Spatiotemporal variability of heavy metals concentration in an industrialized estuarine-bay ecosystem in Northern Persian Gulf

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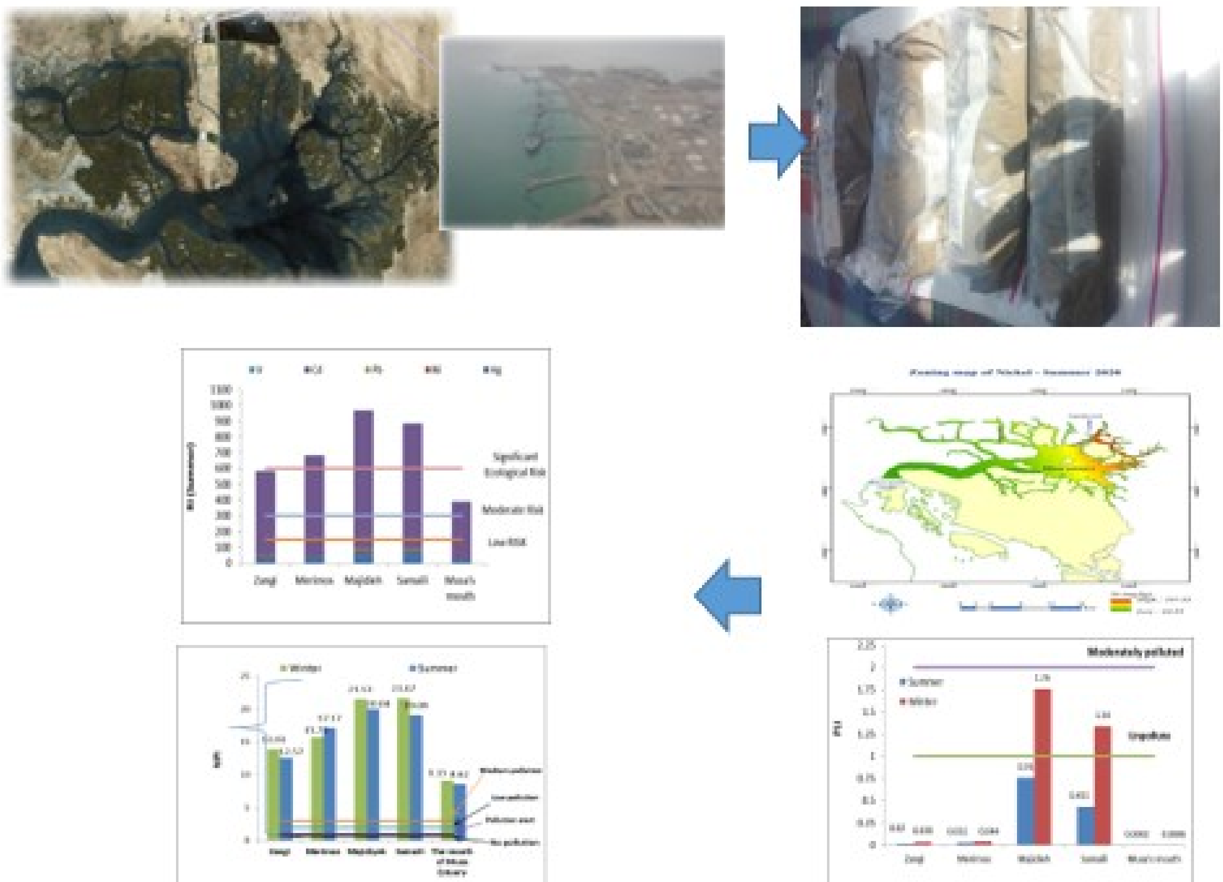
GRAPHICAL ABSTRACT



ABSTRACT

This study aimed to evaluate the pollution level of Ni, Pb, Cd, V, and Hg in the sediments of Musa Estuary and its four main tributaries namely Majidieh, Zangi, Semaili, and Merimos (collectively known as Mahshahr estuaries) in summer and winter 2019 using sediment quality indices. In each tributary, 4 stations were chosen to represent the estuary condition. One control point was also considered at the mouth of the estuary (17 stations in total). 51 sediment samples were collected using the Van Wien Hydrobius grab sampler and measurements were performed utilizing the ICP-MASS device. The heavy metals concentration was ordered as Ni> Pb> Cd> V> Hg and tributaries followed the order of Majidiyeh> Samaili> Zangi> Merimos> Musa's mouth (control point) in terms of heavy metal concentration in both seasons. Heavy metal concentrations were positively and significantly correlated with electrical conductivity and pH ($P < 0.05$). The potential ecological risk (Er) was low for Ni, Pb, and V, moderate for Cd (except for Musa's mouth in summer (27.33)). Hg had low risk in Zangi Tributary and Musa's mouth in summer and medium risk in other tributaries in winter. The ecological risk index (RI) showed considerable risk in Zangi Tributary and Musa's mouth in both seasons and high risk in Merimos, Majidieh, and Samaili tributaries. According to the pollution load index (PLI), the sediments of Mahshahr estuaries showed no pollution in summer and moderate pollution in Majidiyeh and Smaili tributaries in winter. The Nemro Integrated Pollution Index ranged from 8.62 to 20.04 in summer and 9.15 to 21.53 in winter Mahshahr estuaries which indicate its high pollution potential. The findings of this research and those of previous studies alarm increasing levels of heavy metals' content in the sediments of Mahshahr estuaries over the past years.

Keywords: Mahshahr estuaries, spatio-temporal analysis, pollution indices, heavy metals.

1. Introduction

Heavy metals are a major worldwide pollution threat due to their toxicity and dispersion characteristics that can be enriched in living organisms (Bessa et al. 2018; Ekoa Bessa et al. 2018). In aquatic environments, the main sources of heavy metal entry are storm and erosion (Hanif et al. 2016), atmospheric sediments, wastewater effluents, fertilizers, and pesticides (Sun et al. 2018; Zhou et al. 2018; Jiang et al. 2019), land surface runoff and chemicals from municipal, industrial and agricultural activities (Xiao et al., 2013; Zahra et al., 2014; Mimbo et al., 2018; Al-Hadithy et al., 2018; Chen et al., 2019). Sediments are a source of nutrients, micro and macro flora and fauna that play an important role in aquatic habitats (Jain et al., 2004; Guo et al., 2010; Bat and Ozkan, 2019; Gao et al., 2019). Moreover, they have proven effective as pollution exposure indicators and environmental pollution screening tools in surrounding ecosystems (Xiao et al., 2013; Zahra et al., 2014; Ekoa Bessa et al., 2018). High sediment heavy metal levels can increase the risk of adverse human health effects due to their easy assimilation and bioaccumulation potential in biota and food chain (Dessalew et al., 2018; Bhattacharyya et al., 2019).

The Musa Estuary is the largest in the Persian Gulf. It is located in the northwest of the Persian gulf in Khuzestan Province, southwestern Iran and is composed of several tributaries known as Khur. The Musa Estuary is of special importance due to its geographical characteristics. It is situated next to the largest inland wetland of Shadegan and the Mahshahr Special Economic Zone and is the recipient of wastewater from various industries such as the large-scale Petrochemical Complex of Imam, Razi, and Farabi. This multitude of stressors has caused multiplicative adverse impacts on the resilience of the sensitive and high-potential fishing coastal ecosystem. Hence, it is of high importance to determine the biological potentials and their associated hazards of the aquatic ecosystem.

Sediment quality guidelines (SQGs) provide a well-known procedure for heavy metal pollution assessment in aquatic environments. In combination with various other statistical techniques and indicators, SQGs can be applied for ecological risk assessment as well as pollution severity classification and zoning of different habitats (Zhang et al., 2017; Bezerra da Silva et al., 2019). Sediment indicators such as SQGs are useful tools for assessing the potential biological effects of heavy metals on living organisms, determining water quality, and evaluating residual reactions and material storage in sediments (Petrosyan et al., 2019; Wojciechowska et al., 2019). This study aimed to determine the concentration, distribution, and zoning of heavy metals (Ni, Hg, Pb, Cd and V) in sediments of Mahshahr estuaries (Zangi, Merimos, Majidieh and Semili tributaries) and the impact of human activities and on sediment quality using sediment quality indicators in different seasons (summer and winter).

2. Materials and methods

Sampling was performed in the summer and winter of 2019 in Mahshahr Estuary (Zangi, Marimos, Majidieh and Samaili tributaries). Four stations with a distance of about 1.5 km from each other were set up in each tributary. The control station was situated at the mouth of the Musa estuary (16 stations in total + 1 control station). Table 1 shows the coordinates of the sampling stations in each tributary and Figure 1 represents their spatial distribution. 51 surface sediment samples (depth of 0-5 cm and area of 0.45 m^2) were collected using the Van Wien Hydrobius bottom sampler (made in Germany) operated by a boat. The sediments were packed in cellophane bags, labeled by sample specifications, date and station number and stored in ice-cooled refrigerator boxes until transport to the laboratory.

Table 1- Coordinates of sampling stations

Geographical coordinates		Khur name	Station number	Geographical coordinates		Khur name	Station number
E longitude	N latitude			E longitude	N latitude		
´45,6´´1149°	´46,28´´30°	Majidieh	9	´55,3´´0349°	´25,2´´2830°	Zangi	1
´44,8´´1149°	´22,5´´2830°		10	´48,8´´0349°	´45,5´´2930°		2
´35,6´´1149°	´22,9´´2830°		11	´43,5´´0349°	´3,1´´2930°		3
´59,4´´1049°	´51,4´´2830°		12	´38,8´´0349°	´22,9´´2930°		4
´5,1´´1149°	´49,4´´2830°	Samaili	13	´59,8´´0349°	´18,2´´2430°	Merimos	5
´10,9´´1149°	´41,3´´2830°		14	´30,5´´0449°	´46,1´´2430°		6
´28,1´´1149°	´25,4´´2830°		15	´53,1´´449°	´54,9´´2330°		7
´45,6´´1149°	´46,6´´2830°		16	´14,4´´549°	´44,9´´2330°		8
´92,6´´03 39°	´85,2´´6433°	Musa's mouth	17				

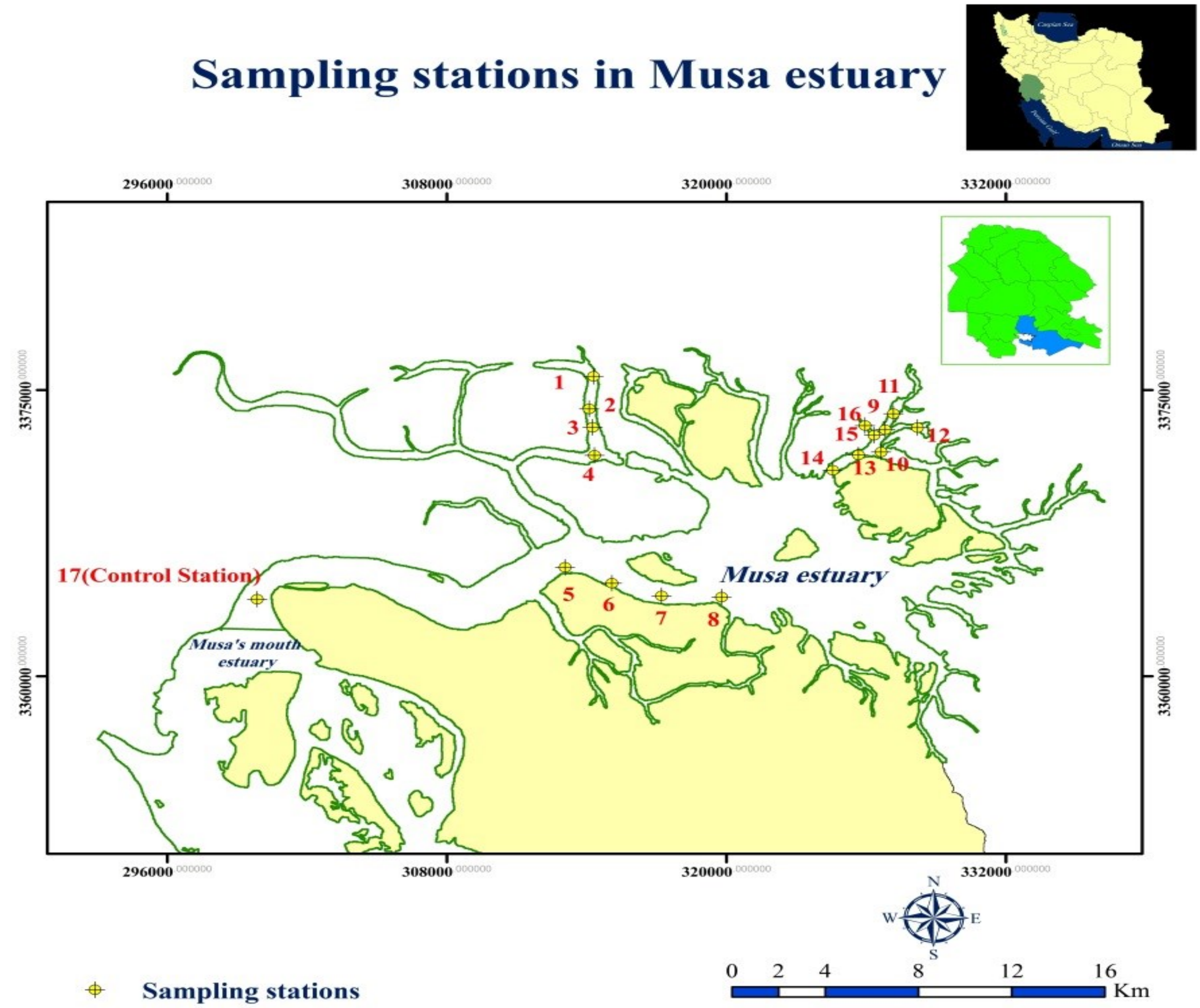


Figure 1: Location of sampling stations in Musa Estuary

In the laboratory, the sediment samples were dried at 52 ° C and passed through sieve No. 230 (mesh size 63 microns). Ni, V, Pb and Cd were analyzed by acid digestion. 1 gram of dried sediment was transferred to the digestion tube and mixed with 7 ml concentrated nitric acid (HNO_3) and Hydrochloric acid (HCL) in a ratio of 1:3. Samples were placed on a hot plate for three hours. After cooling at room temperature, 5 ml HF was added. The digested solutions were transferred to a 25 ml volumetric flask and diluted with normal HCl. The samples were passed through Whatman 42 filter paper and analyzed at ppm level using the ICP-MASS Integra model device made by GBC Korea (Vaezi et al., 2014).

To measure the sediment mercury content, 1 g of each sample with 4 ml concentrated HNO and 2 ml of concentrated sulfuric acid were added into a digestion tube. They were kept at room temperature for one hour and placed on a hot plate for 3 hours at 90 ° C. After cooling, 0.5 ml of 10% potassium dichromate solution was added to the digested samples, passed through a filter paper and reached a volume of 25 ml in a volumetric flask (MOOPAM, 2010). The Hg content of the samples was measured employing the cold steam technique using a graphite furnace atomic absorption apparatus (Perkin Elmer 100B) with an accuracy of 0.5 ng/g and at ppb level (Glasby and Szefer, 1998). Sediment properties including pH and electrical conductivity (in a mixture with distilled water in a ratio of 1: 5) were measured using a portable HACH device (Clesceri et al., 1989).

2.1. Potential Ecological Risk and Ecological Risk Index

The sediment ecological risk of the sediments was measured using Equations 1 and 2 (Hakanson, 1980)

where C_f is the contamination factor (the metal concentration in a location to that of the reference place), T_r is the metal ecological risk, RI is the ecological risk of all metals. Hakanson (1980) indicated the value (TR) an indicator of the toxicity of heavy metals. Measurements were classified into four groups as given in Table 2.

Eq.1: $E_r = T_r \times C_f$

Eq.2: $RI = \sum E_r$

Table 2 – Classification of Er and RI (Hakanson 1980)

E_r	Ecological risk	RI	Risk
E_r <40	Low	150< RI	**Low**
40<E_r <80	Moderate	300 < RI ≤ 150	**Moderate**
80<E_r <160	Considerable	600 <RI ≤ 300	**Considerable**
160<E_r <320	Very high	600 ≥ RI	**High**
E_r > 320			**Very high**

2.2. ***Pollution Load index***

This indicator estimated the metal contamination level in sediments (Equation 3)(Tomlinson et al. 1980). Where CF is the contamination factor. PLI values of less than 1 indicate no pollution, 1 to 2 indicate moderate pollution, 2 to 3 indicate high pollution and 3 to 4 indicate very high pollution (Liu et al. 2014).

$$\text{Eq.3} \qquad PLI = \sqrt[5]{CF_V \times CF_{Cd} \times CF_{Pb} \times CF_{Ni} \times CF_{Hg}}$$

2.3. Nemro Integrated Pollution Index

This index is calculated using Equation 4:

The quality of sediments are categorized into five classes according to NIPI values: no pollution (NIPI≤0.7), pollution warning (0.7<NIPI≤1), low pollution (1<NIPI≤2), moderate pollution (2<NIPI≤3), and high pollution (NIPI>3) (Zhang et al., 2018).

$$\text{Eq4.} \qquad NIPI = \sqrt{\frac{Cf_{average}^2 + Cf_{max}^2}{2}}$$

2.4. Data processing

The normality of the data was checked using the Shapiro-Wilk test. One-way analysis of variance was applied to compare the concentrations of heavy metals and the supplementary test was used to distinguish between groups with significant differences. Statistical analysis and data editing were performed using SPSS 23 and Exel 2013 software.

3. Results and Discussion

normality of the data was checked using the Shapiro-Wilk test. One-way analysis of variance was applied to compare the concentrations of heavy metals and the supplementary test was used to distinguish between groups with significant differences.

Table 3. Comparison of mean heavy metals concentrations (mg/kg) in sediments of Mahshahr estuaries in two seasons of summer and winter 2019.

	Hg		**Ni**		**Pb**		**Cd**		**V**	
	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	**Winter**
Zangi	0.32^{Acd}	0.40^{Ac}	64.99^{Ac}	64.81^{Ac}	16.86^{Ac}	16.85^{Ac}	4.93^{Ab}	5.28^{Ac}	0.64^{Ab}	$\mathbf{0.77^{Ab}}$
Merimos	0.40^{Bc}	0.48^{Ab}	51.8^{Ad}	53.72^{Ad}	10.61^{Ad}	10.25^{Ad}	6.52^{Aa}	5.93^{Ac}	1.1^{Aa}	$\mathbf{1.25^{Aa}}$
Majidieh	0.67^{Aa}	0.73^{Aa}	102.52^{Aa}	112.02^{Aa}	20.55^{Ab}	25.21^{Ab}	7.9^{Bc}	8.75^{Aa}	1.14^{Aa}	$\mathbf{1.25^{Aa}}$
Samaili	0.55^{Ab}	0.74^{Aa}	81.61^{Ab}	82.46^{Ab}	24.37^{Aa}	29.25^{Aa}	6.95^{Aa}	7.91^{Ab}	1.14^{Aa}	$\mathbf{1.31^{Aa}}$
Musa's mouth	0.23^{Ad}	0.28^{Ad}	44.85^{Ae}	40.92^{Ae}	4.50^{Ae}	4.96^{Ae}	2.73^{Ac}	3.56^{Ad}	0.53^{Ab}	$\mathbf{0.56^{Ac}}$

Different lowercase letters indicate significant differences ($p \leq 0.05$) in heavy metal’s concentrations between tributaries in each season and different uppercase letters indicate significant differences ($p \leq 0.05$) in heavy metal’s concentrations between seasons

Electrical conductivity and pH samples of sediments differed significantly between tributaries but insignificantly between summer and winter ($P < 0.05$). The lowest Electrical conductivity and pH were observed in the mouth of Musa estuary and Zangi tributary. Electrical conductivity varied between 27.36 and 17.46 ms/cm and pH ranged from 8.21 to 8.9 with the highest value recorded in the Semali tributary (Table 4). Correlation results showed strong associations between sediment parameters and heavy metal concentrations (Table 5).

Table 4. Comparison of mean electrical conductivity and pH in the sediments of Mahshahr estuaries in summer and winter of 2019

	EC (ms/cm)		pH	
	Summer	Winter	Summer	Winter
Zangi	18.6±0.74 [Ad]	18.88±0.71 [Acd]	8.3±0.11 [Ac]	8.43±0.15 [Ac]
Merimos	20.11±0.28 [Ac]	20.13±0.35 [Ac]	8.21±0.06 [Ac]	8.32±0.12 [Ac]
Majidieh	22.33±0.78 [Ab]	22.56±0.77 [Ab]	8.64±0.09 [Ab]	8.68±0.03 [Ab]
Samaili	26.76±1.12 [Aa]	27.36±1.13 [Aa]	8.89±0.04 [Aa]	8.9±0.05 [Aa]
Musa's mouth	17.46±0.14 [Ad]	17.92±0.16 [Ad]	8.31±0.08 [Ac]	8.38±0.18 [Ac]

SD, Standard deviation

Different lowercase letters indicate significant differences ($p \leq 0.05$) in between tributaries in each season and different uppercase letters indicate significant differences ($p \leq 0.05$) between seasons

Table 5. Correlation results between heavy metal concentration, electrical conductivity and pH in the sediments of Mahshahr estuaries

pH	**pH**						
Ec	**0.829	Ec					
Hg	**0.781	**0.782	Hg				
Ni	**0.610	**0.657	**0.860	Ni			
Pb	**0.829	*0.810	**0.842	**0.819	Pb		
Cd	**0.719	**0.583	**0.861	**0.831	**0.785	Cd	
V	**0.725	**0.508	**0.789	**0.586	**0.601	**0.858	V

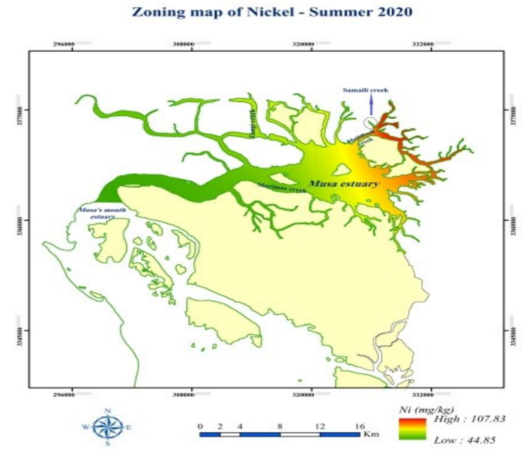
** Corelation at 0.01

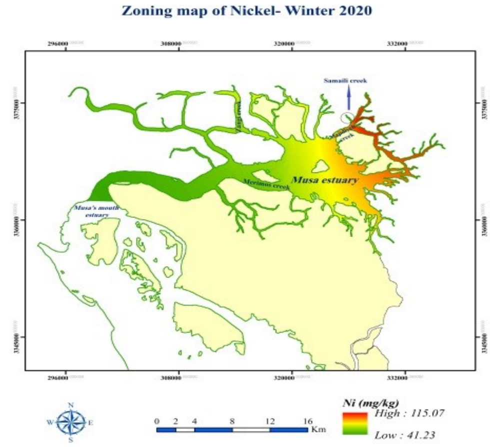
According to Table 6, RI showed low Ni, Pb and V risk whereas showed very high Cd risk in winter and summer except for the Musa's mouth. Hg had low ecological risk potential in summer and winter. In terms of Er, Zangi and Musa tributaries showed significant ecological risk and Merimos, Majidieh and Samaili tributaries showed very high ecological risk.

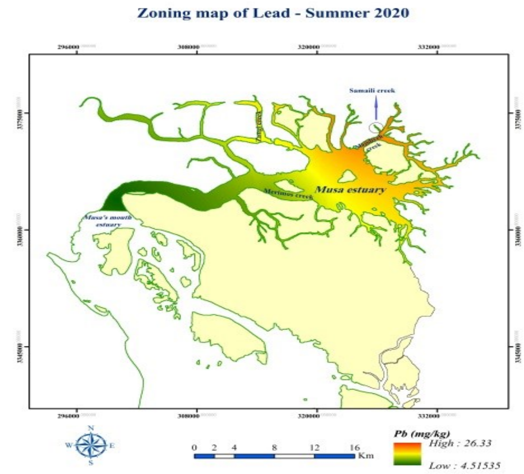
Table 6. Results of Er and RI in sediments of Mahshahr estuaries in summer and winter 2019

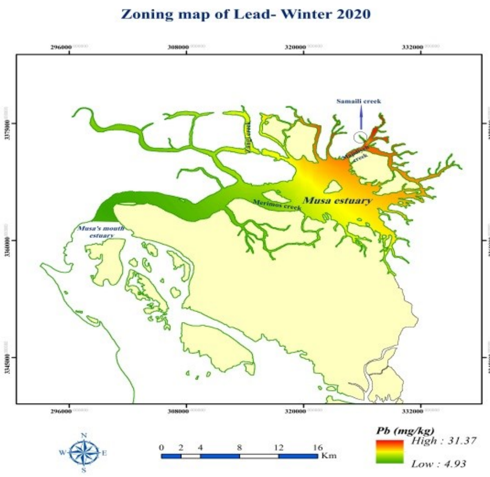
Season	station	Er					RI	
		Hg	Ni	Pb	Cd	V		
Summer	Zangi	32.5	6.49	4.22	493.41	0.14	536.79	Considerable

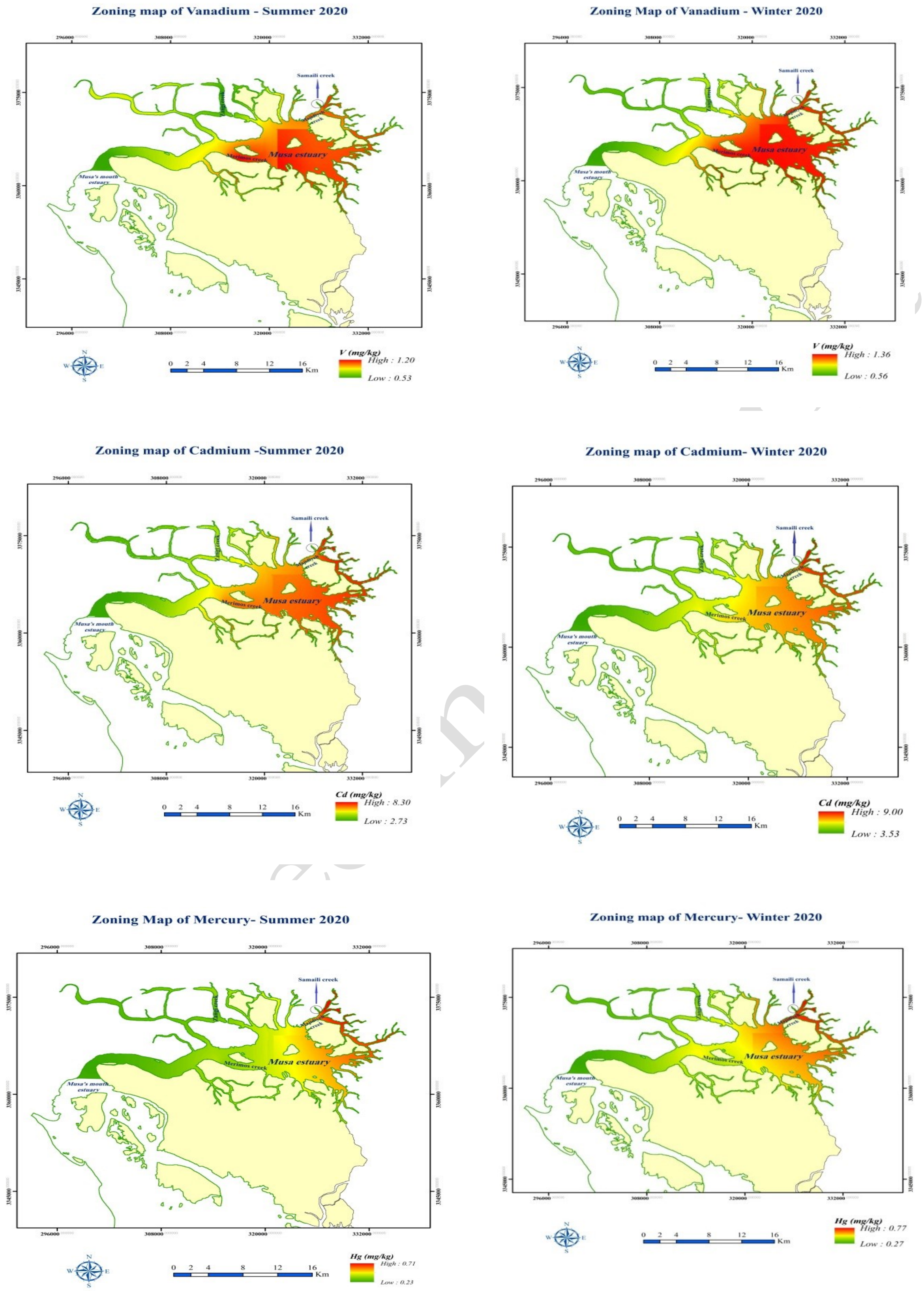
	Merimos	40.66	5.18	2.65	652.5	0.25	701.25	Very high
	Majidieh	67.08	10.25	5.13	790.83	0.26	873.56	
	Samaili	55	8.16	6.09	695.83	0.26	798.21	
	Musa's mouth	22.66	4.5	1.12	273.33	0.12	301.74	Considerable
Winter	Zangi	42.66	6.36	4.10	506	0.15	579.66	Considerable
	Merimos	48.33	5.29	2.60	624.16	0.27	649.47	Very high
	Majidieh	72.83	11.20	6.3	875	0.29	965.62	
	Samaili	74.33	8.23	7.31	790.83	0.3	881.01	
	Musa's mouth	27.33	4.12	1.23	353.33	0.12	386.15	Considerable
Very high	High		Moderate		Low			







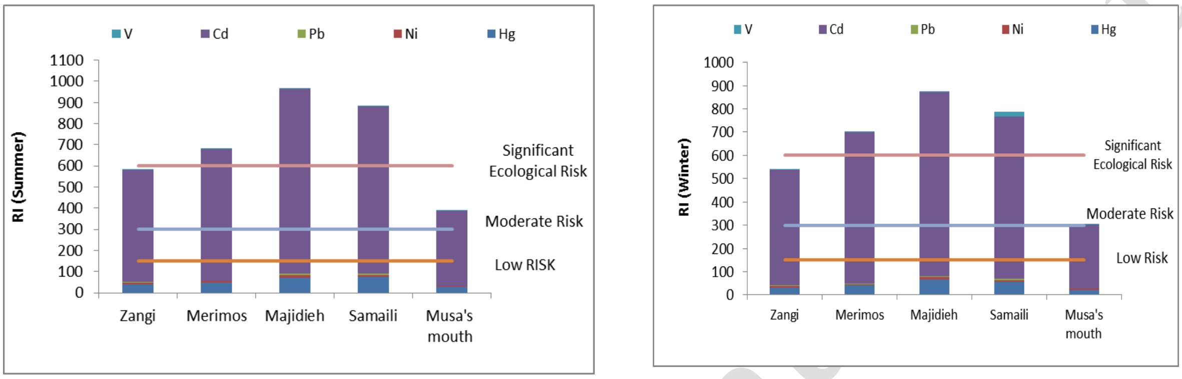




Summer | Winter

Figure 1. Zoning of heavy metal concentrations in the Musa Estuary

Figure 3 shows RI results of sediments in summer and winter. Cd had the largest contribution to RI in both summer and winter. The index values were higher in winter than in summer in all stations. Except for the control point and the Zangi tributary, a very high ecological risk was observed in Marimos, Majidiyeh and Samaili tributaries.



a. b.

Figure 3. Results of RI index in Mahshahr estuaries in summer and winter 2019

a. summer b. Winter

According to the results of the PLI, the sediment pollution load was higher in winter compared to summer. In Majidiyeh and Smaili estuaries, the index was non-polluted in summer whereas moderately polluted in winter. The lowest PLI was measured at the mouth of Musa Estuary, followed by Zangi Estuary (Figure 4).

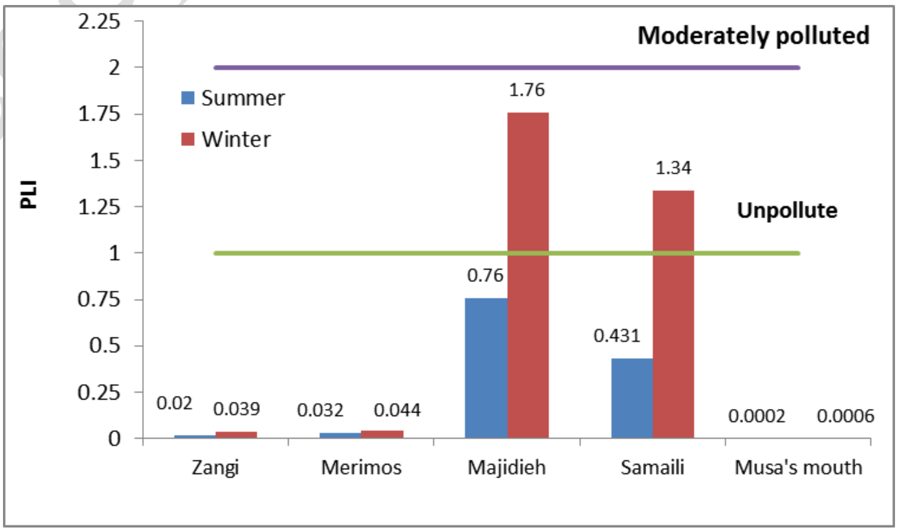


Figure 4. Comparison of pollution load index in Mahshahr Estuaries in summer and winter 2019

The NIPI ranged from 8.62 to 20.04 in summer and 9.15 to 21.53 in winter (Table 7), indicating that Mahshahr estuaries are at a high level of pollution (Figure 5).

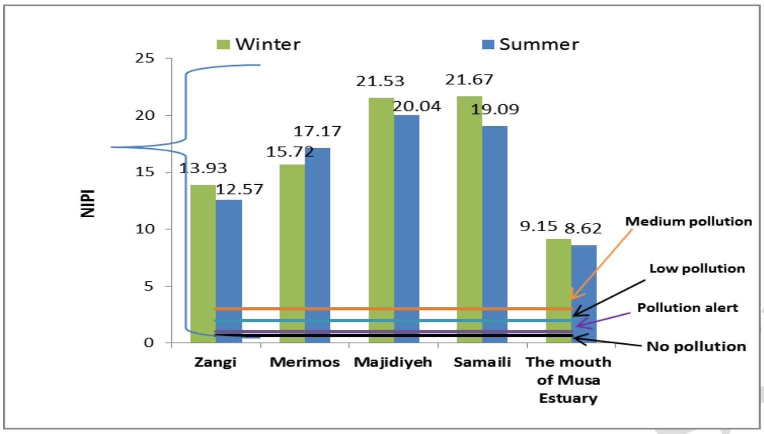


Figure 5- Results of NIPI in Mahshahr estuaries

Heavy metals are environmental pollution indicators and determining their levels and spatial distribution is essential to better understand the sources of pollution and their dangers to the environment and human health. The order of heavy metals concentrations was Ni> Pb> Cd> V> Hg. The concentrations of Hg, Ni, Pb and Cd in the sediments were notably greater than those of Shale: Hg of 0.4 mg/L, Ni of 50 mg/L, Pb of 20 mg/L and Cd of 0.3 mg/L)(European Union Standards 2002) which indicate human manipulation of the environment and anthropogenic sources of pollution. The high concentration of Ni and Cd can be attributed to oil and petrochemical-related operations in the region as they are found in large quantities of oil (Huang et al. 2020). Seifi et al (2019) emphasized the role of human industrial activities in sediment enrichment. Visual inspection of zone maps also confirmed high Cd and V concentrations in large parts of the estuary. The V concentrations, however, were below the standard shale value of 130 mg/kg (European Union Standards 2002) and were considered as a no-risk metal in Mahshahr estuaries. The presence of Pb in the environment is often associated with diesel and gasoline production in adjacent petrochemical facilities and maritime traffic because even unleaded gasoline products contain some lead (below 5 mg / L) (Wojciechowska et al. 2019). The results showed an insignificant increase in the concentration of heavy metals in winter compared to summer ($P < 0.05$) which can be attributed to an increase in the level and displacement of pollution in the low-slope end of the Musa Estuary. These higher winter pollution levels were also evident in the RI and Er seasonal results.

Tributaries with the highest Ni concentrations followed the order of Majidieh> Smaili> Zangi> Merimos such that three tributaries of Majidieh, Smaili and Zangi were represented by red in the Ni zone map. Also, NIPI categorized these three tributaries as the most polluted areas in both winter and summer. Another noteworthy point in the zone maps is that although Ni had a relatively high concentration in Mahshahr estuaries, its red zone had lower mobility and covered a smaller area as compared to Pb and Cd. Qeshlaqi and Rostami (2016) showed more than 50% of Pb and Cd compounds were related to the cation exchangeable form in the sediments of Siahroud riverbed whereas Ni was associated with the residual part of the sediments (stable from), causing it to be less mobile in the soil. A similar finding was also reported by Absieh et al (2019) in Mousa Estuary. The higher concentration of heavy metals in the Majidiyeh tributary can be due to its specific location at the end of the Musa estuary which receives high loads of upstream pollution transported by tidal currents (Absieh et al. 2019). The order of tributaries was Majidiyeh> Semaili> Merimos> Zangi for vanadium, Majidiyeh> Semaili> Merimos> Zangi for Cd and Semaili> Majidiyeh> Zangi> Merimos for Pb. In agreement with results, Manouchehr et al. (2008) reported lower levels of Cd and Pb in the sediments of the Zangi tributary as compared to other tributaries of Musa Estuary and coastal sediments of the northern Persian Gulf. Despite close adjacency to petrochemical facilities, another explanation for lower concentrations of heavy metals in Zangi tributary (colored green or blue in the zone maps with low and medium-range of pollution) is the high rate of water movement in the estuary which causes elimination and transport of pollution to other estuaries. The Merimos tributary, however, has limited shipping activities due to its shallow water depth and slow water flow (Taghizadeh et al., 2014) and probably resulted in a low concentration of heavy metals compared to other tributaries (Majidieh> Samaili> Zangi> Merimos> Musa). The high Cd and V levels in this tributary (represented by red in the zone maps) can be due to its adjacency to Petrochemical facilities and receiving water coming from the Zangi tributary. The concentration of heavy metals was found to be strongly correlated with pH and electrical conductivity. High levels of these two factors, especially alkaline pH, provide the necessary conditions for significant associations between metals and sediment particles (Louhi et al. 2012) which, in addition to natural conditions, confirms the high concentration of heavy metals in the Majidieh and Semali estuaries.

Another point that relates the concentrations of Ni, Cd, V and Pb to human activities is the high pollution levels of Majidiyeh and Semaili tributaries as export ports for oil, gasoline and petroleum products in which the chemicals are released into seawater during loading and stored in bed sediments. Bastami et al. (2015), Pourang et al. (2005) and Mohammadi Golangesh et al. (2015) reported oil pollution, discharge of ballast tank water and oil as the causes of high Ni and V levels in sediments. In Majidieh tributary, the mean concentration of Hg was found to be 410 ng/g by Babadi et al. (2015) and 99-490 ng/g by Mori Bazofti et al. (2017). The higher mean Hg concentration in this estuary (0.67 mg/kg) compared to that of the Zangi (0.32 mg / kg), Merimos (0.40 mg / kg) and Samaili tributaries (0.55 mg / kg) indicates increasing Hg concentration over time and corroborates previous findings.

In the studies of Mori Bazofti et al. (2017) and Goudarzi et al. (2006), the petrochemical estuary (one to the tributaries of the Musa Estuary) was implicated as the source of high Hg concentration in the region. Moreover, the Hg concentration was high in its adjacent tributary of Merimos due to having shallow waters and very limited shipping and dredging activities which cause further accumulation of heavy metals in the sediments and differing concentrations in the Zangi tributary.

According to Hg concentration, the tributaries were ordered Majidiyeh> Smaili> Merimus> Zangi. Babadi et al. (2015) found high Hg levels in sediments of Majidiyeh tributary. Faghiri et al. (2011) reported that repair, dyeing and washing of ships in this port are causes of increasing Hg levels in the Jafari Estuary (part of the Musa Estuary). Therefore, the high level of Hg in the Majidiyeh and Semail tributaries can be related to their extensive oil-export and ship maintenance activities.

In the study of Mori Bazofti et al. (2017), Petrochemical, Ghazaleh and Majidiyeh tributaries were categorized as mercury-contaminated estuaries due to being located at the end of Mahshahr estuaries where pollutants remain stable for relatively long terms which confirms the Hg concentration order of (Majidiyeh (29.05 mg/kg)> Smaili (22.92 mg/kg)> Zangi (17.58 mg/kg)> Marimos (14.08 mg/kg)> Mousa estuary (10.59 mg/kg)). The Majidiyeh tributary which is located at the end of the estuary had the highest level of pollution whereas Merimus tributary, which is located near the estuary mouth, had the lowest metal concentrations. A comparison of the heavy metals concentrations measured in this research with those of previous studies conducted in the Musa Estuary and other parts of the world is shown in Table 5.

Table 5. Comparison of sediment heavy metals concentrations in the present study with other studies conducted in the study area

Refrence	V	Cd	Pb	Ni	Hg	
This Study	0.64-1.31	4.93-7.91	10.61-29.25	51.8-112.02	0.32-0.74	**Mahshahr estuaries, Persian Gulf (mg/L)**
Manuchehri et al., 2008		2.21	3.71			**Zangi estuaries, Persian Gulf (mg/L)**
Faqyani et al., 2011					2.15	**Musa estuaries, Persian Gulf (µ g/L)**
Parvaneh et al., 2011		2.52	18.64	119.91	4.76	**Musa estuaries, Persian Gulf (µ g/L)**
Vaezi et al., 2014				62		**Musa estuaries, Persian Gulf (mg/L)**
Mori Bazofti et al., 2017					90-490	**Mahshahr estuaries, Persian Gulf (ng/L)**
Mohammadi Roozbahani et al., 2017			6.68			**Musa estuaries, Persian Gulf (mg/L)**
Pejman et al., 2014	119.47	0.85	5224	124.43	124.43	**Musa estuaries, Persian Gulf (mg/L)**
Jahangiri and , 2016 Janadeleh		0.23	7.73	34.05		**Northern Coast of the Persian Gulf (mg/L)**
El Tokhi et al., 2017	60	5.02	9	497.46		**Oman Beaches, United Arab Emirates(mg/L)**
Zarezadeh et al., 2017		2.63	7.94	80		**Coasts of Qeshm Island, Persian Gulf (mg/L)**
Mandeng et al., 2019		0.099-0.128	5.85-5.97	34.77-217.28	0.068-106	**Abiete-Toko goldKameron (mg/L)**
Silva et al., 2019		0.16-0.27	36.27-302.22	3.01-16.62	0.02-0.08	**Brazilian (mg/L)**

Except for the mouth of Musa Estuary in summer, RI and Er results indicated very high risk for Cd in the tributaries. The effects of human activities on pollution levels were season-independent with similar ecological risk status such that Merimos, Majidiyeh and Samaili tributaries had very high ecological risk and Zangi Estuary and the mouth of the Musa Estuary had considerable ecological risk. These findings can be rationalized considering the transfer of pollution from Zangi tributary to other parts of the estuary and also the distance of Musa Estuary from the pollution sources but an important point is the transport of pollution to the mouth of the Musa estuary as a non-petrochemical activity area. Based on the PLI results, the sediments of Mahshahr Estuaries showed no pollution in summer and winter except for Majidiyeh and Samaili tributaries which can be related to continuous dredging of bed sediments for shipping. In winter, however, the pollution level increases due to increasing sea-level rise and water flow rate in inner parts of the estuary, including Majidiyeh and Samaili tributaries. This interpretation can also be supported by the results of PLI. According to Tomlinson et al. (1980), PLI provides insufficient information regarding the conjoint effect of all pollutants (simultaneous presence of several metals). Therefore, due to the importance of sediments in habitat and nutrition provisioning, ecological indicators have been developed to evaluate the effects of these pollutants on living organisms. According to the ecological risk index, Zangi tributary showed considerable ecological risk Merimos, Majidieh and Smaili tributaries showed very high ecological risk. Despite the high water flow rate at the mouth of the Musa Estuary, this region showed moderate ecological risk which indicates the impact of this pollution on living organisms. Cd (very high risk) and Hg (low ecological risk and moderate ecological risk in other estuaries) had the highest ecological risk while V, Ni and Pb had the lowest ecological risk in Mahshahr estuaries. In a study in Musa Estuary, Janadeleh and Jahangiri (2016) showed that heavy metals had low ecological risk in the order of Pb> Cd> Ni> Zn in Musa Estuary. The NIPI was used to summarize the contamination status of the Musa Estuary. The index values were high, especially in winter, which indicates the significant impact of human activities on the estuary.

3.1Conclusion

According to the results of this study, tributaries were ordered as follows: Majidieh> Smaili> Zangi> Marimus> Musa estuary in terms of metal concentrations. Ni and Pb were also the most contaminating metals in the estuary. Moreover, the concentrations of heavy metals were season-independent. According to the ecological risk potential index, Cd had a high risk, Hg in the three tributaries of Merimos, Majidieh and Samaili had a medium risk and Pb, Ni and V had low risk. PLI contamination index was categorized as moderate in Majidieh and Samaili tributaries in winter and no-pollution in other tributaries in summer. The NIPI showed a high level of pollution in four tributaries of Zangi estuary, Merimos, Majidieh and Samaili and also at the mouth of Musa Estuary in both seasons. The findings of this research and those of previous studies indicated increasing levels of heavy metals content in the sediments of Musa Estuary. Due to the continuous discharge of these pollutants, seasonal changes had no significance on the metal accumulation and biological indicators.

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