



Role of Salinity and Oxidation-Reduction Potential in Mobility of Heavy Metals in Suspended Sediments at Estuarine Zone

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ABSTRACT

Effect of salinity and oxidation-reduction potential (ORP) in the mobility of metals bound to suspended sediments at estuarine zone is investigated. Saline and freshwater samples as well as suspended sediments from estuarine zone of the Chalus River and the Caspian Sea, have been collected. Two series of four aquaria sets (natural and ORP-augmented conditions) containing turbid water with salinities of 0.25, 0.75, 1.5 and 2.5 psu were arranged. An increasing pattern of exchangeable-phase of all studied metals contents (at higher salinities) was observed under natural and ORP-modified conditions. Furthermore, the exchangeable-phase metal contents under ORP-modified conditions are higher (or equal) when compared with natural conditions. The overall trend of metals mobility potential might be evaluated as: Cd > Pb > Mn > Cu > Zn > Co > Ni. Findings of this research confirm the direct effect of both salinity and ORP parameters in mobility of metals bound to suspended particles in estuarine zones.

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INTRODUCTION

For their non-degradable nature, heavy metals have always been categorized as high-risk pollutants (Shariati et al., 2019). Non-degradability enables heavy metals to be transported far away from their source of generation (Nasrabadi et al., 2016). Surface water bodies are considered among the most vulnerable locations as they are directly exposed to surface run-offs (urban, industrial, and agricultural), direct and indirect wastewater discharges as well as wet/dry deposition from the atmosphere (Bastami et al., 2018; Talebi et al., 2019). Bed and suspended sediments are considered as reliable representatives for declaration of metallic pollution in a water body while they are not affected by instantaneous variations. On the other hand, their bilateral role as simultaneous sink and source of metals introduce suspended sediments as significant determinants for metallic fate and transport schemes (Esmailzadeh et al., 2017; Nasrabadi et al., 2018). Estuaries as intensified habitats for a wide range of flora and fauna including humans are considered as the most threatened surface water bodies (Ye et al., 2015). Flocculation of metals (conversion of metals from dissolved to colloid form due to saline and freshwater mixing) is a significant phenomenon which plays a key role in controlling metals behavior at estuarine zones (Sholkovitz, 1976). Besides flocculation, chemical partition of metals bound to sediments (exchangeable, carbonates, organics, sulfides, Fe-Mn oxides, silicates) also affects the final destiny of metals at estuaries (Nasrabadi et al., 2010; Mohseni-Bandpei et al.,

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2017). Some researchers have tried to develop standard methods for investigating the mobility of metals by proper natural (estuarine water) extracting agents (De Vallejuelo et al., 2014). There are various approaches towards the studies on mobility of metals during estuarine mixing. Some researchers opine that topography of water bodies might play significant role in the mobility of metals (Du Laing et al., 2009) while others know oxic–anoxic conditions as the main governing factor (Richard, 2012).

A variety of studies have focused on the effects of physico-chemical parameters like pH, salinity, dissolved oxygen (DO), electrical conductivity (EC) and ORP on flocculation rate of dissolved metals at estuaries during last decades (Fox and Wofsy, 1983; Hunter, 1983; Featherstone and O’Grady, 1997; Karbassi and Marefat, 2017). The gap in the current state of knowledge however is the final fate of the generated flocs. Accordingly, the main goals of this study is to evaluate the role of salinity and ORP levels of river waters at estuarine zones on concentration of metals bound to exchangeable fractions of suspended sediments and further to assess the mobility potential of suspended particle-bound metals (ratio of exchangeable-phase to total concentrations) under above-mentioned conditions.

MATERIALS AND METHODS

With an approximate length of 180 kilometers, Chalus River drains a catchment with a surface area of around 1550 km². It is a major river of central–northern Iran. It originates in the Central Alborz mountain range, flows northward, passes through the city of Chalus in Mazandaran Province and empties into its river mouth on the southern Caspian Sea (Fig.1).

In order to collect pure saline seawater and fresh river water, two distinct sampling points one in the Caspian Sea around 16 kilometers away from the river mouth and the other in the upstream of the river mouth (ca. 16 kilometers) were selected. Water samples were collected in acid-washed (a mixture of HCl and HNO₃) 20-liter polyethylene buckets.

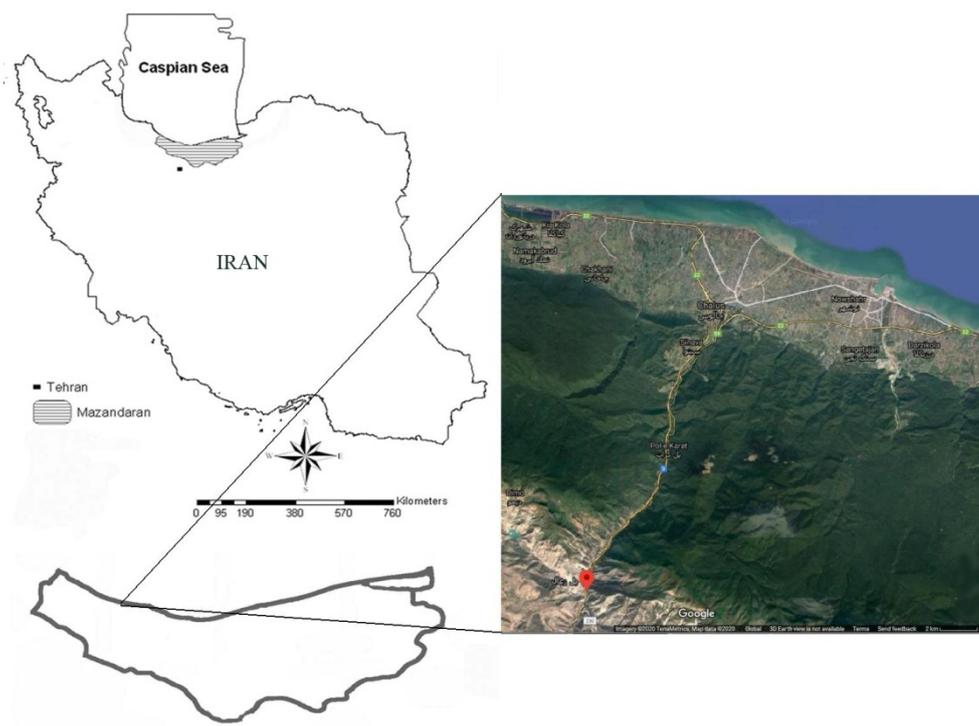


Fig. 1. Location of study area in Iran and Mazandaran Province

Table 1. Characteristics of different aquaria contents (saline and freshwater volumes)

Aquarium No.	Freshwater volume (ml)	Seawater volume (ml)	Salinity (psu)
1	500	0	0.25
2	500	30	0.75
3	500	80	1.50
4	500	190	2.50

Having been filtered (through 0.45 μm Millipore AP and HA filters) in the lab, fresh and saline water samples were mixed in aquaria (Sholkovitz, 1976) according to a scheme to create specific salinities (Table 1). Potential flocculation of dissolved metals occurs mostly in lower salinities (Fazelzadeh et al., 2012; Valikhani Samani et al., 2015; Karbassi and Marefat, 2017) and accordingly the range of salinities is selected in a way to cover the most efficient span.

The initial salinity of fresh and saline waters was 0.25 and 11.17 psu, respectively. To provide suspended sediments, turbid water samples from upstream of the river were collected, filtered through 63 μm filters and dried in the room temperature. Equal amounts of filtered suspended sediments were added to both aquaria series. A residence time of 24 hours was considered for aquaria to guarantee the maximal probable flocculation of dissolved metals. During the first hour of residence period the contents were stirred by a glass rod every fifteen minutes (Sholkovitz, 1976). To create ORP-modified conditions two aluminum electrodes were installed in the specified aquaria and a 3-amp electrical current with a voltage difference of 600 mV was implemented for a period of one hour. Water contents of all aquaria were then emptied and the sediments were air-dried. To detect total and exchangeable-phase concentration of metals a mixture of 8 mL of HNO_3 , 65 %; 5 mL of HCl, 37 %; 1.5 mL of HF, 40 % (Milestone, 2003) and acetic acid 25 % v/v at pH =5 (Chester and Hughes, 1967) were used, respectively. Metal contents were analyzed by inductively coupled plasma (ICP) with a mass spectrometer.

Random duplicates and blanks were analyzed in parallel with samples in the same way to assure the analytical data quality. The analytical errors were less than 5% for all elements. A portable multi-parameter device (BANTE 900) was used to measure ORP, pH, EC, salinity and dissolved oxygen (DO).

RESULTS

Values of physico-chemical parameters of water samples in both series of aquaria (natural and ORP-modified) are shown in Tables 1 and 2. As it is seen no distinguished difference is observed between the values of parameters ORP, EC, pH, salinity, temperature and DO at the start of the test (time = 0 hour) and after 24 hours in all four aquaria under natural conditions (Table 1). In ORP-modified aquaria however a total increase of ORP between 9.3 and 17.6% is seen after 24 hours of the residence time. In this period EC and salinity values are also increased up to 8.9 to 12.1% and 10.5 to 19.2%, respectively. Other parameters (pH, temperature and DO) indicate no tangible differences (Table 2).

Total concentration of metals in suspended sediments of river freshwater as well as exchangeable -phase concentrations of sediments in both series of aquaria are shown in Table 3. Furthermore, the overall trend of exchangeable-phase concentration of each metal at different salinities is also illustrated (Figs. 2 to 8). There are two distinguished characteristics regarding the overall trend of all eight metals: First of all an increasing pattern of exchangeable-phase concentration is seen from the lowest salinity value (0.25 psu) to the highest one (2.5 psu); the order of such increase (in proportion to initial values at the lowest salinity) for different metals may be classified as the following schemes: Mn (77.8%) > Cu (76.5%) > Ni (75%) > Pb

Table 1. Physico-chemical parameters of samples in aquaria under natural conditions

<i>Parameter</i>	<i>Time (hour)</i>	<i>Aquarium No. 1</i>	<i>Aquarium No. 2</i>	<i>Aquarium No. 3</i>	<i>Aquarium No. 4</i>
<i>ORP (mV)</i>	0	217	215	193	184
	24	216	195	190	175
<i>EC (mS)</i>	0	0.54	1.53	3.01	4.82
	24	0.56	1.59	3.03	4.93
<i>pH</i>	0	7.87	7.82	7.81	7.80
	24	7.73	7.68	7.68	7.61
<i>Salinity (psu)</i>	0	0.26	0.77	1.57	2.58
	24	0.27	0.80	1.58	2.64
<i>Temp (°C)</i>	0	23.7	23.7	23.9	23.7
	24	24.3	23.5	23.5	23.3
<i>DO (mg/l)</i>	0	6.07	6.21	6.15	6.11
	24	6.07	6.21	6.14	6.09

Table 2. Physico-chemical parameters of samples in aquaria under ORP-modified conditions

<i>Parameter</i>	<i>Time (min)</i>	<i>Aquarium No. 1</i>	<i>Aquarium No. 2</i>	<i>Aquarium No. 3</i>	<i>Aquarium No. 4</i>
<i>ORP (mV)</i>	0	217	215	193	184
	24	242	235	227	207
<i>EC (mS)</i>	0	0.54	1.53	3.01	4.82
	24	0.60	1.72	3.32	5.25
<i>pH</i>	0	7.87	7.82	7.81	7.80
	24	7.75	7.74	7.74	7.83
<i>Salinity (psu)</i>	0	0.26	0.77	1.57	2.58
	24	0.31	0.87	1.74	2.85
<i>Temp (°C)</i>	0	23.7	23.7	23.9	23.7
	24	23.8	23.7	23.9	23.7
<i>DO (mg/l)</i>	0	6.07	6.21	6.15	6.11
	24	6.05	6.17	5.99	6.01

(47.4%) > Co (40%) > Cd (33.3%) > Zn (30%) under natural conditions and Mn (78.7%) > Cu (77.3%) > Cd (66.7%) > Ni (50%) > Pb (42.9%) > Zn (31.3%) > Co (14.3%) under ORP-modified conditions. The specified pattern of exchangeable-phase concentrations in sediments under natural conditions is somehow in accordance with the flocculation pattern of dissolved metals in the same river water achieved from a former study (Karbassi and Heidari, 2015) as follows: Pb (24.32%) < Zn (24.38%) < Cd (40%) < Cu (64.71%) < Ni (68%) < Mn (76.47%). Secondly, the exchangeable-phase concentrations at each salinity value under ORP-modified conditions are higher (or equal) in comparison with pertinent concentrations under natural conditions. The order of such increase for mean concentration values (all four salinities) obeys the following order: Cu (41.9%) > Co (29.2%) > Mn (19.1%) > Pb (15.7%) > Cd (14.3%) > Zn (3.5%) > Ni (0%).

Mobility potential of metals in sediments (suspended and bed) of water bodies is a crucial characteristic in determination of environmental risk levels (Nasrabadi, 2015; Eghbal et al., 2019). Such potential directly relates to amount of metals bound to loose fractions of their carriers. Exchangeable-phase concentration (extraction at pH = 5) of metals is considered as an unbiased representative of mobility potential of metals in solid matrixes. Risk assessment code

Table 3. Concentration of metals (in mg/kg otherwise indicated) in suspended sediments (total and exchangeable-phase) of aquaria under natural and ORP-modified conditions (No. 1 to 4 correspond to salinities .25, .75, 1.5 and 2.5 psu)

Element	Total concentration	Exchangeable-phase concentration in Aquaria (natural conditions)				Exchangeable-phase concentration in Aquaria (ORP-modified conditions)			
		No. 1	No. 2	No. 3	No. 4	No. 1	No. 2	No. 3	No. 4
Zn	43.7	3	3.6	3.9	3.9	3.2	3.5	4	4.2
Pb	14.6	1.9	1.9	2.3	3	2.1	2.5	2.7	3.2
Ni	29.3	0.2	0.4	0.5	0.8	0.4	0.4	0.7	1.1
Mn	802	78	93	104	119	105	120	127	134
Cu	20.2	1.5	2	2.6	3	2	3.4	3.7	3.9
Co	11	0.4	0.5	0.5	0.6	0.7	0.8	0.8	0.8
Cd	0.16	0.03	0.03	0.04	0.04	0.03	0.04	0.04	0.05
Fe (%)	2.48	ND*	ND	ND	ND	ND	ND	ND	ND

*Not detected

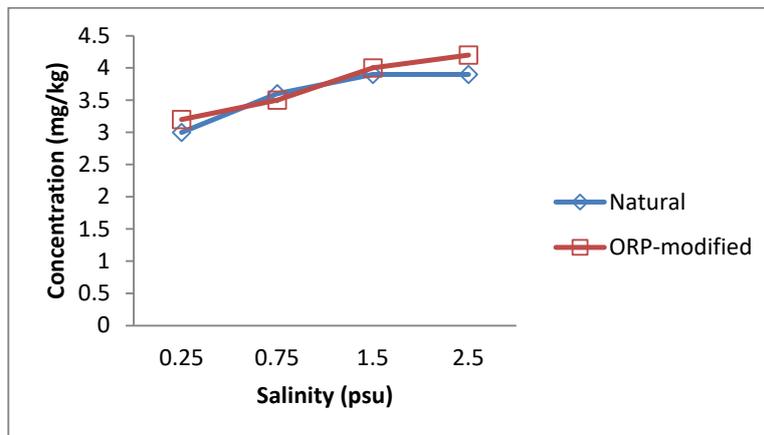


Fig. 2. Exchangeable-phase concentration of Zn in sediments at different salinities under natural and ORP-modified conditions

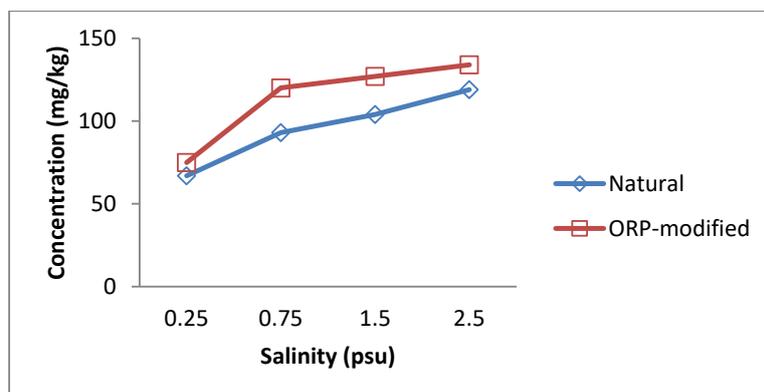


Fig. 3. Exchangeable-phase concentration of Mn in sediments at different salinities under natural and ORP-modified conditions

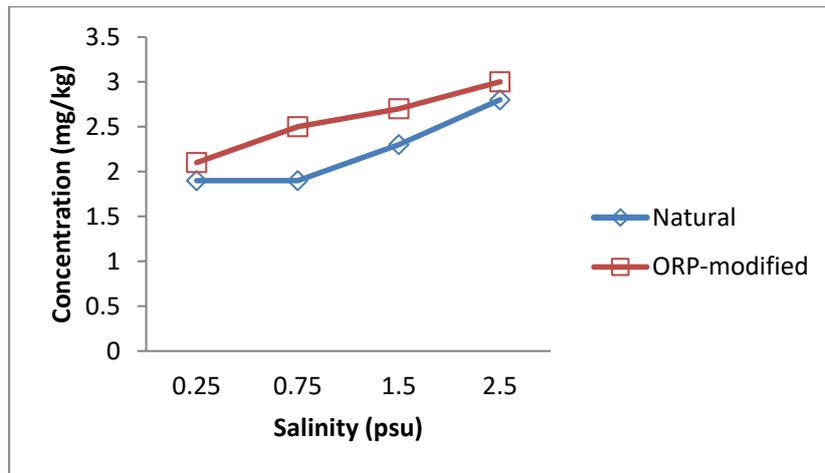


Fig. 4. Exchangeable-phase concentration of Pb in sediments at different salinities under natural and ORP-modified conditions

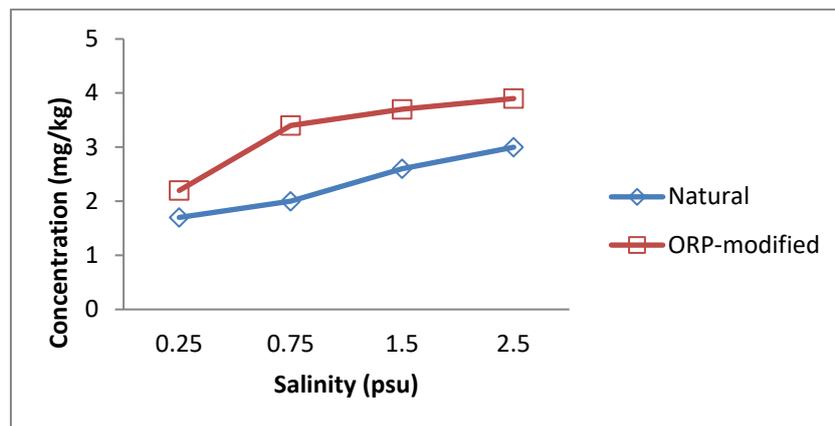


Fig. 5- Exchangeable-phase concentration of Cu in sediments at different salinities under natural and ORP-modified conditions

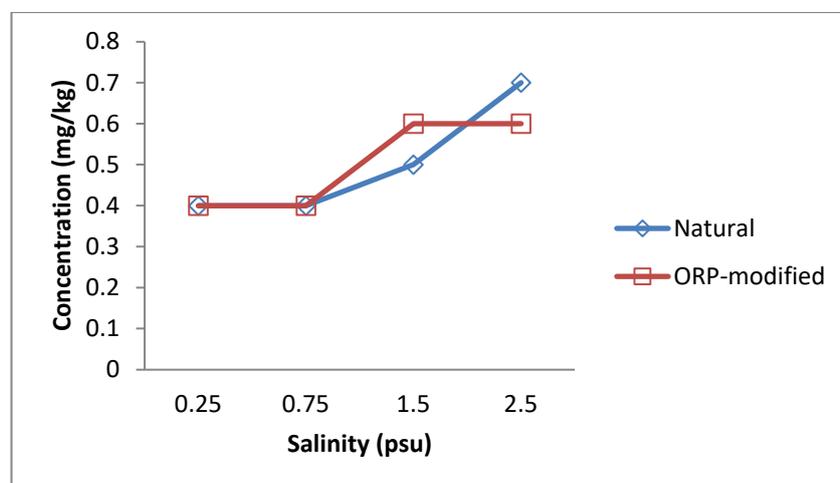


Fig. 6. Exchangeable-phase concentration of Ni in sediments at different salinities under natural and ORP-modified conditions

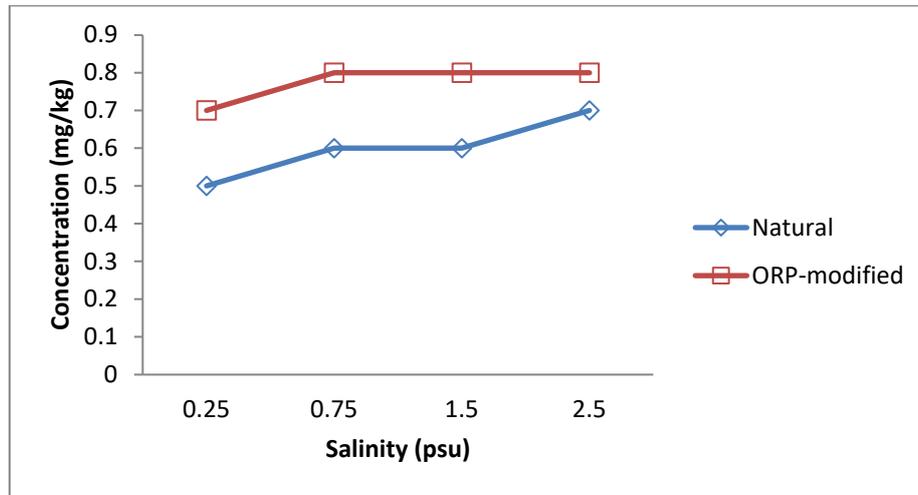


Fig. 7. Exchangeable-phase concentration of Co in sediments at different salinities under natural and ORP-modified conditions

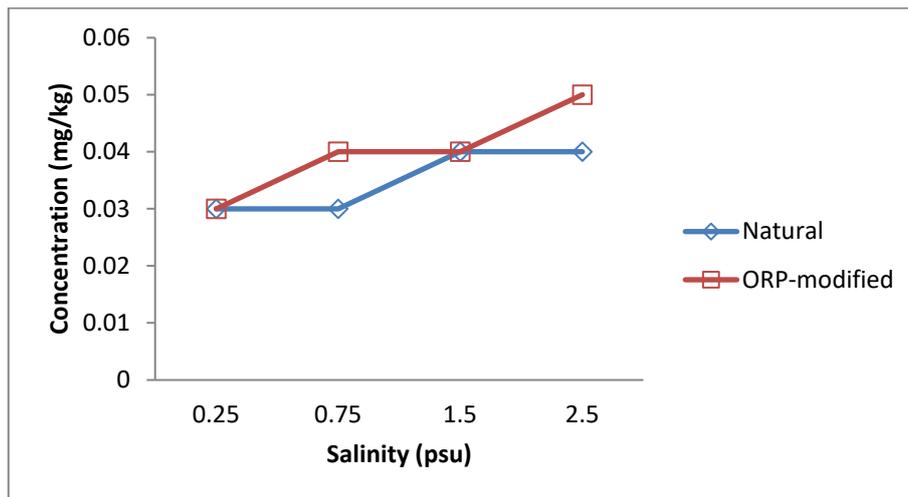


Fig. 8. Exchangeable-phase concentration of Cd in sediments at different salinities under natural and ORP-modified conditions

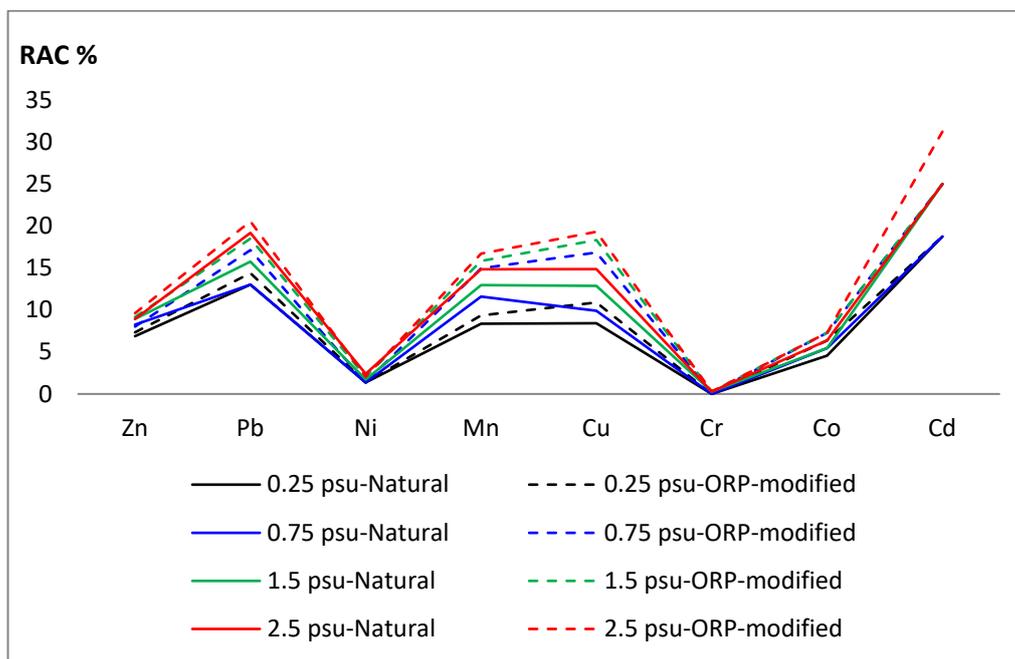
(RAC) defined as the ratio of exchangeable-phase to total concentration of each metal has been widely used to quantify the above-mentioned risk levels (Perin et al., 1985). Interpretation of different risk levels are described in Table 4.

Calculated values of RAC for each metal at different salinities and under both natural and ORP-modified conditions are shown in Fig. 9. Case-wise comparisons indicate that despite the slight increase of RAC values under ORP-modified conditions the overall values obey the following order under both conditions: Cd > Pb > Mn > Cu > Zn > Co > Ni. According to the RAC classification ladder (Table 4) metals Zn, Ni and Co are categorized in low risk level while Cd, Pb, Cu and Mn are considered to expose medium risk level to the surrounding environment.

Changes in ORP value mostly affect the fraction of total metal contents bound to sulfides and Fe–Mn oxides. It is reported by some researchers that metals bond with Fe–Mn oxides or even sulfides might be converted into the soluble form under some environmental changes (Li

Table 4. Risk assessment code (RAC) classification scheme (Perin et al., 1985)

Risk level	Percent of total metal concentration in loosely bound fractions
No risk	< 1
Low risk	1 - 10
Medium risk	11 - 30
High risk	31 - 50
Very high risk	> 50

**Fig. 9.** RAC values of suspended sediments in different aquaria (with different salinities) under natural and ORP-modified conditions

et al., 2000). For instance, it could be changes in pH and Eh of water body. The hydroxides of Fe and Mn on the surface of suspended particulates can be considered as major scavenger of heavy metals in aquatic environment. The degree of adsorption of metal on to the Fe–Mn however differs from one place to another (Badarudeen et al., 1996).

CONCLUSIONS

In this study the effect of salinity and ORP in mobility of metals bound to suspended sediments at estuarine environments has been taken into consideration. To this end saline and freshwater samples as well as suspended sediments from estuarine zone of Chalus River in southern Caspian Sea basin have been collected and aquaria analysis was run at lab scales. Higher exchangeable-phase concentration for all metals observed at higher salinity values. Higher generation rate of dissolved metal flocs at higher salinities (especially within the range of 0.2 to 3 psu) has been widely reported in the literature (Karbassi and Najafpour, 1996; Karbassi et al., 2008; Biati et al., 2010; Shamkhali Chenar et al., 2013). Such increase in flocs density ends

in higher concentrations in solid phase. Among different solid phase fractions (exchangeable, reducible, oxidizable and residual) exchangeable phase has the most affinity to bind the floccs. This would also justify higher mobility potential of metals in suspended sediments (higher RAC values) at higher salinities. An increase in ORP value (the case in this study) has terminated in partial release of sulfide and Fe–Mn oxides-bound metals from the solid matrix into the water phase (partial increase in EC and salinity values as indicated in Table 2). Not all of them however are released into water phase but some have been absorbed by particles through exchangeable bonds. This is the main reason of increased exchangeable-phase concentration of metals (Figs. 1 to 8) and consequently increased RAC values (Fig. 9) in sediments under ORP-modified conditions in comparison with natural conditions. Findings of this study confirm the direct effect of both salinity and ORP parameters in mobility of metals bound to suspended particles in estuarine zones. Future works however should be focused on details regarding the concentration of metals in different fractions (acid-soluble, carbonates, Fe-Mn oxides, Sulfides, organics and residuals) of suspended sediments at estuarine zones under ORP-modified conditions.

GRANT SUPPORT DETAILS

The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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