Pollution, 5(4): 803-819, Autumn 2019
DOI: 10.22059/poll.2019.280504.620
Print ISSN: 2383-451X Online ISSN: 2383-4501
Web Page: https://jpoll.ut.ac.ir, Email: jpoll@ut.ac.ir

Lake Hydro Geochemistry: An Implication to Chemical Weathering, Ion-exchange Phenomena and Metal Interaction

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Received: 05.04.2019 Accepted: 26.06.2019

ABSTRACT: Present study aims to unravel the hydro geochemical interaction of sediment and water of Saheb bandh lake, West Bengal, India with an emphasis on heavy metal assessment. Lake water belongs to Ca^{2+}–HCO_3^- type hydro geochemical faces and water-rock interaction primarily controls the lake water chemistry. Based on different Hydro chemical characteristics it is suggested that silicate weathering is the major hydro geochemical process operating in Saheb bandh lake water. Regarding point source contribution of pollutants the average value of NO_3-N, TP and Hg are much higher in inlet water (7.5 mg/L, 1.29 mg/L and 8.5 µg/L) than the lake water (1.5 mg/L, 0.05 mg/L and 0.42 µg/L). Risk assessment indices suggest advanced decline of the sediment quality. Water-sediment interaction of heavy metals reveals that Cd, As, Pb and Hg metals enter into lake water as a result of not only natural processes but also of direct and indirect activities of humans. This study recommends that continuous monitoring of these metals in water and sediment and other aquatic biota of Saheb bandh should be directed to assess the risk of these vital heavy metals in order to maintain the safe ecology in the vicinity of this lake.

Keywords: Hydro geochemistry, Heavy metal assessment, Water-rock interaction, Risk assessment, Saheb Bandh Lake.

INTRODUCTION

Most of the lake water resources are gradually getting polluted due to the addition of foreign materials from the surroundings (Karnataka State Pollution Control Board, 2002). These include organic matter of animal and plant origin, washing from land surface and industrial and sewage effluents (Ajayan & Kumar, 2016; Abida et al., 2008, Thorat & Sultana, 2000). Rapid urbanization and industrialization with improper environmental planning often lead to discharge of industrial and sewage effluents into lakes (Singare et al., 2010). Heavy metals are refractory through natural processes in the aquatic environment; they can be chemically altered by organisms and converted into organic complexes, some of which may be more hazardous to animal and other human life (Das Sharma, 2019; Xie et al., 2010; Taghinia et al., 2010; Rogival et al., 2007). In case of shallow lakes heavy metals are more likely to be re-suspended and cause secondary contamination to the water environment as sediments can act both as a sink and a source for metals in the aquatic ecosystems (Yao & Xue, 2010, Liang & Wong, 2003), bottom sediments have been largely researched, especially in industrialized and congested urbanized

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regions (Shen et al., 2007; Karbassi & Amirnezhad, 2004; Xie & Chen, 2002). Heavy metals such as Cu, Fe, Zn, Ni and others are important for proper functioning of ecosystems (Lokhande et al., 2010a, 2010b, 2010c; Lokhande et al., 2009a, 2009b); however their excess could lead to a number of disorders. The fate of metals transported to aquatic body exhibits two properties of particular concern (i) bioaccumulation in organisms and passed upwards to man through food and drinking water (Chi et al., 2007) and (ii) the close relationship of metals and sediments in which adsorption and sedimentation are important mechanisms for purification of the water column in a lake water body.

Saheb bandh (23º20'42"N latitude and 86º21'37"E longitude) (Fig. 1) is an important wetland ecosystem of Purulia district, West Bengal for its biological diversity, aesthetic beauty, recreation and multipurpose features having a total catchment area of 82 acres, out of which water portion occupies approximately 60 acres (Dutta et al., 2014). Saheb bandh is a vulnerable source of water for the drought-prone town of Purulia. Saheb bandh, located at the valley zone, accumulates runoff water from catchment area of the town. Other than this, surface water stored at this lake includes water from springs and rock fracture conduits. These rock fractures actually facilitate to discharge the underground water from weathered mantle in the concerned micro watershed. Depth of the weathered mantle is extended upto a depth of ~20 feet below which there is a layer of hard massive granite gneiss rock which stops seepage of accumulated water of Saheb bandh. These technical factors stated above actually make this lake as perennial in nature (MED, 2008). Both inflow and outflow of lake water are regulated by five inlets (at the rate of ~277m^3/hr) and two outlets surrounding the lake. Recently, a number of motor repairing shops, garages, nursing home, private apartments, housing complexes, hotels, bathing ghats, amusement park *etc.* have been cropped up within the lake catchment area, contributing continuous discharge of wastewater into lake. The main objectives of this research are to find out the trend of temporal variation of lake hydro geochemistry and to assess the heavy metal content and its interaction between surface water and Lake Bottom sediment.

MATERIALS AND METHODS

Altogether 40 surface water and respective bottom sediment samples were collected from Saheb bandh lake during pre-monsoon (April) and post-monsoon (December) season of 2016 – 2018. Thereafter samples were stored in 500 ml polyethylene bottles which were pre-washed with dilute HCL and distilled water and followed by rinsing twice with the lake water sample before filling it up to capacity. Wastewater samples were also collected from five inlets and two outlets. pH, Electrical Conductivity (EC), Total Dissolved Solids (TDS) were measured at the sampling sites by using ion selective electrode (Thermo Orion Star 081010MD) whereas other parameters, *viz.*, total hardness (TH), calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), bicarbonate (HCO_3^-), chloride (Cl^-), sulfate (SO_4^{2-}), total iron (total Fe), total phosphorus (TP), nitrate-nitrogen (NO_3^--N) were analyzed in the laboratory by following standard methods (APHA, 1998). Lake bottom sediment samples were collected from pre-selected locations by Grab sampler and thereafter immediately transferred into pre-cleaned polythene bags and placed in cool and dark place. Sediment samples were furthered prepared by drying (70°C for 24 hours), grinding, sieving (2 mm) and the processed powdered samples were stored in desiccators for further chemical analysis. Organic matter was analyzed according to Walkley and Black, 1934 method. Available nitrogen, available phosphorus and available potassium were measured by Subbiah and Asija, 1956, Bray and Kurtz, 1945 and Black, 1965 methods, respectively.

In this investigation, nine heavy metals *viz.*, arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), manganese (Mn), lead (Pb), and zinc (Zn) were analysed both from surface water and bottom sediment samples of the studied lake. Prior to analysis, water and sediment samples were completely digested according to Kouadio & Trefry, 1987. Thereafter digested samples were analysed for aforementioned heavy metals by Atomic Absorption Spectrophotometer (AA 8000, AA500AFG, LABINDIA) using flame-AAS and hydride generator system. Different pollution indices in respect of heavy metals of water and sediment samples are referred which are represented in Table 1.

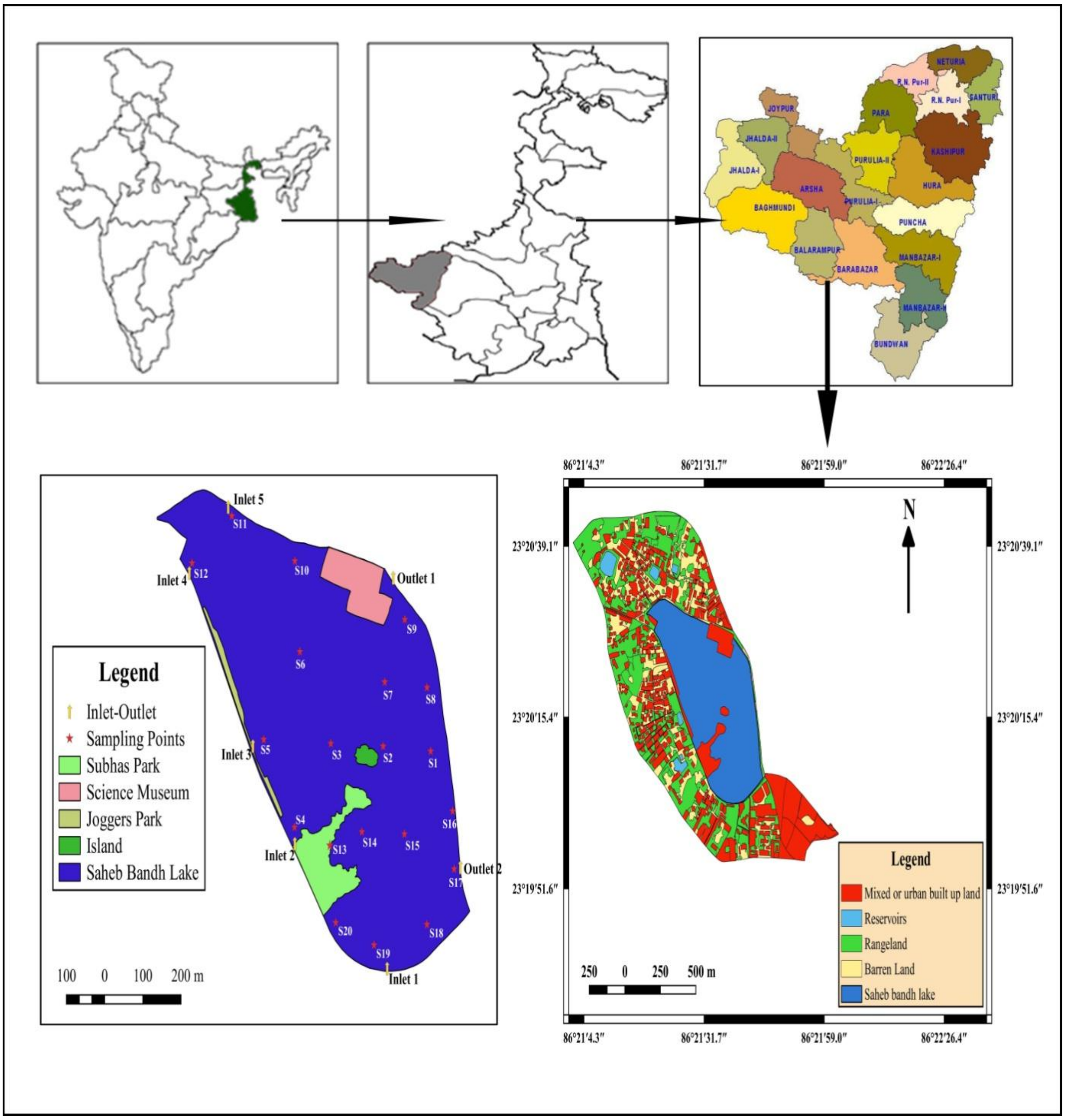


Fig. 1. Study area map

Table 1. Pollution evaluation indices in respect of heavy metals

Pollution Indices	Equation	References
Water		
Degree of contamination (C_{deg})	$C_{deg} = \sum_{i=1}^{n} C_{fi}$ $C_{fi} = \frac{C_{Ai}}{C_{Ni}} - 1$	Bhuiyan et al. (2010)
Heavy metal evaluation index (HEI)	$HEI = \sum_{i=1}^{n} \frac{H_C}{H_{MAC}}$	Bhuiyan et al. (2010)
Sediment		
Contamination factor (Cf)	Cf = Ms/Mb	Hakanson (1980) and Turekian &Wedepohl (1961)
Contamination degree (Cd)	$Cd = \sum_{i=0}^{n} Cf$	Hakanson (1980)
Index of geo-accumulation (I_{geo})	$I_{geo} = Log2 \left(\frac{C_n}{1.5\ B_n}\right)$	Muller (1981)
Enrichment factor (EF)	$EF = (C_M/C_X)_{sample}/(C_M/C_X)_{Earth's\ crust}$	Sinex & Helz (1981)
Pollution load index (PLI)	PLI for a site = $n^{th} \sqrt{CF_1 * CF_2 * \ldots CF_{nt}}$ PLI for a zone = $n^{th} \sqrt{site_1 * site_2 * \ldots site_n}$	Tomlinson et al. (1980)
Potential ecological risk index (PERI)	$RI = \Sigma E_r^i$ $E_r^i = T_r^i \times C_r^i$ $C_r^i = C_{surface}^i / C_n^i$	Hakanson (1980) and Guo et al. (2010)
Assessment of possible biological effects	$m\text{-}PEL\text{-}Q = \frac{\sum_{i=1}^{n}(C_i/PEL_i)}{n}$	Long & MacDonald (1998)

Quality control measures were performed to assess contamination and dependability of the analyzed data. Mark reagents (AR grade) and standard solutions were used for physico-chemical parameters of water and sediment samples. In case of heavy metal analysis, the instrument calibration standards were made by diluting standard (1000 ppm) supplied by LOBA Chemie. For quality control purposes, proper care was taken during sample collection and preservation and for every experimental procedure and analytical precision. Each water and sediment samples were analysed three times in order to ensure the accuracy of the analytical results. A blank was also prepared at the same time during experimental procedure and no detectable contamination was found when aliquots of reagents were processed and analyzed with the samples. Double distilled deionised water was used throughout the experimental procedure.

In this research it is assumed that the underlying distribution of the studied parameters are normal because of sample size >15. Pearson's correlation analysis was performed to evaluate potential relationships among the various physico-chemical parameters of water and sediment samples. Cluster Analysis (CA) was also performed to distinguish natural and anthropogenic contributions of incoming pollutants into the lake water system based on the level of association of the variables.

RESULTS AND DISCUSSION

Hydro chemical analysis (Supplementary table 1) reveals that pH of the lake water varies between 7.87 and 7.69 whereas electrical conductivity (EC) is found to be decreased from pre-monsoon (382 µS/cm) to post-monsoon (283 µS/cm). The highest TDS value is recorded during pre-monsoon (325 mg/L). Other parameters such as TH, Ca^{2+} and Mg^{2+} are noticed to be decreased from pre- to post-monsoon in the range of 173 – 164, 44 – 42 and 14 – 12 mg/L, respectively. In contrary, Na^+ and K^+ content is observed to increase from 67 to 69 and 6 – 8 mg/L which may be attributed to the deposition of Na^+ and K^+ salts and other dissolved solids through surface run-off from lake catchment area, discharge of wastewater from inlets *etc.* and also due to lower photosynthetic activity during this season. Bicarbonate (HCO_3^-) ion decreases in pre-monsoon (272 mg/L) as compared to post-monsoon season (235 mg/L). Increased Cl^- concentration in pre-monsoon could be due to maximum growth of phytoplankton and bottom biota (Verma & Shukla, 1970) that precipitate Cl^- and also due to low water level and other anthropogenic sources. As compared to post-monsoon (8 mg/L), sulphate concentration is slightly higher in pre-monsoon (9 mg/L) which may be due to metabolism of increased organic matter by constant input of domestic waste and sewage (Tyagi, 2008). Maximum Fe concentration is found in pre-monsoon (0.60 mg/L) followed by post-monsoon season (0.45 mg/L) which may be attributed to human settlement (residential area) and increased diffusion of ferrous ion released from sediment at lower concentration near the sediment surface. The maximum concentration total phosphate is noticed (0.06 mg/L) during post-monsoon season which, may be attributed to runoff from the catchment area with rain water and utilized thereafter for the growth of algae and other types of flora within the lake. The NO_3^--N concentrations in lake water is found highest in post-monsoon (2 mg/L). Similar kind of seasonal pattern of both the nutrients (TP and NO_3^--N) is corroborated to high water flow, inflow of high phosphorus and nitrogen loaded wastewater through inlets and from non-point pollution source *viz.*, washing, and bathing, and sewage runoff, accumulation of nutrients and huge growth of aquatic macrophytes. The arrangement of major ions in the studied lake follows the sequence of $HCO_3^- > Cl^- > Na^+ > Ca^{2+} > Mg^{2+} > SO_4^{2-} > K^+$ in pre-monsoon and $HCO_3^- > Na^+ > Cl^- > Ca^{2+} > Mg^{2+} > K^+ > SO_4^{2-}$ during post-monsoon.

Generally, point source pollution is responsible for decline of water quality in Saheb bandh lake. Investigation result reveals that the main sources of the lake water pollution are waste and wastewater of automobile repairing shops, domestic sewage, hotels, apartments, housing, complexes, *etc.* The average value of NO_3-N in inlet water (7 and 8 mg/L) is higher than the lake water (1 and 2 mg/L) in both pre- and post-monsoon season. Waste/wastewater generated from domestic and hospital source are the point source pollutant of nitrate in lake water. Among all the heavy metals, Hg is very much higher in inlet water (9 and 8 µg/L) than lake water (0.21 and 0.62 µg/L) in both pre- and post-monsoon season. In case of present study, chemical waste and residential waste may be the possible source of mercury. The present study observed that the concentration of total phosphorus (TP) in inlet water (1.25 and 1.32 mg/L) is higher than lake water (0.04 and 0.06 mg/L). According to the results of field investigation, NO_3-N, TP and Hg loadings are significantly contributed by point source pollution in Saheb bandh lake.

According to Pearson correlation analysis pH shows significant negative correlation with Ca^{2+} in both the pre- and post-monsoon seasons (Table 2) because at a given temperature, pH is controlled by the dissolved chemical compounds (Chapman, 1996). Electrical conductivity shows significant positive correlation with COD in pre- and post-monsoon seasons because of high rate accumulation of dissolved solids and nutrients. Total hardness shows significant positive correlation ($r = 0.727$) with total Fe in pre-monsoon season which may be corroborated to the presence of maximum concentration of total Fe during this season. Calcium exhibits the significant positive correlation (Table 2) with sulphate ($r = 0.509$) in post-monsoon season as both of them have natural origin in contributing hardness in the water. Magnesium exhibits significant positive correlation with Na^+, K^+, TP, NO_3^--N because in pre-monsoon season highest Mg^{2+} concentration is recorded. Sodium shows significant positive correlation with TP ($p < 0.05$) and NO_3^--N (p

< 0.01) in both the seasons and K^+ exhibits significant positive correlation with TP (r = 0.642) and NO_3^--N (r = 0.555) in pre-monsoon season. During post-monsoon seasons, bicarbonate and total alkalinity are positively correlated with Na^+ and NO_3^--N which, may be attributed to their contribution to alkalinity. Chloride shows significant positive correlation with NH_4-N and NO_3^--N with a coefficient value of 0.511 and 0.454 in pre- and post-monsoon, respectively because Cl^- concentration is varied according to mineral content of the earth. Nitrate shows significant positive correlation with ammonia (r_{pre} = 0.539 and r_{post} = 0.687) in both the seasons because nitrates are naturally occurring anion that is a part of nitrogen cycle and major limiting nutrients and ammonia is important animal excretory product and easily used in form of nitrogen. Nitrate shows significant positive correlation with TP (r = 0.450) in pre-monsoon season. Total phosphorus possess significant positive correlation with K^+ (r = 0.642) that may be due to increased bacterial decomposition of organic matter originated by dead plants.

Table 2. Pearson correlation matrices during (a) pre-monsoon and (b) post-monsoon

Pre-monsoon														
Variables	**pH**	**EC**	**TDS**	**TH**	**Ca^{2+}**	**Mg^{2+}**	**Na^+**	**K^+**	**HCO_3^-**	**Cl^-**	**SO_4^{2-}**	**Total Fe**	**TP**	**NO_3-N**
pH	1													
EC	-0.593	1												
TDS	-0.223	0.294	1											
TH	-0.164	0.136	0.435	1										
Ca^{2+}	-0.449	0.504	0.242	0.211	1									
Mg^{2+}	0.186	-0.010	0.644	0.195	0.047	1								
Na^+	-0.173	-0.102	0.448	0.297	0.097	0.597	1							
K^+	0.039	0.236	0.713	0.310	0.203	0.626	0.315	1						
HCO_3^-	-0.376	0.040	0.276	0.178	-0.033	-0.144	0.159	-0.108	1					
Cl^-	0.189	0.287	0.177	-0.114	0.104	0.375	0.207	0.127	-0.116	1				
SO_4^{2-}	-0.322	0.327	0.362	0.714	0.434	-0.039	0.320	0.311	0.436	0.103	1			
Total Fe	-0.198	0.301	0.377	0.727	0.440	0.276	0.580	0.337	0.009	0.331	0.707	1		
TP	-0.276	0.223	0.771	0.339	0.283	0.480	0.509	0.642	0.283	0.229	0.463	0.403	1	
NO_3-N	0.066	-0.029	0.489	0.460	0.076	0.667	0.649	0.555	0.062	0.314	0.336	0.523	0.450	1
(b) Post-monsoon														
Variables	pH	EC	TDS	TH	Ca^{2+}	Mg^{2+}	Na^+	K^+	HCO_3^-	Cl^-	SO_4^{2-}	Total Fe	TP	NO_3-N
pH	1													
EC	-0.235	1												
TDS	-0.389	0.524	1											
TH	-0.099	0.162	0.257	1										
Ca^{2+}	-0.467	0.555	0.642	0.045	1									
Mg^{2+}	-0.047	0.081	0.283	0.535	-0.137	1								
Na^+	-0.117	0.171	0.631	0.201	0.574	0.081	1							
K^+	0.249	-0.099	0.213	-0.216	-0.182	0.206	0.064	1						
HCO_3^-	-0.420	0.388	0.628	0.387	0.568	0.354	0.701	-0.080	1					
Cl^-	-0.181	-0.153	0.315	0.099	0.169	0.107	0.195	-0.075	0.042	1				
SO_4^{2-}	-0.279	0.252	0.429	0.411	0.509	0.065	0.424	-0.192	0.327	0.010	1			
Total Fe	0.104	0.240	0.627	0.165	0.620	0.097	0.745	0.138	0.382	0.312	0.520	1		
TP	-0.050	0.283	0.697	0.170	0.321	0.081	0.557	0.361	0.410	0.346	0.196	0.453	1	
NO_3-N	-0.281	0.106	0.547	0.020	0.401	0.152	0.641	0.211	0.449	0.454	0.245	0.632	0.297	1

Values in bold are different from 0 with a significance level alpha=0.05

Dendrogram of the sampling sites, obtained by Ward's method is represented in Fig. 2. In pre-monsoon season two statistically significant clusters are formed. Cluster 1 (sampling sites 4 – 5, 11 – 12, 18 – 20) is mainly comprises of inlet points, located at south-west direction of lake whereas Cluster 2, comprises of sampling sites 1 – 3, 6 – 10, 13 – 17, is located at the middle and north-eastern direction of the lake, near about outlet points. In post-monsoon season, no significant classification is observed in respect of sampling sites. The four inlets out of five are situated at south-western portion of the lake which is more influenced by external discharge consisting of wastewater from automobile repairing shops, garages, hospitals, nourishing home, runoff from municipal sewage, *etc.* Another one inlet point is located at north direction of lake which is more influenced by also external discharge consisting of waste and wastewater from hotels, apartments, complex and catchment area of lake.

To understand the actual mechanism which controls the lake water quality among precipitation, rock weathering, and evaporation, Gibbs's diagram (Gibbs, 1970) is used. It describes the ratio between TDS toward vertical axis versus $(Na^+ + K^+)/(Na^+ + K^+ + Ca^{2+})$ and $Cl^-/(Cl^- + HCO_3^-)$ on horizontal axis. Fig. 3a and b reveal that lake water quality is primarily controlled by the weathering of surrounding rocks during both the seasons.

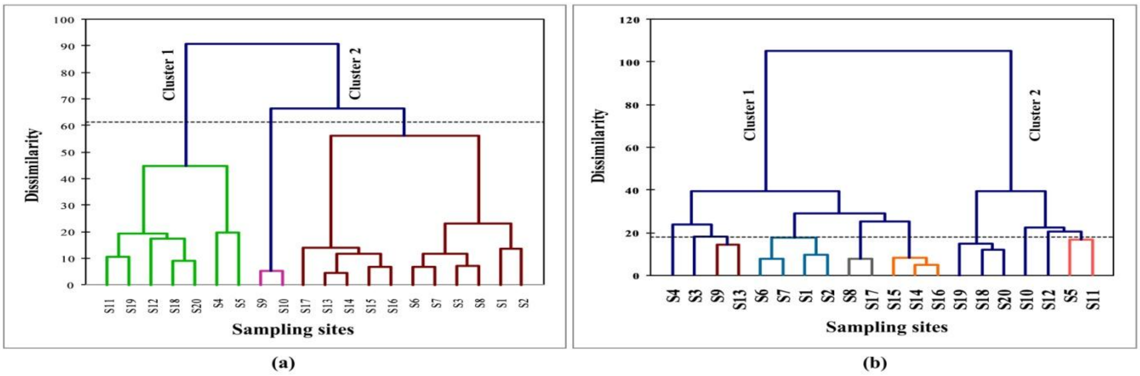


Fig. 2. Dendrogram of the dissimilarity between the different sampling sites of Saheb bandh lake based on water quality parameters in (a) pre- and (b) post-monsoon season. The dotted line denotes the truncation line that represents the stations that are somewhat homogeneous

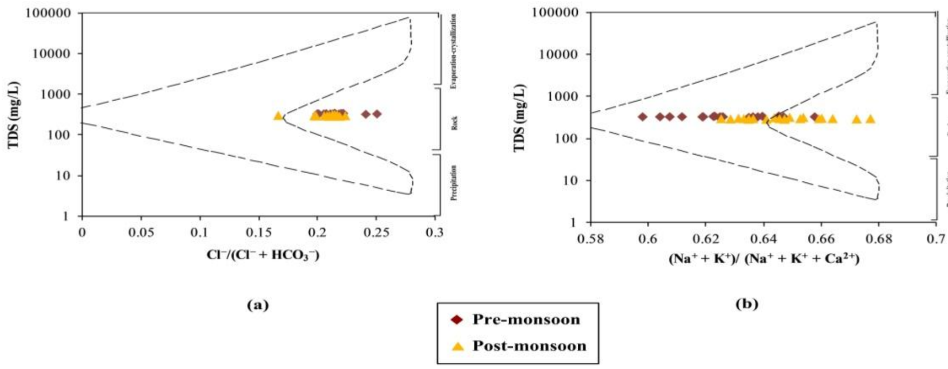
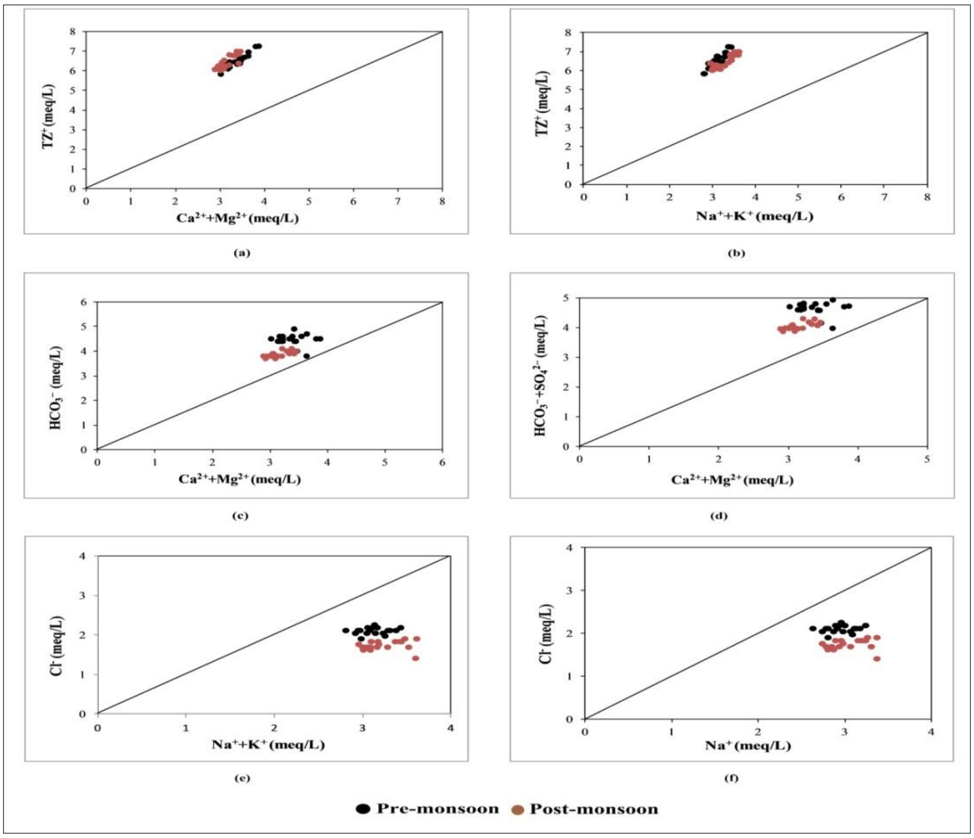


Fig. 3. Gibb's (1970) diagram showing a water-rock interaction through (a) TDS *vs* $Cl^-/(Cl^- + HCO_3^-)$ and (b) TDS *vs* $(Na^+ + K^+)/(Na^+ + K^+ + Ca^{2+})$

Hydro chemical characteristics of ions in lake water are studied using 1:1 aquiline diagrams. The comparison among alkaline earth metals $(Ca^{2+} + Mg^{2+})$ and TZ^+ (Fig. 4a) reveals that that all the plotted points of lake water samples fall much above the aquiline, reflecting an increasing contribution of Na^+ and K^+ with increasing dissolved solids in both the pre- and post-monsoon seasons. The plot for Na^++K^+ *vs* TZ^+ (Fig. 4b) of both the seasons shows high ionic ratio of $(Na^++K^+)/TZ^+$ (0.46 – 0.51) and all the samples fall far above the 1:1 aquiline, suggesting that the cations in lake water might have been derived from weathering of alumino-silicates (Stallard and Edmond, 1983). The plot of $(Ca^{2+}+Mg^{2+})$ *vs* HCO_3^- for this study area shows that all the sampling points fall above 1:1 trend in both the pre- and post-monsoon seasons which suggest that excess of HCO_3^- alkalinity should be balanced by the alkalis (Fig. 4c). The comparison between $(Ca^{2+}+Mg^{2+})$ *vs* $(HCO_3^-+SO_4^{2-})$ (Fig. 4d) shows that all the points fall in the HCO_3^- $+SO_4^{2-}$ side suggesting that silicate weathering is the major hydro geochemical process operating in Saheb bandh lake water. The excess of $(HCO_3^-+SO_4^{2-})$ over $(Ca^{2+}+Mg^{2+})$ suggest that significant contribution from non-carbonate source and demanding the required portion of the $(HCO_3^-+SO_4^{2-})$ to be balanced by the alkalis (Na^++K^+). The scatter plot of Na^++K^+ *vs* Cl^- (Fig. 4e) in both pre- and post-monsoon seasons shows that all the plotted points of lake water samples fall below the aquiline, indicating silicate weathering is the source of alkali metals, not the precipitation and other atmospheric sources. The sources of Cl^- are probably the dissolution of evaporites as well as anthropogenic activities. The Na^+ *vs* Cl^- plot (Fig. 4f) of the Saheb bandh lake water samples also suggests silicate weathering is the major contributor of Na^+ ion in lake water.

Fig. 4. The scatter plots between (a) Ca^{2+} + Mg^{2+} *vs* total cations, (b) Na^+ + K^+ *vs* total cations, (c) Ca^{2+} + Mg^{2+} *vs* HCO_3^-, (d) Ca^{2+} + Mg^{2+} *vs* HCO_3^-+ SO_4^{2-}, (e) Na^+ + K^+ *vs* Cl^- and (f) Na^+ *vs* Cl^-

The geochemical nature and relationship between dissolved ions in natural water may also be evaluated by plotting the analytical value on Piper (Piper, 1953) tri-linear diagram. The studied lake water is of Ca^{2+}–HCO_3^- type in both the seasons except 5% of samples in pre-monsoon season are of Ca^{2+} – Mg^{2+} – SO_4^{2-} type (Fig. 5). Weak acid (HCO_3^{2-}) is found to surplus over strong acid (SO_4^{2-}) as well as alkaline earth metals $(Ca^{2+}$ + $Mg^{2+})$ over alkali metals $(Na^+$ + $K^+)$ from pre to post-monsoon season.



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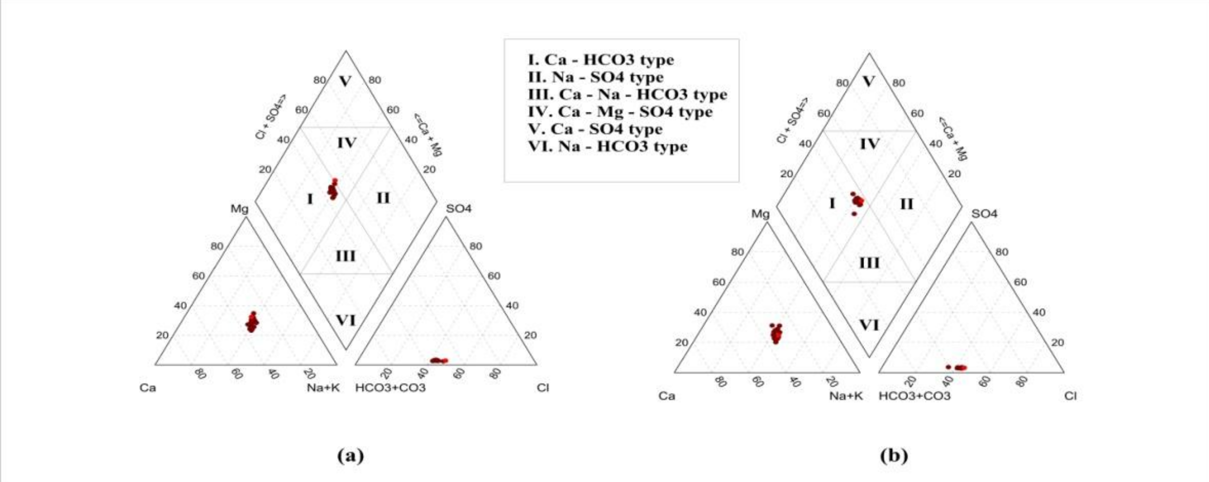

Fig. 5. Piper diagram (1953) showing types of water and concentration of different parameters during (a) pre- and (b) post-monsoon season

Detailed physico-chemical analysis of lake bottom sediments in pre- and post-monsoon season (2016 – 2018) are presented in Supplementary table 2. During pre-monsoon, pH of the sediment sample ranges from 6.9 – 8.05 with a mean value of 7.55 whereas in post-monsoon season it varies from 6.10 – 7.25 with a mean of 6.79 (Fig. 6a). The lower value of EC (202 µS/cm) is observed during pre-monsoon as compared to post-monsoon (229 µS/cm) season (Fig. 6b) which may be attributed to the utilization of ionic minerals by the producers. Organic carbon content of the lake bottom sediments fluctuates between 0.08 – 1.88% with a mean of 0.54% during pre-monsoon whereas, in post-monsoon it varies from 0.07 – 0.98% with a mean of 0.32% (Fig. 6c). The average concentration of available nitrogen is much higher in post-monsoon (503 ± 136 mg/kg) than pre-monsoon (318 ± 74 mg/kg) (Fig. 6d) due to enhanced microbial effect. Presence of significant positive correlation between available nitrogen and organic carbon ($r = 0.652$, $p > 0.01$) suggests that most of the nitrogen comes from organic carbon as probably bound to it (Table 3). Available phosphorus concentration in the sediment samples varies from 3 to 35 mg/kg during pre-monsoon and 9 to 87 mg/kg during post-monsoon (Fig. 6e). There is a good correlation between nitrogen and phosphorus ($r = 0.842$) and organic carbon and phosphorus ($r = 0.645$, $p<0.01$), suggesting that both the nutrients become released from the common source. The mean values of available potassium in sediment are 165 and 170 mg/kg in pre-monsoon and post-monsoon season, respectively (Fig. 6f).

The average data of heavy metal concentration in lake water in both pre- and post-monsoon season (2016 – 2018) is presented in Table 4. Seasonal variation of concentration reveals that all the examined heavy metals are high in post-monsoon rather than pre-monsoon except As and Cd. Because, under anaerobic condition inorganic As is released into the lake water from lake bottom sediments under anaerobic conditions in pre-monsoon, while As(V) is adsorbed onto Fe/Mn oxides and then settle down into the sediments in post-monsoon season. Maximum Cd is found in pre-monsoon due to flushing or dissolution of non-lithogenic material by infiltrating water. Apart from geogenic input anthropogenic input due to transport of waste leachate with the runoff of rainwater and sewage waste through inlets from the surrounding catchment area are also responsible for heavy metal concentration in the studied lake.

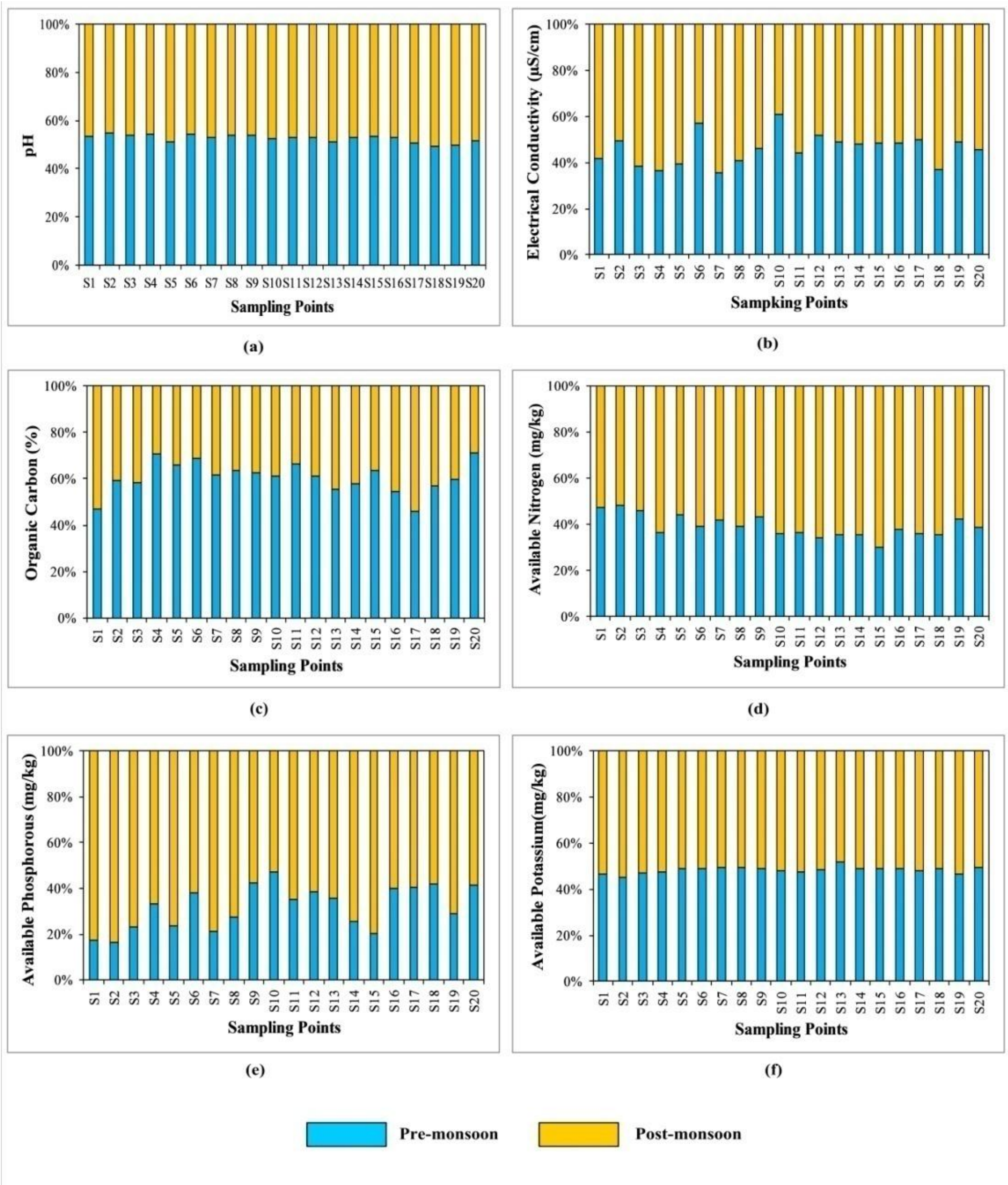

Fig. 6. Seasonal distribution pattern of physico-chemical parameters within sediment samples

Table 3. Pearson correlation coefficient (r) for various physico-chemical parameters of lake sediment

Variables	pH	EC	OC	N_{AVL}	P_{AVL}	K_{AVL}
pH	1					
EC	*0.550	1				
OC	0.179	0.246	1			
N_{AVL}	-0.022	0.305	**0.652	1		
P_{AVL}	0.097	0.351	**0.645	**0.842	1	
K_{AVL}	-0.185	0.116	0.365	**0.780	**0.678	1

**Correlation is significant at the 0.01 level; *Correlation is significant at the 0.05 level

Table 4. Concentration of heavy metals in Saheb bandh lake water and bottom sediment

Heavy metals	Lake water (µg/L)			Bottom sediment (mg/kg)			
	Pre-monsoon (mean)	Post-monsoon (mean)	WHO (2006)	Pre-monsoon (mean)	Post-monsoon (mean)	Shale standard[a]	Earth crust[b]
Fe	38.60	79.15	-	35472.1	36705.6	47200	56300
Mn	9.55	12.58	500	126.006	147.589	850	950
Cr	1.56	2.40	50	54.59	58.90	90	100
Zn	5.77	13.36	3000	19.00	20.91	95	70
Cu	1.67	4.03	2000	9.33	11.20	45	55
Pb	14.26	17.28	10	5.81	7.28	20	12.5
As	2.93	2.19	10	5.24	6.20	13	1.8
Cd	5.97	0.62	3	0.35	0.51	0.3	0.2
Hg	0.21	0.62	1	0.28	0.31	0.4	0.08

a: Turekian and Wedepohl (1961); b: Taylor (1964)

The metal concentrations in the various compartments of the lake generally decrease in the order of suspended matter > bottom sediment > surface water (Luck et al., 2008). Most of the reactions take place in the aquatic ecosystems at the sediment – water interface. In Saheb bandh Fe and Mn concentrations are found as low as compared to standard value in both pre- and post-monsoon seasons (WHO, 2006; Turekian & Wedepohl, 1961; Taylor, 1964) which may probably caused by Eh, pH of the total aquatic ecosystem and other geogenic sources. Iron concentration varies between different sampling sites due to redox condition of the lake water which is further supported by the discharge of wastewater from Lake catchment area. The low concentration of Fe, Mn and its combination with their remobilization into the lake water and the release of heavy metals may take place due to anoxic condition of near bottom layer (Salomons & Baccini, 1986). Major share of manganese may derive from granite gneiss basement rock of Saheb bandh lake. Lead concentration in the studied lake water exceeds the standard value of WHO, 2006 due to its higher solubility in the pH range of 7.6 – 7.9. The concentrations of Pb in bottom sediment are low as compared to shale and earth crust value in both pre- and post-monsoon seasons (Turekian & Wedepohl, 1961; Taylor, 1964). Lead is accumulated in bottom sediments (Rickard & Nriagu, 1978; Kabata-Pendias & Pendias, 2000) and direct link exists between the Pb content in bottom sediments with their granulometric and mineralogical composition, and also with the origin of the bedrock, emissions from road vehicles, automobile industry and other anthropogenic sources. In both the seasons, Zn and Cr concentration are found as low as compared to standard value (Table 4) due to average pH of 7.6 – 7.9 and 6.7 – 7.6 with moderate dissolved oxygen and organic matter in water and bottom sediment portion, respectively. At pH less than 7, Zn is generally present as a divalent ion, which readily forms complexes with organic and inorganic compounds thereby rapidly adsorbed on the surface of suspended particulate matter and bottom sediments (Bertling et al., 2006). Low concentration of Cr is found in water because Cr does not persist for long period in the dissolved state and is precipitated as a suspension (mostly hydroxides) in natural aquatic ecosystems. Presence of low levels of Cu in lake water and bottom sediment suggests no contamination of Cu loaded compound by the human activities. The dissolution of Cu is favoured by the presence of nitrates and dissolved oxygen in the lake water and insoluble compounds of Cu make up 40 – 90% of its total amount in surface waters (Moore et al., 1996). The concentrations of Cd, Hg and As in the bottom sediment exceed the earth crust value (Taylor, 1964) which may primarily caused by the weathering of Cd, Hg and As bearing minerals from granite gneiss rock. The concentration of these heavy metals in lake water is low as compared to WHO, 2006 except Cd. This discrepancy is probably due to its greater dependence on adsorption processes and pH. Bacteria play a major role in binding Cd in sediments, often causing the metal to precipitate as a sulfide (Kabata-Pendias, 2000). Cadmium in sediments neither dissolves nor passes into the lake water due to pH of the water is neutral to basic. Higher concentration of Cd in lake depends on different natural factors but primarily related to discharge of solid waste, domestic waste, human pressure, particularly, land-use/land-cover change in the Lake catchment area and air pollution (Rzetala, 2016).

The estimated pollution evaluation indices for the selected metals (Cd, Cr, Cu, Mn, Pb, Zn) in the water samples during pre- and post-monsoon seasons are shown in Table 5. Degree of contamination (C_{deg}) is classified into three categories (Edet & Offiong, 2002; Backman et al., 1997) such as low ($C_{deg} < 1$), medium (C_{deg} = 1–3) and high ($C_{deg} > 3$). The proposed HEI criteria for the surface water samples are as follows: low (HEI < 150), medium (HEI = 150–300) and high (HEI > 300) (Bhuiyan et al., 2010). In case of studied lake water samples, the average values of C_{deg} and HEI indices during pre- and post-monsoon seasons are -0.42, 1.95 and -0.66, 2.00 respectively, indicating water samples are contaminated with low degree of pollution by all the selected heavy metals.

Table 5. Descriptive statistics of pollution evaluation indices for selected heavy metals (µg/L) in lake water samples

Metals	Pre Mean	Post Mean	C_{Ni}[a]	MAC[b]	Pre-monsoon C_{deg}	Pre-monsoon HEI	Post-monsoon C_{deg}	Post-monsoon HEI
Cd	5.97	0.62	3.00	3.00	0.99	1.99	-0.79	0.21
Cr	1.56	2.40	50.00	50.00	-0.97	0.03	-0.95	0.05
Cu	1.67	4.03	2000.00	1000.00	-1.00	0.00	-1.00	0.00
Mn	9.55	12.58	500.00	50.00	-0.98	0.19	-0.97	0.25
Pb	14.26	17.28	10.00	1.50	0.43	9.50	0.73	11.52
Zn	5.77	13.36	3000.00	5000.00	-1.00	0.00	-1.00	0.00
Average					-0.42	1.95	-0.66	2.00

a: Upper permissible concentration from WHO (2006)
b: Maximum admissible concentrations (MAC) from Siegel (2002)

The estimated pollution evaluation indices for the selected metals (Cd, Cr, Cu, Fe, Al, Mn, Pb, As, Zn, Hg) in the sediment samples during pre- and post-monsoon seasons are discussed below (Karbassi et al., 2016; Karbassi et al., 2014; Afkhami et al., 2013). The mean contamination factor (Cf) values of all examined metals are less than 1, indicating low contamination factor whereas, only Cd shows moderate contamination in both the seasons. The degrees of sediment contamination (Cd) with the examined heavy metals are less than 8 indicating low degree of contamination, whereas few sampling points in both seasons indicate a moderate degree of contamination with these metals because in these sites untreated waste water is directly discharge through inlets. Geo-accumulation index values for all investigated heavy metals fall under 'one class', indicating uncontaminated to moderately contaminated in nature and its corresponding contamination intensity are illustrated in Fig. 7A. According to Enrichment factor (EF), Saheb bandh lake bottom sediment is not contaminated (EF < 2) with the examined metal except, Cd and Hg in both the seasons at few sampling points (Fig. 7B), which suggest that lake bottom sediment is moderately contaminated with Cd and Hg during both seasons. The lake bottom sediment is anthropogenically contaminated with Cd and Hg because according to Zhang & Liu , 2002) EF values between 0.5 – 1.5 indicates the metal enrichment entirely from crustal materials or natural processes; whereas EF values greater than 1.5 suggest that the sources are more likely to be anthropogenic. The Pollution load index (PLI) values of sediments in the different sites of studied lake ranged from 0.17 to 0.62 with average of 0.30 in pre-monsoon and 0.22 – 0.71 with average of 0.35 in post-monsoon (Fig. 7C). The PLI of the zone or the whole examined area of the lake is 0.01 for both the seasons which confirmed that the lake bottom sediments are not polluted (Table 6).

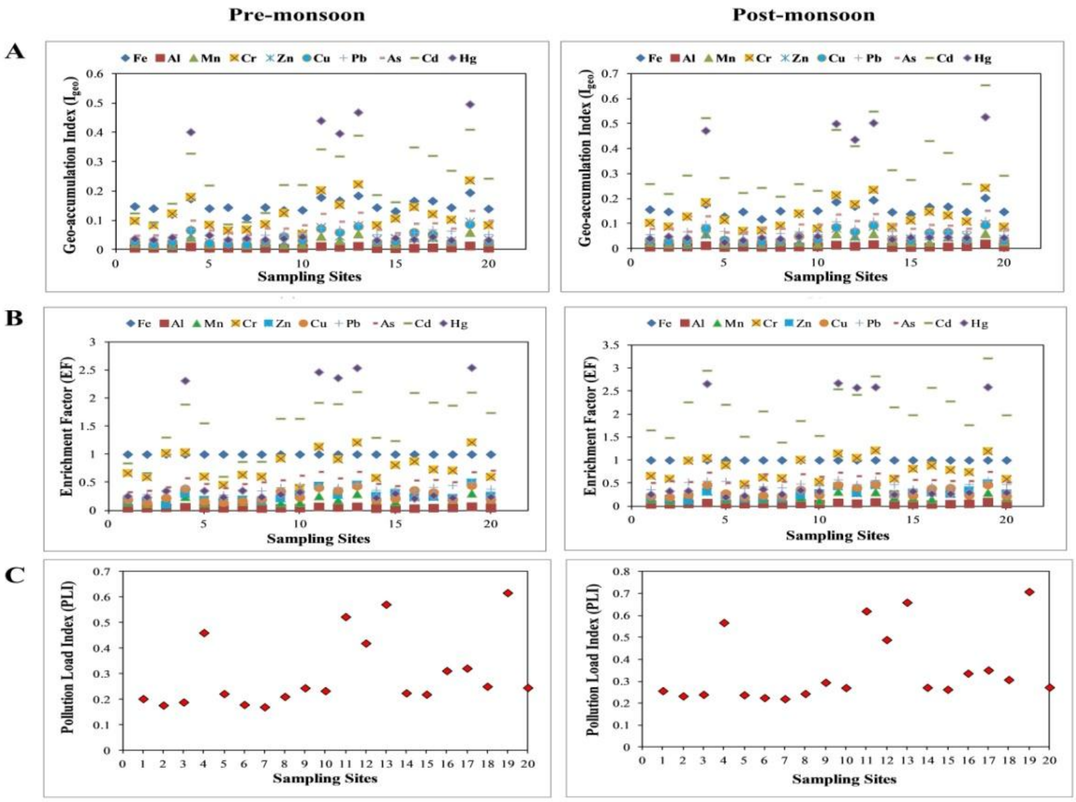

Fig. 7. (A) Index of geoaccumulation (I_{geo}), (B) Enrichment Factor (EF) and (C) Pollution Load Index (PLI) values of measured heavy metals in Saheb bandh lake bottom sediments

Table 6. Terminologies for pollution classes to assess the lake bottom sediment

I_{geo} classes[a]			Cf classes[b]		Cd classes[c]		EF classes[d]		PLI classes[e]	
I_{geo}	I_{geo} class	Pollution	Cf	Pollution	Cd	Pollution	EF	Pollution	PLI	Pollution
$I_{geo} \leq 0$	0	Uncontaminated	Cf < 1	Low contamination factor	Cd < 8	Low degree of contamination	EF<2	Depletion to mineral	0	Perfection
$0 < I_{geo} < 1$	1	Uncontaminated to moderately contaminated	1 ≤ Cf < 3	Moderate contamination factor	8 ≤ Cd <16	Moderate degree of contamination	2 ≤ EF <5	Moderate	<1	Baseline levels
$1 < I_{geo} < 2$	2	Moderately contaminated	3 ≤ Cf < 6	Considerable contamination factor	16 ≤ Cd <32	Considerable degree of contamination	5 ≤ EF < 20	Significant	>1	Polluted
$2 < I_{geo} < 3$	3	Moderately to strongly contaminated	Cf ≥ 6	Very high contamination factor	Cd ≥ 32	Very high degree of contamination	20 ≤ EF < 40	Very high		
$3 < I_{geo} < 4$	4	Strongly contaminated					EF > 40	Extremely high		
$4 < I_{geo} < 5$	5	Strongly to extremely contaminated								
Igeo < 5	6	Extremely contaminated								

a: Muller (1981); b: Hakanson (1980); c: Sutherland (2000); d: Tomlinson et al., (1980) The potential ecological risk indices (PERI) of Mn, Cr, Zn, Cu, Pb, As, Cd, Hg (mean value) are lower than 40 during pre- and post-monsoon except Cd in post-monsoon season (Supplementary table 3). This suggests that the sediment has slightly potential ecological risk for seven metals and moderately potential in respect of Cd. The main metal that can cause the ecological risk to the lake water is Cd which has recorded the highest average value of E_r^i as 34.95 and 50.83 in pre- and post-monsoon season, respectively. The average RI values of the 20 sampling sites are 70.69 and 91.01 in pre- and post-monsoon season, respectively. According to the evaluating standard, in general, the sediment samples of the lake have low ecological risk level (RI ≤ 150) in both the seasons. In present study, the consensus based Sediment Quality Guidelines (SQGs) developed for freshwater ecosystem is used to assess the potential ecotoxic effects of heavy metals in Saheb bandh lake. The strategy includes a threshold effect level (TEL), below which adverse effects on sediment-dwelling organisms are not expected to occur and a probable effect level (PEL), above which adverse effects on sediment-dwelling organisms are likely to be observed by MacDonald et al. (2000). A subsequent ranking of sites of potential concern has been proposed: lowest priority for m-PEL-Q < 0.1; medium-low priority for 0.11 < m-PEL-Q < 1.5; medium-high priority for 1.51 < m-PEL-Q < 2.3; and high priority for m-PEL-Q > 2.3. The concentrations of all the metals are below the SQG-PEL except the mean value of Hg is higher than SQG-TEL in both the seasons and Cd is higher than SQG-TEL in pre-monsoon which suggest that sediment-dwelling organisms are expected to occur due to Hg and Cd (Supplementary table 4). According to mean PEL quotients (m-PEL-Q) sediment samples ranked at the medium-low priority level.

CONCLUSIONS

The hydrochemistry of the Saheb bandh lake is found to be varies during both pre- and post-monsoon seasons. Both Ca^{2+} and Na^{+} are observed as the dominant cations and HCO_3^- and Cl^- are noticed as the dominant anions during both seasons. Gibbs's plot has indicated that the rock weathering as the key factor, which controls the water quality of the lake to a major extent. The requirement of cations from weathering of silicate rocks suggesting that silicate weathering is the major hydro geochemical process operating in Saheb bandh lake water. The tri-linear water-type (Piper diagram) diagram reveals that Ca^{2+}–HCO_3^- is the dominant hydro geochemical faces in both pre- and post-monsoon seasons of the lake water samples. In the present study, concentrations of Cd (pre-monsoon) and Pb (pre- and post-monsoon) in lake water and As, Cd and Hg (both seasons) in bottom sediment are higher than the safe values and earth crust values, respectively which indicates that the lake is polluted by these metals and may create an adverse effect on this lake ecosystem. Risk assessment associated with EF, Cf, Cd, I_{geo}, PLI and PERI expose that sediments are unpolluted to moderately pollute by heavy metals and Cd can cause the ecological risk to the lake water. The overall result indicates that the lake is getting polluted due to anthropogenic factors such as untreated effluents or waste from automobile shops, hospitals and domestic sewage, etc. Moreover different remediation measures should be taken promptly to remove excising pollution and metal contamination e.g., aeration, diversion of point sources of pollutants, use of algaecides, plantation in buffer zone, water quality should be monitored at regular intervals, dumping of waste should be restricted, etc.

ACKNOWLEDGEMENTS

The authors are thankful to Department of Higher Education, Science & Technology, Biotechnology (DHESTB) Govt. Of West Bengal [45(Sanc.)/ST/P/S&T/1G-60/2017] and Department of Science and

Technology, Govt. Of West Bengal for providing financial support through minor project and FIST programme respectively. Authors are highly indebted to Purulia Municipality Authority for their co-operation and endless support in carrying out sampling from this national lake.

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