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Spatiotemporal distribution of potentially toxic elements in the lower Gangetic delta and their implications for non-carcinogenic health risk management

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Abstract

River Hooghly, a tributary of river Ganges is one of the major rivers of Asia having traditional, social, economic, religious, and spiritual values. Water samples were collected from 18 sampling locations of river Hooghly during summer (dry), monsoon (wet), and winter (cold) seasons. The samples are analysed for basic physicochemical properties and abundance of selected potentially toxic elements (PTEs) are measured. Several PTEs, e.g., Al, Fe, Ni, and Pb, were found to be above the permissible limits, prescribed by national and international guidelines for safe human consumption. The trend of variation in the mean PTE concentrations showed the following order: Cd < Pb < Co < Cr < Ni < Cu < Zn < Mn < Fe. Due to the presence of high total dissolve solid (TDS) and PTE contents, the water quality of river Hooghly is not suitable for direct human consumption. The evaluated Water Quality Index (WQI) value showed a distinct spatio-temporal variation indicating very severe condition of water quality, which is deteriorating gradually from upstream to downstream. In summer, monsoon, and winter, the highest WQI values were observed in Maushuni Island (S15), Petuaghat (S18), and Tapoban (S17), respectively. However, the non-carcinogenic human health risk in terms of Hazard Quotient and Hazard Index values of PTEs indicates no immediate adverse impact on human health due to exposure of PTE contaminated water from river Hooghly through ingestion or dermal route. Though, these risk values for children were higher than adults warranting the adoption of a long-term management plan to cope with potential human health risks. The result suggests implementation of a combination of stringent socio-legal regulations and numerical models for sustainable water related health risk management in river Hooghly.

Keywords: Toxic elements, Water quality index, Seasonal variation, Contaminants, Physicochemical attributes, River Hooghly

Introduction

Riverine freshwater is a major natural resource for ecological sustainability (Rai 2008). Organic and inorganic contaminants or pollutants may enter in the riverine or estuarine systems from a wide range of anthropogenic sources, e.g., industrial and domestic effluents (Amman et al. 2002), storm and surface water run–off (Bhattacharya et al. 2015), agriculture and aquaculture run-off (Ghosh et al. 2016; Mitra et al. 2018a) and

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natural processes like biogeochemical cycle (Garrett 2000), chemical leaching of bedrocks and water drainage basins (Zhou et al. 2008a, b), sediment resuspension, and ground water inflow (González-Ortegón et al. 2019). Among the organic and inorganic contaminants of water, potentially toxic elements (PTEs) are drawing added attention due to their non-biodegradable nature and accumulation potential through tropic levels causing a long-term effect on the ecosystem. PTEs refer to both metals and non-metals having a range of environmental significance (Nieder et al. 2018). Some PTEs have nutrient-like profiles, such as Copper (Cu) and Cadmium (Cd), suggesting their correlation with biological cycles (Boyé et al. 2012), whereas some other PTEs such as Lead (Pb) possess scavenger like behaviour (Flegal and Patterson 1983). Some of the PTEs such as Cobalt (Co), Copper (Cu), Iron (Fe), Manganese (Mn), and Zinc (Zn) take part in several significant biochemical reactions and act as terminal electron acceptor and micro nutrients (Munoz-Olivas and Camara 2001), but show toxic effects in excess quantities (Low et al. 2015). Some other elements such as Arsenic (As), Cadmium (Cd), and Mercury (Hg) show toxicity in minute quantity (Alves et al. 2014). Thus, use of PTE contaminated water in irrigation may have detrimental effects on local biodiversity including invertebrate and microbial communities hampering ecological balance and sustainability (Kar et al. 2008; Tom et al. 2014; Bhattacharya et al. 2015; Ferreira et al. 2016; Allinson et al. 2017). Moreover, the water qualities of the rivers and estuaries are also regulated by constant influx of contaminated water from several point or non-point sources from upstream making difficult to regulate the water quality (Mitra et al. 2018a).

The estuarine region behaves like a natural filter or buffer zone where the PTEs are adsorbed by the suspended solids and/or might get bio-accumulated in aquatic organisms, e.g., phytoplanktons, zooplankton, benthos, invertebrate, fish, etc. (Tao et al. 2012; Karbassi et al. 2015). Dissolved organic matter (e.g., humic acid, fulvic acid, carbohydrates) also plays a crucial role in regulating PTE concentration in natural streams by forming metal complexes or chelates (Philippe and Schaumann 2014). In the past few decades, public health policy-makers and researchers have focused on increasing anthropogenic input resulting exposure of aquatic habitats to hazardous contaminants due to their toxicity and persistence in natural environment (Upadhyay et al. 2006; Zhou et al. 2008a, b). Toxic elements also had the probability of human health risk (carcinogenic and non-carcinogenic) even at concentration below permissible limit and can be estimated following United States Environmental Protection Agency (USEPA) methods (USEPA 2004; Gao et al. 2019). Seasonal or temporal variations in intensity of agricultural and industrial activity, aquaculture, storm water drainage, atmospheric deposition, and climatic events can have strong influences on the status of river water quality (Singh et al. 2004; Ouyang et al. 2006; Li and Zhang 2010). River Hooghly, a tributary of river Ganges is one of the major rivers of Asia having traditional, social, economic, religious, and spiritual values. About 0.5 billion peoples are directly or indirectly dependent on Ganges river system for their livelihood (Bharati et al. 2016). The region experiences 80% of its annual rainfall between June and September from southwest monsoon (Mukhopadhyay et al. 2006). This strong monsoonal effect might have strong consequences on PTEs inputs to river Hooghly during rainy season. Hence, characterization of seasonal variability of contaminants in this river water is essential for proper evaluation of water quality and assessment of long-term impacts on public health and human welfare. This approach can help to formulate proper policy for reducing contaminants and safeguarding human health and hygiene in the Hooghly river region of the lower Gangetic basin. Although there are several studies (e.g., Sekhar et al. 2005; Mukhopadhyay et al. 2006; Henderson et al. 2007; Sarkar et al. 2007; Pertsemli et al. 2007; Li et al. 2008; Li and Zhang 2010; Mitra et al. 2018a, b), none has emphasised the implication of seasonal and geospatial variations of PTEs for the management of river water quality. To bridge the existing knowledge gap, the present study aims to evaluate geospatial and seasonal water quality and potential non-carcinogenic health risk associated with PTEs in river Hooghly.

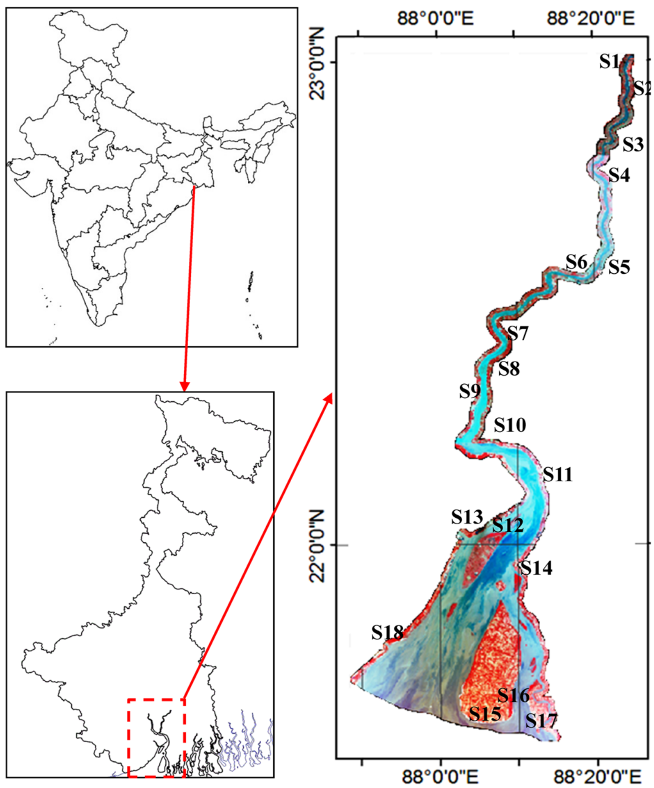
Materials and methods

Study area and sampling

River Ganges divides into two major distributaries; Bhagirathi (India) and Padma (Bangladesh) at Mithipur village in Murshidabad district, West Bengal. The tidal regime of river Bhagirathi starting from the downstream of Nabadwip city is known as river Hooghly (Rudra 2014; Ghosh et al. 2016), and serves as a navigable waterway for Kolkata and Haldia ports (Fig. 1). Water samples were collected using clean plastic sampling bottles from a depth of 10 cm in triplicate from 18 sampling stations covering ~ 200 km of River Hooghly in summer (March–May), monsoon (June–September), and winter (November–January) during low tide in 2015–2016 to avoid marine dilution of PTEs and other parameters, readily transferred to the laboratory in ice box and processed (Fig. 1).

Characterization of physicochemical properties and PTEs of water

pH and electrical conductivity (EC) were measured on field using HANNAH Multi parameter (HI-9829-13102).



Sl. No.	Sampling Location	Station Code	Latitude	Longitude
1	Tribeni	S1	22°59'42.02"N	88°24'22.10"E
2	Halisahar	S2	22°56'25.40"N	88°24'29.70"E
3	Panihati	S3	22°42'11.27"N	88°21'56.56"E
4	Kashipur	S4	22°37'33.21"N	88°21'59.66"E
5	Babughat	S5	22°33'48.83"N	88°20'15.27"E
6	Shibpur	S6	22°33'11.10"N	88°17'59.74"E
7	Bata Nagar	S7	22°30'53.48"N	88°12'52.86"E
8	Birlapur	S8	22°26'5.08"N	88° 8'15.15"E
9	58 Gate	S9	22°20'53.78"N	88° 5'19.79"E
10	Falta	S10	22°14'43.13"N	88° 5'14.74"E
11	Diamond Harbour	S11	22°11'10.70"N	88°11'15.77"E
12	Nayachar	S12	22° 1'24.62"N	88° 8'31.17"E
13	Haldi Estuary	S13	22° 0'07.76"N	88° 2'58.16"E
14	Lot 8	S14	21°53'06.85"N	88°09'52.85"E
15	Maushuni Island	S15	21°35'57.98"N	88°12'26.31"E
16	Chemaguri	S16	21°39'27.25"N	88° 9'19.80"E
17	Tapoban	S17	21°37'27.79"N	88° 5'34.34"E
18	Petuaghat	S18	21°47'25.18"N	87°53'31.38"E

Fig. 1 Study area

Dissolved oxygen (DO) concentration was measured following Winkler's method (Winkler 1888; APHA 2017). In laboratory, surface water quality parameters such as total dissolved solid (TDS), salinity, hardness, alkalinity, and chemical oxygen demand (COD) were analysed following protocols as described in APHA (2017). PTEs in water were measured using inductively coupled plasma optical emission spectrometer (ICP-OES) (Thermo Fisher iCAP 7400 ICP-OES). Analytical procedure, accuracy, and precision data have been provided in Additional file 1: Table S1.

Geospatial analysis

A multivariate interpolation method, i.e., inverse distance weighed (IDW) process, was used in ArcGIS software (V10.2) to plot the seasonal geospatial map of studied PTEs. The data were projected to WGS 1984, UTN zone 45 N.

Statistical analysis

General statistical analyses of physicochemical properties of water were conducted to understand the variation in physicochemical characteristics of water. Pearson product correlation co-efficients, analysis of variance (ANOVA) single factor method followed by post hoc comparison test, i.e., least significant difference (LSD test), and principal component analysis (PCA) between PTEs were done using software SPSS (V16.1) to understand the source and association of the PTEs. Shapiro–Wilk test was also applied to evaluate the normality of the dataset, whereas Kaiser–Meyer–Olkin (KMO) Measure and Barlett's Test of Sphericity were used to find data adequacy for PCA (Kaiser 1958; Ul-Saufie et al. 2013).

Evaluation of water quality indices (WQI) and risk assessment

Water quality indices (WQI) can be described as a rating that reflects the combined impact of different water quality parameters (Şener et al. 2017; Gao et al. 2019). To calculate WQI, different weights were assigned to each of the measured chemical parameters (Ramakrishnaiah et al. 2009; Yidana and Yidana, 2010). However, risk assessment of the PTEs is a multi-step procedure based on the exposure to and tendency of the PTEs to bioaccumulate within the human body. Human body can be exposed by PTEs from water through consumption/ingestion or dermal routes. Thus, the examined PTEs were compared with reference dosages, and the mean daily intake ($MDI_{ingestion}$ and MDI_{dermal}) were estimated for both children and adults as per USEPA Risk

Assessment Guidance for Superfund (RAGS) standards (USEPA 1989, 1991, 2004, 2011; Wu et al. 2009; Li and Zhang 2010; Mitra et al. 2018b; Singh et al. 2018; Saleem et al. 2019; Gao et al. 2019). Toxicological profile of the studied elements indicates that all PTEs have toxic carcinogenic or non-carcinogenic human health effects (Luo et al. 2012). Here, the potential degree of non-carcinogenic risk to human population due to ingestion of PTE contaminated water was evaluated as hazard quotients (HQ). Detailed calculation of WQI, risk assessment, and HQ is given in Additional file 1: Table S2.

Results

Physicochemical properties of water

In river Hooghly, physicochemical properties of water in summer, monsoon, and winter season varied between 7.22 and 7.80, 7.23 and 8.00, and 7.19 and 8.00 for pH; 308.0 and 3120, 122 and 3100, and 135 and 3200 µS/cm^2 for EC; 568.2 and 2250, 274.6 and 1668.3, and 503.4 and 2023.6 mg/l for TDS, 0.04 and 26.80, 0.09 and 19.80, and 0.08 and 21.78 for salinity; 80.6 and 1881.0, 19.8 and 1485.0, and 22.3 and 1650.4 mg/l for hardness; 136.5 and 1173.0, 126.1 and 1044.3, and 133.2 and 1085.5 mg/l for alkalinity; 3.1 and 4.9, 3.7 and 5.1, and 3.1 and 5.5 mg/l for DO; 16.3 and 52.1, 20.5 and 42.1, and 29.3 and 51.3 mg/l for COD, respectively (Additional file 1: Table S3). In summer, highest value of pH was recorded in Lot 8 (S14), EC, salinity, and DO in Maushuni Island (S15), TDS, hardness, and alkalinity in Tapoban (S17), and COD in Bata (S7). During monsoon, highest value of pH and DO was observed in Panihati (S3), EC, TDS, salinity, alkalinity in Tapoban (S17), and hardness and COD in Chemaguri (S16), whereas in winter, highest value for pH was observed in Shibpur (S6), EC and Salinity in Tapoban (S17), TDS and alkalinity in Petuaghat (S18), hardness in Chemaguri (S16), and DO in 58 Gate and COD in Bata Nagar (S7).

Spatiotemporal distribution of PTEs

The vast study area in River Hooghly is regularly exposed to different sources of natural and anthropogenic inputs and experiences dynamic river processes resulting in varied distribution of PTEs. The average range of concentrations for PTEs in summer, monsoon, and winter varied between 5401.7 and 15,488.5, 4710.8 and 16,988.6, and 6146.6 and 16,287.4 µg/l for Al, 3.7 and 14.1, 2.6 and 9.3, and 4.3 and 18.2 µg/l for Cd, 14.7 and 57.0, 10.6 and 30.7, and 14.4 and 61.6 µg/l for Co, 17.4 and 54.1, 10.7 and 41.5, and 16.0 and 68.4 µg/l for Cr, 34.6 and 76.3, 24.0 and 61.0, and 33.6 and 85.9 µg/l for Cu, 4436.4 and 17,552.1, 4597.9 and 15,111.8, and 6546.9 and 21,461.2 µg/l for Fe, 122.5 and 334.5, 84.0 and 300.8, and 130.9 and 422.5 µg/l for Mn, 24.9 and 63.5, 18.2 and 71.6, and 28.3 and 107.3 µg/l for Ni, 7.9 and 29.8, 6.3 and 25.5, and 10.1 and 35.8 µg/l for Pb, and 37.7 and 101.1, 30.2 and 78.3, and 45.3 and 121.4 µg/l for Zn (Additional file 1: Table S4), respectively, as depicted on the geospatial maps developed using ArcGIS. The colour gradient from blue to red represents the lowest to highest concentration of PTEs (Figs. 2, 3, 4). In summer, highest concentrations of Al, Cd, Cr, and Cu were observed in Nayachar (S12), Co in Tapoban (S17), Fe, Mn, and Pb in Falta (S10), Ni in Shibpur (S6), and Zn in Maushuni Island (S15). During monsoon, maximum concentrations of Cd were found in Petuaghat (S18), Co in Lot 8 (S14), Cr in Haldi estuary (S13), Al and Cu in Nayachar (S12), Fe and Pb in Falta (S10), Mn in Halisahar (S2), Ni in Bata Nagar (S7), and Zn in Shibpur (S6), whereas in winter, the highest concentration of Cd was observed in Haldi estuary (S13), Co in Tapoban (S17), Cr in Petuaghat (S18), Al, Cu, Fe, and Pb in Falta (S10), Mn in Diamond Harbor (S11), Ni in Birlapur (S8), and Zn in Maushuni Island (S15).

Discussion

Regulation of physicochemical parameters of river Hooghly

The physicochemical profile of a water body is regulated by an interplay of multitude of biological, physical, and anthropogenic processes (Singh et al. 2004; Ouyang et al. 2006; Li and Zhang 2010; Zhang and Gao 2015; Mitra et al. 2018b). pH is a significant physicochemical parameter indicative of the usage of water for drinking and irrigation purpose (Şener et al. 2017), as it can regulate the alkalinity, hardness, and solubility of PTEs in water column (Osibanjo et al. 2011; Şener et al. 2017). A higher pH reduces the solubility of PTEs, while lower pH enhances release of their ions (Singh and Kumar 2017). However, irrespective of the season and sampling stations, the range of pH observed in river Hooghly varies between neutral to sub-alkaline range (7.19–8.00) that is within the WHO guidelines and Indian Standards for safe drinking water (WHO 2008, 2011; BIS 2012).

In general, rivers which are relatively narrow in the upstream and have funnel-shaped wide mouth in the downstream show a steep salinity gradient across the river. A similar pattern of steep salinity gradient is also evident in river Hooghly, as it is a funnel-shaped wide mouthed macro-tidal river (Mukhopadhyay et al. 2006; Rudra 2014). This salinity gradient might be regulating the flocculation of dissolved PTEs, which affects the elemental composition of the river (Samani et al. 2015). Irrespective of the season, the salinity of samples collected from Babughat (S5), the sampling station near Kolkata varies between 0.09 and 0.12. The salinity is found to be lowest in summer, in Babughat (S5) when tidal magnitude or influx is expected to be higher compared to other

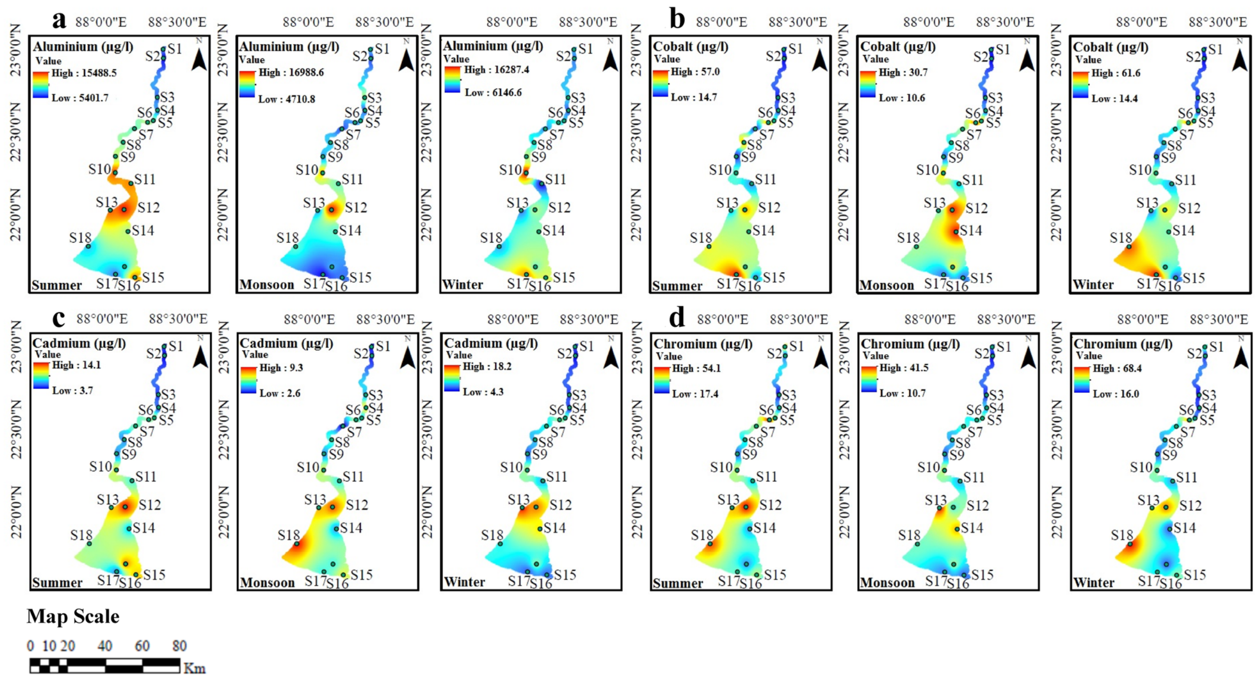


Fig. 2 Geospatial distribution of dissolved Aluminium (Al), Cadmium (Cd), Cobalt (Co), and Chromium (Cr) in river Hooghly. **a** Al, **b** Cd, **c** Co, and **d** Cr

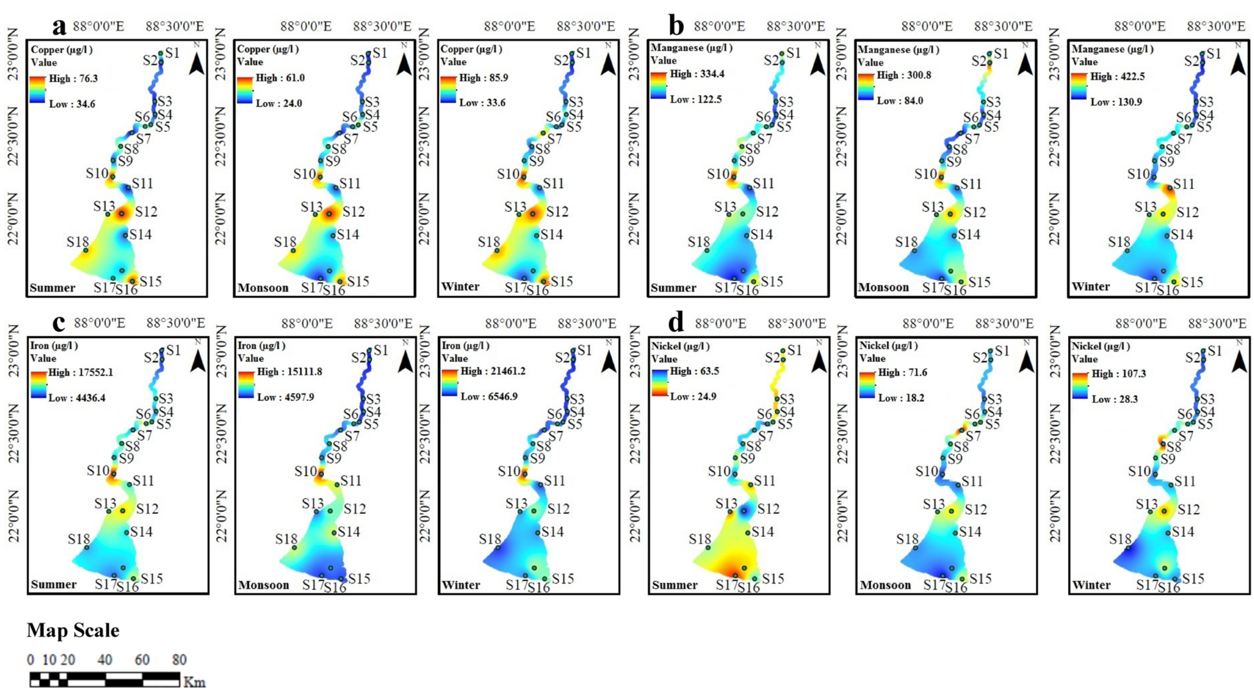


Fig. 3 Geospatial distribution of Copper (Cu), Manganese (Mn), Iron (Fe), and Nickel (Ni) in river Hooghly. **a** Cu, **b** Mn, **c**) Fe, and **d** Ni

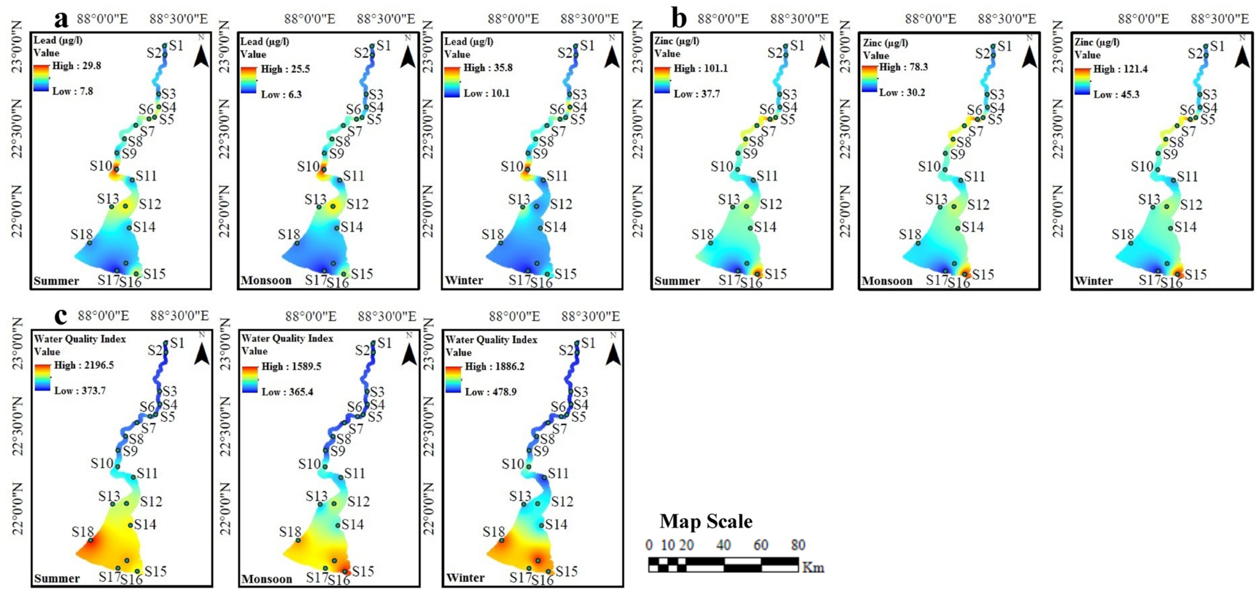


Fig. 4 Geospatial distribution of Lead (Pb), Zinc (Zn), and Water Quality Index (WQI) in river Hooghly. **a** Pb, **b** Zn, and **c** WQI

seasons (Sadhuram et al. 2005; Mukhopadhyay et al. 2006). This indicates that increased magnitude of tidal influx rarely influences salinity of river Hooghly near Kolkata (Samanta et al. 2018) and flocculation of dissolved PTEs at lower saline regime (Samani et al. 2015).

The mean values of EC, TDS, salinity, hardness, and alkalinity show wide range of variation throughout the river, but have direct relation with each other. The post hoc analysis indicates significant statistical variation of Hardness, DO, COD, and TDS (LSD Test; $p<0.05$). It was evident from the observed data that the monsoonal downpour has reduced the TDS, EC, salinity, alkalinity, hardness, and COD level of river Hooghly, but DO was increased within the same timeframe. Measured salinity, EC, and TDS show an increasing trend towards mouth of the river Hooghly, which also complements the study of Mitra et al. (2018b). In river Hooghly, the mean values of salinity and TDS are comparatively higher in summer than those in monsoon and winter, which might be due to the combined effect of higher rate of evaporation, higher water temperature, and lower precipitation (Rajasegar 2003; Mukhopadhyay et al. 2006; Mitra et al. 2018b). High TDS concentration in downstream might also be due to dissolved clay particles and sediment resuspension from wide mud flats along both banks of river Hooghly (Batabyal et al. 2014; Ghosh et al. 2019a). The elevated alkalinity of river Hooghly was found to be twice or higher of the permissible limit which indicate the prevalence of bicarbonates (Ghosh et al. 2019a). Irrespective of the sampling station and seasons, water of river Hooghly is found to be very hard (BIS 2012), and due to high TDS, it is not fit for direct human consumption without treatment (WHO 2004, 2008, 2011). The DO values indicate that hypoxic condition does not prevail in river Hooghly (Satpathy et al. 2013) and complements the study of Kazi et al. (2009). However, the observed DO value suggests that water of river Hooghly was suitable for drinking only after proper treatment and disinfection, but might be used directly for the propagation of wildlife and fisheries (BIS 2012). The mean COD values are found to be highest in winter and lowest in summer which might be due to the abundance of microbial population in river Hooghly in summer season as reported by Basu et al. (2013). Moreover, higher COD values in upstream of river Hooghly are due to inflow of domestic and municipal sewage along with agricultural waste and effluents from adjoining industries (Kazi et al. 2009; Pati et al. 2014).

Regulation of geospatial distribution of PTEs

River Hooghly is comparatively well mixed and there is little stratification in the water column (Mukhopadhyay et al. 2006; Samanta and Dalai 2018). In general, irrespective of sampling locations and seasonal changes in river Hooghly, the mean concentrations of PTEs are found to be in the following order Cd < Pb < Co < Cr < Ni < Cu < Zn < Mn < Fe < Al (Table 1) (LSD test; $p<0.05$). PTEs like Al, Cd, Fe, Ni, and Pb in water of river Hooghly exceed respective permissible limit prescribed by World Health Organization and Indian Standard

Table 1 Distribution of PTEs in river Hooghly

	Parameter	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Summer	Mean	8306.5	6.5	25.0	27.8	41.0	7659.7	158.7	35.1	13.2	52.0
	Std. deviation	4721.2	3.9	16.0	15.8	22.9	4516.9	93.1	18.8	7.9	28.2
	25th Percentiles	6132.4	4.1	15.3	19.1	33.8	5060.4	124.0	27.1	8.9	41.3
	50th Percentiles	8493.0	6.4	23.1	28.6	40.4	7783.2	165.8	36.8	12.7	56.5
	95th Percentiles	15,197.0	14.7	54.7	54.4	76.4	16,429.0	308.9	63.2	27.4	99.4
Monsoon	Mean	7471.6	5.7	21.0	21.8	34.9	6667.8	132.4	29.0	10.8	42.0
	Std. deviation	5409.0	4.3	16.7	16.7	26.1	4969.7	103.0	20.6	8.3	29.6
	25th Percentiles	915.1	1.1	3.3	3.5	3.6	1068.9	11.3	4.5	1.9	4.5
	50th Percentiles	7981.5	5.7	19.9	22.4	37.4	7136.1	129.4	31.4	11.8	51.7
	95th Percentiles	15,454.0	14.7	52.4	52.6	76.4	16,429.0	310.4	60.1	27.4	84.8
Winter	Mean	9482.90	8.69	30.87	32.89	51.18	9183.40	193.18	45.34	15.78	66.38
	25th Percentiles	3885.38	4.21	15.78	16.84	22.32	4132.27	92.92	23.14	7.64	27.60
	50th Percentiles	7572.70	5.61	18.68	20.75	38.78	7293.00	145.54	34.11	11.98	56.91
	95th Percentiles	9236.80	8.39	29.45	31.47	46.50	8563.20	184.50	42.30	14.42	66.03
	25th Percentiles	15,882.00	17.71	61.16	66.46	88.15	15,645.00	375.47	93.88	31.69	113.77

All values are in μg/l

(WHO 2008, 2011; BIS 2012). Rapid and unplanned urbanization along with industrialization have homogenized the sources and tidal influx in river Hooghly throughout the year, which might have played a crucial role behind the similar mean spatial distribution or concentration of PTEs in all three seasons (Stucker and Lyons 2017). However, adsorption, flocculations, formation of oxides and/or hydroxides, alumina-silicates, organic chelates, and river water chemistry are major factors which regulate the distribution of PTEs in the water column (Takayanagi and Gobeil 2000). A large quantum (4×10^8 m^3) of PTE containing sewage is entering regularly into the river from adjoining urban settlements (Mukhopadhyay et al. 2006). The sewage from the adjoining cities, industrial discharge, and agricultural run-off contains colloidal materials or particles like dissolved organic carbon (DOC) which can form complexes with PTEs by organic ligands (Wen et al. 1999). Moreover, the downstream of river Hooghly is also dominated by mangrove forests, which acts as a major source of DOC. Samani et al. (2015) pointed out that DOC (hydrophobic humic materials) plays a crucial role in the flocculation of PTEs because of salinity gradient. Higher concentrations of Al and Fe are evident as dissolved toxic elements in all of the sampling stations of river Hooghly as Al and Fe are major constituents of the earth crust (Mitra et al. 2018b). PTEs (Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn) might have sourced in river Hooghly from different industrial units comprising paper and pulp, iron and steel, thermal power plant, brick kiln, welding industries, and battery industries, which have been operating on both the banks of the river (Karar and Gupta 2006; Govil et al. 2008; Ghosh et al. 2016, 2019b; Bakshi et al. 2017, 2019; Mitra et al. 2018b).

When compared with other rivers and estuaries around the globe, concentrations of toxic elements measured in river Hooghly are observed to be much higher than Costa Concordia wreck, Han river, Yangtze river, Padma river, Yangzhong water system, and Ghana stream rivers (Asante et al. 2007; Zhou et al. 2008a, b; Wu et al. 2009; Li and Zhang 2010) but lower than Odiel River and Sydney estuary (Olias et al. 2004; Birch and Lee 2018). The PTEs concentration of river Hooghly are also found to be higher than river Padma, Bangladesh (Jolly et al. 2013) which might be due to greater fresh water input in river Padma from river Brahmaputra and river Meghna. While comparing with different rivers of India like Gomti river, Manjira river, Mahanadi river, and Subarnarekha river, the concentration of toxic elements were observed to be higher in river Hooghly, except for Cu which was higher in Manjira river, and Pb and Zn for Gomti river (Senapati and Sahu 1996; Konhauser et al. 1997; Gaur et al. 2005; Krishna et al. 2009). However, the concentration of toxic elements like Cd, Co, and Zn showed a temporal increment when compared with previous studies on the same river (Table 2). Thus, river Hooghly has became a major route or drain for discharge of toxic elements originating from different point and non-point sources like municipal and urban wastes, industrial effluents, and agricultural run-off to the Bay of Bengal.

Table 2 Comparison with previous studies conducted across the globe, India, and river Hooghly

	Al	Bi	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	U	V	Zn
Global Rivers and Estuaries														
Odiel River, Olías et al. (2004)					7600	23,470	13,700			207				
Ghana stream rivers, Asante et al. (2007)			1.3	0.52	2.65		682			0.85	1.4			
Yangzhong water system, Zhou et al. (2008a, b)		0.043	0.117	0.319	6.414			82.634			2.585	0.8	2.79	114.6
Yangtze River, Wu et al. (2009)			4.7	20.9	20.9	10.7	239.8	5.4	11.7	13.4	55.1		10.5	9.4
Han River, Li and Zhang, (2010)			2.45	3	12.25	15.9	36.82	31.15			1.71		70.2	
Costa Concordia wreck, Schintu et al. (2018)			0.006		0.048	0.142				0.154	0.003		2. 7	
Padma River, Jolly et al. (2013)				2.0	3.0	20.0	199.0	15.0		8.14	1.5			7.26
Sydney Estuary, Birch and Lee, (2018)				84	303	689	47,000	6965		86	642			1127
Other rivers in India														
Subarnarekha River, Senapati and Sahu, (1996)					1.13	15.88				15.75	19.13			
Mahanadi River, Konhauser et al. (1997)					9.8	5.9				7.2	2.68			23
Gomti River, Gaur et al. (2005)			0.69		5.6	22.4	3054.6	116.8		17.1	22.3			84.4
Manjira River, Krishna et al. (2009)				16.8		161.8	72.9	26.7		2.1				11
Previous studies in River Hooghly														
River Hooghly, Sarkar et al. (2007)						20.2	185.8	204.4			29.3			190.3
River Hooghly and Sundarban coastal water, Bhattacharya et al. (2015)			0.64		41.56	56.05				48.4	21.25			58.94
River Hooghly, Mitra et al. (2018b)			1.98	17.15	60.88	46.32	55,163.3	813.79		41.08	30.09		107.2	63.24
Present study														
Summer	10,197.8		7.9	30.4	33.9	50.4	9331.4	196.2		42.9	16.2			63.9
Monsoon	8489.4		5.41	18.90	21.00	36.98	7316.6	159.5		36.4	12.54			50.40
Winter	9895.2		9.0	31.2	34.3	54.4	10,048.0	219.1		53.7	18.0			76.3
Mean	9527.5		7.4	26.9	29.7	47.3	8898.7	191.6		44.3	15.6			63.5
WHO and Indian Standards (WHO, 2008, 2011; BIS, 2012)	200		3	40	50	2000	300	500		20	10			3000

All values are in μg/l

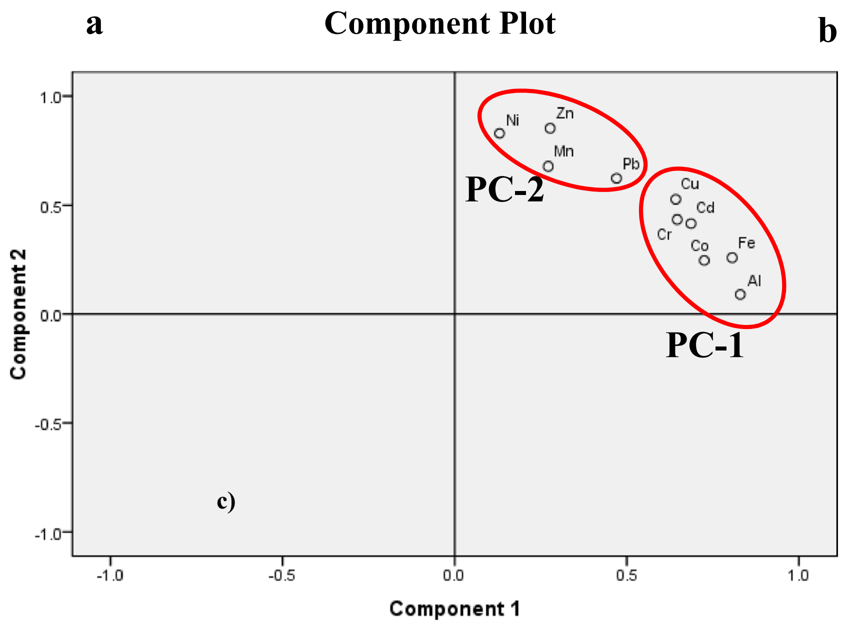
Statistical analysis to identify potential sources of PTEs

As evident from Additional file 1: Table S5, in river Hooghly throughout the year, the spatial distributions of all potentially toxic elements are correlated with each other ($r = 0.217–0.737$; $n = 162$; $p < 0.01$) except for Ni, which is correlated with Al ($r = 0.197$; $p < 0.05$). Cd, Co, Cr, Mn, and Zn show weak positive correlation with Al ($r < 0.422$) which indicates lesser role of clay minerals in geochemical cycling of these PTEs in river Hooghly. Cu, Fe, and Pb show strong positive correlation with Al suggesting previous association of these elements with clay minerals. However, inter-elemental associations among PTEs suggest that the elements are cycled mostly with a common phase of Fe–Mn oxyhydroxides (Samanta et al. 2017). The positive correlations between PTEs also indicate that they might originate from identical natural/riverine and/or anthropogenic sources having similar mode of movement in the river Hooghly. Higher concentrations of PTEs like Al, Fe, Mn, Co, and Cr can be attributed to various sources, which may be both natural erosion and weathering (Ghosh et al. 2018, 2019a, b; Mitra et al. 2018b) and/or anthropogenic activities; for example, Al can be sourced from foils, garbage, electrical wires, alloy industries (Mitra et al. 2018b); Cd can be originated from fossil fuel, thermal power plants, fertilizer, industrial waste incineration (Caruso and Bishop 2009; Reza and Singh 2010; Raknuzzaman et al. 2016; Mitra et al. 2018b); Co from metal alloys run-off from navigating ships (Mitra et al. 2018b); Cr from textile industries, dyes, pigments; Cu from insecticides, smelting industries, and shipping and boating activities (Shazili et al. 2006; Ghosh et al. 2016, 2019a, b; Ismail et al. 2016); Fe from iron and steel industries, thermal power plant, fossil fuel (Mahato et al. 2014; Mitra et al. 2018b); Mn from paper and pulp industry, power plant, and welding industries (Giri and Singh 2014); Ni from glass and ceramic industries, power plants, automobiles batteries, alloys, and smelting industries (Tariq et al. 2006; Govil et al. 2008); Pb from batteries, fossil fuels, chemical fertilizers (Jumbe and Nandini 2009; Wuana and Okieimen 2011); Zn from fertilizers, synthetic paints, and immersion of idols (Wu et al. 2009; Bhattacharya et al. 2015).

The ANOVA results suggest a statistically significant variation in distribution of toxic elements at all 18 sampling stations and between seasons in river Hooghly at 99.995% confidence level (Additional file 1: Tables S6 and S7). Post hoc analysis additionally reveals significant statistical variation in the distribution of elements amidst the seasons, more specifically between summer and monsoon (LSD test; $p < 0.05$); except Cr, among monsoon and winter (LSD Test; $p < 0.05$). However, Co, Ni, and Zn shows statistical significant variation in their distribution between winter and summer (LSD test; $p < 0.05$). Shapiro–Wilk test was applied to evaluate the normality of the experimental data after transforming the data by taking the base 10 logarithms. Kaiser–Meyer–Olkin (KMO) Measure and Barlett's Test of Sphericity were used to find data adequacy for PCA. The KMO measure value (0.874) is greater than 0.500, indicating that the data are sufficient and Barlett's measure of sphericity ($p < 0.001$) for all examined data shows a higher degree of relationship among the PTEs, suggesting suitability of the data set for performing PCA ($p < 0.001$) (Kaiser 1958; Ul-Saufie et al. 2013). The result of PCA (VARIMAX rotation mode) suggests that eigen values more than 1 represent 65.9% of the total variance, indicating that distinctive controlling components or sources are responsible for the distribution of dissolved PTEs in river Hooghly. PCA for the PTEs shows two different sources or components in which first principal component (PC-1) have strong positive loadings on Al, Cd, Co, Cr, Cu, and Fe, having 35.6% variability, while second principal components (PC-2) extracted accounted for 65.9% of variability and strong positive loadings among the PTEs like Mn, Ni, Pb, and Zn in river Hooghly (Fig. 5). The components of PC-1 showed correlation with each other indicating similar sources of their origin. They might be the product of natural weathering and erosion of upstream alumina-silicate (quartz, feldspars, mica) and clay minerals containing catchment rocks as Al and Fe is abundant in earth crust (McDonough and Sun 1995; Dalai et al. 2002); Cr is the product of extensive chemical weathering of bed rocks in plains; Cu, Cd, and Co are associated with the carbonate and Fe–Mn oxyhydroxides containing mineral particles (Achyuthan et al. 2002; Ghrefat and Yusuf 2006; Ghosh et al. 2016; Samanta and Dalai 2016; Manon et al. 2019). Fe–Mn oxyhydroxides also play a significant role in geochemical cycling of PTEs in the water column of river Hooghly (Samanta et al. 2017). The components of PC-2 indicate anthropogenic sources of origin as they are found predominantly in municipal and domestic sewage, agricultural run-off, and effluents from industries (Govil et al. 2008; Wuana and Okieimen 2011; Giri and Singh 2014; Bhattacharya et al. 2015; Ghosh et al. 2016, 2018, 2019a, b). The results of both correlation analysis and PCA suggest that the sources of PTEs in water of river Hooghly are combination of both natural and anthropogenic processes.

Evaluation of water quality index

In this study, the water quality of river Hooghly has been evaluated for drinking and other purposes by comparing with permissible or acceptable limits fixed by WHO and Indian standards of drinking and surface water quality (IS 10500: 2012 and IS 2296: 1982) (ISI 1991; WHO 2008, 2011; BIS 2012). Both basic physicochemical parameters a



b

Component Matrix

	Component	
	1	2
Al	**0.830**	0.089
Cd	**0.687**	0.415
Co	**0.725**	0.246
Cr	**0.647**	0.434
Cu	**0.642**	0.527
Fe	**0.807**	0.259
Mn	0.272	**0.677**
i	0.131	**0.829**
Pb	0.471	**0.624**
Zn	0.278	**0.852**

Fig. 5 Principal component analysis of PTEs. **a** Component plot (VARIMAX rotation); **b** component matrix

like pH, EC, salinity, hardness, alkalinity, DO, COD, TDS, and PTEs like Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn were considered to evaluate the WQI. The relative weight (W_r) values are shown in Additional file 1: Table S8. In summer, monsoon, and winter, the WQI values of river Hooghly are found to vary between 373.7 (S2) and 2196.5 (S15), 365.4 (S2) and 1589.5 (S18), and 478.9 (S2) and 1886.2 (S17), respectively (Fig. 4c). WQI values showed increasing trend towards downstream due to high salinity in the mouth of the river and strong tidal amplitude (Mukhopadhyay et al. 2006; Rudra 2014). Irrespective of sampling locations and seasonal changes, the evaluated WQI value of river Hooghly indicates "very severe" condition of water quality which is unsuitable for direct human consumption. This poor condition of water quality might be due to both natural processes like upstream erosion causing influx of sediment loads (Rudra 2014) and anthropogenic activities (Ghosh et al. 2018, 2019a, b; Bakshi et al. 2018, 2019). Moreover, this mangrove dominated estuarine system acts as a source and sink of nutrients and PTEs. Their flow in river Hooghly has been regulated by the input of litter fall and nutrients associated with the sediment particles, which are released during estuarine transport (Mukhopadhyay et al. 2006).

Evaluation of non-carcinogenic human health risk

Non-carcinogenic health risks in terms of $MDI_{ingestion}$, MDI_{dermal}, $HQ_{ingestion}$, HQ_{dermal}, and HI for summer, monsoon, and winter for adult and child are summarized in Table 3. The $MDI_{ingestion}$ and MDI_{dermal} values of Al are found to be highest in summer and monsoon and for Fe in winter, whereas $MDI_{ingestion}$ and MDI_{dermal} values of Cd are observed to be lowest irrespective of season and age groups. However, the mean $HQ_{ingestion}$ and HQ_{dermal} values for Co and Cr are observed to be highest, whereas mean $HQ_{ingestion}$ and HQ_{dermal} values of Zn are found to be lowest for both age groups throughout the year. The $HQ_{ingestion}$, HQ_{dermal}, and HI value for both adult and children are well below the unity, i.e., safe limits suggesting no immediate adverse non-carcinogenic effect on human health due to ingestion or dermal contact of water from river Hooghly. Moreover, the $MDI_{ingestion}$, MDI_{dermal}, $HQ_{ingestion}$, HQ_{dermal}, and HI values for children are higher in comparison with adults suggesting necessity of long-term measures to mitigate non-carcinogenic human health risks. As evident from Table 3, findings of our study complement with Wang et al. (2017), Xiao et al. (2019), and Gao et al. (2019), and indicate that the children are much more susceptible and vulnerable than adults to PTE exposure. It is also evident from our results that humans are getting PTE exposure predominantly via oral or ingestion route rather than dermal pathways in the study area of River Hooghly where bathing is widely practiced since ages mostly due to religious beliefs, and prawn seed/crab collection is being conducted for the sustenance of livelihood especially for the riverine community of lower Bengal delta. It can also be concluded that at the upstream of river Hooghly, the local inhabitants might be at higher non-carcinogenic risk, as they are consuming river water after filtration and disinfection

Table 3 Seasonal $MDI_{ingestion}$ (adult and child), MDI_{dermal} (adult and child), HQ (adult and child), and HI (adult and child)

Seasons	Parameters	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Summer	MDI ingestion (adult)	3.64E−01	2.81E−04	1.09E−03	1.21E−03	1.80E−03	3.33E−01	7.01E-03	1.53E−03	5.78E−04	2.28E−03
	MDI ingestion (child)	5.30E−01	4.09E−04	1.58E−03	1.76E−03	2.62E−03	4.85E−01	1.02E−02	2.23E−03	8.42E−04	3.33E−03
	MDI dermal (adult)	3.37E−05	2.60E−08	4.02E−07	2.24E−07	1.66E−07	3.08E−05	6.49E−07	5.67E−07	2.14E−07	1.27E−07
	MDI dermal (child)	3.16E−03	2.44E−06	3.77E−05	2.11E−05	1.56E−05	2.90E−03	6.09E−05	5.32E−05	2.01E−05	1.19E−05
	HQ ingestion (adult)	3.64E−04	5.61E−04	3.62E−03	4.04E−04	4.50E−05	1.11E−03	3.50E−04	7.66E−05	4.13E−04	7.61E−06
	HQ ingestion (child)	5.30E−04	8.17E−04	5.27E−03	5.88E−04	6.55E−05	1.62E−03	5.10E−04	1.12E−04	6.01E−04	1.11E−05
	HQ dermal (adult)	1.69E−07	5.20E−06	6.70E−06	1.50E−05	1.39E−08	6.86E−07	8.11E−07	1.05E−07	5.10E−07	2.11E−09
	HQ dermal (child)	1.58E−05	4.88E−04	6.29E−04	1.40E−03	1.30E−06	6.44E−05	7.61E−05	9.86E−06	4.78E−05	1.98E−07
	HI_{Adult}	3.64E−04	5.67E−04	3.63E−03	4.19E−04	4.50E−05	1.11E−03	3.51E−04	7.67E−05	4.13E−04	7.61E−06
	$HI_{Children}$	5.46E−04	1.31E−03	5.90E−03	1.99E−03	6.68E−05	1.68E−03	5.86E−04	1.21E−04	6.49E−04	1.13E−05
Monsoon	MDI ingestion (adult)	3.03E−01	1.93E−04	6.75E−04	7.50E−04	1.32E−03	2.61E−01	5.70E−03	1.30E−03	4.48E−04	1.80E−03
	MDI ingestion (child)	4.41E−01	2.81E−04	9.83E−04	1.09E−03	1.92E−03	3.80E−01	8.29E−03	1.89E−03	6.52E−04	2.62E−03
	MDI dermal (adult)	2.81E−05	1.79E−08	2.50E−07	1.39E−07	1.22E−07	2.42E−05	5.27E−07	4.81E−07	1.66E−07	1.00E−07
	MDI dermal (child)	2.63E−03	1.68E−06	2.35E−05	1.30E−05	1.15E−05	2.27E−03	4.95E−05	4.52E−05	1.56E−05	9.38E−06
	HQ ingestion (adult)	3.03E−04	3.86E−04	2.25E−03	2.50E−04	3.30E−05	8.71E−04	2.85E−04	6.50E−05	3.20E−04	6.00E−06
	HQ ingestion (child)	4.41E−04	5.62E−04	3.28E−03	3.64E−04	4.81E−05	1.27E−03	4.15E−04	9.47E−05	4.66E−04	8.74E−06
	HQ dermal (adult)	1.40E−07	3.57E−06	4.17E−06	9.26E−06	1.02E−08	5.38E−07	6.59E−07	8.92E−08	3.95E−07	1.67E−09
	HQ dermal (child)	1.32E−05	3.36E−04	3.91E−04	8.69E−04	9.56E−07	5.05E−05	6.19E−05	8.37E−06	3.71E−05	1.56E−07
	HI_{Adult}	3.03E−04	3.90E−04	2.25E−03	2.59E−04	3.30E−05	8.72E−04	2.85E−04	6.51E−05	3.20E−04	6.00E−06
	$HI_{Children}$	4.55E−04	8.98E−04	3.67E−03	1.23E−03	4.90E−05	1.32E−03	4.77E−04	1.03E−04	5.03E−04	8.89E−06
Winter	MDI ingestion (adult)	3.53E−01	3.21E−04	1.12E−03	1.22E−03	1.94E−03	3.59E−01	7.82E−03	1.92E−03	6.43E−04	2.72E−03
	MDI ingestion (child)	5.15E−01	4.68E−04	1.62E−03	1.78E−03	2.83E−03	5.22E−01	1.14E−02	2.79E−03	9.36E−04	3.97E−03
	MDI dermal (adult)	3.27E−05	2.97E−08	4.13E−07	2.27E−07	1.80E−07	3.32E−05	7.24E−07	7.10E−07	2.38E−07	1.51E−07
	MDI dermal (child)	3.07E−03	2.79E−06	3.88E−05	2.13E−05	1.69E−05	3.12E−03	6.80E−05	6.66E−05	2.23E−05	1.42E−05
	HQ ingestion (adult)	3.53E−04	6.43E−04	3.72E−03	4.08E−04	4.86E−05	1.20E−03	3.91E−04	9.58E−05	4.59E−04	9.08E−06
	HQ ingestion (child)	5.15E−04	9.36E−04	5.41E−03	5.94E−04	7.07E−05	1.74E−03	5.70E−04	1.39E−04	6.68E−04	1.32E−05
	HQ dermal (adult)	1.64E−07	5.95E−06	6.89E−06	1.51E−05	1.50E−08	7.38E−07	9.05E−07	1.31E−07	5.66E−07	2.52E−09
	HQ dermal (child)	1.54E−05	5.58E−04	6.46E−04	1.42E−03	1.41E−06	6.93E−05	8.50E−05	1.23E−05	5.32E−05	2.37E−07
	HI_{Adult}	3.54E−04	6.49E−04	3.73E−03	4.23E−04	4.86E−05	1.20E−03	3.92E−04	9.59E−05	4.60E−04	9.08E−06
	$HI_{Children}$	5.30E−04	1.49E−03	6.06E−03	2.01E−03	7.22E−05	1.81E−03	6.55E−04	1.52E−04	7.21E−04	1.35E−05

but getting exposure to dissolved PTEs. A regular exposure to PTEs at this level will be toxic towards human causing long-term irreversible health effects (Singh et al. 2018). Moreover, long-term PTE exposure might also lead to bio-magnification and bio-accumulation of PTEs causing different diseases like cardiovascular problems, damage of kidney, renal cortex and liver, osteoporosis, and developmental retardation (Oyem et al. 2015; Paul 2017; Mitra et al. 2018b).

Conclusions

The deterioration of water quality of river Hooghly due to different natural and anthropogenic processes of varied nature is coupled with a combination of biological and/or physicochemical processes. The pH and other physicochemical properties of river Hooghly such as alkalinity and hardness are in direct relation with each other. Salinity, EC, and TDS show an increasing trend towards mouth of the river Hooghly. The river water is found to be unsuitable for direct human consumption as indicated by the WQI. Increased concentration of PTEs is observed especially near the industrial belt and urban centres surrounding the river belt. The varied accumulation of PTEs at different sampling locations might be due to local tidal amplitude, magnitude of discharge of industrial effluent and municipal sewage, and sedimentation. The concentrations of PTEs like Cd, Co, and Zn show temporal increment in concentration compared to other available reports on river Hooghly. The evaluation of non-carcinogenic human health risk of PTEs indicates no immediate adverse impact due to ingestion or dermal contact of water through bathing or drinking from river Hooghly. However, children are much more susceptible to non-carcinogenic health risks than adults. The results suggest implementation of a combination of legislative regulation, awareness campaign among stake holders, monitoring data, and software based models for risk and vulnerability assessment can be used as a useful tool for improvement of water quality in river Hooghly, which serves as a lifeline of the lower Gangetic delta and supports livelihood of millions.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s40562-021-00189-5.

Additional file 1: Table S1. Analytical procedure, Accuracy and Precision data of Standard Reference Materials of (SRM 1643f). **Table S2**. Detailed calculation of WQI, risk assessment and HQ. **Table S3**. Seasonal physico-chemical properties of water of river Hooghly ($n=3$ for each site and season). **Table S4.** Seasonal distribution of PTE in river Hooghly ($n=3$ for each site and season). **Table S5.** Pearson's Correlation analysis between toxic elements in water of river Hooghly ($n=162$). **Table S6.** ANOVA co-efficients of toxic elements between sampling locations in different seasons. **Table S7.** ANOVA co-efficients of toxic elements in river Hooghly considering three seasons. **Table S8.** Relative Weight (Wr) of studied water quality parameters.

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Authors' contributions
PC provided supervision and financial support for this study, and checked the manuscript. JKB checked the manuscript and helped in statistical interpretation. SB and TG helped in analysis. SM helped in sampling, laboratory analysis and manuscript writing. MB helped in laboratory analysis, manuscript writing, data curation, and statistical analysis. SG collected samples, laboratory analysis, writing original draft and its review, data curation, formal analysis, validation, and GIS analysis. All authors read and approved the final manuscript.

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Availability of data and materials
The research data of this study are given in the additional tables and can be obtained by requesting the corresponding author.

Declarations

Competing interests
The authors declare that they have no conflict of interests.

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