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THEMATIC ISSUE

Seasonal variation in heavy metal contaminations in water and sediments of Jamshedpur stretch of Subarnarekha river, India

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Abstract Subarnarekha river, a rain fed peninsular river, passes through the city of Jamshedpur is an important industrial centre of Jharkhand, India. The aim of the study was to investigate the seasonal variability in the water and sediment quality status and to assess the impact of metals contamination in various seasons due to mining and industrial discharge. Concentrations of Zn, Pb, Cr, Cd, Ni, and Cu were determined using ICP-MS. Concentrations of Pb, Ni, and Cd were found quite higher than Indian drinking water standards. Heavy metal concentrations in sediment were found much higher than river water, which was further supported by high contamination factors ($C_f > 6$) for Cd and Pb for all sites in pre-monsoon whereas, high degree of contamination ($C_d > 24$) further ensured heavy pollution. Pollution load index values were found >1 which suggest progressive deterioration of sites whereas, geoaccumulation index recommended sediment quality towards moderately to very strongly polluted condition. Principle component analysis (PCA) was conducted separately for pre- and post-monsoon seasons. PCA (with eigen value >1 focused on two components, irrespective of seasonal variation with 55.21–58.77 % variance) indicated that high metals loading are due to anthropogenic origin, and Zn, Pb, Ni, and Cr have significant loading. Coupling of bioremediation technology with other economically efficient methods could help in solving the problem.

Keywords Sediment · Water · Heavy metal · Pollution · Pollution load · Contamination factor · Geoaccumulation index · Principle component analysis

Introduction

The deterioration of river water quality due to indiscriminate discharge of untreated sewage, industrial waste and other anthropogenic activities like urbanization, population growths and land development along river basin is one of the major concerns throughout the world (Kumar and Maiti 2015). River system provides historical records of metal pollution. Metal pollution analysis allows an establishment of correlation between metal partitioned into the sediment and present in dissolved state in water. Release of heavy metals in river can occur in both particulate and dissolved state, of which the particulate form gets deposited in the river sediment under favourable hydraulic conditions. Interaction between sediment and water can also be responsible for the deposition of metals in sediments (Singh et al. 2005; Wan et al. 2012; Silva et al. 2014). Rivers in urban areas passing through industrial zones are associated with water quality problems because of the unregulated discharge of domestic and industrial waste water into the water bodies that lead to increased levels of heavy metal in the rivers (Lin et al. 2013; Silva et al. 2014). Continuous discharge of toxic heavy metals and oxygen-demanding substances along with solids and colored wastes

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into river water changes the physico-chemical characteristics such as temperature, pH, total dissolved solid (TDS), biochemical oxygen demand (BOD), chemical oxygen demand (COD).

There are basically three reservoirs of metals in aquatic environment: water, sediment and biota (Maiti and Chowdhury 2013). Metal levels in each of these three reservoirs are dominated by a complex dynamic equilibrium governed by a various physical, chemical and biological factors (Saha et al. 2001). Variation in the concentrations of heavy metal content in sediment and water depend on few important parameters like temperature, pH and solute concentration. The behaviour of metals in natural water is the function of the substrate sediment composition, suspended sediment components and water chemistry. Therefore, it is very important to study the status of river water quality in relation with pollution load in order to safeguard public health and to protect the valuable fresh water resources.

The river carries maximum pollution load discharged from upstream while downstream population is suffering to a great extent as they do not have any alternate sources of water (Banerjee et al. 2015). Heavy metal contamination in the sediment is a significant environmental problem because of their toxicity, non-degradability and easy bioaccumulation potential in biota and food chain (Sundaray et al. 2014). The sediments act as sink of contaminants, receive and absorb pollutants resulting from these sources (Liu et al. 2003), and have been recognized as an important indicator of water pollution. Heavy metals are inert in the sediment environment and often considered as conservative pollutants, while they can be released to water in response to certain disturbances and pose potential threats to ecosystems and human health (Moghaddas et al. 2013; Kumar and Maiti 2015; Zhang et al. 2015). Common heavy metal pollutants are Cr, Ni, Cu, Zn, Cd and Pb. These can easily accumulate in the finely grained sediment, remain in water bodies and eventually carried downstream through the river. Thus sediments act as a major sink of heavy metal pollutants in the river bodies.

Pollution indexes like pollution load index (PLI), contamination factors (C_f), and geo-accumulation index (I_{geo}) are widely used to evaluate the pollution in soil and water bodies.

Principal component analysis (PCA) have been widely used to identify the conglomeration of metals and hence is a very effective statistical tool to identify origin of pollutants in a particular environment (Yan et al. 2010; Bu et al. 2010; Candeias et al. 2011; Zhang et al. 2015).

In the recent years, several research articles addressing the pollutant load, pollution pattern and chemical composition of surface waters have been published in India (Sarin and Krishnaswami 1984; Sundaray et al. 2014). Some researchers published papers on water quality of Subarnarekha river (CPCB 2008; Giri et al. 2013) mainly focussed either on physico-chemical parameters (like pH, TDS, conductivity, BOD, COD, organic matter) or on some heavy metals concentration in sediment to address the level of the water pollution. However, in spite of very huge population and several environmental problems, no comprehensive report/research is available on the assessment of seasonal variations in physicochemical characteristics and heavy metal pollution load in water and sediment of Subarnarekha river to identify the source of pollution. The objectives of the present study are to assess the seasonal variations in the water quality of Subarnarekha river and to assess the variations of metal contamination both in water and sediment with pollution indicating indices.

Materials and methods

Study area

Out of 14 major river basins in India, the Subarnarekha basin is one of the smallest (1.9 million hector) but important (dwelling 7 million people) basin which passes through the mineral-rich industrial belt of Jharkhand, West Bengal and Orissa, India. The Subarnarekha river, a rain-fed peninsular river, originates near Nagri village in the Ranchi district, travel 269 km in Jharkhand, 64 km in West Bengal and 62 km in Orissa. The river drains a total area of 19,296 sq km with wet months from June to September. Subarnarekha passes through an important industrial belt of Jharkhand (TATA Steel, TATA motors and other associate companies at Jamshedpur and Adityapur, India) and is often highly polluted due to the industrial effluent, particularly during dry periods. Two major industrial towns Tatisilwai and Jamshedpur with huge population and mining belt Ghatsila are the major polluters of Subarnarekha river. Jamshedpur is situated at the confluence of the rivers Kharkai and Subarnarekha.

Sampling location

Central Pollution Control Board (CPCB), India started national water quality monitoring in 1978 under Global Environmental Monitoring System (GEMS). Parallel to GEMS, in 1984, a National Programme of Monitoring of Indian National Aquatic Resources System (MINARS) selected total 1113 stations spread over ten river basins which are further in 2007 reduced by CPCB into 870 stations on rivers, lentic and sub surface water bodies. For Subarnarekha basin, out of six recommended sampling stations, five sampling locations were selected for the present study (Table 1). The sampling stretch used for the

Table 1 Description of the sampling locations

Site no.	Station name	Location	Description of the sampling location
S1	Kanderbera	23.028 N, 86.119E	About 20 km upstream of river Subarnarekha from industrial city Jamshedpur just touching the southern demarcation line of the basic igneous rock layer
S2	Domohani	22.804 N, 86.202E	Located at the confluence point, where river Kharkai discharges industrial waste from Adityapur, Gamaria and part of Jamshedpur
S3	Mango Bridge	22.836 N, 86.178E	Centrally located in Jamshedpur city, where water is abstracted for potable water production
S4	Asanbani,	22.704 N, 86.321E	Located approximately 18 km at downstream of the Jamshedpur city
S5	Galudih	22.644 N, 86.410E	Located approximately 35 km of downstream of the Jamshedpur city

study was approximately 50 km long. The geographic locations of sampling stations are presented in Fig. 1.

Collection of water and sediment samples

To study the seasonal pollution load, water and sediment samples were collected from five sampling stations during three seasons, i.e. summer (March to May, 2013), rainy (July to September, 2013) and winter (November 2013 to January 2014) for both physico-chemical and heavy metal analysis.

Water sample processing and analysis

Grab sampling of water was done in triplicate and mixed to get a composite sample for each site. Collected samples were kept in polyethylene bottles (1 L capacity). For heavy metal analysis, 500 mL of water sample was preserved by acidifying with concentrated (conc.) HNO_3 [nitric acid, Analytic reagent (AR) grade] to $pH < 2$. For bacteriological analysis, 200 mL water sample was collected in 250 mL pre-sterilised borosilicate glass bottle (i.e., ¾ full) as per APHA (2012). Samples were kept on ice box and brought to the laboratory, stored in refrigerator (4 °C) for further analysis. All the bacteriological analyses were started within 6 h from the time of sample collection.

Parameters like temperature, pH, and conductivity were measured in situ. The temperature was measured with calibrated mercury filled glass thermometer, turbidity by turbidity meter (Eutech) and pH with potable pH-meter (HACH, Sension-1). For dissolved oxygen (DO) estimation, samples were collected in a 300 mL of BOD bottle, and fixed in the field by addition fixing reagents (1 mL of $MnSO_4$, followed by 1 mL of alkaline-iodide-azide solution). Other physico-chemical parameters like turbidity, TDS, DO, COD, BOD, chloride, alkalinity, hardness, and bacteriological analysis were analysed in laboratory as per APHA (2012).

For heavy metal (Cu, Cd, Zn, Ni, Cr and Pb) analysis, 100 mL of acid treated samples was heated with 5 mL conc. HNO_3 on hot plate at 95 °C, reduced to a final volume of 10 mL. The beaker and watch glass were washed down with deionised water and the whole solution was filtered through membrane filter paper soaked in 1 N HNO_3 and metals were analysed by inductively coupled plasma-mass spectrophotometer (ICP-MS), (PerkinElmer Sciex, Elam DRC-e, Analytical West Inc., Corona, California, USA).

Sediment sample processing and analysis

Composite grab-samples of sediment were collected in ten numbers (each with three replicates) at a depth of 0–15 cm using plastic spoon to avoid metal contamination from metallic spoon, transferred in a polyethylene air tight zip-lock bags and transported in an ice box to the laboratory. Samples were air dried for 5 days and sieved through <1000 µm stainless steel sieve to remove large rock debris, mollusc skeletons and organic debris. The homogenised sediment samples were further sieved through <63 µm size nylon sieve and dried at 105 °C in hot air oven until the constant weight was achieved. For analysis of metals (Cu, Cd, Zn, Ni and Cr), accurately weighed 0.2 g of sample was taken in Teflon beaker, digested with mixture of conc. HF (Hydrofluoric acid, 40 %, Merck), 1:1 H_2SO_4 (Sulphuric acid, 98 %, Merck) and 0.5 N HCl (Hydrochloric acid, 35 %, Merck) in a ratio of 8:5:5 (v/v/v) on a hot plate until the complete dissolution of sample was achieved. Similarly, Pb was extracted using mixture of HF and 3 % HNO_3 (Nitric acid, 69–71 %, Merck) in the ratio of 3:5 (v/v). The digested samples were filtered through membrane filter and were analyzed by ICP-MS.

Sediment pollution load analysis

Heavy metal contamination in sediments reported by several researchers by computing statistical indices; (1) contamination factors (C_f) and (2) geoaccumulation index (I_{geo}) (Suthar et al. 2009; Kumar et al. 2013). Depending on contamination factor further calculation has been carried out to find degree of contamination (C_d) and pollution load

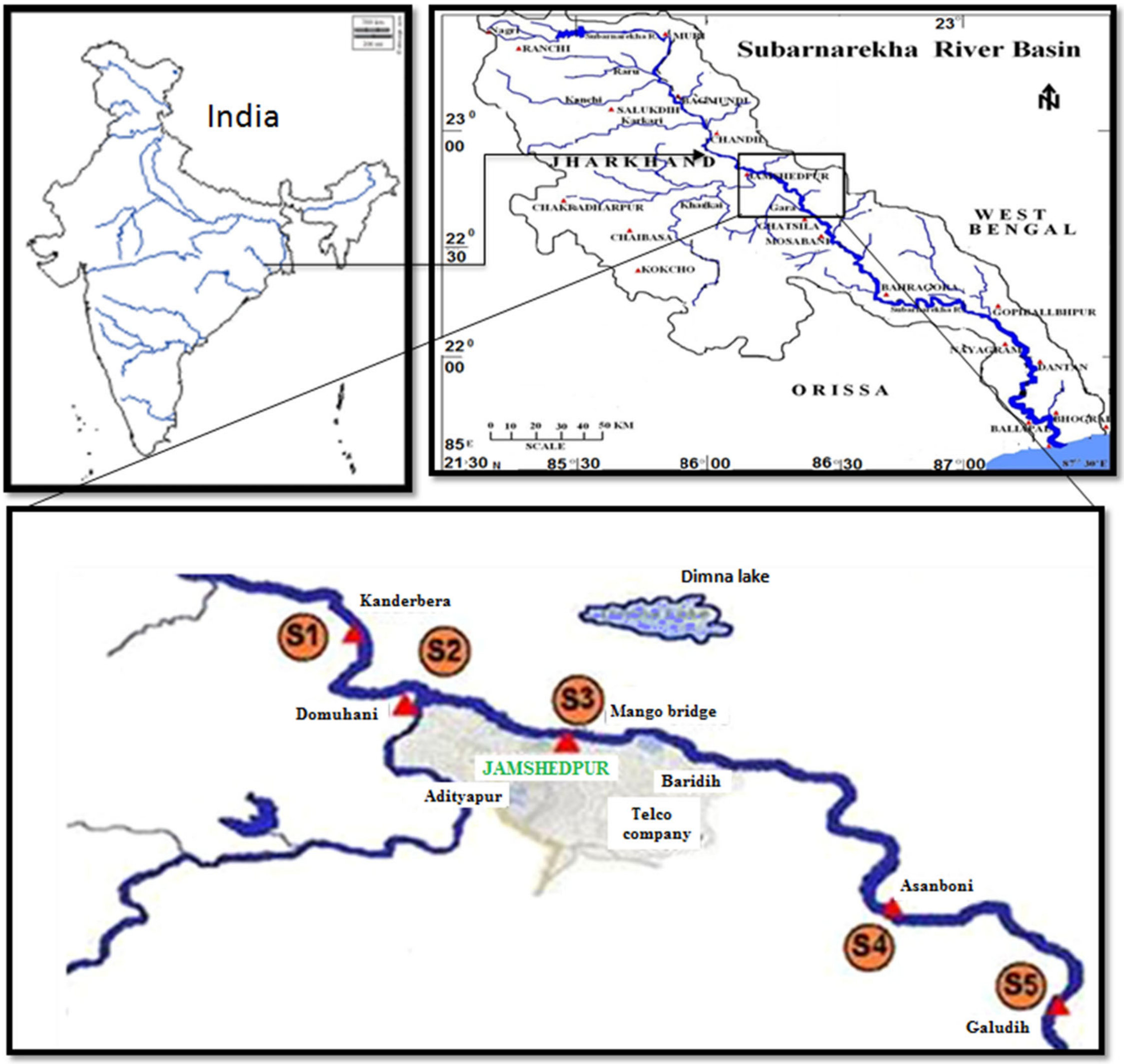

Fig. 1 Location map of study area showing Subarnarekha river basin with respect to India. Refer to Table 1 for description of sampling sites

index (PLI) which helps in determining the magnitude of heavy metal deposition in river bed sediment for each sampling site (Table 2).

The C_f is the ratio of heavy metal concentration in the sediment to the average shell value of earth crust (Hakanson 1980). For background concentration of metals, the average shale values for different metals are used for Pb: 20 mg/kg, Ni: 68 mg/kg, Cd: 0.3 mg/kg, Cr: 90 mg/kg, Zn: 95 mg/kg and, Cu: 45 mg/kg (Turekian and Wedepohl 1961). Similarly, Geoaccumulation (I_{geo}) is the assessment of contamination by comparing the current and past concentrations originally used with bottom sediment and were determined using Muller's equation (Muller 1979).

The C_d can be calculated by sum of all C_f values divided by total number of C_f, whereas PLI is integrated index which combines all heavy metals to one index and allows to compare the pollution status of various sites together without considering the grain-size and natural geochemical variability (Tomlinson et al. 1980).

Quality assurance, quality control and statistical analyses

For quality assurance and quality control (QA/QC), analytical reagent (AR, Merck, Germany) grade, NIST traceable chemicals, reagent and double distilled and Milli-Q Millipore water were used for preparation of all reagents and calibration standards. Method validation was performed with NIST traceable standard reference material NIST-1646a (Estuarine sediment), reagent blanks, duplicates and spiked samples. Detection limits of six metals (mg/kg) were: Cr: 0.02; Cu: 0.02; Pb: 0.02; Cd: 0.01; Ni: 0.02; and Zn: 0.02 with agreement levels ranging from 90.8

Table 2 Description of factors and indices for the assessment of metal pollution in sediment

Factor/index	Formula	Notations	Threshold values
Contamination factor (C_f)	$C_f = [C]_{\text{sediment}}/[C]_{\text{shale}}$	$[C]_{\text{sediment}}$ = metal concentration in sediment $[C]_{\text{shale}}$ = average shell value of earth crust	CF = 0: none; CF = 1: none to medium; CF = 2: moderate; CF = 3: moderate to strong; CF = 4: strongly polluted; CF = 5: strong to very strong; CF = 6: very strong
Degree of contamination (C_d)	$C_d = \frac{1}{N}\sum_{i=1}^{N} C_f i$	$\sum_{i=1}^{N} C_f i$ = sum of all contamination factors; N = number of contamination factors	$C_d < 6$: low degree of contamination; $6 \le C_d < 12$: moderate degree of contamination; $12 \le C_d < 24$: considerable degree of contamination; and $C_d > 24$: high degree of contamination
Geoaccumulation index (I_{geo})	$I_{\text{geo}} = \log 2\left(\frac{C_{\text{sediment}}}{1.5\ B_n}\right)$	C_{sediment} = metal concentration in sediment; Bn = geochemical background metal concentration; 1.5 = correction factor for the background matrix due to lithogenic effects	$I_{\text{geo}} \le 0$: practically uncontaminated; $0 < I_{\text{geo}} < 1$: uncontaminated to moderately contaminated; $0 < I_{\text{geo}} < 2$: moderately contaminated; $2 < I_{\text{geo}} < 3$: moderately to heavily contaminated; $3 < I_{\text{geo}} < 4$: heavily contaminated; $4 < I_{\text{geo}} < 5$: heavily to extremely contaminated; $5 < I_{\text{geo}}$: extremely contaminated
Pollution load index (PLI)	$\text{PLI} = (C_f 1 \times C_f 2 \times C_f 3 \times \cdots C_f \text{N})^{1/N}$	N = number of metals; C_f = contamination factor	PLI < 0: unpolluted; 0 < PLI ≤ 1: baseline levels of pollutant present; 1 < PLI ≤ 10: polluted; 10 < PLI ≤ 100: highly polluted; PLI > 100: progressive deterioration of environment

to 101.5 %. To calibrate the instrument, sample blank and standards were run after every ten readings. The relative standard deviation (RSD) of each group of replicate samples was <10 % for all the metals maintaining calibration coefficients at ≥0.999.

The mean and standard deviations were calculated using MS Excel 2007 (Microsoft Inc.). Hierarchical cluster analysis (CA) was performed on the normalized dataset of heavy metals to group the metals using Ward's method with Euclidean distances as measure of similarity. Principal component analysis (PCA) was performed separately for summer and winter seasons.

Results and discussion

Seasonal variation of river water quality

The effect of industrial discharge in river water quality can be assessed by seasonal monitoring of physico-chemical characteristics of water (Table 3). The mean pH value ranged between 7.6 and 8.0. Among all the sites, pH varied between 7.6–7.8 during rainy and winter seasons due to high volume of water flow as a result of abundant rainfall. Overall pH values were found within acceptable limits of all classes of designated use of river water (CPCB 2008). The conductivity values varied between 135 and 305 µS/cm for all the seasons. Chloride content found highest during winter (22–26 mg/L) and lowest in rainy (6.5 mg/L) season which was far below the maximum permissible concentration of 250 mg/L (CPCB 2008). DO, BOD_5 and COD are good indicator of degree of organic pollution in a river (Maiti 2001). DO contents varied between 6.0 mg/L during summer to 7.5 mg/L during winter. During winter, lower temperature facilitates oxygen dissolving capacity of water. The highest BOD value (4.5 mg/L) was found for S3 (just d/s of confluence point) during summer season which was below tolerable range of 5–20 mg/L. While in all other sites, values ranged between 2.2 mg/L for S1 and 3.8 mg/L for S4 site. Data analysis showed that BOD was predominant in confluence point and within 1 km d/s (sites S2 and S3) in all the seasons which may be because of discharge of domestic waste in and around these points of the river. Average BOD values are complying with class *C* water as prescribed by CPCB (2008). Similar trend was found for COD and ranged between 20 and 28 mg/L in summer while in other seasons the values were found low and ranged between 7 and 8 mg/L. Subarnarekha being a rain fed river, during pre-monsoon season, water flow was found minimum and quantity of effluent discharge was approximately same as it was observed in other seasons.

Table 3 Seasonal variation in physicochemical parameters (mg/L) of water sample of Subarnarekha river

Parameter	Summer					Rainy				Winter				
	S1	S2	S3	S4	S5	S1	S2	S3	S4	S1	S2	S3	S4	S5
Turbidity(NTU)	4.3 ± 0.10	7.0 ± 0.06	8.0 ± 0.15	3.2 ± 0.1	2.6 ± 0.1	100 ± 1.0	198 ± 2.64	175 ± 3.60	112 ± 4.35	5.5 ± 1.1	6.0 ± 1.1	9.0 ± 0.27	8.0 ± 0.2	4.0 ± 0.15
pH	7.8 ± 0.58	7.9 ± 0.1	8.0 ± 0.1	7.7 ± 0.06	7.8 ± 0.1	7.7 ± 0.1	7.7 ± 0.06	7.8 ± 0.1	7.6 ± 0.22	7.8 ± 0.13	7.8 ± 0.07	7.8 ± 0.1	7.7 ± 0.2	7.7 ± 0.2
Conductivity (mho/cm)	151 ± 1.0	242.6 ± 0.6	248.3 ± 0.6	205 ± 1.1	194 ± 0.58	152 ± 2.0	135 ± 3.6	168 ± 2.6	147 ± 3.0	150 ± 4.1	243 ± 1.0	225 ± 0.1	305 ± 0.87	286 ± 2.0
Chloride (mg/L)	12.0 ± 0.12	16.0 ± 0.15	18.0 ± 0.31	13.0 ± 0.12	15.1 ± 0.3	6.5 ± 0.36	7.0 ± 0.17	7.2 ± 0.13	8.0 ± 0.15	13 .0 ± 1.5	14.0 ± 0.53	15.0 ± 1.0	25.5 ± 0.81	22.0 ± 0.44
DO (mg/L)	6.4 ± 0.10	6.2 ± 0.25	6.0 ± 0.12	6.4 ± 0.06	6.7 ± 0.1	7.0 ± 0.2	6.0 ± 0.27	7.1 ± 0.36	7.2 ± 0.17	7.2 ± 0.1	7.1 ± 0.26	7.4 ± 0.43	7.3 ± 0.46	7.5 ± 0.3
BOD5 (mg/L)	3.2 ± 0.06	4.1 ± 0.12	4.5 ± 0.1	3.07 ± 0.12	3.5 ± 0.15	2.2 ± 0.53	3.2 ± 0.2	3.4 ± 0.46	3.0 ± 0.26	2.4 ± 0.46	3.1 ± 0.43	3.0 ± 0.30	3.8 ± 0.27	2.7 ± 0.27
Ammonia Nitrogen (mg/L)	2.0 ± 0.06	2.5 ± 0.15	2.0 ± 0.11	0.63 ± 0.06	1.1 ± 0.1	1.6 ± 0.1	2.5 ± 0.27	2.0 ± 0.27	1.2 ± 0.17	1.7 ± 0.17	3.0 ± 0.20	2.80 ± 0.17	1.2 ± 0.2	1.5 ± 0.27
COD (mg/L)	20.1 ± 0.11	24.1 ± 0.23	28.0 ± 0.11	21.4 ± 0.53	24.1 ± 0.3	7.7 ± 0.25	8.1 ± 0.25	7.4 ± 0.55	7.1 ± 0.11	7.16 ± 0.57	7.57 ± 0.29	59 ± 3.12	7.53 ± 0.25	7.03 ± 0.21
Faecal Coliform (MPN/100 mL)	16000	92000	160000	13000	47666	35000	33000	33000	28000	24000	92000	160000	160000	160000

Therefore, the dilution of pollutants in river water was minimum during summer season, which impart high COD.

The fecal coliform (FC) density ranged between 16,000 and 24,000 MPN/100 mL in S1 site, while in summer it exceeded the standards (Maiti 2001; CPCB 2008). Several researchers had reported very high value of coliform density for Indian river (Maiti 2001; Vinay et al. 2005; Derwich et al. 2011). In summer, FC density was found highest (160,000 MPN/100 mL) at S3 site which indicates untreated sewage discharge and its high multiplication during summer season. Similarly, for S4 and S5 sites, the FC density ranged between 92,000–160,000 MPN/100 ml during summer and winter seasons which further indicates the anthropogenic activities (cattle bathing, washing, and human bathing) as well as waste discharge (particularly faeces of cattle, sheep, goats, etc.).

Seasonal variation of heavy metals in river water

Seasonal variation of heavy metal concentrations in water of Subarnarekha river are shown in Table 4. Considering all the heavy metal contents in water, Zn was observed as predominant metal in river water. Maximum concentration of 0.49 mg Zn L^{-1} was found in S2 site followed by S3 and S4 during rainy season whereas minimum concentration of 0.07 mg Zn L^{-1} was found during winter season. Average Zn content was found high during rainy season and ranged between 0.28–0.49 mg L^{-1} whereas for summer and winter, it ranged between 0.22–0.33 mg L^{-1} and 0.07–0.16 mg L^{-1}, respectively which were found below the prescribed concentration of 15 mg L^{-1} (CPCB 2008). High Zn content was found in S2 site (influx of Kharkai river into Subarnarekha river) may be due to often discharge of considerable amount of untreated industrial effluents in Kharkai river from small scale galvanizing steel and iron industries, rubber manufacturing units of Adityapur.

The second most predominant metal in water was Cu which was found higher in summer season as compared to two other seasons and ranged between 0.01 and 0.03 mg L^{-1}. Maximum concentration of 0.03 mg L^{-1} was found in S2 site during summer which is comparatively much lower than that of prescribed CPCB standard (1.5 mg L^{-1}). During rainy and winter seasons, the Cu concentration varied between 0.01 and 0.02 mg L^{-1} indicating its maximum concentration in S2 site which specifies insignificant source of pollution. On the other hand, though the

Table 4 Seasonal variation in heavy metal concentrations (mg L^{-1}) in water samples of Subarnarekha river

Metal	Summer					Rainy					Winter				
	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5
Cd	bdl	bdl	bdl	bdl	bdl	bdl	bdl	0.01 ± 0.001	0.01 ± 0.001	bdl	bdl	0.01 ± 0.002	0.02 ± 0.004	0.01 ± 0.002	0.01 ± 0.002
Cu	0.01 ± 0.001	0.03 ± 0.001	0.02 ± 0.001	0.02 ± 0.001	0.02 ± 0.002	0.01 ± 0.004	0.02 ± 0.003	0.01 ± 0.001	0.02 ± 0.003	0.02 ± 0.002	0.01 ± 0.004	0.02 ± 0.002	0.02 ± 0.004	0.01 ± 0.007	0.01 ± 0.007
Pb	bdl	bdl	bdl	bdl	0.17 ± 0.01	bdl	bdl	bdl	bdl	bdl	0.02 ± 0.003	0.04 ± 0.017	0.38 ± 0.12	0.08 ± 0.002	0.05 ± 0.001
Ni	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	0.01 ± 0.001	0.02 ± 0.008	0.02 ± 0.002	0.03 ± 0.01	0.01 ± 0.002
Zn	0.22 ± 0.020	0.33 ± 0.015	0.27 ± 0.015	0.26 ± 0.006	0.25 ± 0.026	0.28 ± 0.02	0.49 ± 0.006	0.421 ± 0.003	0.38 ± 0.01	0.23 ± 0.011	0.07 ± 0.01	0.08 ± 0.017	0.16 ± 0.02	0.09 ± 0.017	0.08 ± 0.002
Cr	0.01 ± 0.001	0.02 ± 0.003	0.02 ± 0.001	0.01 ± 0.001	0.019 ± 0.001	bdl	bdl	bdl	bdl	bdl	0.01 ± 0.02	0.02 ± 0.04	0.02 ± 0.02	0.01 ± 0.02	0.02 ± 0.003

bdl below detection limit

maximum concentration of Cr did not exceeded the CPCB standard (0.05 mg L^{-1}), Cr showed higher range (0.01–0.023 mg L^{-1}) in winter followed by summer and at below detectable limit (bdl) during rainy season. Maximum concentration of 0.023 mg L^{-1} was found in S3 site followed by S2 (0.02 mg L^{-1}) in winter which further indicate possibilities of point source of pollution due to discharge of industrial effluent at confluence point and within 1 km d/s (S2 and S3 sites).

Metals like Cd, Pb, and Ni showed their presence particularly in winter season. Highest concentration of Cd (0.02 mg L^{-1}) and of Pb (0.38 mg L^{-1}) was found in S3 site and both the metals were found exceeding CPCB (2008) standard limits (Cd: 0.003 mg L^{-1} and Pb: 0.01 mg L^{-1}). It further supports the chances of direct anthropogenic pressure (increased industries and sewage discharge) in river nearby these sites. The overall concentration of heavy metals in river water showed an elevated trend in comparison with earlier studies. However, in the current study, metal concentrations were found high in S2 and S3, though a decreasing trend was observed in downstream sampling sites, which may be due to increased urbanisation (expansion of residents, roads and subsequent disposal of industrial and domestic wastes in water) near S2 and S3 sites.

Seasonal variation of heavy metals in river sediment

Seasonal variations of heavy metal concentrations in sediment of Subarnarekha river are shown in Table 5. Analysis of sediments revealed that the concentration of heavy metals in sediment were considerably far higher than those determined in water samples from same sampling sites. Considering pre- and post-monsoon seasons, metal concentrations in sediment were found in the order of Zn >Ni >Pb >Cr >Cu >Cd. Except Zn, all heavy metals showed higher range of concentration during summer. High metal concentrations in the study area may be because of excessive industrial discharge from industries like iron, steel, fertilizers, pesticides, heavy vehicles as well as discharge of domestic sewage coupled with natural geogenic processes (Mathivanan and Rajaram 2014).

During winter, maximum concentration of Zn was found in S3 site (1.25 g kg^{-1}) followed by S2 site (1.13 g kg^{-1}), whereas in summer season, highest concentration of 1.05 g kg^{-1} was found for S2 site. The concentrations of Zn in other sites varied between 0.09 g kg^{-1} (S5 site in winter) to 1.02 g kg^{-1} (S3 site in summer). The possible reason for the presence of high Zn content in the sediment samples of river may be due to the huge discharge of industrial waste from metal plating industries and mixing of runoff with fertilizers residue (Guo and He 2013; Mathivanan and Rajaram 2014; Benhaddya et al. 2014).

Highest concentration of 0.17 g Cr kg^{-1} was found in S3 site followed by 0.16 g Cr kg^{-1} in S2 during summer whereas minimum for S5 site (0.048 g kg^{-1}) during winter season. Average Cr concentration was found higher in summer as compared to winter season. High concentrations of Cr in S2 and S3 sites suggested the possibilities of significant discharge and accumulation of effluent from dye

Table 5 Seasonal variation in heavy metal concentrations (mg L^{-1}) in sediment samples of Subernarekha river

Metal	Summer					Winter				
	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5
Cd	0.015 ± 0.002	0.02 ± 0.003	0.035 ± 0.001	0.03 ± 0.03	0.025 ± 0.001	bdl	bdl	bdl	bdl	bdl
Cu	0.06 ± 0.001	0.08 ± 0.001	0.1 ± 0.001	0.11 ± 0.001	0.1 ± 0.002	0.019 ± 0.004	0.06 ± 0.002	0.055 ± 0.004	0.025 ± 0.007	0.053 ± 0.007
Pb	0.52 ± 0.002	0.62 ± 0.003	0.805 ± 0.002	0.66 ± 0.002	0.795 ± 0.01	bdl	bdl	bdl	bdl	bdl
Ni	0.24 ± 0.002	0.32 ± 0.002	0.43 ± 0.002	0.34 ± 0.002	0.38 ± 0.002	0.01 ± 0.002	0.04 ± 0.001	0.04 ± 0.002	bdl	bdl
Zn	0.34 ± 0.02	1.05 ± 0.015	1.03 ± 0.015	0.36 ± 0.006	0.32 ± 0.026	0.59 ± 0.01	1.13 ± 0.17	1.25 ± 0.02	0.77 ± 0.017	0.09 ± 0.004
Cr	0.11 ± 0.001	0.17 ± 0.003	0.17 ± 0.001	0.06 ± 0.001	0.14 ± 0.001	0.075 ± 0.002	0.135 ± 0.004	0.165 ± 0.002	0.049 ± 0.002	0.048 ± 0.003

bdl below detection limit

and chrome plating industries (Sarkar et al. 2011; Mathivanan and Rajaram 2014; Benhaddya et al. 2014).

Overall concentration of Cu was found high in summer season may be because of the high pH value which facilitates its accumulation in sediment (Singh et al. 2005). High concentrations of Cu were found in S3, S4, and S5 sites and ranged between 0.1 and 0.105 g kg^{-1} in summer season whereas lowest value of 0.025 g kg^{-1} was observed in S4 site during winter season. Because of the presence of Cu mines, the concentrations of Cu were found high in the downstream samples of S4 and S5 sites (Giri et al. 2013). Similarly, concentration of Ni was found high and ranged between 0.23 and 0.42 g kg^{-1} in summer while during winter it ranged between 0.001 and 0.04 g kg^{-1}. The maximum Ni content was found for S3 (0.42 g kg^{-1}) site followed by S5 (0.37 g kg^{-1}), and S4 (0.33 g kg^{-1}) sites. A typical trend for both Pb and Cd was observed as these metals were found low in concentration during winter season for the all sampling sites, whereas showed higher concentration in summer season. Maximum content of 0.27 g Pb kg^{-1} was found in S3 site while lowest concentration was observed in S1 (0.17 g Pb kg^{-1}) for summer season. Similarly, Cd was found maximum in S3 (0.0087 g kg^{-1}) followed by S4 (0.0075 g kg^{-1}) site while minimum concentration of 0.0037 g Cd kg^{-1} was found in S1 site during summer season. Low pH releases free metal ions into water column from sediment and therefore concentration of metal ion decreased in sediment. The reverse condition seems to be happened when pH of water rises (Kumar et al. 2013). This trend was observed in accumulation of heavy metals in sediment of Subarnarekha river. Mean pH value was found high in summer than rainy and winter seasons which may facilitate the accumulation of heavy metals in sediment.

Multivariate analyses for metal pollution source identification

Principal component analysis (PCA) has been widely used to identify the conglomeration of metals and a very effective statistical tool to identify origin of pollutants in a particular environment. It helps in identifying a reduced set of underlying variables or factors that focus on the variance of a combined data set to explain the correlation matrix of the data. This new set of data is known as principal component (PC) which is a linear combination of the initial variables (Zhang et al. 2013; Guo and He 2013; Wang et al. 2014).

Normalized variables (original variables) were transformed into the rotated components to extract the significant principal components (reducing the contribution of variables with minor significance). Both the PCAs indicated correlated variables under two principal components

PC1 and PC2 and plays an important role in explaining metal pollution of this study area with seasonal variation (Table 6). During summer season, the first component PC1 showed 58.77 % variance out of total variance and consists of heavy metals like Cd, Cu, Pb and, Ni whereas PC2 contributed 34.65 % of the total variance and included Cr (>0.94) and Zn (0.92). Considering winter season, high loading values were found for Cr, Ni and Zn under PC1 with 55.21 % total variance, whereas PC2 specifically included Cu and Pb with 28.53 % of total variance. Therefore, high loading of these metals in PC1 for both the seasons seems to be associated with anthropogenic origin and may be originated from similar pollution sources. Similar trend was also noticed in hierarchical CA dendograms (Figs. 2, 3). A cluster of Pb, Ni, Cr, Zn was obtained in one cluster, Cu and Pb in second cluster whereas C_d was independently associated. The results were found similar to PC's of both summer and winter, proving the anthropogenic origin of these metals. Discharge of industrial effluent, mining activities, sponge iron, fabrication, galvanising, painting and electroplating industries along the bank of river seems to be associated with high concentration of these metals especially during summer season. Metal loading in PC2 is assumed to be related to the agricultural activities as well as municipal and industrial wastewater discharge and to some extent originated from the erosion and weathering of rock and mineral. It is notable that high loading of Zn, Cr, Ni, and Pb in PC1 and PC2 with seasonal variation is indicative of both geogenic and anthropogenic pollution as primary sources.

Sediment pollution load analysis

The values of C_f, C_d, and PLI for all the sites during summer and winter season are depicted in Table 7. The calculated C_f for Cr and Cu during summer were ranged between $1 \leq C_f < 3$, indicating towards moderate contamination for all sampling sites, except in d/s of Jamshedpur city (S4). Contrarily, during winter season C_f of same metals were <1 for all sites except S2 (confluence point of Kharkai river) and S3 (stressed with anthropogenic activities) where C_f were ranged between $1 \leq C_f < 3$ pointing towards moderate contamination. Similarly, C_f value of Ni during summer ranged between $3 \leq C_f < 6$ i.e., considerable contamination for all sites. However, reverse condition found during winter where C_f of Ni was <1 showing low contamination for all sites. This trend relatively endorses the metal deposition process in sediment from water due to pH variation during summer as compared to winter.

Table 6 Rotated component matrix of principal component (PC) for the sediments samples of Subarnarekha river during summer and winter season

Metal	Summer		Winter	
	PC1	PC2	PC1	PC2
Cr	0.070	**0.945**	**0.956**	0.235
Cd	**0.961**	−0.262	−0.655	−0.180
Cu	**0.967**	−0.070	0.478	**0.824**
Ni	**0.925**	0.288	**0.980**	0.183
Pb	**0.898**	0.428	−0.080	**0.943**
Zn	0.017	**0.921**	**0.880**	−0.152
Eigen values	3.526	2.079	3.313	1.712
Total % of variance	58.775	34.655	55.219	28.534
Cumulative variance (%)	58.775	93.430	55.219	83.753

Rotated component solutions above 0.8 are indicated in bold

Notably, C_f for Cd and Pb portrayed $C_f > 6$ for all sites during summer season which pointed towards very high contamination. C_f values for Zn were found highest in S3 (13.2) followed by S2 (11.9) during winter season comprising $C_f > 6$ for all sites and indicates towards very high contamination. Even in summer, C_f values of S2 and S3 sites were >6 whereas rest of the sites it showed between $3 \leq C_f < 6$. Considering metals shell value, high C_f of Cd, Pb and Zn for all the sites leads towards assumption of serious industrial, agricultural and anthropogenic pollution.

The overall values of C_d for all the sampling sites during summer season were found between $6 \leq C_d < 12$ which suggests moderate degree of contamination. During premonsoon period, maximum C_d values were found for S3 (10.62) followed by S4 (7.93) and S5 (7.78). On the other hand, during winter C_d values were <6 for all sites specifying low degree of contamination. High values of C_d in S3 for both the seasons may be because of considerable anthropogenic activities and industrial discharge in upstream or nearby this site where the metals get carried out and accumulated in sediment of this site.

This study revealed that PLI values are comparatively higher in all the sites during summer than winter. During summer, most of the sites showed PLI > 1 which represents progressive deterioration of sites and showed baseline levels of pollutants (PLI ≤ 1) during winter. PLI was found highest in summer at S3 (6.92) followed by S2 (5.54) which confirms the same trend. Pb, Cd and Zn are the major contributors for high PLI value which confirms that the sediment quality of Subarnarekha river is under progressive deterioration which may be a result of discharge of industrial effluent in Subarnarekha and Kharkai rivers along with influx of high volume of domestic sewage from Jamshedpur city.

For both summer and winter, I_{geo} values of all sites for Cr and Cu were found <0 to <1 describing the sites are unpolluted to moderately polluted (Table 8). Contrarily, these sites are found with high I_{geo} values of Cd, Pb, Zn, and

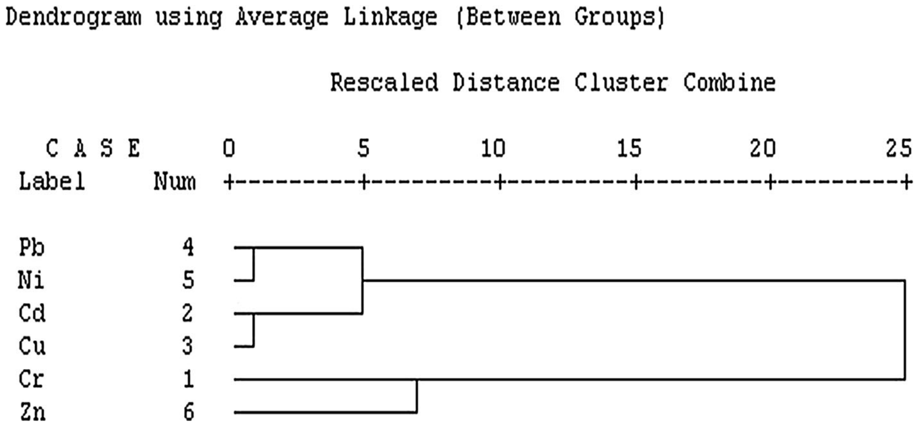

Fig. 2 Dendogram showing cluster of metals during summer season

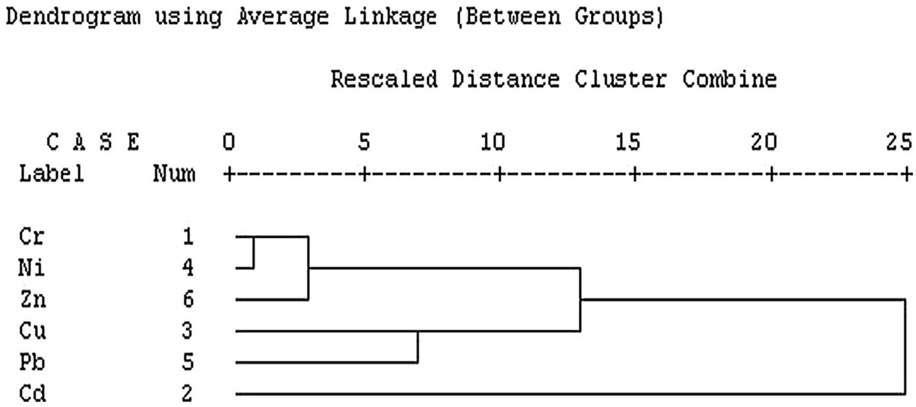

Fig. 3 Dendogram showing cluster of metals during winter season

Table 7 Metal contamination factor (C_f), degree of contamination (C_d) and pollution load index (PLI) of river sediment at five sampling sites during summer and winter seasons

Metal/index	Summer					Winter				
	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5
Cr	1.16	1.83	1.88	0.61	1.55	0.83	1.52	1.83	0.54	0.53
Cd	**12.5**	**16.6**	**29.16**	**25.0**	**20.83**	**3.00**	**3.33**	**3.33**	**5.00**	**3.33**
Cu	1.33	1.77	2.22	2.33	2.22	0.42	1.33	1.22	0.55	1.17
Pb	**8.66**	**10.33**	**13.41**	**11.0**	**13.25**	0.006	0.007	0.009	0.007	0.009
Ni	**3.45**	**4.70**	**6.25**	**4.92**	**5.51**	0.18	0.51	0.59	1.00	1.00
Zn	**3.52**	**11.05**	**10.78**	**3.73**	**3.31**	**6.21**	**11.89**	**13.21**	**8.15**	**0.95**
C_d	5.10	7.72	10.62	7.93	7.78	1.79	3.12	3.39	2.56	1.19
PLI	3.55	5.54	6.92	4.39	5.08	0.436	0.812	0.896	0.663	0.510

Bold values indicate high contamination of metals based on contamination factor

Ni in summer season. Cd displayed highest I_{geo} values in S3 (4.28) followed by S4 and S5 indicating these sites are very strongly polluted whereas, its values for same sites during winter belongs to class 2 (moderately polluted). I_{geo} values of Pb for all sites in summer season ranged between 2.52 (S1) to 3.16 (S3) and showed the sites are moderately to strongly polluted. Significantly, during winter I_{geo} values of Pb for all these sites restricted in class 0 ($I_{geo} \leq 0$) pointing towards unpolluted condition. In case of Zn, considering both the seasons, all the sampling sites belongs to class 2 and 3 which indicates moderate to strong pollution (except S5: class 0 during post monsoon period). S3 site was found with highest I_{geo} values for summer and winter (2.84 and 3.14, respectively) followed by S2 which indicates Zn as a major pollutant for bed sediment. I_{geo} values of Ni falls in class 2 ($I_{geo} < 2$) with highest I_{geo} value of 2.05 at S3 site indicating moderately polluted status during summer season while in winter it belongs to class 0. On a whole, I_{geo} values suggested that the sediment quality is indicating towards moderate to very strong pollution.

Table 8 Geoaccumulation index (I_{geo}) of heavy metals in the river sediment at five sampling sites during summer and winter seasons

Metal	Summer					Winter				
	S1	S2	S3	S4	S5	S1	S2	S3	S4	S5
Cr	−0.37	0.286	0.325	−1.29	0.047	−0.853	0.021	0.286	−1.47	−1.5
Cd	**3.05**	**3.47**	**4.28**	**4.06**	**3.79**	1.00	1.15	1.15	1.73	1.15
Cu	−0.173	0.238	0.565	0.635	0.565	−1.83	−0.173	−0.298	−1.44	−0.358
Pb	**2.52**	**2.78**	**3.16**	**2.87**	**3.14**	−7.96	−7.74	−7.38	−7.74	−7.38
Ni	1.2	1.64	2.05	1.71	1.87	−3.05	−1.56	−1.35	−0.585	−0.585
Zn	**1.23**	**2.88**	**2.84**	**1.31**	**1.14**	**2.05**	**2.99**	**3.14**	**2.44**	−0.66

Bold values indicate high contamination of metals based on geoaccumulation index

The sediment pollution load analysis showed high degree of metal contamination with a trend of increased values of C_d and PLI in the downstream of Subarnarekha river. Similar results on sediment quality were also previously reported (Giri et al. 2013). A comparison of metal concentrations of present study with other Indian river and sediments are presented in Supplementary Tables.

Conclusions

The present study evaluated the seasonal variation in the river water quality in respect of heavy metal contamination level in Subarnarekha river while passing through a major industrial town. Heavy metals like Zn, Cu, and Cr were found predominant in water with seasonal variation however they did not exceeded the CPCB standard. Contrarily, Cd, Pb, and Ni were found particularly in winter season and exceeded the CPCB standard in few sites due to various anthropogenic activities like industrialization, urbanization and mining activities. Heavy metal concentrations in sediment were found much higher than river water, which indicate towards alarmingly strong polluted condition. High contamination factors and degree of contamination were obtained for Cd and Pb for all sites in summer. In addition, PLI values of river bed sediment were found > 1 that represents progressive deterioration of sites whereas, I_{geo} recommended sediment quality towards moderately to very strongly polluted condition. PCA analysis suggested that high metal loading seems to be stemmed out from anthropogenic origin with significant loading of metals like Zn, Pb, Ni, and Cr. It is clear from the study that the metal concentrations in river water and sediment are getting higher trend day by day. The river water is being used for drinking purpose by the wide population settled along the bank of river which could exert toxic effect on human health.

Recommendations

High metal loaded sediment is a serious concern as it has great potential for severe impact on aquatic organisms and human being. An unremitting system of river water and sediment quality monitoring may be developed to capture the pollution status in long term basis. Further, studies are recommended on effect of these metals on different food chains of aquatic ecosystem of Subarnarekha river as well as the effectiveness of self purification process of river by river water quality modelling. Water treatment plants with huge storage capacity should be developed to provide safe water for local population. Coupling of bioremediation technology with other economical conventional methods will definitely help in solving this devastating problem. Our previous study has presented contamination problem in phytoplankton, molluscs and fishes in Subernrekha river (Banerjee et al. 2015). The study could be utilised by various authority for questing a long term solution for better management of this river water and may formulate policy to minimise the discharge loads from various industries with regulatory action and using green technology.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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