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MINI-REVIEW

Role of Natural Flocculation in Eliminating Toxic Metals

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Abstract
Metals as the most common environmental pollutants derive from different sources and have far-reaching harmful impacts on flora, fauna and human health. Moreover, metals cause irreversible damages to marine ecosystems. Estuaries are most productive ecosystems for living creatures and act as a transporting corridor for exchanging materials from river to water bodies including oceans, seas and lakes. One of the most important processes in this region is flocculation. Not only does flocculation process convert a huge percentage of metals from dissolved phase to particulate phase in providing micronutrients to aquatic organisms, more importantly, but it also eliminates metals from aquatic ecosystems and gives aid to the pollution of water bodies to be on the decrease. Moreover, the chemical mass balance between river and sea is substantially influenced by flocculation process. Salinity, pH, dissolved oxygen, dissolved organic carbon and sodium hypochlorite as important factors affect the flocculation of metals during estuarine mixing of river water with seawater. It is vital to make use of natural processes in eliminating pollutants. Thus, natural processes need to be recognized and promoted by various means.

Metals

The accumulation of trace metals has enormously increased in the past decades (Ashraf et al. 2017; Ahmed et al. 2015; Bonanno and Raccuia 2018; Censi et al. 2006; Dhaliwal et al. 2018; de Mora et al. 2004; Hwang et al. 2018; Orani et al. 2018; Vaezi et al. 2016; Zheng et al. 2018). The essential and toxic elements are considered as two types of trace metals (Ahmed et al. 2015). Essential elements at high concentration along with toxic elements at low concentration create several problems for marine organisms and human health (Alamdar et al. 2017; El-Kady and Abdel-Wahhab 2018; Mudgal et al. 2010). Metals derived from different point and nonpoint sources are carried by rivers into oceans, seas and lakes (Fakhraee et al. 2015; Heidari et al. 2017; Heidari and Heidari 2015; Karbassi et al. 2007, 2008a, b, c, 2016a; Meybeck 1988; Vaezi et al. 2014).

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Estuary

Estuaries are well known for supporting biogeochemical cycling, maintenance of biological production, controlling nutrient modifications and movement of allochthonous nutrients (Farajnejad et al. 2017; Hassani et al. 2008; Karbassi et al. 2013; Karbassi and Heidari 2015; Meire et al. 2005; Samarghandi et al. 2007). Estuaries have high recovering capacity where terrigenous metals undergo various changes (Breuer et al. 1999; Fakhraee et al. 2015; Sharp et al. 1982). In other words, estuaries as a semi-enclosed coastal body of water affect the chemical forms of trace metals during mixing of river water with seawater (Chiffoleau et al. 1994; Karbassi et al. 2016a). A comprehensive investigation of the variability of the existing forms of microelements in estuary contributes to understanding their behavior in passing through the geochemical barrier and their impacts on estuarine ecology (Achterberg et al. 2003; Anikiev and Gorychev 1991; Biati and Karbassi 2010).

Flocculation Process

One of the most important processes that occur in the estuary is flocculation (Karbassi et al. 2016a). The conversion of trace metals from dissolved phase to particulate phase is defined as flocculation process (Boyle et al. 1977; Eckert and Sholkovitz 1976; Karbassi et al. 2015; Karbassi and Marefat 2017). Flocculation as a physicochemical process affects the chemical mass balance between rivers and seas or lakes (Farajnejad et al. 2017; Karbassi et al. 2013, 2014). Moreover, such a process is renowned for providing micronutrients to the aquatic organisms in conjunction with decreasing the pollution load of marine ecosystems (Biati and Karbassi 2012; Biati et al. 2010a, b; Saeedi et al. 2003). The results of some investigations have demonstrated that flocs are formed in the estuary (Biddle and Miles 1972; Eisma et al. 1983; Gibbs 1983; Gibbs and Konwar 1986; Karbassi et al. 2016b; Kranck 1975, 1979, 1984; Krone 1972; Owens et al. 1974; Schubel and Kana 1972; Sheldon 1968; Zabawa 1978). Different parameters such as surface area, density, settling velocity and deposition rates of sediment are influenced by flocculation process in the estuary (Droppo and Ongley 1989). Although influences of colloidal stability, surface characteristics, humic acids, salinity, DO, DOC, NaClO, electrical potential and pH have been studied, there are still many unknown mechanisms that influence elemental flocculation (Chenar et al. 2012; Featherstone and O'grady 1997; Fox and Wofsy 1983; Karbassi and Nadjafpour 1996; Zhiqing et al. 1987).

Effects of Metals on Fauna, Flora and Human Health

Metals refer to elements which have atomic density greater than 4 g/cm^3, or 5 times or more, higher in comparison with water, and their accumulation over time in the bodies of marine organisms causes irreparable damages (Duruibe et al. 2007; Hesami et al. 2018; Nagajyoti et al. 2010). Accumulation of metals in marine sediments adversely affects the quality of coastal ecosystems and aquatic organisms (Eggleton and Thomas 2004; Wang et al. 2018).

Copper (Cu)

Copper originating from petroleum refining and the manufacture of antifouling chemical products negatively affects bacteria, plants, fish and benthic invertebrates (Couet et al. 2018; Campana et al. 2012). Conducting investigation on the mechanisms of accumulation of copper in water body plays a vital role in the management of the marine ecosystem (Zhang et al. 2018).

Manganese (Mn)

The dissolved fraction of manganese as a naturally occurring element is strongly impacted by different factors such as pH, DO, reduction potential, organic matter, acidic water, land use and municipal wastewater discharges (Howe et al. 2004).

Nickel (Ni)

The toxicity of high concentration of nickel to aquatic organisms is influenced by concentration, water chemistry and organism physiology (Pyle and Couture 2012; Tamzin and Erin 2017).

Lead (Pb)

Lead as a cumulative toxic metal finds its way into water systems through runoff, industrial and sewage waste streams (Afshan et al. 2014). Increasing the accumulation of lead in the marine ecosystem can cause irreversible damages to some aquatic life, anemia, impairment of the function of the liver, spinal deformities, reduction in the bioturbation activity and death (Afshan et al. 2014; Blankson et al. 2017; Holcombe et al. 1976).

Zink (Zn)

In marine environments, the concentration of Zn as a ubiquitous and mobile metal is greatly influenced by water chemistry, depth and silica level (Biller and Bruland 2013; Raoult et al. 2018).

The effects of metals on human health, marine organisms and plant are summarized in Table 1.

Different Types of Estuaries

Estuaries as buffer zones between river and ocean environments provide sheltered habit, nursery and spawning areas for fish, crabs, prawns and shellfish. Hence, estuaries are among the highly productive ecosystems where organic and mineral nutrients from the land and sea accumulate (Currie and Small 2005; Dobson and Frid 2008; Karbassi et al. 2016b). On the other hand, estuaries can act as a geochemical reactor changing the chemical forms of continental-derived trace metals in the mixing zone of fresh and salt waters. According to different factors such as water balance, geomorphology, vertical structure of salinity and hydrodynamics, estuaries are classified into various types (Table 2).

The Removal of Metals Due to Flocculation Process During Estuarine Mixing of River Water with Seawater in Estuary

A remarkable percentage of dissolved metals come to particulate phase due to the estuarine mixing of freshwater from the river with saline water from ocean, seas or lakes

Table 1 Effects of meals on marine plant, marine organisms and human health

Metals	Harmful impacts		
	Human health	Marine organisms	Marine plants
Cu	Metabolic disorders, anemia, growth inhibition, blood circulation, etc.	Lipid peroxidation, harmful effects on enzymatic activity, restriction on potassium regulation and osmotic equilibrium, etc.	Accumulation in plant roots, reduction in biomass and seed production, decreasing plant height and root growth plant mortality, root malformation, etc.
Mn	Kidney disease, hypertension, confusion, muscle weakness, etc.	Accumulation in glands and blood, reduction in bacterial capacity, confusion, muscle weakness, making organisms vulnerable to various infections, etc.	Reduction in shoot length, root length, chlorophyll, carotenoid content, photosystem II activity, photosynthetic O_2 evolution activity, etc.
Pb	Kidney anemia, nerve and brain damages, decreasing appetite and physical fitness, constipation, fatigue, abdominal pains, etc.	Reduction in food and growth, spinal deformity, detrimental effect on gill function, etc.	Reduction in germination, growth, plant biomass, protein content, height, number of leaves, etc.
Ni	Lung nose and larynx cancers, respiratory failure, birth defects, heart disorders, dizziness, asthma, chronic bronchitis, etc.	Excess blood, swelling, hemorrhage, ataxia, iand loose stools, lung cancer, nasal sinusitis, etc.	Reduction in chlorophyll content, enzyme activity, plant nutrient acquisition, shoot yield, root growth, etc.
Zn	High concentration of Zn contributes to impairment of immune response and reduction in high-density lipoproteins	Reduction in function of olfactory receptor neurons, appetite, food intake, growth, etc.	Impairment of growth and reproduction

Bradl (2005), Calabrese (1998), Carbonell and Tarazona (1994), Eisler (2004), Frazier (1979), McIntyre et al. (2008), Pyle and Couture (2012), Schroeder et al. (1966), Tamzin and Erin (2017) and Wright and Welbourn (2002)

Table 2 Classification of estuary based on various factors

Factor	Description	Estuary	Description
Water balance	According to the balance between volume inputs (precipitation, river discharge as well as thawing) and outputs (evaporation and freezing)	Positive	Freshwater input > evaporation
		Inverse	Water losses > water inputs
		Low in flow	Evaporation is in the high level, and a low volume of freshwater finds its way into the estuary
Geomorphology	According to the geologic origin of the basin	Coastal plain	Forming due to incising sea level throughout Pleistocene era
		Fjord	Relating to high latitudes where glacial acts intensely
		Bar-built	Forming due to building up sandbars along the coastline
		Tectonic	Forming due to earthquakes and fractures of the crust of the Earth
Vertical salinity structure	According to the water column stratification	Salt wedge	Weak mixing of freshwater and saltwater
		Weakly stratified	Partial mixing of freshwater and saltwater
		Strongly stratified	Complete mixing of freshwater and saltwater
Hydrodynamics	According to the estuarine hydrodynamics non-dimensional parameters	Circulation parameter	Ratio of near-surface flow speed to sectionally averaged flow
		Stratification parameter	Ratio of the top-to-bottom salinity difference to the mean salinity over an estuarine cross section

Cameron and Pritchard (1963), Curray (1969), Dalrymple (1992), Hansen and Rattray (1966), Pritchard (1955) and Valle-levinson (2010)

(Karbassi et al. 2016a, b). Flocculation process is capable of eliminating toxic metals from marine ecosystems, on the one hand, and plays a significant role in self-purification of metals (Hassani et al. 2017). The occurrence of such a process at the estuary of the rivers enriches the standard of the biological conditions of marine ecosystems by converting metals from dissolved phase to micronutrients (Karbassi and Marefat 2017). Consequently, the chemical mass balance between river and ocean, sea or lake is affected by flocculation process (Fakhraee et al. 2015; Samani et al. 2014). The results of some investigations on flocculation of metals are presented in Table 3.

Pb was entirely removed from marine ecosystem because of flocculation process (Karbassi et al. 2014). Karbassi et al. (2016a) indicated that 88% of Cu comes to particulate phase during estuarine mixing. 97.31% of Ni, 97.21% of Zn and 74% of Mn were converted to micronutrients (Karbassi et al. 2016a, b; Samani et al. 2014). Throughout the mixture of river water with seawater, in field and laboratory, iron is eliminated from dissolved phase because of flocculation process (Droppo and Ongley 1989; Roux et al. 1998; Sholkovitz et al. 1978). The elimination of metals from marine ecosystems is influenced by a wide variety of factors including salinity, pH, DO, DOC, NaClO, etc. (Table 4) (Fakhraee et al. 2015; Karbassi et al. 2016a; Saeedi et al. 2003).

Salinity (S)

The maximum elimination of metals from marine ecosystem occurs in the upper part of the estuary where lower salinity regimes are found (Karbassi et al. 2016b; Saeedi et al. 2003). The vast majority of researchers have found that salinity is a major factor in controlling the colloidal metal flocculation process in estuarine mixing (Boyle et al. 1977; Eckert

Table 3 Flocculation rate of metals during estuarine mixing

A	Ni (25%) > Zn (18.59%) > Cu (16.67%) > Mn(5.83%) > Pb(4.86%)
B	Cu (88%) > Ni (86%) > Pb (84%) > Mn (74%)
C	Ni (97.31%) > Cu (41.10%) > Zn (48.94%) > Pb (44.7%) > Mn (49.65%)
D	Pb (22.6%) > Zn (7.6%) > Ni (3.62%) > Mn (3.6%) > Cu (2.8%).
E	Zn (97.21%) > Ni (93.02%) > Pb (83.15%) > Mn (63.60%) > Cu (41.23%)
F	Pb (100.00%) > Zn (78.26%) > Ni (62.50%) > Mn (37.50%) > Cu (15.15%)
G	Zn (59.3%) > Pb (47.6%) > Mn (37.5%) > Cu (29.2%) > Ni (27%).

A: Hassani et al. (2017), B: Karbassi et al. (2016a), C: Karbassi et al. (2016b), D: Fakhraee et al. (2015), E: Samani et al. (2014), F: Karbassi et al. (2014), G: Farajnejad et al. (2017)

Table 4 Effective factor in controlling the flocculation of metals

Metals	Effective factors				
	Salinity	pH	DO	DOC	NaClO
Cu		*			
Mn	*	*	*	*	*
Pb		*	*		
Ni	*				

[*] This symbol shows the effective factor in flocculation process of metals

Fakhraee et al. (2015), Karbassi et al. (2007, 2014, 2015), Karbassi and Heidari (2015) and Saeedi et al. (2003)

and Sholkovitz 1976; Meybeck 1988). Salinity showed a significant role in the flocculation process of Zn and Mn (Biati et al. 2010b). The highest flocculation of Zn and Mn occurred at the salinity of 2.9 ppt (Biati et al. 2010b). Furthermore, karbassi et al. (2014) showed that the removal efficiency of Mn and Ni is controlled by salinity (Fig. 1a).

pH

In the wetland, the flocculation is enhanced due to increasing different factors, especially pH (Karbassi et al. 2014). The flocculation rate of Pb, Cu and Mn was governed by pH, while pH did not play any role in flocculation of Ni (Fig. 1b, c) (Karbassi et al. 2014). The maximum flocculation occurred at pH = 7, 7.5 and 8 for Pb, at pH = 7.5 for Cu and at pH = 7.5 and 8 for Mn. The removal efficiency of Cu, Zn and Ni was substantially controlled by pH (Karbassi et al. 2007; Saeedi et al. 2003).

Dissolved Oxygen (DO)

Karbassi and Heidari (2015) demonstrated that dissolved oxygen has a significant role in the removal of metals. The maximum removal of Pb and Cd occurred at DO = 8.36 mg/L, and their removal was controlled by DO (Fig. 1d) (Karbassi and Heidari 2015).

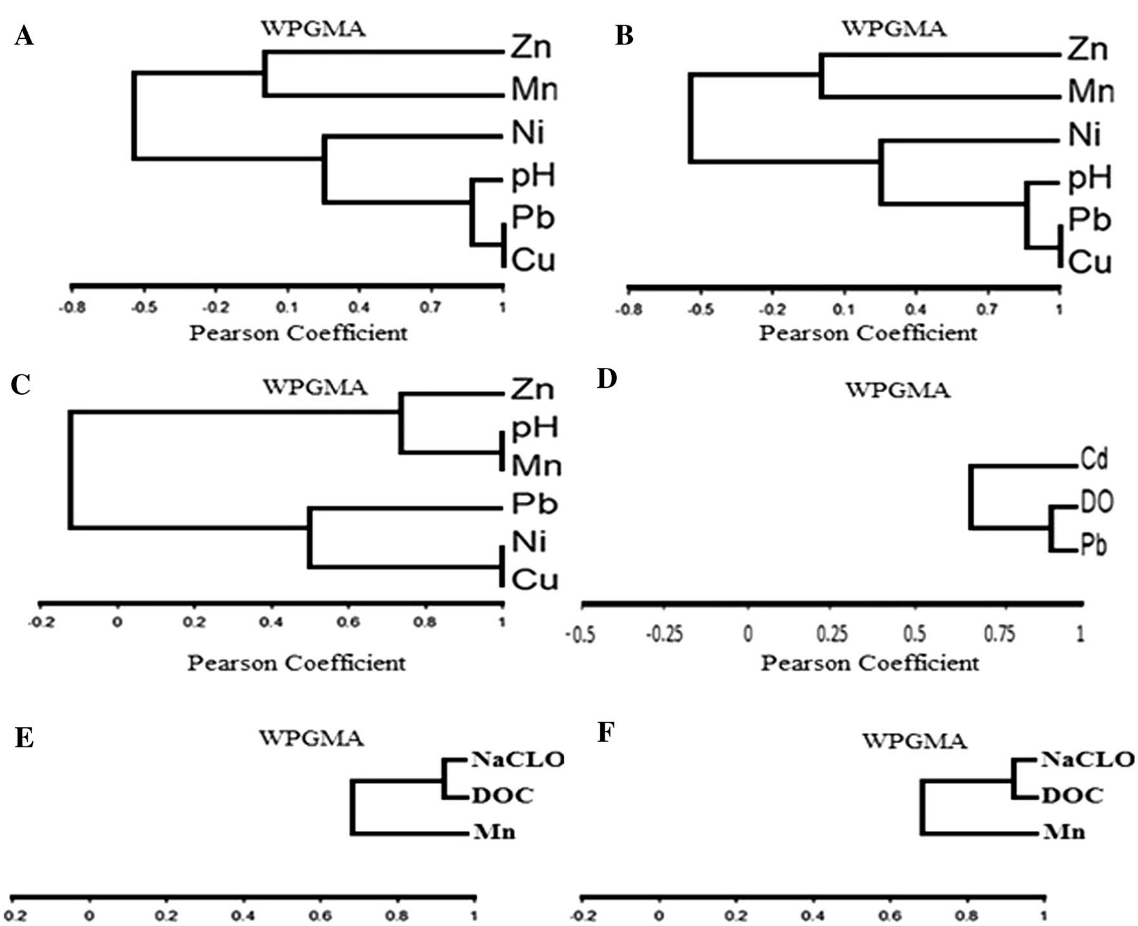

Fig. 1 Cluster analysis of metals in natural flocculation (Fakhraee et al. 2015; Karbassi et al. 2014; Karbassi and Heidari 2015)

Dissolved Organic Carbon (DOC)

In the estuary, source and behavior of DOC in estuary can be evaluated by spectral absorption and other optical techniques appropriately (Gardner et al. 2005; Guo et al. 2007; Laane 1981; Rochelle-Newall and Fisher 2002). Some studies have shown DOC as the main governing reason for the flocculation of metals (Mantoura and Woodward 1983; Meyer and Tate 1983; Sholkovitz et al. 1978). Maximum elimination occurred at DOC = 3.2 mg/L for Cu, Mn, Ni and Pb and at DOC = 7.32 mg/L for Zn (Samani et al. 2014). Fakhraee et al. (2015) indicated that the flocculation rate of Mn is governed by DOC (Fig. 1e).

Sodium Hypochlorite (NaClO)

During the mixing of freshwater with saline water, more detailed investigations on formation of NaClO are needed (Karbassi et al. 2008a, b, c). NaClO played an important role in governing flocculation process of Mn during estuarine mixing (Fig. 1f) (Karbassi et al. 2015). Moreover, Mn underwent maximum removal at NaClO = 50,000 ppm.

Conclusion

The accumulation of metals in marine sediments has harmful effects on marine ecosystems. Flocculation is considered as an important process during the mixing of freshwater from river with saline water from sea. This process provides micronutrients to aquatic environments along with enriching the conditions of marine ecosystems in an appropriate way. Micronutrients (trace metals) are vital for supporting and sustaining primary and secondary productions in aquatic ecosystems, on the one hand, and play a significant function in electron transport processes associated with photosynthesis, respiration, and nitrogen fixation, as well as the reduction of nitrate and nitrite to ammonium. Different factors in flocculation process give aids to the removal of metals. Such factors cause the flocculation rate of metals to be on the increase which plays a noteworthy role in reducing the pollution load of ocean, sea and lake. Various investigations have been conducted on eliminating metals from marine ecosystem during mixing of river water with seawater. Nevertheless, the individual effects of a wide variation of salinity, pH, dissolved oxygen (DO), dissolved organic carbon (DOC), sodium hypochlorite (NaClO), Na, Ca, Cl, Mg, SO4, HCO_3, CO_3, temperature (T) and electrical potential on flocculation of metals should be investigated. Future research should focus on the utilization of natural flocculation in treating various wastewaters. This task should preferably be carried out at very low salinity regimes (0.5–0.75 ppt) so that discharge of saline water would not leave an adverse effect on environment. Speciation of metals can have a significant role in overall performance of flocculation processes. Therefore, more emphasis should be paid to the role of Eh/pH in future investigations.

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