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Geochemical behaviour and risk assessment of trace elements in a tropical river, Northwest Borneo

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Abstract

By convention, dissolved trace elements in the river water are considered to be the fraction that passes through a 0.45 µm filter. However, several researchers have considered filtration cut-off other than 0.45 µm for the separation of dissolved trace elements from particulate fraction. Recent research indicated that trace elements could exist in particulate form as colloids and natural nanoparticles. Moreover, the trace elements in the continental dust (aerosols) constitute a significant component in their geochemical cycling. Due to their high mobility, the trace elements in the micron and sub-micron scale have biogeochemical significance in the coastal zone. In this context, this study focuses on the highly mobile fraction of trace elements in particulates (< 11 µm) and dissolved form in the Lower Baram River. A factor model utilizing trace elements in the dissolved and mobile phase in the particulates (< 11 µm) along with water column characteristics and the partition coefficient (K_d) of the trace elements indicated a more significant role for manganese oxyhydroxides in trace element transport. Perhaps, iron oxyhydroxides play a secondary role. The factor model further illustrated the dissolution of aluminium and authigenic clay formation. Except for Fe and Al, the contamination risk of mobile trace elements in particulates (< 11 μm) together with dissolved form are within the permissible limits as per Malaysian water quality standards during monsoon (MON) and post-monsoon (POM) seasons.

Keywords: River; Estuary; Trace Elements; Particulates; Partition Coefficient; Iron-Manganese Oxides.

1. Introduction

The riverine network across the globe play a vital role in weathering the rocks on the continent and in transferring them to the world oceans in dissolved and particulate form (Chester and Jickells, 2012; Gaillardet et al., 2014; Qu et al., 2019). The warm and humid climate of the tropics with intense rainfall enhances weathering in the river basins (Dessert et al., 2003; Godderis et al., 2009; Goudie and Viles, 2012; Hartmann et al., 2014; Macdonald et al., 2019). The prevailing high energy conditions in the tropical river basins can rapidly remove the products of weathering as solutes and particulates from the source area to the sea in a relatively shorter residence time compared to a more temperate climate (Sultan et al., 2011). Thus, the contribution of trace elements from tropical river systems to the coastal oceans is significant, and estuarine regions of such river systems are ideal places to explore the trace element geochemistry (Sultan et al., 2011; Kilunga et al., 2017; Borah et al., 2018; Prabakaran et al., 2019; Wu et al., 2019; Zhang et al., 2019; Zhou et al., 2019). Asian rivers with less anthropogenically influenced basins may present an insight into the weathering and erosion control over aquatic chemistry (Zhang and Huang, 1993).

The estuaries are the delivery points of weathered products, and they also serve as an interface between the river and the ocean with gradational changes in their water column characteristics that render a unique environment (Price et al., 1999; Bianchi, 2007; Fernandes et al., 2011; Samanta et al., 2018). The geochemical processes occurring within the estuarine regions are unique and essential to understand the fate of trace elements transported through the rivers. These trace elements influence the coastal water quality and the health of the coastal ecosystem (Looi et al., 2013; Gopal et al., 2018).

River induced changes to the marine environment associated with increased sediment load, nutrients and pollutants are a severe threat to coral ecosystems. Asia is renowned for its global hotspot status for coral biodiversity especially in the South China Sea and the Coral Triangle, supporting a diverse population of marine life including coral, algae, reef fishes and other invertebrates (Roberts, 1993; Bellwood and Hughes, 2001; Hoeksema, 2007; Huang et al., 2015; Heery et al., 2018). Despite being resilienet, the Miri-Sibuti Coral Ecosystem in the sub-aqueous Baram delta region exhibits signs of adverse effects owing to poor water quality and sediment load that endangered the coral health (Browne et al., 2019). Moreover, fishes in the coastal Miri exhibits trace metals accumulation but within the permissible limits (Anandkumar et al., 2017, 2018, 2019). However, long-term trends are not known. Therefore, knowledge of the behaviour of trace elements in the tropical river systems, especially in their estuarine region, becomes essential. This work, carried out in the Lower Baram River of Malaysian Borneo, is one such attempt to understand the geochemical processes that govern the behaviour of trace elements in the water column.

1.1 Partitioning of trace elements between dissolved and particulate form

The Baram River originates in the Kelabit Highlands of Borneo and flows through a dense rainforest before reaching the South China Sea. The river water pH is slightly acidic that can be ascribed to the presence of dissolved organics (Oliver et al., 1983) which enhances weathering (Walther, 1996) and results in the formation of dissolved, particulate and metal-organic complexes that finally reaches the estuary (Tipping and Heaton, 1983; Guo and Macdonald, 2006; Gaillardet et al., 2014).

The partitioning of trace elements between dissolved and particulate form in the estuaries has ramifications for their transport to the ocean, interaction with biota, biogeochemical cycling, and elemental budgeting (Huang et al., 2012; Jin et al., 2010; Wang and Liu, 2003, 2008; Xiao et al., 2014). Chemically both the dissolved and particulate form of trace elements exists in their different speciation state. While dissolved trace elements form organic and inorganic complexes, the particulate form of the trace elements exists mainly as adsorbed species on clay particles, colloids and natural nanoparticles that serve as a host for their transport (Dai et al., 1995; Kretzschmar and Schafer, 2005; Plathe et al., 2013; Tepe and Bau, 2014). There exists a dynamic equilibrium between dissolved and particulate forms of trace elements. The prevailing water column characteristics namely pH and redox potential govern such an equilibrium. This dynamic equilibrium results in a continuous exchange of the trace elements between dissolved and particulate form. The equilibrium constant that is unique to the prevailing environmental conditions of the concerned trace element is defined as the "partition coefficient" (O'Connor and Connolly, 1980; Shi et al., 1998; Lu and Allen, 2001). Theoretically, the partition coefficient is expected to be a constant for the given environmental conditions. Thus, it has been used as a probe to ascertain whether a given trace element has more affinity to remain in dissolved or particulate form. Such a piece of knowledge has helped us to understand the behaviour of trace elements in the coastal regions (Tang et al., 2002; Oursel et al., 2014; Wang et al., 2017). However, practical observations have always yielded a range of values for the partition coefficient due to the involvement of various influencing factors (Boyer et al., 2018; Tomczak et al., 2019) in the water column. Even then the fact remains that if the chemical composition of the water column, the particle nature and the environmental conditions are identical, the partition coefficient of a trace element is expected to remain within a very narrow range in all estuarine systems. On the other hand, the partition coefficient can be related to the water column characteristics to infer the factors or geochemical processes that influence the trace elements concentration. This work revolves around these concepts for the interpretation of geochemical characteristics of the trace elements in the estuarine region. However, we have introduced modifications to the definition of the "partition coefficient" to include reactive trace elements in the particulates along with the dissolved form. That, we believe, is essential to the understanding of the role of nanoparticles, colloids and particulates on the trace elements behaviour in the estuaries (Supplement –I). Therefore, for trace elements analysis, the samples were filtered through 11 µm filter (Whatman No.1) using a polycarbonate filtration assembly and acidified to pH < 2 (refer Section-3.1 for more details).

2. Study Area

Borneo is the third-largest island in the world situated along the equator, comprising of the two Malaysian states along with the nation of Brunei and Indonesia. Sarawak is one of two Malaysian states situated on the northwest coast of the island of Borneo, bordering the Malaysian state of Sabah to the northeast, Indonesia to the south, and surrounding Brunei. Extending to the length of around 466 km stretch, the Baram River is the second-longest river in Sarawak next to the Rajang River (Anandkumar, 2016). Originating in the Kelabit Highlands, the Baram River initially flows westwards through tropical rainforests and then turns northward to drain into the South China Sea at Miri (near Kuala Baram). The Baram River covers a catchment area of about 22,800 km^2 and delivers an estimated freshwater of 1590 m^3/s and 2.4×10^{10} kg of sediment/year to the South China Sea (Sandal, 1996; Straub and Mohrig, 2009). The study area (Figure-1) covers the lower stretch of the Baram River, extending to a length of 111 km from Baram River mouth to Marudi. A transitioning water column with a gradational salinity increase from Marudi to Kuala Baram characterises the study area. The geology of the Baram River Basin consists predominantly of meta-sedimentary to sedimentary rocks aged from Paleogene to Recent. Oligocene, Miocene and Eocene meta-sedimentary to sedimentary rocks primarily cover the upstream region. In contrast, the downstream region primarily consists of the Quaternary river and coastal alluvium as the Baram River mouth was initially located near Marudi but has extended towards the north over the past 5000 years (Caline and Huong, 1992). For more details on regional geology, climate, tectonics, weathering and coastal processes, please refer earlier works (Calvert et al., 1991; Caline and Huong, 1992; Hiscott, 2001; Hutchison, 2005; Wang et al., 2011; Nagarajan et al., 2014, 2015a, b, 2017a, 2019; Yan et al., 2015; Ramkumar et al., 2018; Prabakaran et al., 2019; Wurster et al., 2019).

3.Methodology

3.1. Choosing the filtration cut-off

In the coastal areas, earlier studies have taken the nominal size of particulates as 0.2 to 10 μm (Hurst and Bruland, 2007), 0.4 to 10 μm (Weinstein and Moran, 2004) or, greater than 0.4 μm (Lambert et al., 1984; Helmers, 1996; Weinstein and Moran, 2004) though custom made filters that approximate the 0.45 μm filter has also been used for large volume sampling in Atlantic and Pacific oceans for separating particulates (Krishnaswami and Sarin, 1976; Krishnaswami et al., 1976). Forest fires in Borneo and surrounding other Southeast Asian regions are a good source of organic aerosols apart from the continental dust (Durkee et al., 1991; Rasch et al., 2001; Idris et al., 2005; I-I Lin et al., 2007; Langner et al., 2007; Gabey et al., 2010; 2011; Lee et al., 2017; Bikkina and Sarin, 2019). Thus, atmospheric fallout is one of the sources of trace elements in the Baram River Basin. The global atmospheric dust range in size from 100 nm to 100 μm (Pye, 1987; Formenti et al., 2011 and the references cited therein) and the long transported aerosol particles deposited in the ocean is finer than 10 μm (Pye, 1987; Bikkina and Sarin, 2019). Gabey et al. (2011) have shown that in Borneo the non-fluorescent aerosols peaked at 0.8 to 1.2 μm while the fluorescent primary biological aerosol peaked at 3 to 4 μm. Ohta et al. (2006) have found that the nominal size of aerosol in the Asian region is 11 μm. To take into account the trace elements associated with the atmospheric dust fallout and the primary biological aerosol, a filtration cut-off of 11 μm is considered optimal. Apart from the aerosols, the flocs are another carrier of trace elements in the estuarine region of the Baram River. There is an increasing salinity in the river water from Marudi to Kuala Baram and the dissolved iron and manganese are expected to flocculate and adsorb the dissolved trace elements (Sholkovitz, 1978). Moreover, in the forested soil mobilisation of Al has been reported by several earlier studies (Evans, 1986; Mulder and Stein, 1994; Van Hees et al., 2004). The aluminium is amphoteric (Pacioglu et al., 2016) and exists as $Al(OH)_3$ and polynuclear complexes (Palleiro et al., 2018) that serve as a potential adsorbent for many trace elements akin to iron and manganese oxyhydroxides (Singh et al., 1984). Thus, most of the riverine dissolved trace elements are expected to remain as adsorbed species with a dynamic equilibrium between their dissolved and particulate forms (Hurst and Bruland, 2007). By filtering the river water through 11 μm filters, the trace elements associated with Fe, Mn oxyhydroxides and Al hydroxides could be extracted.

Several studies documented the role of colloids in the bioavailability of trace elements to aquatic organisms (Ferreira et al., 2008; Zhang and Davison, 2000; Pan and Wang, 2002; Wang and Guo, 2000, Guo et al., 2002; Seah et al., 2017). Moreover, the colloidal-sized organic matter, are known to adhere firmly with certain trace metals and trace organics, thereby influencing the bioavailability of trace metals to aquatic organisms (Sigleo and Means, 1990; Santschi et al., 1997, 1999; Wen et al., 1999; Carvalho et al., 1999; Doblin et al., 1999; Wang and Guo, 2000). Therefore, filtering of water samples through a 0.45 μm filter will not serve the purpose of estimating the quantum of available trace elements to aquatic biota, because colloids play a significant role in the pathway of metals into the food chain (Farag et al., 2007). Hence, while considering bioavailability, the quantum of metals associated with the colloidal phase must also be considered as aquatic organisms are likely to consume colloidal particles. Therefore taking a filtration cut-off of 11 μm will provide additional knowledge towards trace elements bioavailability to aquatic populations.

Thus, the trace elements are measured in 11 µm filtered (Whatman No.1) river water instead of using the 0.45 µm as in many earlier studies (Karbassi et al., 2008; Stolpe and Hassellöv, 2010). After filtration, all water samples were acidified to pH < 2 to release labile forms of trace elements from the particulates. Acidification of the water samples shall release trace elements transported by the river in the form of colloids (Duarte and Cacador, 2012) natural nanoparticles (Wigginton et al., 2007) and particulates of size < 11 µm. While the results do not represent the truly dissolved form of the trace elements, it should be noted that the methodology is designed to desorb the highly labile fraction of trace elements from the particulates.

3.2. Sample collection, analysis and data processing

Water samples were collected during January and April of 2015, representing monsoon and postmonsoon seasons, respectively. Thirty two river water samples were collected in acid cleaned 1000 mL polyethylene containers from the Lower Baram River (Kuala Baram to Marudi) using an acid-cleaned non-metallic aqua-trap water sampler. All the samples were brought to the laboratory within 12 hours after collection. For trace analysis, the samples were filtered through an 11 µm filter (Whatman No.1) using a polycarbonate filtration assembly and acidified to pH < 2. The acidified filtrates were stored under refrigeration (< 4° C) until analysis for the trace elements. The water column properties such as temperature, pH, salinity, total dissolved solids (TDS), electrical conductivity (EC) and redox potential (Eh) were measured in the field using necessary probes (Thermoscientific Orion Star Plus, USA). Dissolved oxygen was measured in the field by using a DO meter (YSI Pro) and turbidity was measured using Hach-2100Q portable turbidimeter. The major ions and nutrients analyses were carried out following standard procedures (Supplementary Table-1). Trace elements were analysed using a Flame AAS (Perkin Elmer AA400). Analytical control was assured by preparing the reagent blanks and utilizing standard solutions as quality control at every five-sample interval. The precision of AAS analytical results are within 5%, and accuracy is below 10%.

3.3 Statistical analysis

In order to reveal the significant variations of the analysed parameters between seasons, a paired t-test was performed (Supplementary Table-2). The partition coefficient (K_d) values were calculated from the reported concentration of trace elements in the sediment (Prabakaran et al., 2019) and water column. The data, subjected to Principal Component Analysis (PCA) and varimax rotation using SPSS software version 17, provided factor loadings for further interpretation. Because the units of measured parameters were not the same, the entire data set was standardised before carrying out the factor analysis.

3.4 Risk assessment indices

The Contamination Index (C_d) and Heavy metal Evaluation Index (HEI) were used to evaluate the water quality of the Lower Baram River. The Contamination Index (C_d) of Backman et al. (1998) evaluated the degree of contamination of Fe, Mn, Al, Cu, Zn, Pb, Ni, Cr, Cd, Co and Hg by using the following equation.

$$C_d = \sum_{i=1}^{n} C_{fi}$$

$$Cfi = \frac{CAi}{CNi} - 1$$

Where,

C_{fi} = contamination factor for the ith component

C_{Ai} = analytical value for the ith component

C_{Ni} = upper permissible concentration of the ith component (N denotes the normative value).

The resultant C_d values comprise three categories as low ($C_d < 1$), medium ($C_d = 1$ to 3) and high ($C_d > 3$).

The HEI revealed the overall water quality with respect to heavy metals (Edet and Offiong, 2002). This index was calculated using the following equation:

$$\mathrm{HEI} = \sum_{i=1}^{n} \mathrm{Hc/Hmac}$$

Where, H_c is the monitored value of the ith parameter, and H_{mac} is the maximum admissible concentration of the ith parameter.

4. Results and discussion

The descriptive statistics of the analysed parameters including physicochemical parameters, major and minor ions, nutrients and trace element concentrations are given in Table-1. From the results of the paired t-test, all the physicochemical parameters vary significantly ($P < 0.01$) between seasons except resistivity. Similarly, all the major and minor ions such as chloride, carbonate, bicarbonate, sodium, potassium, calcium and magnesium varied significantly ($P < 0.01$; < 0.05 for bicarbonate) between the seasons. Nitrate also showed a significant difference ($P < 0.01$), between seasons. Among the trace elements, Fe, Mn, Al, Cr, B, Zn and Pb varied significantly ($P < 0.01$; $P < 0.05$ for Al) between seasons while Cu, Co, Ni, Cd and Hg were not significantly different ($P < 0.05$).

4.1 Geochemistry of trace elements in the Baram River surface water during the MON season

Factor analysis was carried out to understand the geochemical processes occurring in the water column. As the ionic composition of the water column, pH, dissolved oxygen and temperature always have interaction with trace elements (Gundersen and Steinnes, 2003; de Souza Machado et al., 2016), they have also been included in this factor model to elucidate the geochemical processes operating within the Lower Baram River. The component matrix with six factors (Supplementary Table-3) has eigenvalue > 1 and reveals 81.32% of the variance in the data.

Factor-1 is loaded with Pb, Zn, Mn, Ni, Cu, Co, Hg and Al and explains 28.16% variance in this factor model. The association of these elements suggests the dissolution of particulate trace elements adsorbed onto manganese oxyhydroxides. (Sholkovitz et al., 1978; Means et al., 1978; Johnson, 1986; Dong et al., 2000; Li et al., 2001; Kay et al., 2001; Feng et al., 2007; Zhong and Wang, 2008; Suda and Makino, 2016). As observed in Supplementary Table-4, a high factor score of 4.96 for the sample BES-26 and significantly lower values for all other samples implies that such a dissolution is predominant at the confluence of Sungai Bakong (low end tributary) with the Baram River. The positive loading of Al under factor-1 and its high concentration in the water column at the confluence of Sungai Bakong (Sample BES-26) indicates the dissolution of aluminium. Such dissolution is possible by the low pH of organic-rich water entering the Baram River from Sungai Bakong. Moreover, during the fieldwork, a strong swirling was observed at this confluence point, which might aid in thorough mixing and promote dissolution. While it has been well established that iron and manganese oxyhydroxides are primary carriers of trace elements, this factor model does not exhibit such a role for iron. Indeed, it is the strong interaction of Mn-oxyhydroxide surface with Al in the presence of adsorbed organics (Tipping and Heaton, 1983), which is thought to play a significant role in the formation of metal-organic complexes that serve as major carriers of trace elements.

Factor-2 explained 16.57% variance with the loading of sulphate, B, Fe, K, Al and temperature. Of these, temperature, sulphate and boron have negative loading and Fe, K and Al have a positive loading. Though the loading of temperature, sulphate, boron and Al are on the lower side, they have been taken into account because only their association provides a meaningful geochemical interpretation. The association of temperature with sulphate implies the temperature-controlled jarosite dissolution. The jarosite is a alunite series mineral with the formula $K.Fe_3(SO_4)_2(OH)_6$ and is a member of the aluminium-phosphate-sulphate (APS) minerals with the general formula $AB_3(XO_4)_2(OH)_6$, where A is a large cation (Na, U, K, Ag, NH_4^+, Pb, Ca, Ba, Sr, REE's), and B is one of the cations in the group Al, Fe, Cu and Zn (Dill, 2001). Alunite can form at a pH higher than 3.3. The primary source for Al is the dissolution of aluminium silicates during microbially mediated pyrite oxidation, which releases sulphuric acid (Sánchez-España et al., 2016). The tropical climatic conditions are most suitable for the formation of APS minerals (Dill et al., 2002), and peraluminous parent rocks enriched in sulphur and phosphorus are a prerequisite (For a review, refer Dill, 2001). However, the Baram river basin consists of turbidites and the occurrence of peraluminous rocks is not reported.

The acid sulphate soils are a source of jarosite. Acid sulphate conditions occur in sand, peat, and more extensively in clays (Dent, 1992). The acid sulphate soils are the source of jarosite. It is a common observation that in peat and organic-rich mangrove sediments, pyrite oxidation results in the formation of jarosite (Furukawa, 1988; Shamshuddin et al., 2004; Mohamad et al., 2016). Of the 13 million ha of acid sulphate soil worldwide, around 6.7 million ha is in the Southeast Asian region (Shamshuddin et al., 2004). In Malaysia, the acid sulphate soil covers 0.5 million ha (500,000 ha) of which Peninsular Malaysia accounts for 110,000 ha

(Kanapathy, 1973; Abdul Halim et al., 2018). Of this, the very acidic soil occurs in 25,000 ha of swamps, where *Nipa fruticans* grows (Kanapathy, 1973). Some of the plant species that thrive in acidic soils include *Meluleuca leucadendron*, *Rhizophora mucronata* and *Nipa fruticans* (Rahman et al., 2018). The *Nipa fruticans* also occurs in the Lower Baram River apart from *Sonneratia caseolaris*, and no other mangrove species thrive here. Andriesse et al. (1973) report the occurrence of acid sulphate soils in the mangrove swamps along the Sarawak river, near Kuching, in the State of Sarawak. They also occur along the coastal plains of Sarawak (Teng, 2005), the Liang Formation of Brunei basin (Wu, 1994), Kota Samarahan-Asajaya area of West Sarawak, Malaysia (Mohamad et al., 2016), and Tukau Formation adjacent to the study area (Nagarajan et al., 2017b).

Though the mineralogical studies were not carried out for the identification of aluminium-phosphate-sulphate (APS) minerals, this cannot preclude the chances for its formation in a terrain where volcanic, sedimentary, and metamorphic processes have taken place, and alunite has an excellent chance of formation in all these environments (Dill, 2001). Given the fact that the Baram River basin mainly consists of turbidites and shale, which are a rich source for the sedimentary pyrite, its dissolution might release sulphuric acid into the environment resulting in the formation of alunite group of minerals.

The river carries the aluminium bearing minerals towards downstream in suspension. The negative loading of sulphate and boron under factor-2 indicates the dissolution of alunite, whereas the positively loaded Fe, K and Al under the same factor imply the formation of authigenic clay minerals. Another factor model using the partition coefficient (K_d) of the elements (Supplementary Table-5) confirms this proposition. The positively loaded K_d values of Fe and Al, and SO_4 points out that in the absence of sulphate reduction (suboxic conditions) higher dissolved concentrations of Fe and Al are favoured (Rahman, 2016). Then authigenic clay minerals form by consuming the dissolved potassium in the water column which is implied by its negative factor loading (Mackin and Aller, 1986; Mackenzie and Kump, 1995; Rahman, 2016; Church, 2016). Such a formation of authigenic clay minerals is rapid in tropical and sub-tropical deltas (Michalopoulos and Aller, 2004; Presti and Michalopoulos, 2008; Loucaides et al., 2010; Rahman, 2016) possibly by enhanced forward reaction under increasing temperature (endothermic). The formation of authigenic minerals in estuarine and coastal areas is a widely disputed topic. Therefore, further investigation is necessary.

Factor-3 accounted for 11.08% variance. The EC, Na^+ and Cl^- exhibit positive loading, implying the influence of seawater. Eventhough there is no measurable salinity during MON season due to flooding of the river, the loading of these parameters on factor-3 demonstrates seawater incursion in the river. The Baram River has no distributaries and behaves like a salt-wedge estuary. The density difference drives the seawater underneath the freshwater of the river. The sinuosity of the Baram River is lesser downstream Marudi and seawater incursion shall be facilitated in this stretch. This salinity incursion is also observable in the factor model given in Supplementary Table-5. Moreover, authigenic clay mineral formation occurs only in the presence of saline water (Mackenzie and Kump, 1995), confirming the validity of our interpretation. Another source of sodium and chloride ions is sea salt. Worldwide, the annual sea salt aerosols production is 10×10^{15} to 11.7×10^{15} g/yr [Mæller, 1990; Gong et al., 1997] but estimates for the Baram River basin is not available. However, sea salt can also form a significant contribution to the dissolved load of this river. The distinction between the contribution of Na^+ and Cl^- from the sea salt and saline water could not be made due to lack of available data.

Factor-4 explained 9.20% variance and shows the loading of pH and DO, illustrating the role of water column productivity on the pH and dissolved oxygen (Feely et al., 2010). The water column productivity increases the pH and releases oxygen to the water column. Water column productivity is often affected by turbidity in tropical river systems (Lee, 1990; Davies et al., 2008). Despite their moderate contribution to overall production, algae (periphyton and phytoplankton) appear to be the prominent drivers of aquatic food webs in tropical river systems (Lewis et al., 2001; Winemiller, 2004; Douglas et al., 2005). Factor-5 accounted for 8.80 % variance and loaded with Ca^{2+}, Mg^{2+}, Cl^- and HCO_3^-. This implies ion exchange reactions involving Na^+, Ca^{2+} and Mg^{2+} (Cerling et al., 1989). The majority of the water samples are not showing up an excess of Na^+ over Cl^- expected to result from the weathering of shale terrains (Cerling et al., 1989) (Supplementary Figure-1). Moreover, due to lack of excess (Ca^{2+} + Mg^{2+}) over (SO_4^{2-} + alkalinity), it is inferred that silicate weathering might contribute to excess alkalinity and Na^+ and K^+ ions (Cerling et al., 1989) (Supplementary Figure-2).

Factor-6 accounted for 7.59% variance in the factor and loaded with orthophosphate and nitrate, indicating the remobilisation of the nutrients from the sediments to the water column (Zhang et al., 1997). Such a remobilisation is possible upon the degradation of sedimentary organic matter (Emerson et al., 1980; Nedwell et al., 1994; Morford et al., 2005). Such degradation is likely to release low molecular weight organics into the water column, (Nedwell et al., 1994; Beck et al., 2008), which may form stable metal-organic complexes (Beck et al., 2008) and serve as an essential repository for some of the elements like Cu and Zn (Widerlund, 1996). The factor model in Supplementary Table-5 indicates such an association. The other possible source of orthophosphate and nitrate is agricultural activities in the river catchment area (Sim et al., 2017).

4.2 Geochemistry of trace elements in the Baram River surface water during POM season

Supplementary Table-6 presents the factor model for the POM season. Six factors accounted for 82.61% of the variance. Factor-1 exhibits loading of salinity, Cl^-, SO_4^{2-}, B, Pb, Co, Cd, Na^+, Ca^{2+} and Mg^{2+}. The $Na^+/Cl^- < 1$ implies possible ion exchange reactions involving Na^+, Ca^{2+} and Mg^{2+} (Cerling et al., 1989) (Supplementary Figure-3). Further, an excess of ($Ca^{2+} + Mg^{2+}$) over ($SO_4^{2-} + HCO_3^-$) indicates ion exchange reactions where Na^+ replaces Ca^{2+} and Mg^{2+} in the clay minerals (Cerling et al., 1987; El-Sayed et al., 2012) (Supplementary Figure-4). Correlation between ($Na^+ + K^+$) and SO_4^{2-} indicates sulphide oxidation through the dissolution of jarosite (Mermut and Arshad, 1987) (Supplementary Figure-5). Factor-2 is accounted for 16.43% of the variance with positive loadings of pH and DO, and negative loading by orthophosphate and ammoniacal-nitrogen. Orthophosphate is remobilized from the sediments only under reducing conditions (Sundby et al., 1986) whereas ammonia is a by-product of anaerobic bacterial degradation of organic matter (Canfield et al., 1993; Baric et al., 2002). With increasing seawater influence, the pH and DO increase and the production of orthophosphate and ammonical nitrogen diminishes.

Factor-3 shows positive loading of K and negative loading of Fe that explains 9.20% of the variance indicating either loss of K from Fe bearing clay minerals (Craw, 1981) or loss of iron from biotite (Acker and Bricker, 1992) that exists in particulate form in the < 11 μm fraction. Factor-4 accounts for 6.27% of the variance and is explained by the positive loading of Hg and negative loading of Cu which indicates two different sources. Factor-5 is loaded with nitrate and accounts for 5.27% variance, indicating nitrification in the water column (Scott and Abumoghli, 1995; Cébron et al., 2003; Strauss et al., 2004). Factor-6 has accounted for 4.99% of the variance with Zn and Al exhibiting positive and negative loadings, respectively.

The K_d-Zn exhibits a positive loading under factor-5 (Supplementary Table-7), while nitrate is negatively loaded, indicating Zn toxicity to water column nitrification (Hu et al., 2004; Zarcinas and Rogers, 2002).

4.3 Risk assessment indices

The Contamination Index (C_d) for the Baram River water during the MON and POM seasons is presented in Supplementary Table-8. The river water quality falls into the high contamination category ($C_d > 3$) for both seasons. The C_d values range from 28.49 to 643.36, with a mean of 176.19 during the MON. During POM, the C_d varies from 36.29 to 92.60 with a mean of 47.26 during the POM season. The calculated HEI values (Supplementary Table-8) ranged from 39.49 to 654.36 with a mean of 187.19 during the MON season and 47.29 to 103.60 with a mean of 58.26 during the POM season. The river water samples were filtered using 11 μm filters that contained both purely dissolved and particulate trace elements. Thus, the water samples showed a high contamination category as revealed from the risk assessment indices. Moreover, when compared with the Malaysian water quality standards (Supplementary Table-9), the majority of the elements fall between the Class I and IV, however, Fe and Al fall under Class V.

5. Conclusions

Seasonally significant ($P < 0.05$) difference exists for temperature, pH, DO, EC, TDS, salinity, turbidity, redox potential, Cl^-, HCO_3^-, SO_4^{2-}, NO_3^- Na^+, K^+, Ca^{2+}, Mg^{2+}, B, Pb, Zn, Mn, Fe, Cr and Al. Whereas, the water column concentrations of PO_4^{3-}, NH_3-N, Ni, Cu, Cd, Co and Hg do not show any significant variation between seasons. During the monsoon, the desorption of Pb, Zn, Ni, Cu, Co, Hg and Al from manganese oxyhydroxides carried in the particulate form by the Baram River is dominant. Jarosoite dissolution, authigenic clay mineral formation, water column productivity, ion exchange and remobilisation of the nutrients are other inferred geochemical processes. During post monsoon, ion exchange is the primary geochemical process followed by the remobilization of orthophosphate and the production of ammonical nitrogen. The Contamination Index (C_d) shows that water samples are highly contaminated. However, such a result needs careful inspection since the particulate-bound trace elements in < 11 μm fraction is taken up for the calculation. The water quality is within the safer limits as per the Malaysian water quality standards, except for Fe and Al.

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Conflict of Interest

The authors declare that there are no conflicts of interest.

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Figure

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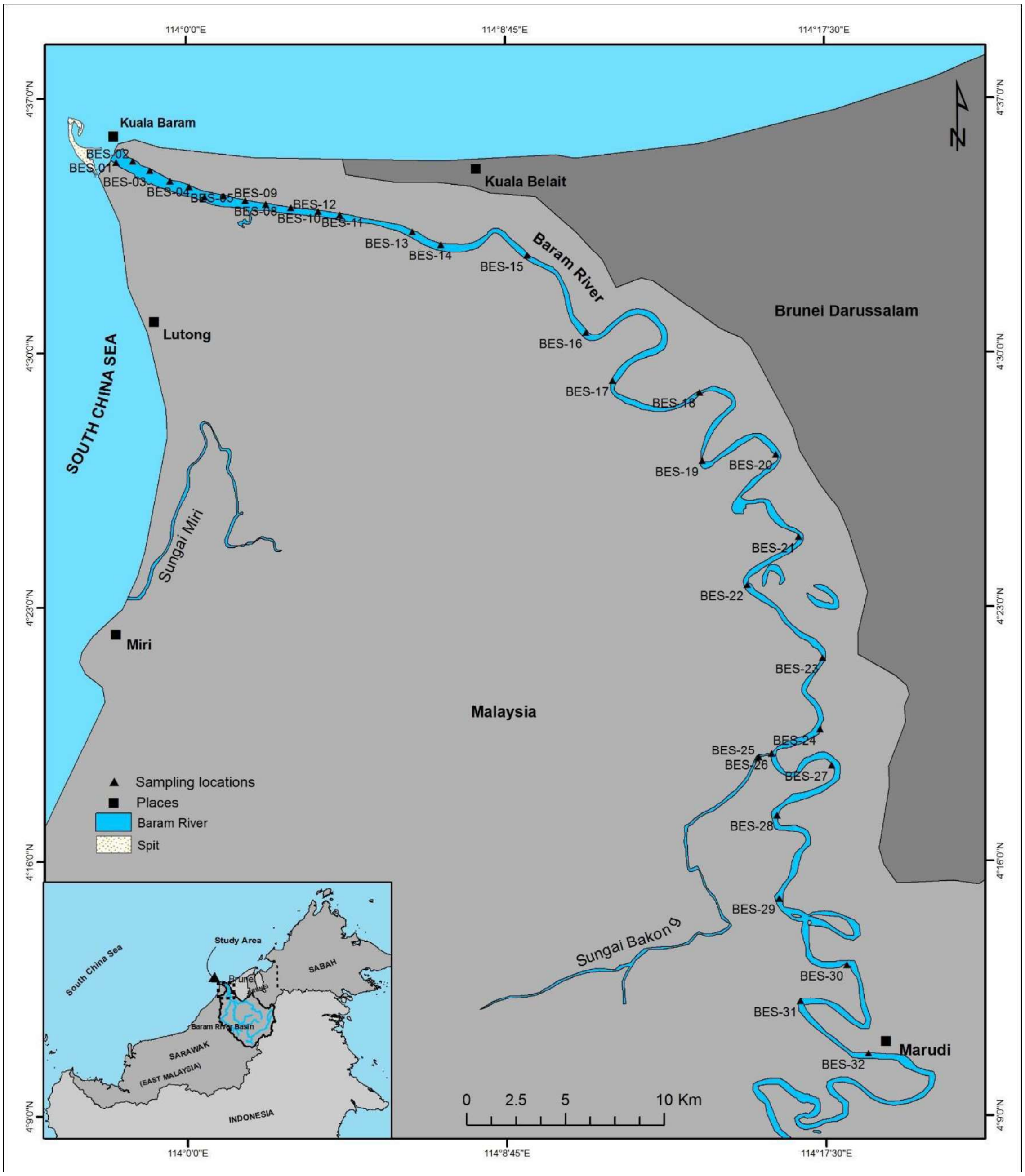

Figure 1 Study area map showing sampling locations (after Prabakaran et al. 2019)

Table
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Table 1 Descriptive statistics of the physicochemical parameters, nutrient, major and minor ions and trace elements levels of surface water during MON and POM seasons (n=32 for all parameters; turbidity (MON) – n=28

Parameters	Range (Mean with Std. Deviation) MON Season	Range (Mean with Std. Deviation) POM Season
Temp. °C	25.6-27.8 (26.7±0.6)	28.7-32.0 (30.13±0.72)
pH	4.3-6.1 (5.8±0.3)	4.6-7.5 (6.82±0.59)
DO (mg L^{-1})	1.3-5.8 (4.1±0.7)	0.4-5.6 (4.53±0.87)
EC (µS cm^{-1})	0.0-112.9 (43.7±22.0)	39.5-9060.0 (3178.16±3392.73)
TDS (mg L^{-1})	0.0-55.0 (21.31±10.7)	19.0-4440.0 (1557.22±1662.40)
Salinity (ppt)	BDL	0.0-5.1 (1.64±1.88)
Resistivity (Ω·cm)	0.01-100.0 (3.15±17.7)	0.0-0.03 (0.01±0.01)
Turbidity (NTU)	77.3-993.0 (627.76±209.7)	45.5-284.0 (114.77±53.63)
Redox (mV)	45.2-154.4 (63.6±19.0)	-22.2-138.0 (13.68±32.15)
PO_4^{-3}	BDL-1.7 (0.24±0.3)	0.2-0.9 (0.2±0.1)
SO_4^{2-}	BDL-5.0 (1.31±1.5)	BDL-410.0 (131.1±145.1)
(NH_3-N^-)	BDL	BDL-300.0 (9.4±53.0)
NO_3^- (µg L^{-1})	BDL-50.0 (2.50±9.5)	BDL-0.3 (0.04±0.05)
Cl- (mg L^{-1})	4.0-26.0 (12.25±6.0)	BDL-2940.0 (966.6±1096.1)
CO_3- (mg L^{-1})	BDL	BDL
HCO_3- (mg L^{-1})	18.3-42.7 (32.79±6. 9)	12.2-54.9 (27.7±9.2)
Na^+ (mg L^{-1})	2.3-14.2 (5.54±2.5)	2.6-1317.0 (377.0±437.6)
K^+ (mg L^{-1})	1.8-10.7 (4.67±2.2)	1.6-279.2 (61.5±77.3)
Ca^{2+} (mg L^{-1})	0.1-4.3 (1.87±1.1)	0.4-301.0 (34.4±60.853)
Mg^{2+} (mg L^{-1})	2.3-6.9 (4.00±0.1)	1.9-445.2 (77.3±101.5)
Cu (mg L^{-1})	0.04-0.2 (0.06±0.02)	0.04-0.1 (0.06±0.02)
Zn (mg L^{-1})	0.03-0.4 (0.09±0.08)	0.02-0.2 (0.04±0.02)
Pb (µg L^{-1})	BDL-78.0 (3.8±14.8)	BDL-0.04 (0.01±0.02)
Co (mg L^{-1})	0.02-0.1 (0.03±0.02)	0.02-0.06 (0.03±0.01)
Ni (mg L^{-1})	0.1-0.3 (0.2±0.03)	0.1-0.2 (0.2±0.01)
Cd (µg L^{-1})	BDL-5.0 (1.8±1.5)	BDL-7.00 (2.6±2.0)
Mn (mg L^{-1})	0.1-3.1 (0.4±0.5)	0.06-0.1 (0.08±0.01)
Fe (mg L^{-1})	3.4-43.3 (15.6±12.6)	2.3-10.8 (4. 9±1.7)
Al (mg L^{-1})	0.7-88.2 (17.1±24.1)	0.4-6.2 (2.3±1.0)
Cr (mg L^{-1})	0.2-0.4 (0.3±0.06)	0.1-0.3 (0.2±0.05)
Hg (µg L^{-1})	BDL-1.7 (0.3±0.3)	BDL-0.5 (0.2±0.1)
B (mg L^{-1})	BDL-2.2 (0.3±0.4)	BDL-1.6 (0.8±0.4)