ± 1.8^{b}	2.6 ± 0.54^b
	Range	25.5-29.1	$7.5 - 7.6$	25.0-49.5	5.4-7.6	41.8-78.5	$0.11 - 2.9$	34.2-198	26.9-136	$1.3 - 5.6$	$2.1 - 3.4$
Standard limit		35 (FEPA) 1991)	$6.5 - 8.5$ (WHO) 1993)	1.0 (WHO) 1993)	5.0 (FEPA 1991)	10 (FEPA) 1991)	1.5 (WHO) 2007)	200 (WHO 2007)	400 (WHO) 2007	20 (FEPA 1991)	5.0 (FEPA 1991)

Note: In a column, different letters a and b indicates statistically significant difference among the station $(p<0.05)$

NTU (Nephelometric turbidity units)

amount of total dissolved salts (APHA [2005](#page-12-0)). The EC values at the surface water varied from 30.8 to 50.7 mS/cm (44.2±5.6 mS/cm) at Cox's Bazar Coast, 34.7 to 48.9 mS/cm (42.0±5.5 mS/cm) at Chittagong Coast, 33.1 to 51.8 mS/cm (43.6±5.8 mS/cm) at Meghna Estuary, and 25.0 to 46.5 mS/cm (36.4 \pm 7.6 mS/cm) at Sundarbans, respectively (Table [1](#page-4-0)). The value of EC in all the sampling stations were above the maximum limit (1.0 mS/cm) given by WHO [\(1993\)](#page-14-0). High value of EC in the study area could be attributed to the intrusion of seawater and stormy suspension of sediment particle by the ocean and others from effluents of conducting species such as chloride, phosphate, and heavy metals present in the sampling stations (Okorafor et al. [2013\)](#page-13-0). EC showed a significant $(p<0.01)$ positive correlation with fluoride ($r = 0.864$), chloride ($r = 0.888$), and sulfate $(r = 0.767)$ and negatively correlated with turbidity $(r = -0.861)$ (Table [4\)](#page-8-0). The value of EC showed a significant difference $(p<0.05)$ for Sundarbans with the other three sampling sites.

Dissolved oxygen (DO) is an important parameter in the aquatic system since it is virtually required by all organisms for their metabolism. Chapman [\(1996\)](#page-12-0) stated that DO levels below 5.0 mg/L adversely affect the aquatic life while concentration below 2.0 mg/L may lead to death for most fishes. The mean \pm SD value of DO in surface water was 6.8 \pm 0.71, 6.8±0.60, 6.4±0.84, and 6.6±0.63 mg/L at Cox's Bazar Coast, Chittagong Coast, Meghna Estuary, and Sundarbans, respectively (Table [1\)](#page-4-0). The level of DO in water samples were higher than the permissible limit (\geq 5.0 mg/L) recommended by Chapman ([1996\)](#page-12-0) and FEPA ([1991\)](#page-12-0) throughout the sample stations which could be as a result of influx of fresh water from the Bakkhali estuary, Ganges, Yamuna, Padma, Jamuna, and Karnaphuli Rivers as well as precipitation to the sea and estuaries of the Bengal coast of Bangladesh. The inflow of fresh water and sea water makes the sea a more productive natural habitat, resulting in high of nutrients (DO inclusive) in the water column. Turbidity is an important parameter for maintaining water quality that can indicate the presence of dispersed suspended solids, organic materials, algae, microorganisms and many more. The mean \pm SD value of turbidity at the surface water was 56.8 ± 10.9 , 57.6 ± 13.8 , 54.1 ±9.9, and 66.2±10.8 NTU for Cox's Bazar Coast, Chittagong Coast, Meghna Estuary, and Sundarbans, respectively (Table [1\)](#page-4-0). Turbidity values recorded from the sample stations were above the permissible limit (10 NTU) given FEPA [\(1991\)](#page-12-0) which could be attributed to the presence of suspended matter such as silts, clay, finely divided inorganic, and organic matter from heavy dredging activities, residential discharges, and disposal of waste from hatchery and port area. Turbidity showed significant $(p<0.01)$ negative correlation with fluoride $(r = -0.803)$, chloride $(r = -0.807)$ and sulfate $(r = -0.666)$.

Nitrate is known to be the most stable and predominant inorganic form of nitrogen in seawater which are produced naturally as part of the nitrogen cycle. The mean \pm SD value

of nitrate (NO_3^-) in this investigation was 5.1 ± 2.4 , 6.5 ± 2.4 , 2.9±1.3, and 3.7±1.8 mg/L for Cox's Bazar Coast, Chittagong Coast, Meghna Estuary, and Sundarbans, respectively (Table [1\)](#page-4-0). Generally, the concentrations of nitrate at all the sample stations were within the permissible limit set by FEPA (20 mg/L) and could be due to utilization by phytoplankton or to the neritic water dominance, which contained negligible amount of nitrate (Govindasamy et al. [2000;](#page-12-0) Ramalingam et al. [2011](#page-13-0)). Phosphates are essential for the growth of organisms so they could be considered as a limiting nutrient for the growth of phytoplankton. The cycling of phosphorus within the aquatic water body is dynamic and complex; involving adsorption and precipitation reactions, interchange with sediments and uptake by aquatic biota (Mohamed [2005](#page-13-0)). The mean \pm SD value of phosphate (PO₄³⁻) in this investigation was 3.4±0.96, 3.9±1.3, 2.6±1.0, and 2.6±0.54 mg/L for Cox's Bazar Coast, Chittagong Coast, Meghna Estuary, and Sundarbans, respectively (Table [1\)](#page-4-0). Phosphate levels in the study area are likely to result from rock weathering and sand dredging activities that release soluble alkali phosphate together with inputs from domestic sewage and industrial effluents (Balogun and Ajani [2015](#page-12-0)). Phosphates values are within the maximum limit (5 mg/L) prescribe by FEPA ([1991](#page-12-0)) in all the sample stations.

Distribution of heavy metals in waters of the Bengal coast

The distribution of heavy metal concentrations in the Bay of Bengal coast are presented in Table [2](#page-6-0). Average concentration of heavy metals in waters followed the decreasing order of Fe $> Zn > Cu > Mn > Cr > Pb > As > Ni > Cd$. Considering Cr concentration in surface water, elevated level of Cr was observed CT3 site $(257\pm35.5 \,\mu g/L)$ of Chittagong coast followed by CT4 site $(234\pm19.0 \text{ }\mu\text{g/L})$ of the Chittagong coast. During our sampling period we observed huge amount of Cr was used in stainless steel for polished silvery mirror coating, chrome plating, metal ceramics, shiny finish in dyes and paints, and leather tanning in the ship breaking area of the Chittagong coast. In the natural water systems virtually all chromium ore is processed via hexavalent chromium, specifically the salt sodium dichromate which is toxic and in higher concentration to be carcinogenic (ATSDR [1998;](#page-12-0) Ustaoğlu and Tepe [2019](#page-13-0); Ustaoğlu [2021](#page-13-0); Tokatlı and Ustaoğlu [2020;](#page-13-0) Islam [2021\)](#page-12-0). The mean concentration of Cr in waters of the Bay of Bengal coast was far higher than the drinking water quality criteria and freshwater quality criteria for the protection of aquatic life (USEPA 2020 a,b; Ustaoğlu et al. [2020a](#page-14-0), [b](#page-14-0)) (Table [3\)](#page-6-0). Nickel has been reported to be associated with the effluents from electroplating, breakdown of old batteries, land disposals, sewage sludge, paint and dyes, effluents from crude oil transportations, fertilizers application, and vehicular emissions (Popoola et al. [2015;](#page-13-0) Tokatlı [2017](#page-13-0), [2019](#page-13-0), [2021](#page-13-0); Aydın

Table 2 Heavy metals concentration (μg/L) in water sample collected from the coastal sea and estuarine areas of Bangladesh (mean±SD, from each station, $n=3$)

Sampling sites	site	Cr	Ni	Cu	As	Cd	Pb	Fe	Mn	Zn
Cox's Bazar Coast	CX1	132 ± 20.7	35.4 ± 1.6	337 ± 8.3	73.0 ± 20.1	41.8 ± 5.8	89.7 ± 15.8	694 ± 48.5	187 ± 15.5	353 ± 6.1
	CX ₂	132 ± 10.4	49.9 ± 3.1	358 ± 31.2	92.5 ± 28.4	45.0 ± 9.0	104 ± 6.6	823 ± 77.6	175 ± 14.0	256 ± 7.4
	CX3	128 ± 9.6	28.2 ± 12.1	159 ± 25.9	61.5 ± 21.9	39.3 ± 5.1	18.8 ± 6.1	820 ± 236	226 ± 10.6	174 ± 32.9
	CX4	130 ± 2.1	42.3 ± 2.4	143 ± 21.0	69.4 ± 13.3	34.7 ± 7.5	21.5 ± 4.3	568 ± 44.0	195 ± 27.5	139 ± 20.0
Chittagong Coast	CT1	219 ± 15.6	39.0 ± 1.1	257 ± 28.0	76.7 ± 19.3	31.3 ± 21.2	80.8 ± 15.8	1084 ± 75.8	195 ± 64.6	169 ± 56.9
	CT ₂	224 ± 15.3	62.2 ± 6.0	300 ± 46.9	116 ± 4.7	31.3 ± 23.1	94.7 ± 7.4	1194 ± 63.6	177 ± 15.5	166±49.1
	CT ₃	257 ± 35.5	50.8 ± 5.0	280 ± 27.0	119 ± 11.3	63.8 ± 7.5	109 ± 10.4	1189 ± 123	185 ± 14.4	380±34.0
	CT4	234 ± 19.0	45.0 ± 5.6	317 ± 31.7	138 ± 5.6	70.7 ± 11.1	114 ± 9.2	1328±283	259 ± 25.1	443 ± 8.5
Meghna Estuary	ME1	83.0 ± 1.0	33.9 ± 4.4	101 ± 7.9	55.1 ± 3.6	17.0 ± 10.7	50.4 ± 19.0	695 ± 145	141 ± 22.1	143 ± 4.0
	ME ₂	109 ± 27.7	35.4 ± 2.2	68.2 ± 0.75	45.5 ± 18.4	8.2 ± 0.89	37.9 ± 19.1	566 ± 21.7	162 ± 5.7	166 ± 45.7
	ME3	133 ± 10.4	38.6 ± 1.0	53.5 ± 4.1	33.2 ± 6.2	4.9 ± 3.7	$52.7 + 4.7$	520 ± 71.2	142 ± 20.6	160 ± 28.7
Sundarbans	SN ₁	82.0 ± 3.6	28.9 ± 4.4	71.8 ± 20.5	62.4 ± 12.7	15.1 ± 1.6	43.1 ± 12.4	867 ± 33.6	116 ± 16.1	159±12.4
	SN2	104 ± 9.0	33.5 ± 3.7	97.3 ± 12.8	$66.8{\pm}4.7$	19.9 ± 1.3	57.0 ± 5.5	910 ± 46.5	141 ± 4.0	151 ± 18.8
	SN ₃	136 ± 19.3	33.9 ± 9.9	63.6 ± 20.5	74.1 ± 17.1	34.3 ± 8.4	61.3 ± 7.5	943 ± 64.9	188 ± 24.0	245 ± 2.1

et al. [2021\)](#page-12-0). At the Cox's Bazar Coast, the mean±SD concentration of Ni was observed as 35.4±1.6, 49.9±3.1, 28.2±12.1, and 42.3 ± 2.4 μ g/L for CX1, CX2, CX3, and CX4, respectively; in the Chittagong Coast, Ni was observed as 39.0±1.1, 62.2±6.0, 50.8±5.0, and 45.0±5.6 μg/L for CT1, CT2, CT3, and CT4, respectively; in the Meghna Estuary,

Ni was observed as 33.9±4.4, 35.4±2.2 and 38.6±1.0 μg/L for ME1, ME2, and ME3, respectively and in the Sundarbans, Ni was observed as 28.9±4.4, 33.5±3.7, and 33.9±9.9 μg/L for SN1, SN2, and SN3, respectively (Table [2\)](#page-6-0). Nickel values at all the sampling sites in water samples are below the maximum limit of 470 μg/L (acute) and 52 μg/L (chronic) as pre-scribed by (USEPA [2020a,](#page-13-0) [b](#page-13-0)) (Table [3](#page-6-0)). In water samples, Ni showed a significant positive correlation with other metal except Zn and Mn (Table [4\)](#page-8-0).

Copper is a very common element that occurs naturally but Cu in the aqueous systems received attention mostly because of its toxic effects on biota. In water, even at very low concentrations, Cu can be toxic. It is known to cause brain damage in mammals (Fatoki et al. [2002;](#page-12-0) Ustaoğlu and Aydın [2020\)](#page-13-0). The descending order of mean Cu concentration $(\mu g/L)$ in the sampling sites were of CX2 (358) > CX1 $(337) > CT4$ $(317) > CT2$ $(300) > CT3$ $(280) > CT1$ $(257) >$ $CX3 (159) > CX4 (143) > ME1 (101) > SN2 (97.3) > SN1$ $(71.8) > ME2 (68.2) > SN3 (63.6) > ME3 (53.5)$. Among the four sampling sites, elevated level of Cu was observed at the Cox's Bazar Coast and the Chittagong Coast which might be due to the effects from hatchery operation, electrical, and metal processing, antifouling paints in the marine environment, used as coatings for ships, buoys, underwater, and decking pollutants (Raknuzzaman et al. [2016\)](#page-13-0). During the sampling campaign, we observed that the local peoples are commonly used form as copper sulfate, which is available either as a crystal or a powder often known as "Bluestone" or "Blue powder" (Watson and Yanong [1989;](#page-14-0) Shamsuzzaman and Biswas [2012](#page-13-0)).

Arsenic is a ubiquitous element in the environment and is mobilized through a combination of natural processes such as weathering reactions, biological activity, and volcanic emissions. In the natural environment, As can occur in several oxidation states $(-3, 0, +3, \text{ and } +5)$ but in the natural water systems it is mostly found in inorganic form as oxyanions of trivalent arsenite [As(III)) or pentavalent arsenate (As(V)]. Among fourteen sampling points of four coastal sites, elevated levels of As was observed at three stations of Chittagong sea port area like CT2 (116±4.7 μg/L), CT3 (119±11.3 μg/L), and CT4 (138±5.6 μg/L), respectively). During sampling campaign, we observed the movement ships for loading and unloading of containers, huge application of arsenical pesticides for crop production besides the Chittagong sea port connecting Karnaphuli River (Shao et al. [2016](#page-13-0)), used chromated copper arsenate (CCA) for wood treatments to protect from rotting due to insects and microbial agents (Pravin et al. [2012;](#page-13-0) Islam [2021\)](#page-12-0) and arsenic (III) sulfide to make infrared-transmitting glass, pigments, electrical semiconductors, and tanning agents (Bhuiyan et al. [2011](#page-12-0); Aydın et al. [2021\)](#page-12-0) at the Chittagong sea port area. The concentration of As in water samples of the Bengal coast was higher than the freshwater and drinking water quality criteria (USEPA 2020

a,b) (Table [3](#page-6-0)) indicating As in water may create ecological and health risks to the entire coastal ecosystems of the Bay of Bengal.

Cadmium (Cd) is used in phosphate fertilizers, in PVC products, nickel-cadmium rechargeable battery and pigment alloys which are not recycled but discarded along with household waste, and create deleterious effect on the aquatic organisms and human health (Ekere et al. [2014\)](#page-12-0). It has been established that chronic renal failure is associated with Cd exposure (Hellstrom et al. [2001](#page-12-0)). The descending order of Cd concentration $(\mu g/L)$ in the sampling sites were of CT4 (70.7) > CT3 (63.8) > CX2 (45.0) > CX1 (41.8) > CX3 (39.3) > CX4 (34.7) > SN3 (34.3) > CT1 (31.3) > CT2 (31.3) > SN2 $(19.9) > ME1 (17.0) > SN1 (15.1) > ME2 (8.2) > ME3 (4.9).$ Among the four sites, elevated level of Cd was observed at the Cox's Bazar Coast and the Chittagong Coast which might be due to the effects from different electroplating industries, ship repairing, and battery dumping. Besides, weathering of rocks, manures, municipal sewage sludge, fossil fuel combustion, artificial phosphate fertilizers, and pesticides as $CdSO₄$ also contribute significant amount of Cd in this area (Sheppard et al. [2009](#page-13-0); Raknuzzaman et al. [2016](#page-13-0)).

Lead is a nonessential component for plants and animals and it is often an accumulative poison for the living organisms. It has been a potentially hazardous to most forms of life (USEPA [1986\)](#page-13-0). However, in the water systems, Pb can be carried either dissolved or as waterborne particles where a few Pb compounds dissolve readily in water, though a significant amount is then precipitated and becomes incorporated into sediments at the base of the watercourse. Among the sampling sites, the highest mean±SD concentration of Pb $(114\pm9.2 \text{ µg/L})$ was observed in water samples of the Chittagong Coast (CT4 site) followed by CT3 site (109 ± 10.4 μg/L) (Table [2\)](#page-6-0). The high concentration of Pb in the Chittagong Coast site might be attributed to the acid drainage from the industrial wastes and different chemical activities of hatcheries, fish processing, paint, and ship repairing industries.

Iron, Mn, and Zn are the essential micronutrient for most living organisms and human beings. Among them Fe is a constituent of proteins and many enzymes, including hemoglobin and myoglobin. These essential elements are typically more common in the marine environment than other metals due to its high prevalence on Earth (Edokpayi et al. [2016\)](#page-12-0). The highest mean \pm SD concentration of Fe (1328 \pm 283 μg/L) was observed in water samples of the Chittagong Coast (CT4 site) followed by CT2 site $(1194\pm63.6 \text{ µg/L})$; for Mn was $(259$ ± 25.1 μg/L) at (CT4 site) followed by CX3 site (226 ± 10.6) μ g/L), and for Zn was (443±8.5 μ g/L) at (CT4 site) followed by CT3 site (380±34.0 μg/L) (Table [2\)](#page-6-0). During our sampling campaign, we observed huge tourist presence in this town with longest sandy sea beach, 53 fish hatcheries operation, a fish landing center, activities from ship and boat making

*Correlation is significant at the 0.05 level (2-tailed)

*Correlation is significant at the 0.05 level (2-tailed)

industries, paint industries, salt industries, fish processing, unplanned urbanization, and many hotels at the coast of Chittagong and Cox's Bazar. Therefore, elevated levels of the studied heavy metals were observed in these areas.

The observed concentrations of all studied heavy metals in surface waters from the coast and estuaries of the Bay of Bengal were much higher than the guideline values for fresh-water quality criteria (DoE [1997;](#page-12-0) EC [1998](#page-12-0); WHO [2011](#page-14-0); USEPA [2020a](#page-13-0), [b\)](#page-13-0) (Table [3\)](#page-6-0) indicated that the waters of the Bengal coast may create health hazard. Apart from freshwater quality criteria, the mean concentrations of heavy metals in water samples were much higher than the USEPA maximum contaminant level (MCL) and maximum contaminant level goal (MCLG) (Table [3](#page-6-0)), indicated that waters from the Bay of Bengal coast might pose potential threats to the surrounding coastal ecosystems.

Pollution assessment of heavy metal in surface water

Contamination factor (CF), pollution load index (PLI), Nemerow's pollution index (NPI), degree of contamination (C_d) , and modified degree of contamination (mC_d) of the study area is shown in Figs. 2 and [3.](#page-10-0) At the Cox's Bazar coast, the mean values of CF in waters were 0.90, 1.01, 4.42, 2.70, 9.35, 1.06, 1.32, 1.36, and 1.20 for Cr, Ni, Cu, As, Cd, Pb, Fe, Ma, and Zn, respectively. At the Chittagong coast, CF values were 1.61, 1.27, 5.10, 4.10, 11.5, 1.81, 2.17, 1.42, and 1.50 for Cr, Ni, Cu, As, Cd, Pb, Fe, Ma, and Zn, respectively. At the Meghna estuary, the CF values were 0.75, 0.93, 1.31, 1.63, 2.34, 0.85, 1.07, 1.03, and 0.81 for Cr, Ni, Cu, As, Cd, Pb, Fe, Ma, and Zn, respectively and at the Sundarbans area, the mean values of CF were 0.74, 0.88, 1.37, 2.47, 5.37, 0.98, 1.364, 1.03, and 0.96 for Cr, Ni, Cu, As, Cd, Pb, Fe, Ma, and Zn, respectively. Following the Hakanson ([1980\)](#page-12-0) classification scheme, the heavy metals ranged from low to very high contamination, with Cd showed very high contamination class for the coast of Chittagong and Cox's Bazar ($CF > 6.0$), while other elements recorded low to moderate contamination level (Fig. 2). High CF value for Cd for the coasts of Chittagong and Cox's Bazar could be due to heavy dredging activities, sea port activities, marine debris, and construction of dredgers at the shore sites.

The calculated pollution load index (PLI) values of heavy metals in water presented in Fig. [3A.](#page-10-0) The PLI values were ranged from 0.85 to 2.96 that the water of the studied coastal area was base line level to the high degree of pollution (PLI > 1). Elevated levels of PLI were observed in three sampling sites of the Cox's Bazar coast, Chittagong coast, and Sundarbans Fig. [3A](#page-10-0) which might be attributed to the discharge of different salts and chemicals from hatcheries, fish processing industries, ship breaking activities, and some aqua farms through their underground pipeline outlets to the sea without treatment at these sites (Raknuzzaman et al. [2016;](#page-13-0) Bristi et al. [2021\)](#page-12-0), vehicle and sugarcane farming activities at these sites. The mean values of Nemerow's pollution index (NPI) of the study area were 17.8, 23.2, 7.8, and 11.6 for the Cox's Bazar coast, Chittagong coast, Meghna Estuary, and Sundarbans, indicated moderately to heavily polluted water (Fig. [3B](#page-10-0)). For computing C_d m C_d (Table S3), the guideline values of heavy metals can be suggested on the basis of various standard values. For evaluating the level of heavy metals contamination mc_d was used which revealed that because of examining the contamination by heavy metals using the mC_d , three studied sites contained waters that surpassed the contamination threshold limit (mC_d=1.5). The sites including Cox's Bazar coast (2.6), Chittagong coast (3.4), and Sundarbans (1.7) sites were evaluated as contaminated (Fig. $3D$). The C_d value of heavy metals showed moderate to considerable degree of contamination (Fig. [3C](#page-10-0)). The descending order of the pollution status of the study area is in the order of Chittagong coast > Cox's Bazar coast > Sundarbans > Meghna Estuary.

Source analysis of heavy metals

After acquiring the dataset based on centered log-transformed algorithm, Spearman correlation and Principal component

Fig. 2. Contamination factor (CF) of heavy metal in water samples collected from the coastal sea and estuarine area in Bangladesh. Horizontal dot lines indicate the standard limit of contamination level

Fig. 3. Distribution of risk index value; A Pollution load index (PLI), B Nemerow's pollution index (NPI), Degree of contamination (Cd) and D Modified degree of contamination (mCd) due to heavy metal

analysis (PCA) were adopted to elucidate the inter-elements association, Euclidean differences, and possible sources concerning the geochemical relationship of studied heavy metals in surface water samples of the Bengal coast, Bangladesh. With the communality method Varimax rotation was coupled which was employed to confirm the PCA to get a basic expression of the heavy metals except changing the coordinate system of the elements. Three principal components (PCs) were extracted from 9 elements in water samples and the three PCs together contributed 80.7% of the total variance. In surface waters for Rotated Component Matrix, As, Fe, Cr, and Ni had high loadings in PC1, and can explain 28.9% of the variance (Fig. [4](#page-11-0) and Table S4). Iron is typically more common in the marine environment than other metals due to its high prevalence on Earth (Edokpayi et al. [2016](#page-12-0); Wang et al. [2017\)](#page-14-0). The weathering of calcite and chlorite in loess can provide an amount of Fe to the surface water of the coastal sea and estuaries (Yokoo et al. [2004\)](#page-14-0). Application of arsenicbased pesticides/herbicides during agricultural activities, different industrial activities of stainless steel, ship breaking and repairing, rechargeable batteries, metal ceramics, dyeing, and

Sampling sites

contamination in water collected from the coastal sea and estuarine areas in Bangladesh. Horizontal dot lines indicate the level of contamination for different risk index

leather industries, plating and gas turbine factories, export import zones in the Chittagong and Mongla port areas (Wang et al. [2019](#page-14-0)).

Principal component 2 (PC2) explained about 30.6% of the total variance with the factor loadings of Zn, Cu, Pb, and Cd which were mainly from anthropogenic input such as industry effluents and domestic sewage (Islam et al. [2015](#page-12-0), [2018\)](#page-12-0). The source identification of the present study findings are consistent with previous study by Genthe et al. ([2013](#page-12-0)) where industries like textiles effluents have been identified as prime sources of metallic components (i.e., Cd, Cu, Pb, and Zn) either as suspended or dissolved forms in the natural water. Furthermore, another study by Kibria et al. ([2012](#page-13-0)), applied laboratory prepared synthetic mussels for the detection of heavy metals in the natural waters and found that industrial and agricultural/aquaculture discharges are potential source for higher levels of Cd, Cu, and Pb (Emenike et al. [2020;](#page-12-0) Islam et al. [2020](#page-13-0)). Principal component 3 (PC3) explained about 21.1% of the total variance with the loadings of Mn (Fig. [4](#page-11-0)) which was mainly from the breakdown of parent material through weathering and pedogenic processes. In the

Fig. 4. Principal Component Analysis (PCA) of heavy metals in surface water collected from the coastal sea and estuarine areas in Bangladesh

natural water systems, Fe and Mn may be released via the reductive dissolution and precipitation of Fe-Mn-Oxyhydroxide and releases Fe and Mn contents from pyroclastic sediments (Kumar et al. [2016](#page-13-0); Islam et al. [2017\)](#page-12-0). This suggests that changes in pH and redox state may cause by seasonal groundwater level fluctuations dissolve Mn complexes and release arsenic adsorbed on them into ground and surface water (Gan et al. [2014](#page-12-0); Díaz et al. [2016](#page-12-0)).

Conclusions

In this study, base line information, identification of source and mobilization mechanisms of physicochemical parameters and heavy metals in surface waters from the Bengal coast of Bangladesh were determined. Among the fourteen sampling sites of the Bengal coast, ship breaking, and sea port areas of Chittagong and hatchery site of Cox's Bazaar were severely polluted where elevated concentrations of Cu, As, Cd, and Pb were observed. The concentrations of physicochemical parameters such as EC, DO, turbidity, Fl⁻, Cl⁻, and $PO₄³⁻$ and heavy metals were higher than the safe recommended values, which suggested that waters of the Bengal coast are polluted by heavy metals and might create an adverse effect on these coastal ecosystems. The CF, PLI, NPI, C_d , and mC_d established that the surficial waters were polluted by heavy metals to different levels. PCA showed that essential and toxic elements originated from both anthropogenic and natural sources. This study suggests that integrated aspects could be explored in the future, for instance, the identification of heavy metals in the aquatic water bodies and their distribution and

how they contribute to accumulate in the human body through different exposure pathways.

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Declarations

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