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Geochemical speciation and ecological risk assessment of selected metals in the surface sediments of the northern Persian Gulf



Mahmoud Reza Neyestani ^a, Kazem Darvish Bastami ^{b,*}, Marjan Esmaeilzadeh ^c, Farzaneh Shemirani ^d, Aida Khazaali ^e, Neda Molamohyeddin ^f, Majid Afkhami ^e, Shahram Nourbakhsh ^f, Mohsen Dehghani ^g, Sina Aghaei ^f, Mohammad Firouzbakht ^h

^a Department of Chemistry, Payame Noor University, P.O. Box 19395-3697, Tehran, Iran

^b Iranian National Institute for Oceanography and Atmospheric Science (INIOAS), No. 3, Etemadzadeh St., Fatemi Ave., 1411813389 Tehran, Iran

^c Department of Environmental Science, Faculty of Environment and Energy, Science and Research Branch, Islamic Azad University, Tehran, Iran

^d Department of Analytical Chemistry, University College of Science, University of Tehran, P.O. Box 14155-6455, Tehran, Iran

^e Young Researchers and Elites Club, Bandar Abbas Branch, Islamic Azad University, Bandar Abbas, Iran

^f Department of Marine Science and Technology, Science and Research Branch, Islamic Azad University, Tehran, Iran

^g Department of Environment, Bandar Abbas Branch, Islamic Azad University, Bandar Abbas, Iran

^h Novin Shimyar Chemical Laboratory, P.O. Box 14589-3699, Tehran, Iran

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ABSTRACT

The present study aimed to geochemical speciation of metals in the surface sediments of the northern Persian Gulf. Metal contents in the sediment were observed in the order: Al > Fe > Cr > Ni > V > Zn > Cu > Co > As > Pb > Cd. The results of sequential extraction procedure revealed that all metals were predominantly associated with the residual fraction. Among the metals, Cu and As exhibited higher bioavailability. The risk assessment code (RAC) indicated that Cu, As and Cd had medium environmental risk at some sampling sites. Based on enrichment factor (EF), Cd and As had moderate to significant enrichment.

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Heavy metals are ubiquitous environmental contaminants derived mainly both natural (physical and chemical weathering of parent rocks) and anthropogenic sources (Callender, 2005; Bastami et al., 2015). Complex processes including anthropogenic activities or natural processes including riverine or atmospheric inputs, floor erosion, biological activities, water drainage and discharge of urban and industrial wastewater are important parameters controlling the distribution of metals within the aquatic environments (Xia et al., 2011; Wei and Yang, 2010).

A minor fraction of metals remain dissolved in water whereas large quantities of free metal ions are stored in sediments. Therefore, sediments in aquatic environments can either retain metals or release them to the water column by various remobilization (biological and chemical) processes. Sediment parameters (mineralogy, texture), metal characteristics, pH, organic matter and oxidation–reduction potential are important parameters controlling the accumulation and the availability of heavy metals in the sediment (Buccolieri et al., 2006; ElNemr et al., 2007; Bastami et al., 2012, 2015).

Heavy metals in sediments are mainly associated with silicates and primary minerals and therefore have limited mobility. However, the metals introduced by human activities show greater mobility and are associated with other sediment phases, such as, carbonates, oxides, hydroxides and sulfides (Passos et al., 2010; Heltai et al., 2005).

Total heavy metal concentrations are often used to assess heavy metal pollution in sediments, but the total concentration provides inadequate information about the mobility and bioavailability of the heavy metals (Nemati et al., 2011).

Investigations on chemical partitioning of metals in sediment are important to distinguish the bioavailable fraction. Among the metal fractions, exchangeable and carbonate bound fractions are considered to be bioavailable, and it determines the bioaccumulation and biomagnification in aquatic food web (Tessier and Campbell, 1987; Sundaray et al., 2011; Dhanakumar et al., 2015).

The Persian Gulf is a semi-enclosed and shallow basin which is connected to Gulf of Oman through Strait of Hormuz. The circulation in the Persian Gulf is driven by wind-stress, surface buoyancy fluxes, fresh water runoff, water exchange through the Strait of Hormuz and tides. The Persian Gulf area has about two-thirds of the world's oil sources therefore problems associated with oil pollution appear to be of greater

* Corresponding author.

E-mail addresses: darvish_60@yahoo.com, darvish.bastami@inio.ac.ir (K.D. Bastami).

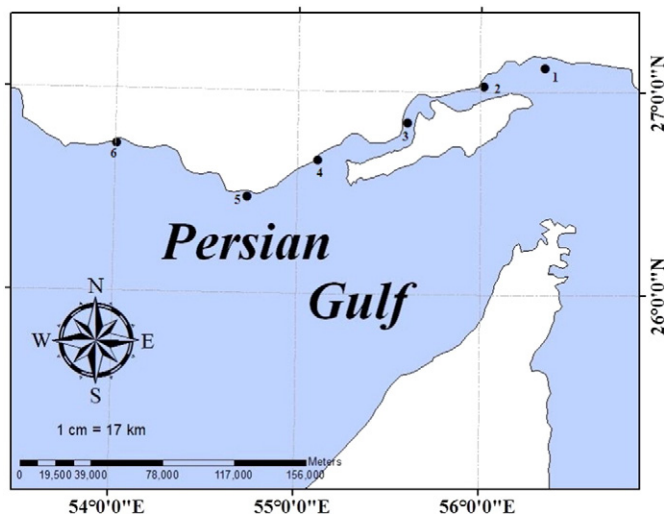


Fig 1. The locations of the sampling sites at the northern Persian Gulf.

significance in the Persian Gulf compared with other regions. Consequently, a combination of tanker traffic, accidental spills, Urbanization, industrialization, port areas and refineries are major sources of pollution in the marine environment.

The objectives of the present work were (i) to assess the dynamics and mobility of heavy metals in different geochemical fractions in the sediments and (ii) to identify the bioavailability and ecological risk of these metals in the surface sediment of the northern Persian Gulf.

Surface sediment samples from six different sites in the northern parts of Persian Gulf were collected using Van-Veen grab for heavy metals analysis during autumn (November) 2015 (Fig. 1). Then, samples were packed and carried to the laboratory in iced-boxes and stored at -4°C until analysis. After drying in an oven, sediment samples were ground by using a hand mortar followed by screening with a 0.5 mm sieve to remove large particles.

For chemical partitioning of metals, sediment samples were analyzed using Tessier sequential chemical extraction procedure (Tessier et al., 1979). The sequential extraction procedure was divided into five operationally defined chemical fractions: (F1) the exchangeable fraction: readily soluble and exchangeable; (F2) the carbonate bound and specifically adsorbed fraction: carbonate-bound, specifically adsorbed and weak organic and inorganic complexes; (F3) the Fe–Mn oxides fraction: bound to iron and manganese oxides (Fe–Mn oxides); (F4) the organic/sulphide fraction: bound to stable organic and/or sulphide (organic) complexes; and (F5) the residual fraction: held in primary and secondary minerals within their crystal structure. The detailed geochemical fractionation procedure of sediment is presented in Table 1a. After each successive extraction, samples were centrifuged at 5000 rpm for 30 min to separate the extract from sediments. For total heavy metal contents, the sediments were digested using a mixed

solution of HF–HCl–HNO₃–HClO₄ according to the ASTM standard practice D5258–92 (ASTM, 2013).

Concentration of Al, Fe, Cd, As, Cu, Ni, Pb, Co, Cr, V and Zn were determined by using Inductively coupled plasma-optical emission spectrometry (ICP-OES; Varian VISTA-MPX). The precision and accuracy of the methods were systematically and routinely checked using standard reference materials. Accepted recoveries range from 94% to 110%.

The recovery rates of the metals, obtained by sequential extraction procedure, were judged by comparing the sum of the five fractions with the total metal concentration (Table 1b).

Also, major element contents (SiO₂, CaO, Fe₂O₃, Al₂O₃, MgO, K₂O, Na₂O, TiO₂, P₂O₅ and MnO₂) were measured by using X-ray fluorescence spectrometer (Bruker Model). For determination of total organic matter, sediment samples were dried at 70 °C for 24 h and then combusted in the electrical furnace at 550 °C for 4 h. Total organic matter, as described by Abrantes et al. (1999), was measured by the following equation:

$$\text{Total organic matter (TOM, \%)} = [(B-C)/B] \times 100 \quad (1)$$

where B and C are the weights of dried sediment before and after combusting in the oven, respectively.

Grain size analysis was performed by air drying and then passing samples through a 63 μm mesh (equivalent to a No. 230 sieve, ASTM E-11).

Enrichment factor is used to determine sedimentary metals source produced by anthropogenic events or natural origin, normalizes metals concentrations according to the sediment texture properties (Selvaraj et al., 2004; Vald'es et al., 2005; Bastami et al., 2014). In this study, Fe has been as a conservative tracer to differentiate natural from anthropogenic components. The formula to calculate the enrichment factor (EF) is:

$$\text{Enrichment Factor} = (H_s/Fe_s)/(H_c/Fe_c) \quad (2)$$

Where H_s and H_c are heavy metal concentrations in sample and background reference, respectively. Fe_s and Fe_c are the iron contents in sample and background reference, respectively. In this study, we used background concentrations of metals in sediment from Iranian waters of the Caspian Sea which are 5.0.16, 150, 10, 26, 64, 53.64, 20, 102.73 ppm and 2.4% for As, Cd, Ni, Pb, Cu, Zn, V, Co, Cr and Fe, respectively (Agah et al., 2012).

To assess the sediment environmental quality, an integrated pollution load index of eight metals was calculated as suggested by Suresh et al. (2011).

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n} \quad (3)$$

where CF metals is the ratio between the content of each metal to the background values,

$$CF_{\text{metals}} = CH_{\text{metal}}/CH_{\text{back}} \quad (4)$$

Table 1

(a) Sequential extraction scheme of Tessier et al. (1979). (b) Recovery rates of metals obtained from sequential extraction.

Step	Fraction	Extraction reagents/conditions							
(a)									
1	Exchangeable – (F1)	8 ml MgCl ₂ (1 M), pH 7, 1 h, room temperature, continuous agitation							
2	Acid-soluble (carbonates) – (F2)	8 ml NaOAc (1 M), pH 5, 5 h, room temperature, continuous agitation							
3	Reducible (Fe–Mn oxide bound) – (F3)	20 ml of 0.04 M NH ₂ OH·HCl in 25% (V/V) HOAc, 6 h, 96 ± 3°C, occasional agitation							
4	Oxidizable (organically bound + sulphide bound) – (F4)	3 ml of 0.02 M HNO ₃ + 5 ml of 30% H ₂ O ₂ , pH 2, 2 h, 85 ± 2°C, occasional agitation. Add 3 ml of 30% H ₂ O ₂ , repeat 3 h, cool and then add 5 ml of 3.2 M NH ₄ OAc in 20% (v/v) HNO ₃ , 0.5 h, room temperature, continuous agitation							
5	Residual – (F5)	HF:HNO ₃ :HClO ₄ = 7:3:1, dryness, again HF:HNO ₃ :HClO ₄ , 1 h, 2 ml conc. HCl, dryness, 10 ml 50% HNO ₃ , make up to 50 ml with distilled water							
(b) Recovery (%)									
Elements	As	Cd	Co	Cr	Cu	Ni	Pb	V	Zn
	83.45–107.13	88.31–110.40	76.59–90.86	89.22–96.60	82.71–93.88	91.45–100.33	78.24–92.68	90.09–97.21	89.90–100.59

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