



Geoenvironment and weathering of silicate minerals in sediments of the Brahmaputra river, India: Implications for heavy metal pollution assessment

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ABSTRACT

We carried out detailed compositional studies on fifteen sediment samples from the Brahmaputra River that were collected in the pre-monsoon period, to obtain the inherent contamination factor. The present study demonstrates that the degrees of metal contamination in the sediments of the Brahmaputra river. Typical metals Fe, Al, Ti, Pb, Zn, Cu, Ni, Co, Mn and Cr, have been evaluated using several parameters like, enrichment factor (EF), contamination factor (CF), geo-accumulation index (I_{geo}) and pollution load index (PLI). The relative distributions of the contamination are found to be: Al > Fe > Ca > Mg > Ti > Mn > Cr > Cu > Zn > Ni > Pb > Co. The suspended sediments have been found to be moderately contaminated by the elements Cu, Pb, Cr, Mn and Ni. The nature of weathering is estimated using plagioclase index of alteration (PIA), chemical index of alteration (CIA), chemical index of weathering (CIW) and index of compositional variation (ICV). The studied suspended sediment samples indicate an intermediate silicate weathering of adjoins area.

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1. Introduction

The river Brahmaputra carries ~73 million tons of dissolved material annually, which accounts for approximately 4% of the total dissolved flux into the oceans (Singh et al., 2005). The dissolved chemical load and sediment flux of the Brahmaputra River has significantly higher rates of physical and chemical weathering than other large Himalayan catchments (Sarin et al., 1989; Harris et al., 1998; Galy and France-Lanord, 1999, 2001; Dalai et al., 2002; Singh and France-Lanord, 2002; Singh et al., 2005). The total sediment budget of Brahmaputra predominantly depends on the nature of weathering of the adjoin areas and erosion of its tributaries. Sediments are an important source and sink of contaminants, including metals through the processes of precipitation, adsorption and chelation (Das et al., 2015). However, the river sediments act as both source and sink for heavy metals. Therefore, contaminants may eventually pass through the food chain and result in a wide range of adverse environmental effects. The sediments have been contaminated by heavy metals when rocks are disin-

tegrated through natural or anthropogenic process (Saikia, 2011; Saikia et al., 2014; Rai et al., 2019). The weathering of silicates exposed on the continents is the largest sink of atmospheric CO₂ on geological time scales (Wallmann, 2001). The weathering and mineralogical studies of sediments are helpful in understanding different sediment sources, environmental parameters influencing the weathering of source rocks, duration of weathering, transportation and post-depositional processes, element distribution pattern and evaluating the environmental conditions existing in an area. The focus on mineralogical, geochemical and geophysical studies and chemical composition of sediments of many Indian rivers were done by many authors (Borole et al., 1982; Subramanian, 1987; Seralathan, 1987; Ramesh et al., 1990; Chakrapani and Subramanian, 1990; Subramanian et al., 1985, 1987; Singh et al., 1997; Kotoky et al., 1997; Dekov, 1998; Singh, 1999; Pattan et al., 2008; Braun et al., 2009). The accumulation and distribution of heavy metals are the most common environmental pollutants, and their occurrence in waters and biota indicate the presence of natural or anthropogenic sources (Cataldo et al., 2001; Hobbelen et al., 2004; Koukal et al., 2004; Okafor and Opene, 2007; Mohiuddin et al., 2010; Chakraborty et al., 2015). However, the unusual phenomenon occurred in the Brahmaputra River turning its colour to muddy indicates the need to monitor water quality and metal pollution as-

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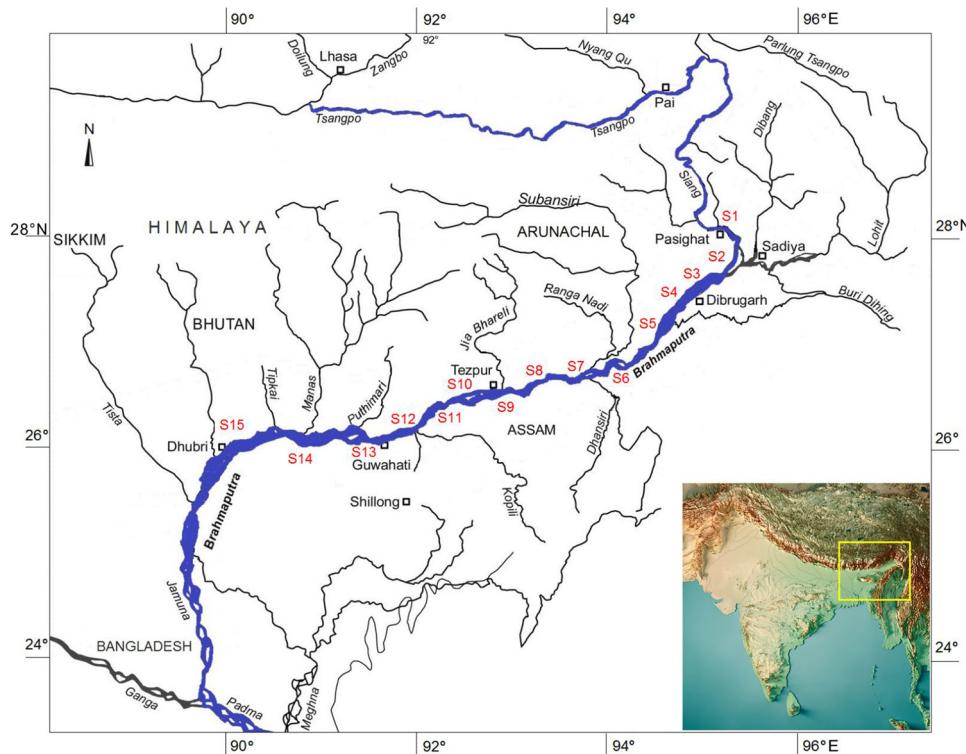


Fig. 1. Location map showing the Brahmaputra River with its tributaries and the sample collection sites.

essment (Tsering et al., 2020). This study is conducted to make a systematic assessment of the heavy metal contaminations and silicate mineral distribution due to weathering in sediments of the Brahmaputra River, which helps to assess the Eco toxic potential of the river sediments.

2. Experimental methods

The samples were collected in pre-monsoon period using grab sampling method from fifteen locations of Arunachal Pradesh and Assam, i.e., Pasighat (S1), Gujan (S2), Raumeria (S3), Dibrugarh (S4), Dikhomukh (S5), Nimatighat (S6) Dhansirighat (S7), Kaziranga (S8), Silghat (S9), Laharighat (S10), Gagalmari (S11), Hatisila (S12), Pandu (S13), Jogighopa (S14) and Dhubri (S15) of the Brahmaputra River from upstream to downstream (Fig. 1) at a depth of 2–5 ft. from the surface of each sampling locations. To eliminate the possibility of bank materials of the local origin, special care is taken on the sample collection by collecting them as far away from the banks as possible.

The suspended particles were separated by gravimetric method using Whatman filter paper (40μ). The wet samples were allowed to dry and the moisture contents were removed by heating the samples at temperature 110°C for 10 min. The heavy metals composition of sediment samples were determined using a Philips MagiX PRO wavelength dispersive X-ray fluorescence spectrometer with a rhodium anode X-ray tube was used, which may operated at up to 60 kV and current up to 125 mA, at a maximum power level of 4 kW. The precision and accuracy of the data is $\pm 2\%$, and average values of three replicates were taken for each determination. The oxides of major elements (SiO_2 , Al_2O_3 , Fe_2O_3 , MgO , MnO , TiO_2 , CaO , Na_2O , K_2O , and P_2O_5) were measured in the homogenized powdered bulk samples after drying to determine the loss of ignition (LoI) by igniting the samples at 850°C . Details of the analytical techniques are described elsewhere (Lozano and Pablo-Bernal, 2005; Verma et al., 2016). The degree of contamination

in the sediments is determined with the help of the parameters: enrichment factor (EF), contamination factor (CF), index of geo-accumulation (I_{geo}) and pollution load index (PLI) using the standard methods discussed elsewhere (Saikia et al., 2014). In order to estimate the nature of weathering intensity in the sediments, we applied commonly used weathering indices: plagioclase index of alteration (PIA), chemical index of alteration (CIA), chemical index of weathering (CIW) and index of compositional variation (ICV) which were proven to be well applicable to lithology (Saikia et al., 2015, 2016).

3. Results and discussion

A comparative metal concentration in the sediment samples of Brahmaputra River with different reference data are shown in Table 1. Average concentrations of all elements are found to be below of their respective reference values except Mg. The concentration of Al, Fe, Mg, Mn, and Cu are slightly higher, whereas Ca and Cr have below the results of the previous worker (Subramanian et al., 1985, 1987). The average concentrations of all observed elements except Si, Mg, Mn, Cu and Pb have less than the world surface rock average as background level. However, the world surface rock represents the average lithology subjected to weathering in the hydrosphere. The world surface rock average of Martin and Meybeck (1979) is used as background value for determination of enrichment factor (EF), contamination factor (CF), index of geo-accumulation (I_{geo}) and pollution load index (PLI) of the sediments samples (Martin and Meybeck, 1979).

The EF is used to assess the level of contamination and the possible anthropogenic impact in sediments. The element which has low occurrence variability is usually considered as reference element. Generally geochemical normalization of the heavy metals data to a conservative element, such as Al, Si and Fe is employed. According to Forstner and Wittmann (1983), in case of Fe, specifically the redox sensitive iron-hydroxide and oxide un-

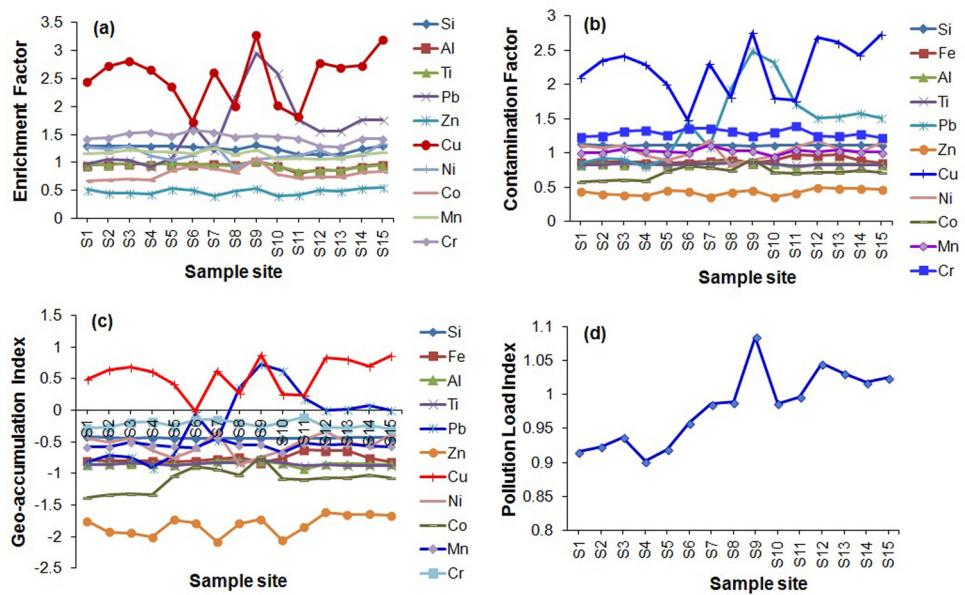


Fig. 2. (a) Enrichment factor, (b) contamination factor, (c) geo-accumulation indices, and (d) pollution load index of heavy metals in sediments of the Brahmaputra River at different sampling sites.

Table 1
Comparative concentration of elements in Brahmaputra River sediments (in ppm).

Elements	Concentration of elements in all site			BBS*	IRSA*	WRA*	WSRA*	WSA*
	Min	Max	Average \pm Standard deviation					
Si	303,433	309,271	304,776.73 \pm 1555.97	–	–	285,000	275,000	330,000
Fe	30,290	34,920	31,925.13 \pm 1589.06	29,000	29,000	48,000	35,900	40,000
Al	54,604	59,769	57,819.33 \pm 1262.76	56,000	–	94,000	69,300	71,000
Ti	3100	3299	3161.40 \pm 52.74	3100	–	4160	3800	5000
Pb	12.8	39.8	22.89 \pm 8.49	–	–	150	16	12
Zn	45.1	62.1	54.43 \pm 5.66	47	16	350	129	90
Cu	47.5	88.3	71.54 \pm 12.64	17	28	100	32	30
Ni	40.3	58.1	50.50 \pm 5.45	47	37	90	49	50
Co	7.5	11.8	9.21 \pm 1.17	–	–	20	13	8
Mn	709	829	766.73 \pm 25.41	600	605	1050	720	1000
Mg	15,857	18,381	17,139.07 \pm 912.78	16,500	–	11,800	16,400	5000
Cr	86.8	98.7	91.44 \pm 3.77	100	87	100	97	70
Ca	18,230	19,900	18,948.07 \pm 492.04	19,300	–	21,500	45,000	15,000
K	9770	12,900	11,457.87 \pm 918.35	12,000	–	14,200	24,400	14,000

* Brahmaputra basin sediment (BBS, Subramanian et al., 1985); Indian river sediment average (IRSA, Subramanian et al., 1987); Worlds river average (WRA); Worlds surface rock average (WSRA, Martin and Meybeck, 1979); Worlds soil average (WSA, Bowen, 1979).

der oxidation condition constitute significant sink of heavy metals in aquatic system (Forstner and Wittmann, 1983). Even a low percentage of Fe(OH)_3 , in aquatic system, has a controlling influence on heavy metal distribution (Rath et al., 2005). In this study we considered Fe as reference element of normalization. The EF [$= (C_{n(\text{Sample})}/C_{ref(\text{Sample})})/(B_{n(\text{Background})}/C_{ref(\text{Background})})$] is calculated as defined by Simex and Helz (1981), where $C_{n(\text{sample})}$ and $C_{ref(\text{sample})}$ are the content of the examined and reference element in the examined environment respectively; $B_{n(\text{background})}$ and $B_{ref(\text{background})}$ are the content of examined and reference element in the reference environment respectively (Simex and Helz, 1981). Due to the unavailability of metal background values for the study area, we used the values from the world surface rocks for analysis. According to Mmola et al. (2011), the categories of enrichment factor are: deficiency to minimal enrichment ($EF < 2$); moderate enrichment ($2 \leq EF < 5$); significant enrichment ($5 \leq EF < 20$); very high enrichment ($20 \leq EF < 40$) and extremely high enrichment ($EF \geq 40$) (Mmola et al., 2011). The enrichment factor of the all observed elements in the Brahmaputra river sediments shows minimal enrichment (Table 2), while Cu (2.40 ± 0.50

shows moderate enrichment in the study area (Fig. 2a). High risk of Cu and Pb in the Brahmaputra river sediments in Assam region was also reported by the previous author (Pallavi et al., 2015).

The level of contamination of sediment by metal is expressed in terms of CF as: $[CF = C_{\text{Sample}}/C_{\text{Background}}]$, where, C_{Sample} is the concentration of the given metal in river sediment, and $C_{\text{Background}}$ is value of the metal equals to the world surface rock average (Martin and Meybeck, 1979). According to Hakanson (1980), the level of contamination can be classified as: low contamination ($CF < 1$); moderate contamination ($1 \leq CF < 3$); considerable contamination ($3 \leq CF < 6$) and very high contamination ($CF > 6$) (Hakanson, 1980). The study area is moderately contaminated by the elements Cu, Pb, Cr, Mn and Ni (Table 2 and Fig. 2b).

According to Muller (1979), the $I_{geo} [= \log_2(C_n/1.5B_n)]$ is characterized by seven grades or classes profile of the geo-accumulation index as: the value of sediment quality is considered as unpolluted (I_{geo} is ≤ 0 , class 0); from unpolluted to moderately polluted (I_{geo} is $0-1$, class 1); moderately polluted (I_{geo} is $1-2$, class 2); from

Table 2

Enrichment factor (EF), contamination factor (CF) and geo-accumulation index (I_{geo}) of the Brahmaputra River sediments.

Elements	Enrichment factor		Contamination factor		Geo-accumulation index	
	Average ± Standard deviation					
Si	1.295 ± 0.060		1.109 ± 0.006		-0.437 ± 0.007	
Al	0.968 ± 0.055		0.829 ± 0.018		-0.847 ± 0.031	
Ti	0.959 ± 0.053		0.821 ± 0.014		-0.851 ± 0.024	
Pb	1.757 ± 0.600		1.506 ± 0.531		-0.160 ± 0.534	
Zn	0.552 ± 0.049		0.473 ± 0.045		-1.815 ± 0.153	
Cu	3.186 ± 0.462		2.728 ± 0.395		0.553 ± 0.269	
Ni	1.344 ± 0.121		1.151 ± 0.111		-0.550 ± 0.159	
Co	0.835 ± 0.112		0.715 ± 0.090		-1.094 ± 0.184	
Mn	1.178 ± 0.062		1.009 ± 0.034		-0.554 ± 0.048	
Cr	1.427 ± 0.083		1.223 ± 0.053		-0.221 ± 0.059	
Fe	-		0.857 ± 0.044		-0.756 ± 0.070	

Table 3

Value of CIA, PIA, CIW, ICV, Al/K, Al/Na, K/Na, Ti/Na and K_2O/Al_2O_3 of the Brahmaputra River sediments.

Sample sites	CIA	PIA	ICW	ICV	Al/K	Al/Na	K/Na	Ti/Na	K_2O/Al_2O_3
S1	65.831	68.488	70.929	1.247	5.831	2.141	0.367	0.118	0.109
S2	65.884	68.561	71.005	1.235	5.817	2.196	0.378	0.121	0.109
S3	64.994	67.633	70.245	1.253	5.538	2.017	0.364	0.112	0.115
S4	68.201	71.957	74.58	1.174	5.078	3.427	0.675	0.188	0.125
S5	64.435	67.593	70.788	1.239	4.573	2.035	0.445	0.111	0.139
S6	63.791	66.878	70.212	1.292	4.441	1.991	0.448	0.111	0.143
S7	65.673	68.881	71.771	1.212	4.923	2.173	0.441	0.116	0.129
S8	65.088	68.131	71.051	1.215	4.942	2.125	0.431	0.114	0.129
S9	63.964	66.755	69.776	1.216	4.891	1.771	0.362	0.098	0.131
S10	65.291	68.507	71.504	1.233	4.785	2.255	0.471	0.124	0.133
S11	63.181	65.984	69.254	1.363	4.589	2.006	0.437	0.114	0.139
S12	66.859	70.181	72.854	1.246	5.174	2.576	0.498	0.139	0.123
S13	67.947	71.591	74.209	1.227	5.128	3.071	0.599	0.167	0.124
S14	67.642	71.126	73.723	1.182	5.221	2.849	0.546	0.154	0.122
S15	67.738	71.251	73.842	1.172	5.219	2.847	0.546	0.155	0.122
Min	63.181	65.984	69.254	1.172	4.441	1.771	0.362	0.098	0.109
Max	68.201	71.957	74.581	1.363	5.831	3.427	0.675	0.188	0.143
Average	65.768	68.901	71.716	1.233	5.076	2.365	0.467	0.129	0.126
SD	± 1.608	± 1.895	± 1.711	± 0.048	± 0.417	± 0.476	± 0.092	± 0.025	± 0.011

moderately to strongly polluted (I_{geo} is 2–3, class 3); strongly polluted (I_{geo} is 3–4, class 4); from strongly to extremely polluted (I_{geo} is 4–5, class 5) and extremely polluted (I_{geo} is >6, class 6) (Muller, 1969). The I_{geo} value of all studied elements shows value less than zero except Cu (Fig. 2c). The I_{geo} value of Cu (0.48 ± 0.29) indicates the sediments are unpolluted to moderately polluted (class 1) by Cu (Table 2). The total index of geo-accumulation (I_{tot}) is defined as the sum of I_{geo} for all trace elements obtain from the site (Bowen, 1979). Therefore, according to Muller's classification, the Brahmaputra river sediments were unpolluted (class 0). The PLI [$= (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n}$], for a particular site can be estimated using the method proposed by Tomilson et al. (1980), where CF is the contamination factor and n is the number of metals (Tomilson et al., 1980). The calculated PLI of the study is (0.971 ± 0.55), which indicates no overall pollution in the Brahmaputra River sediments (Fig. 2d).

The oxide composition of the sediment samples were: SiO_2 (64.77 ± 0.154 wt%), Al_2O_3 (10.99 ± 0.327 wt%), Fe_2O_3 (4.49 ± 0.242 wt%), MgO (2.78 ± 0.214 wt%), MnO (0.09 ± 0.004 wt%), CaO (2.48 ± 0.067 wt%), Na_2O (1.97 ± 0.386 wt%), K_2O (1.46 ± 0.167 wt%), P_2O_5 (0.03 ± 0.016 wt%), TiO_2 (0.51 ± 0.010 wt%) and LOI (2.36 ± 0.067 wt%). Plagioclase is one of the most abundant minerals in the earth's crust and is highly vulnerable to alteration and weathering. In basaltic to andesitic rocks, the plagioclase group ranging sodium feldspar to calcium feldspar as the major constituents. The end members of their constituents demonstrate the parent environment and material. Quantitative estimation of the chemical weathering of silicates by calculating

the values of chemical index of alteration (CIA), plagioclase index of alteration (PIA) and chemical index of weathering (CIW) were widely used to interpret the weathering history of modern and ancient sediments (Nesbitt and Young, 1982; Harnois, 1988; Fedo et al., 1995). PIA values are generally used to quantify the degree of source rock weathering. We determine PIA (= $Al_2O_3/(Al_2O_3 + CaO + Na_2O) \times 100$) using the relation proposed by Fedo et al. (1995). Generally maximum PIA value (equal to 100) indicates completely altered minerals such as kaolinite, gibbsite etc. However, the half of the maximum PIA value indicates unweathered plagioclase (Muller, 1969). The studied sample exhibits PIA value (70.20 ± 1.58), which indicates the weathering nature of the source rocks (Table 3). The PIA suggests almost moderate plagioclase weathering in the source area.

An estimation of the degree of chemical weathering of the sediments is obtained by calculating the CIA (= $[Al_2O_3/(Al_2O_3 + CaO + Na_2O + K_2O)] \times 100$) (Nesbitt and Young, 1982). CIA is a constructive technique to evaluate the progressive alteration of plagioclase and K-feldspars to clay minerals. CIA reflects the changing proportions of feldspar and Al-rich secondary minerals in the depositional environment. The low CIA value indicates little chemical alteration while a high values infers an intensive alteration. The CIA values below 60 display low chemical weathering, between 60 and 80 indicate moderate chemical weathering and more than 80 exhibit extreme chemical weathering (Fedo et al., 1995). The observed CIA (65.02 ± 1.44) of the Brahmaputra River sediment samples indicates moderate chemical weathering (Table 3).

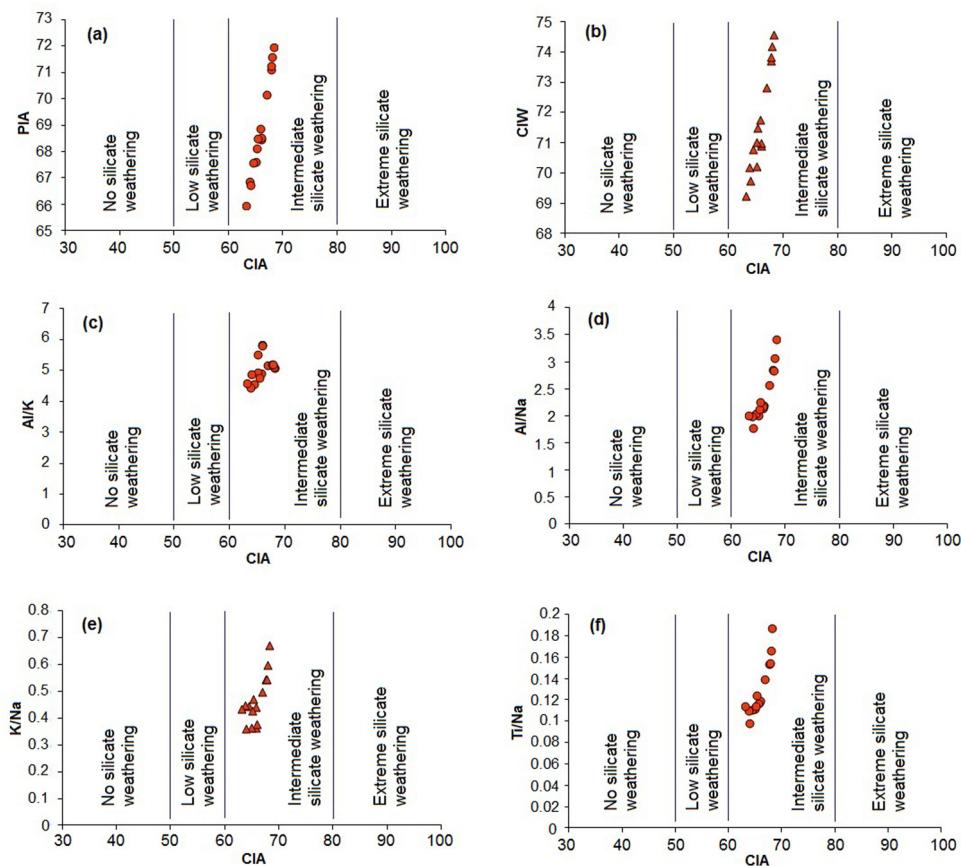


Fig. 3. Scatter plots of chemical index of alteration (CIA) vs. PIA, CIW, Al/K, Al/Na, K/Na, and Ti/Na of the Brahmaputra River sediments.

The composition of non-quartz components of the samples are evaluated by calculating the ICV ($= (\text{Fe}_2\text{O}_3 + \text{K}_2\text{O} + \text{Na}_2\text{O} + \text{CaO} + \text{MgO} + \text{MnO} + \text{TiO}_2)/\text{Al}_2\text{O}_3$) (Cox et al., 1995). The ICV value less than 1 indicates the presence of more clay minerals, while its value greater than 1 indicates more rock forming minerals like plagioclase, alkali-feldspar, pyroxenes etc. (Cox et al., 1995). The calculated ICV value (1.233 ± 0.048) indicates the presence of less clay minerals and more rock forming minerals in the Brahmaputra River sediment samples (Table 3). According to Harnois (1988) the value of CIW can be ascertained as $\text{CIW} = [\text{Al}_2\text{O}_3/(\text{Al}_2\text{O}_3 + \text{CaO} + \text{Na}_2\text{O})] \times 100$ (Harnois, 1988). However, the $\text{K}_2\text{O}/\text{Al}_2\text{O}_3$ ratios indicates how much of alkali feldspar versus plagioclase and clay minerals were present in the original rock. The $\text{K}_2\text{O}/\text{Al}_2\text{O}_3$ ratio is less than 0.3 for clay, while its value 0.3–0.9 for feldspars (Cox et al., 1995). The $\text{K}_2\text{O}/\text{Al}_2\text{O}_3$ ratio (0.126 ± 0.011) of the studied samples indicates predominance of clay minerals. In order to understand the nature of weathering of silicates using the relationship between the calculated indices of chemical weathering and elemental ratios, scatter plots of PIA, CIW, Al/K, Al/Na, K/Na, and Ti/Na against CIA were generated (Fig. 3). Chemical weathering affects the major and trace element geochemistry and mineralogy of the siliciclastic sediments. CIA values of all the samples are linearly related to PIA and CIW. The ratios of Al/Na, Ti/Na and K/Na gradually increase with increasing chemical weathering, however, a lack of linear interrelation with the ratios of Al/K is observed. Generally the ratios of Al/Na and Ti/Na indicate the increasing proportions of clay to primary plagioclases with increasing chemical weathering. Fig. 3 demonstrates the varying degrees of chemical weathering as: no silicate weathering, low silicate weathering, intermediate silicate weathering and extreme silicate weathering (Nesbitt and Young, 1982;

Roy et al., 2008). The interrelation between the indexes reflects the silicate weathering intensity. The studied Brahmaputra River sediment samples exhibit the intermediate silicate weathering.

4 Conclusion

The investigation of Brahmaputra River sediments shows that the order of the mean concentrations of metals as: Al > Fe > Ca > Mg > Ti > Mn > Cr > Cu > Zn > Ni > Pb > Co. The enrichment factor (EF), contamination factor (CF), geo-accumulation index (I_{geo}) and pollution load index (PLI) were applied for assessment of contamination. The EF suggests moderate enrichment of Cu (2.40 ± 0.50) in the Brahmaputra River sediments. Moreover, CF suggests the study area is moderately contaminated by the elements Cu, Pb, Cr, Mn and Ni which have been attributed mainly to dispersion from the mineralized zone of the upper catchment area. The I_{geo} indicates the sediments are unpolluted to moderately polluted by Cu. The PLI indicates sampling sites have no overall pollution. The values of PIA and CIA reflect a moderate plagioclase weathering in the source area. The indices exhibit the studied samples belong to the intermediate silicate weathering.

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