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Geo-accumulation Index and Enrichment Factor of Mercury (Hg) and Methyl mercury (MeHg) in Sediments from some Estuaries, Persian Gulf: Pollution Load Index (PLI)

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Environment geochemistry of mercury and methyl mercury in contaminated sediments from five estuaries along the Imam Khomeini port, northwest part of the Persian Gulf, were studied. The samples were divided into particular grain size fractions and then the content of Hg and MeHg was determined. Environmental indicators such as geoaccumulation index (Igeo) and enrichment factor (EF) for Hg and MeHg in all estuaries were analyzed. According to study results, mean concentrations of Hg in all estuaries were 0.74 for Hendijan, 0.54 Ghanam, 0.37 Meleh, 1.35 Musa and 0.65 µg/g for Zangi, respectively. The mean concentrations of MeHg were 0.61 for Hendijan, 0.40 Ghanam, 0.29 Meleh, 1.01 Musa and 0.47 µg/g for Zangi, respectively. Measuring of geoaccumulation Index show that Meleh and Ghanam estuaries are moderately polluted, Zangi estuary is moderately to strongly polluted and Musa and Hendijan estuaries are strongly to extremely polluted. Enrichment Factor (EF) show metals trace and sources in Ghanam and Meleh estuaries is related to crustal materials or area background, but in Hendijan, Musa and Zangi estuaries is related to anthropogenic impact such as petroleum and petrochemical industries. Finally, the metals accumulated in the estuary sediment as a result of changing chemical conditions such as pH, oxygen content, redox potential, organic carbon and carbonate.

Keywords: Geoaccumulation Index, Enrichment Factor, Mercury, Methylmercury, Persian Gulf

INTRODUCTION

Heavy metals introduced in the aquatic ecosystem are mostly concentrated in estuaries areas, near densely populated and industrialized regions (Vicente-Martorell et al. 2009). These particles are often very small, and can therefore stay in solution for a very long time. In the last decade, there has been an increasing ecological and global public health concern associated with environmental pollution by these metals (Hosseini et al. 2013). Also, human exposure has risen dramatically as a result of an exponential increase of their use in several industrial, specific oil and petrochemical industries and agricultural (Hou et al. 2013). Generally, sources of metals in the environment include industrial, agricultural, pharmaceutical, geogenic, domestic effluents, and atmospheric sources (Ip et al. 2004; Hatje et al. 2008). In the aquatic ecosystem, oil and petrochemical industries are the sources important for metals and enter to environment. After the entering of metals to aquatic environment such as estuary and coast, they can receive by sediments, plant and animals (Kucuksezgin et al. 2008). Therefore, sediments are main source for accumulation of pollutants and they have important role in transport of pollutants to animal and plant (Guo et al. 2011; Hosseini and Sajjadi 2018).

Estuaries, which are regions of active land-ocean interaction, respond sensitively to natural processes and anthropogenic activities (Li et al. 2013). Estuarine sediments are recognized as an important sink for heavy metals and other contaminants (Ip et al. 2004). Incorporation of metals from the weathering of rocks within the catchment of the estuary; materials carried in by the tide; and anthropogenic sources into sedimentary landforms, such as salt marshes or mudflats, at any point within an estuary is further influenced by the interplay of water chemistry (dissolved oxygen, salinity and pH) and hydrological regime (Ranjan et al. 2008; Vilhena et al. 2010). Diagenetic processes, determined by pH, organic matter content, redox conditions, and soil texture, may further influence heavy metal concentrations and distributions once incorporation in a sedimentary landform has occurred (Hatje et al. 2008; Zhao et al. 2017). Nonetheless, high percentages of organic matter or small grains in sediment are generally associated with reduced heavy metal bioavailability and toxicity (Hosseini and Sajjadi 2018).

Heavy metal contamination especially Hg and MeHg in sediments can affect water quality and thus the bioassimilation and bioaccumulation of metals in aquatic organisms, resulting in long-term implications for human and ecosystem health (Ip et al. 2007; Mooraki et al. 2009; Raeisi Sarasiab et al. 2014). The first metal will be accumulating in sediment, followed by algae, zooplanktons, small fish and larger fish. Therefore, sediments are a source and sink for enter of toxic metal in food chain (Hosseini et al. 2013). Therefore, sediments are a source and sink for enter of toxic metal in food chain (Celino et al. 2008; Abdolahpur Monikh et al. 2012; Hosseini and Sajjadi 2018).

Studies on heavy metal pollution along the Persian Gulf have been carried out, but limited data are available on Hg and MeHg pollution in the muddy and sandy sediment of estuaries in the Khuzestan coasts. Therefore, this study, provide new information on the Hg and MeHg pollution in muddy and sandy sediment collected five estuaries in the Khuzestan province along the Persian Gulf in the southwestern of Iran. Also, we studied geo-accumulation index and enrichment factor, the correlation between grain size, organic carbon, nitrogen, carbonate, pH and metal concentration in sediment.

MATERIALS AND METHODS

Sediment samples obtained from 5 estuaries including, Hendijan, Ghanam, Musa, Meleh and Zangi estuaries along the Imam Khomeini port, located in Khuzestan Shore, northwest part of the Persian Gulf (Fig. 1). Surface sediments were collected in July of 2019 by a Van Veen Grab. Subsamples were taken from the uppermost layer of the sediment taking care to minimize contamination. Surface sediments (0–5 cm) were sectioned and stored in pre-combusted glass jars in a freezer (−20°C) until analysis (Carine et al. 2011). Before analysis, sediments were freeze-dried and ground to achieve homogeneity. For determining the relationship between grain size and heavy metal contents, the grain size of surface sediments was measured using a Beckman-Coulter laser particle size analyzer (Model LS 13 320). Briefly, 20 mL deionized water was added to 1 g of freeze-dried sediment in a beaker. After soaking for 24 h, the sediment was subjected to vortex mixing for 5 min to disaggregate loosely-attached aggregates. Neither organic matter nor carbonate was removed for the laser grain size analysis (Bellucci et al. 2003; Anirudh et al. 2009). In this study, the size range of detection for this analyzer is from 0.01 to 1000 μm. The all samples were divided two sample such as sandy (>63 μm) and muddy (5~63 μm) sediment (Hosseini and Sajjadi 2018).

Chemical analysis

All glassware and ceramic ware used in sample processing are combusted at 400□C for at least 4 hours. Samples remain frozen at –20□C until processing. Sediment samples are thawed and homogenized. The sample is dried in an oven at 40□C. A portion of sample is removed, ground and homogenized. An aliquot of dried, homogenized sample is placed in an aluminum-weighing pan and dried at 105□C. The LECO CR-412 Carbon Analyzer is calibrated prior to the analysis of samples. Different amounts of high purity calcium carbonate standard (99.95% purity, carbon content of 12.0%) are used to calibrate the instrument (Hosseini and Sajjadi 2018). The approximate amounts of calcium carbonate used for the six-point calibration are; 0.01 g, 0.05 g, 0.10 g, 0.25 g and 0.50 g. An empty carbon-free combustion boat is analyzed as a blank for the calibration curve. The calibration curve provides an analysis range of approximately 0.0 to 0.06 g total carbon. Each calibration standard must fall within 3% of the known percent carbon value to meet acceptance criteria (Joseph 2001). A continuing calibration check standard (mid-level standard) is analyzed every ten samples and must be within 5% of the known value of the standard. Total

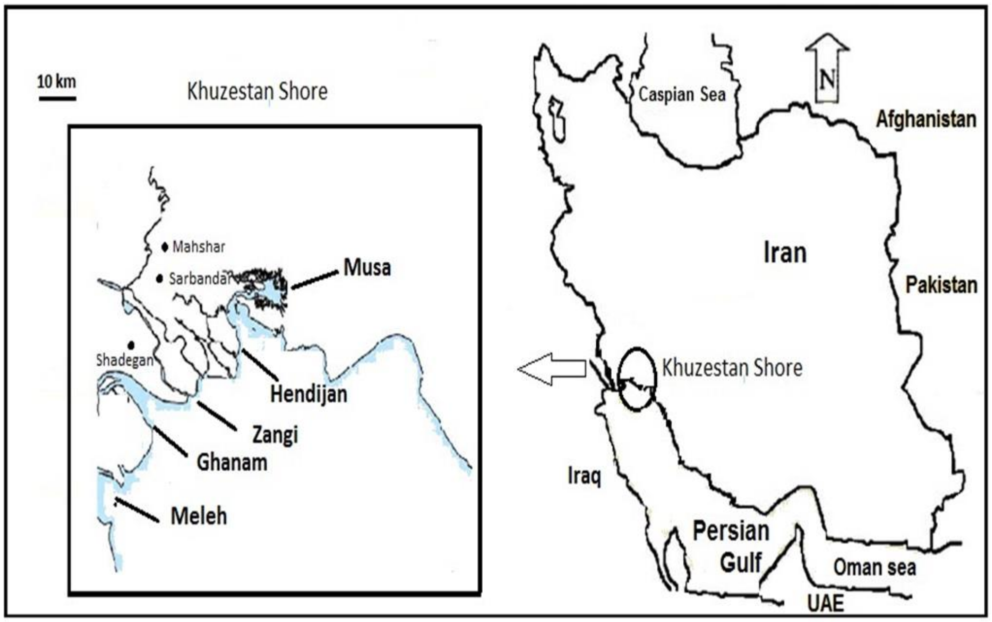

Figure 1: Map of Persian Gulf and study area

organic carbon is analyzed by placing approximately 0.350 g of dried, ground and homogenized sample into a clean, carbon-free combustion boat. Each sample boat is treated with phosphoric acid drop by drop until the sample stops "bubbling" and the sample is completely moist with acid. The sample is placed into an oven set at 40C for 24 hours and then transferred to an oven set at 105C. Once the sample is dry, the boat is placed on the autosampler rack assembly and loaded onto the LECO carbon analyzer (Du Laing et al. 2009). Neither organic matter nor carbonate was removed for the laser grain size analysis. The size range of detection for this analyzer is from 0.01 to 1000 µm. The environmental parameters of sediments collected from all station shown in Table 1. For each sample a known quantity (1 g) of sediment was digested with a solution of concentrated $HClO_4$ (2 ml) and HF (10 ml) to near dryness. Subsequently, a second addition of $HClO_4$ (1 ml) and HF (10 ml) was made and the mixture was evaporated to near dryness. Finally, $HClO_4$ (1 ml) alone was added and the sample was evaporated until white fumes appeared. The residue was dissolved in concentrated HCl and diluted to 25 ml (Abdolahpur Monikh et al. 2012). Recovery varied between 97.8% and 103%. Heavy metal concentrations were determined by a cold vapor Atomic Absorption Spectrometer Leco AMA-254. The accuracy of the analytical procedures was assessed using the certified reference material BCR-1 and yielded results within the reference value range (El Nemr et al. 2007).
The level of pollution in surface sediments can be assessed by the determination of indices such as geo-accumulation index (Igeo). Another approach to assess the contamination level of sediments is the Igeo defined by Muller (1979) according the following formula: $Igeo = \log_2 (C_n/1.5 \times B_n)$
In this formula, Cn = concentration of the examined metal in the sediment sample; Bn = background of a given metal in the geochemical reference. The geoaccumulation index distinguishes 6 classes of quality for sediments that showed in Table 1.
The pollution load index (PLI) is an index for metal pollution assessment, also evaluated to assess the mutual contamination effect of the measured all metals in this study (Adel et al. 2011). This index is expressed as the following equation: $PLI = (CF1 \times CF2 \times CF3 \times \ldots\ldots\ldots\ldots CFn)^{1/n}$
In this formula, n = number of metals; CF = contamination factor (Metal concentration in sediment/background values of the metal) (Table 1).
Enrichment Factor (EF) analyses indicates the sources of metals in sampling area are related to natural and weathering crust or that anthropogenic contribution. In this study, EF was used as a normalizer according to the equation 1:

EF = [Cn/CSc] sample / [Cn/CSc] (shale)

According to Guilherme (2011), if 0.5 <EF < 1.5, the elemental concentration is probably entirely due to crustal or natural weathering origin; values above 1.5 indicate anthropogenic contribution and the higher the EF value the more severe is the anthropogenic contribution.
All data were tested for normal distribution with Shapiro-wilk normality test. One-Way analysis of variance

Table 1 The degree of metal pollution based on different classes of three index pollution

Index	Value	Class designation of sediment quality
EF	0.5 <EF < 1.5	Metal concentration related to earth's crust
	> 1.5	Metal concentration related to anthropogenic contribution
Igeo	0 <	Unpolluted
	0-1	Unpolluted to moderately polluted
	1-2	Moderately polluted
	2-3	Moderately to strongly polluted
	3-4	Strongly polluted
	4-5	Strongly to extremely polluted
	> 5	Extremely polluted
PLI	< 1	No pollution
	> 1	Pollution

Table 2 Organic Carbon (OC), Carbonate ($CaCo_3$) and pH in muddy and sandy sediment

Sediment	Parameter	Station				
		Hendijan	Ghanam	Meleh	Musa	Zangi
Muddy	OC	0.25	0.22	0.14	0.54	0.34
Sandy		0.17	0.20	0.12	0.32	0.31
Muddy	$CaCo_3$	0.55	0.22	0.29	0.74	0.45
Sandy		0.47	0.21	0.23	0.58	0.48
Muddy	Salinity	29	27	30	33	26
Sandy	Salinity	33	33	32	37	32

Table 3 Hg and MeHg concentration (µg/g) in s muddy and sandy sediment

Sediment	Metal	Station				
		Hendijan	Ghanam	Meleh	Musa	Zangi
Muddy	Hg	0.83 ± 0.23^{a}	0.54 ± 0.02^{b}	0.41 ± 0.05^{c}	1.53 ± 0.01^{a}	0.76 ± 0.04^{b}
Sandy		0.67 ± 0.06^{b}	0.46 ± 0.05^{b}	0.38 ± 0.01^{c}	1.17 ± 0.04^{a}	0.54 ± 0.08^{b}
Muddy	MeHg	0.62 ± 0.05^{b}	0.43 ± 0.05^{b}	0.35 ± 0.05^{c}	1.15 ± 0.08^{a}	0.54 ± 0.05^{b}
Sandy		0.53 ± 0.02^{b}	0.37 ± 0.01^{b}	0.23 ± 0.04^{c}	0.85 ± 0.06^{a}	0.39 ± 0.06^{c}

a,b,c show significance difference

(ANOVA) fallowed by Duncan post hoc test was used to compare the data by station. The metal concentration of each sample is expressed in micrograms of metal per gram dry of sediment (µg/g) and a probability of $p = 0.05$ was set to indicate statistical significance.

RESULTS AND DISCUSSION

Table 3 shows the mean and comparison of the Hg and MeHg concentration in muddy and sandy sediment from five estuaries along Khuzestan Shore, north part of the Persian Gulf. According to these data, mean concentrations of Hg were 0.83 ± 0.23 µg/g for Hendijan estuary, 0.54 ± 0.02 µg/g Ghanam estuary, 0.41 ± 0.05 µg/g Meleh estuary, 1.53 ± 0.01 µg/g Musa estuary and 0.76 ± 0.04 µg/g for Zangi estuary in muddy sediment, and 0.62 ± 0.05 µg/g for Hendijan estuary, 0.43 ± 0.05 µg/g Ghanam estuary, 0.35 ± 0.05 µg/g Meleh estuary, 1.15 ± 0.08 µg/g Musa estuary and 0.54 ± 0.05 µg/g for Zangi estuary in sandy sediment. There was significant difference in Hg concentration between different stations (□ < 0.05). The highest concentrations of Hg were detected in sediment of Musa estuary, followed by Hendijan, Zangi and Ghanam and Meleh, respectively. Also, there was significant difference in Hg concentration between muddy and sandy sediment (□ < 0.05). The concentration of Hg in muddy sediments was higher than sandy sediments in all estuaries.

According to these data, mean concentrations of MeHg were 0.62 ± 0.05 µg/g for Hendijan estuary, 0.43 ± 0.05 µg/g Ghanam estuary, 0.35 ± 0.05 µg/g Meleh estuary, 1.15 ± 0.08 µg/g Musa estuary and 0.54 ± 0.05 µg/g for Zangi estuary in muddy sediment, and 0.53 ± 0.02 µg/g for Hendijan estuary, 0.37 ± 0.01 µg/g Ghanam estuary,

Table 4 Correlation matrix of muddy, sandy, organic carbon, carbonate, salinity and metal concentration in sediment sample

	Hg	MeHg	Mud	Sand	OC	$CaCo_3$	Salinity
Hg	1						
MeHg	0.681	1					
Mud	0.722	0.697	1				
Sand	0.394	0.213	0.253	1			
OC	0.776	0.616	0.785	0.457	1		
$CaCo_3$	0.583	0.431	0.627	0.312	0.375	1	
Salinity	0.347	0.292	-0.215	-0.123	-0.143	0.362	1

0.23 ± 0.04 µg/g Meleh estuary, 0.85 ± 0.06 µg/g Musa estuary and 0.39 ± 0.06 µg/g for Zangi estuary in sandy sediment. There was significant difference in MeHg concentration between different stations (□ < 0.05). The highest concentrations of MeHg were detected in sediment of Musa estuary, followed by Hendijan, Zangi and Ghanam and Meleh, respectively. Also, there was significant difference in MeHg concentration between muddy and sandy sediment (□ < 0.05). The concentration of MeHg in muddy sediments was higher than sandy sediments in all estuaries.

Pearson correlation obtained (Table 4) was between Hg and Hg concentration and the finer sediment fractions. Samples with high contributions of muddy fractions presented higher metal concentrations (Hg: r = 0.72; α = 0.001, MeHg: r= 0.69; α = 001), an indication of a highly significant correlation), while the sandy samples presented the lowest metal concentrations (Hg: r = 0.39; α = 0.001, MeHg: r= 0.21; α = 001. There was significant correlation between organic carbon (OC) in the sediment with metal distribution (Hg: r = 0.77; α = 0.001, MeHg: r= 0.61; α = 001). El Nemr et al. (2007) had investigated on concentration of heavy metal in sediment from Egyptian Coast along Mediterranean Sea. They showed there is a significant correlation between metal concentration and particles size of sediment. This means that reducing the size of the sediment particles cause increase metal concentration, because, the organic carbon (OC) content in fine-grained sediments are higher than coarse-grained sediment.

Alagarsamy (2006) studied the metallic and organic contaminants in sediments of Mandovi estuary, west coast of India. Their results showed metal concentrations is higher in muddy sediment than sediment with coarse particles and there is a positive correlation between amount of organic carbon and metal concentration. Hosseini and Sajjadi (2018) studied the Hg accumulation in sediment from Musa estuary, along the Persian Gulf. They results showed there was significant correlation between Hg concentration and organic carbon (OC) in the sediment. The amount of organic carbon is more in the muddy sediment, consequently, accumulation of metal is more in this type of deposit (Anirudh et al. 2009; Qiang et al. 2020).

The carbonate ($CaCo_3$) level in the sediments has significant correlation with Hg and MeHg concentration (Hg: r = 0.58; α = 0.001, MeHg: r= 0.43; α = 001). The $CaCo_3$ content in muddy sediment was higher than the sandy sediment, this means that increases in the carbonate content cause will be increases in the metal concentration. Raeisi Sarasiab et al. (2014) have study on mercury distribution in contaminated surface sediments from four estuaries, north part of Persian Gulf. They showed there is a significant correlation between $CaCo_3$ content with mercury accumulation and the highest mercury concentration was found in sediment with high percentage of carbonate. Also, Mooraki et al. (2009) and Saeedi et al. (2013) have similar results with results of present study and they were found positive correlation between carbonate content and metal concentration.

The comparison between muddy and sandy sediment shows that concentration and pollution level of metal in muddy sediment was higher in all estuaries. Therefore, there is a correlation between the grain size fraction composition of the sediment and the content of metals deposited in it. The contents of these metals increased in the fractions of grain size < 63 µm. This phenomenon can be explained by the mechanism of metal deposition in the coastal sediments. The metals accumulated in the estuary sediment undergo precipitation as a result of changing physical and chemical conditions in the sediment such as pH, oxygen content, redox potential, content of salts, organic carbon and carbonate (Mooraki et al. 2009; Saeedi et al. 2013). In suspended sediment, understanding the particle size distribution and concentration may pave the way for well knowing the concentration and distribution of metals. Particulates adsorb or bind metals and transport together following the hydrological kinetics (Zhong et al. 2012). For an instance, in various runoffs, metal concentration is correlated with suspended sediment content (Saeedi et al. 2013). Besides, Carmen's result indicated that flooding (resulting in more reduced conditions) minimized the availability of metals duo to the biodegradation of fresh organic matters and the production of sulfides (Hernandez Crespo et al. 2012).

Table 5 The Igeo, EF and PLI for Hg and MeHg in muddy and sandy sediment

Index	Sediment	Metal	Station				
			Hendijan	Ghanam	Meleh	Musa	Zangi
Igeo	Muddy	Hg	2.56	1.22	1.39	6.75	2.55
		MeHg	1.62	1.12	1.63	5.42	1.19
	Sandy	Hg	1.87	0.64	0.77	5.52	1.45
		MeHg	0.83	0.78	0.61	5.26	0.59
EF	Muddy	Hg	2.55	0.55	0.75	5.56	2.59
		MeHg	1.82	0.78	0.59	3.61	1.42
	Sandy	Hg	1.66	0.54	0.41	3.76	1.78
		MeHg	0.62	0.57	0.54	2.48	0.85
PLI	Muddy	Hg	3.56	2.12	2.78	4.45	3.73
		MeHg	2.34	0.72	3.54	3.32	2.65
	Sandy	Hg	1.56	0.84	1.34	3.55	1.25
		MeHg	1.23	0.78	1.51	3.43	1.82

Recent studies have also shown the importance of fine grain suspended sediment in the transport of heavy metals through fluvial systems (Chao et al. 2009, Sadeghi et al. 2012). During resuspension, for example, metal in suspended particle matter transformed from stable to labile fractions and their concentrations are higher in the upper of water column than the lower part (Kucuksezgin et al. 2008). Present result possibly be explained by the particles concentration effect, which defined as the downward trend in metal partition coefficients when the suspended particulate matters increases. Adsorption time is a parameter to be considered in distribution and partitioning of heavy metals. For example, aging can redistribute the adsorbed metals to the interior stability of sorption sites of organic and mineral substrates (Guo et al. 2011). The prolonged aging of heavy metals in sediment has been demonstrated to be a major parameter in determining their availability: the exchangeable and carbonate fractions decrease while the refractory fractions (organic and residual phase) increase (Zhong et al. 2012). So it is essential to allow an adequate aging period following trace metal spiking to sediments, especially with regard to the bioavailability of heavy metals.

The salinity level in the sediments has correlation with metal accumulation (Hg: $r = 0.34$; $\alpha = 0.001$, MeHg: $r= 0.29$; $\alpha = 001$). The pH level in sandy sediment was lower than muddy sediment and this means that condition is acidic in sandy sediment and alkaline in muddy sediment. Therefore, accumulation of heavy metal in alkaline environment (muddy sediment) is higher than acidic environment (sandy sediment). The relative high salinity can inhibit the growth and activity of SRB by increasing cell osmotic pressure and repressing metabolic enzymes, and thus affected the reduction of SO_4^{2-} and the decomposition of organic matter in sediment, resulting in increasing metals bioavailability (Hou et al. 2013). Also, the increase of salinity was related to an enhancement in the content of major cations (e.g., Na, K, Ca, Mg) that compete for the sorption sites with heavy metals and decreased the binding of metals to humic acids (Du Laing et al. 2009). When soluble chloride complexes enforced, the mobility of trace metals was improved.

Table 5 shows the geoaccumulation index (Igeo) for Hg and MeHg in muddy and sandy sediment collected from all estuaries. I*geo* was concluded in muddy sediments for Hg moderately polluted in Meleh and Ghanam estuaries, moderately to strongly polluted in Hendijan and Zangi estuaries, and extremely polluted in Musa estuary. In sandy sediment, I*geo* was unpolluted to moderately polluted in Meleh and Ghanam estuaries, moderately polluted in Hendijan and Zangi estuaries and strongly to extremely polluted in Musa estuary. I*geo* was concluded in muddy sediments for MeHg moderately polluted in all estuaries, except Musa estuary that was extremely polluted. In sandy sediment, it was the unpolluted to moderately polluted in all estuaries, but in Musa estuary was extremely polluted. Therefore, result shows that highest Igeo level for Hg and MeHg were found in the Musa Estuary, this estuary was extremely polluted. Also, results shows that pollution level in Ghanam and Meleh estuaries was lower than other estuaries. The pollution load index (PLI) for Hg and MeHg in muddy and sandy sediment showed in Table 5. PLI value were above 1 in Hendijan, Melh, Musa and Zangi estuaries for both metals that means there are Hg and MeHg pollution in four sampling sites. But, in Ghanam estuary Pll value was above 1 for Hg in muddy sediments, and low 1 for MeHg in muddy and both metals in sandy sediments (Table 4). Therefore, Hg and MeHg pollution was in all estuaries except for Ghanam.

Table 6 Hg and MeHg concentrations in sediments from different marine environment

Hg	MeHg	Location	Reference
1.55	1.51	Hendijan Estuary (Persian Gulf)	This study
0.51	0.40	Ghanam Estuary (Persian Gulf)	This study
0.50	0.29	Meleh Estuary (Persian Gulf)	This study
1.84	1.59	Musa Estuary (Persian Gulf)	This study
0.83	0.56	Zangi Estuary (Persian Gulf)	This study
7.8	2.84	California	Joseph (2001)
1–5.6	0.1– 0.4	Persian Gulf	Agah et al. 2008
0.05–2.66	0.02- 0.5	India	Anirudh et al. 2009
13–40	0.5 – 1.2	Bushehr (Persian Gulf)	Agah et al. 2010
0.04 - 0.65	0.03– 0.22	Lebanon	Carine et al. 2011
1.73	1.16	Musa Estuary (Persian Gulf)	Abdolahpour Monikh et al. 2012
0.75	0.55	Meleh Estuary (Persian Gulf)	
1.87	1.41	Musa Estuary (Persian Gulf)	Abdolahpour Monikh et al. 2013
0.54	0.32	Ghanam Estuary (Persian Gulf)	
0.62	0.27	Zangi Estuary (Persian Gulf)	
1.75	1.43	Bahrekan Estuary (Persian Gulf)	Hosseini et al. 2012
2.12	1.72	Musa Estuary (Persian Gulf)	
0.85	0.62	Abadan Estuary (Persian Gulf)	
0.72	0.48	Bushehr coast (Persian Gulf)	
0.65	0.38	Zangi Estuary (Persian Gulf)	Hosseini et al. 2013
0.94	0.67	Arvand River (Persian Gulf)	
1.73	1.21	Musa Estuary (Persian Gulf)	Nabavi et al. 2013
0.95	0.63	Bahrekan Estuary (Persian Gulf)	
0.75	0.53	Ahmadi Estuary (Persian Gulf)	Raeisi Sarasiab et al. 2014
1.81	1.24	Musa Estuary (Persian Gulf)	
0.37	0.23	Ghanam Estuary (Persian Gulf)	
0.54	0.36	Zangi Estuary (Persian Gulf)	
1	1		WHO (2002)
1	1		FAO (2002)
0.5 - 1	0.5		EC (2001)
0.5	0.5		Australia MPC (2000)

The Enrichment Factor (EF) is a convenient measure of geochemical trends and is used for making comparisons between areas (Guilherme et al. 2011). A value of unity denotes neither enrichment nor depletion relative to the Earth's crust. The anthropogenic impact could be quantified by calculating the Enrichment Factor (EF). Table 6 shows the Enrichment Factor (EF) for Hg and MeHg in muddy and sandy sediment from five estuaries along the Persian Gulf. Enrichment Factor (EF) were detected for Hg, a value of $0.55 < EF < 5.56$ and for MeHg value of $0.59 < EF < 3.61$ in muddy sediment. Also, Enrichment Factor (EF) were detected for Hg $0.41 < EF < 3.76$ and for MeHg $0.54 < EF < 3.78$. This result suggests that traces of Hg may be due to crustal materials or natural weathering processes in muddy and sandy sediment in Ghanam (0.55) and Meleh (0.75) estuaries, but in Hendijan, Musa and Zangi estuaries may be due to anthropogenic impact. Enrichment Factor (EF) was detected 2.55 for Hendijan estuary, 5.56 for Musa estuary and 2.59 for Zangi estuary. Therefore, the results suggests that sources of Hg in three estuaries may be to anthropogenic activates such as oil and petrochemical industries. For MeHg in muddy sediment, enrichment factor were detected 0.78 in Ghanam estuary, 0.59 in Meleh estuary and 1.42 in Zangi estuary, therefore, traces of MeHg could be due to crustal materials or geochemical background of area. Enrichment Factor level were detected 1.82 for Hendijan estuary and 3.61 for Musa estuary, therefore, traces of MeHg in both estuaries could be due to anthropogenic activates. In sandy sediment, Enrichment Factor for MeHg were detected ($EF < 1$), except in Musa estuary that was 2.48.

Therefore, traces of MeHg in sandy sediment from all estuaries could be due to crustal materials or geochemical background, except Musa estuary that related to anthropogenic activates.

Finally, this result showed that Enrichment Factor for Hg in sandy and muddy sediment collected from Hendijan, Zangi and Musa estuaries was higher than other estuaries and high concentration of metal related anthropogenic activities. But, Enrichment Factor for MeHg in muddy sediment collected Hendijan, Zangi and Musa estuaries and in sandy sediment collected Musa estuary was more than normal level, and sources of MeHg is related to human activity. Also, Enrichment Factor for Hg and MeHg were less than normal level in sandy and muddy sediment collect from Ghanam and Meleh, and sources of metals in both estuaries related to natural origin and geochemical back ground. Abdolahpour et al. (2012) reported Enrichment Factor for Hg in Musa, Zangi and Ahmadi estuaries from Khuzestan Shore, north part of the Persian Gulf. Their results show that different industries such oil, gas and petrochemical cause into metals to the aquatic environment and those accumulation in sediments, water and organisms. Hosseini et al. (2013) have been reported enrichment factor for Hg and MeHg in Musa estuary. They suggests that sources of metals in Musa estuary related to human and industrial activities such as petrochemical and gas production industries. Raeisi Sarasiab et al. (2014) shows that Enrichment Factor for Hg was higher than normal level in sediment of Musa estuary, and metal sources related to man-made industries, specially, oil industry.

The results showed that pollution level in Hendijan and Musa estuaries is higher than other estuaries and both estuaries are extremely polluted for Hg and MeHg. Because, Musa estuary is near port of Imam Khomeini that located in Khuzestan Province, north-west of the Persian Gulf. There are ships and tanker traffic for transportation of oil and gas and the existence of several industries especially petrochemical industries caused the influx of various organic and non-organic contaminants such as heavy metals into the ecosystem. Also, Musa estuary is surrounded by more than 19 petrochemical units such as chlor-alkali plant and superphosphate plant. The fact that total concentration of some petrochemical-related metals enter to this region and causes increase in the pollution level. Therefore, Musa estuaries is a strong evidence that the heavy metal in this estuary was sourced from petrochemical activities, and not from background geological sources (Abdolahpur Monikh et al. 2012). Also, the Hendijan estuary is the nearest creek to Musa estuary, Mahshahr City and petrochemical units (Mooraki et al. 2009). In addition, Hendijan estuary is one of the biggest ports in Iran is located in the mouth of the Persian Gulf that there are many boats and ships for carry oil and cargo in this estuary. There are many ships making for construction and repair of ships and that they can produce pollutants such as toxic heavy metal, therefore, Hendijan and Musa estuaries receives high level of pollutants from different sources.

An overview of Hg and MeHg concentrations in marine sediment in different marine environments is shown in Table 7. In several coastal areas of the Persian Gulf, (Bushehr, Bahrekan, Abadan and Arvand) and Musa and Hendijan estuaries, Hg and MeHg levels exceeded 1.0 µg/g. In the coastal area of California the average Hg and MeHg concentrations were higher than our study, but in coastal area of Lebanon and India, the average Hg and MeHg concentrations were lower than mean concentration in our study. The average Hg and MeHg concentrations in Musa estuary in all previous studies (Hosseini et al. 2012; Abdolahpour Monikh et al. 2012, 2013; Nabavi et al. 2013; Raeisi Sarasiab et al. 2014) were exceede1.5 µg/g that suggests with results of this study. Also, the average Hg and MeHg concentrations in Ghanam and Meleh estuaries in all previous studies (Hosseini et al. 2012; Abdolahpour Monikh et al. 2012, 2013; Raeisi Sarasiab et al. 2014) were lower 1.5 µg/g that suggests with results of this study. Therefore, the results of this study is suggests with results of all previous studies. In comparison with some standards, the average Hg and MeHg concentrations in Musa and Hendijan estuaries were higher than the all standards (Australia MPC (2000); EC (2001); WHO (2002); FAO (2002). In the Zangi estuary, the mean Hg and MeHg concentrations were lower than WHO (2002) and FAO (2002), but were higher than MPC (2000) and EC (2001). Finally, the mean concentrations of Hg and MeHg in Ghanam and Meleh estuaries were lower than all standards (Australia MPC (2000); EC (2001); WHO (2002); FAO (2002).

CONCLUSIONS

Environment geochemistry of mercury and methylmercury in contaminated sediments from five estuaries along the Imam Khomeini port, northwest part of the Persian Gulf, south Iran, were studied. The results showed there is a correlation between the grain size of the sediment and the content of heavy metals, and concentration and pollution level of mercury and methylmercury in muddy sediment was higher in all estuaries, therefore, the contents of these metals increased in the fractions of grain size < 63 µm. Also, there is a position correlation between chemical conditions in the sediment such as pH, oxygen content, organic carbon and carbonate with content of heavy metal. Muddy sediments have high percentage of organic carbon and carbonate, therefore, they can receive the high concentration of heavy metal in comparison of sandy sediments. There was significant difference different estuaries ($P < 0.05$) , and the highest concentration of heavy metal were detected in Musa estuary, followed by Hindijan, Zangi, Ghanam and Meleh estuary, respectively. The results of geoaccumulation Index showed that Musa and Hendijan estuaries are strongly to extremely polluted. Enrichment Factor show metals trace and sources in Hendijan, Musa and Zangi estuaries is related to anthropogenic impact such as petroleum and petrochemical industries. Finally, it can be concluded that petroleum and petrochemical industries are the most important source for heavy metal pollution around the Imam Khomeini port, so this area need to environmental management of industrial units.

Author statement

Mahnaz Sadat Sadeghi: Conceptualization, methodology, food chain experiment, exposure, measurement, data curation, writing- original draft preparation. Mehdi Hosseini. Methodology, food chain experiment, exposure, writing. Mehrnaz Baniamam: Supervision, editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors declare that the data supporting the findings of this study are available within the article and its Supplementary Information files. Data are also available from the corresponding authors upon reasonable request.

Ethical Statement standards

This article does not contain any studies with human participants or animals performed by any of the authors.

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