Heavy metal enrichment in surface sediments from the SW Gulf of Mexico

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Abstract The south west coastal zone in the Gulf of Mexico is an area with great industrial and agricultural development, which experiences intensive prospecting and extraction of hydrocarbons. After running through industrial, agricultural, and urban areas, waters from both the Jamapa River and La Antigua River arrive here. The rivers' discharge areas of influence were estimated considering the textural and chemical composition of the supplied sediments. The main factors that determine sediment distribution were mineralogy, heavy minerals, carbonates, and anthropic contributions. The presence of metals in excess was evaluated using various pollution indicators, such as the enrichment factor, contamination factor, modified contamination factor, and geo-accumulation indexes. Data from different used contamination indexes show metal enrichments in As, Cu, Zn, Co, Cr, and V in La Antigua; As, Cu, and Cr in Jamapa; and As, Zn, and Pb in the Continental slope area. The adverse effects of metals on aquatic organisms were assessed using sediment quality guidelines that show Ni, As, Cu, and Cr may produce adverse effects on coastal areas. There was no evidence of contamination associated to the oil industry.

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Introduction

Metals are among the most toxic pollutants in aquatic systems. They are of great ecological significance due to their toxicity, resistance to degradation, and tendency to bioaccumulation (Manahan 2005). Dissolved metals in the water column are typically present at very low concentrations, but they may accumulate in the sediments, which act as potential sinks, depending on various factors such as the nature of the sediments (Horowitz and Elrick 1987), the properties of the adsorbed compounds, and the prevailing environmental conditions (Christophoridis et al. 2009). The different impacts that affect coastal areas, such as fluvial and atmospheric inputs, coastal erosion, industrial and urban activities, and marine transportation discharges from ships, can affect the environment quality or produce an increase in metal concentration in the coastal areas, which are of great importance because they host 25 % of the global ocean primary productivity and 90 % of commercial fisheries are present in this area (Crossland et al. 2005).

The geochemical composition of coastal marine sediments depends on source rocks composition, soils and sediments, the physicochemical characteristics of the depositional environment (pH, Eh, O₂, etc.), associated with natural biogeochemical processes (diagenesis,

adsorption/desorption on or from organic matter, precipitation-dissolution of Fe–Mn oxyhydroxides), and, recently, by inputs produced from anthropogenic activities (Selvaraj et al. 2010). When the collection of sediment cores to determine backgrounds and trace the history of contaminant delivery is not possible, several indicators can be used to distinguish if the origin of the metal in surficial sediments is natural or anthropogenic. It is necessary to study the ecological effects of metals in the aquatic media and assess metal provenance. Several indicators have been developed that provide an acceptable, accurate, and fast way to evaluate the conditions of the system. These indicators include enrichment factor (EF) (Abrahim and Parker 2008), degree of contamination (C_d) (Hakanson 1980), geo-accumulation index (I_{geo}) (Loska et al. 1997), and sediment quality guidelines (SQGs) (Long et al. 1995).

EF can be used to discriminate if the observed metal concentration in marine sediments is influenced by anthropogenic sources. A normalization procedure compensates for differences in grain size or carbonate content in the sediments. Different elements have been used for normalization (Al, Fe, Mn, Co) (Abrahim and Parker 2008). Al is widely chosen as a normalizing element because it is considered a proxy of the clay content in the sediments and is not expected to be enriched by anthropogenic sources. EF are obtained by comparing the metal concentrations in the study area of interest with those found in unpolluted reference zones. Different values have been used as baseline reference materials: (1) the average metal content on the upper continental crust (McLennan 2001), (2) the average metal content in shale (Christophoridis et al. 2009), and (3) the least contaminated sample in the study area (Feng et al. 2010) or local background concentration levels (Selvaraj et al. 2010).

The degree of contamination (C_d), developed by Hakanson (1980), is a method used to calculate an overall pollution factor, based on the concentration of seven metals and one organic contaminant. This index has the advantage of providing a general view of the studied area. Abrahim and Parker (2008) proposed the use of a modified degree of contamination (mC_d) based on the contaminant determinations available in each case.

The geo-accumulation index (I_{geo}) takes into account a factor that includes possible variations of the background values due to lithogenic characteristics. It allows for the study area to be mapped and to

differentiate sub-areas according to their pollution degree (Christophoridis et al. 2009).

Finally, SQG were developed because the total concentration of metal in the sediments did not provide sufficient evidence to estimate the potential impact on benthic organisms living in the sediment. SQG indicators use the guideline effects range—low (ERLs) and effects range—median (ERMs) to evaluate the potential effects of the concentration of metals in organisms (Long and Macdonald 1998; Long et al. 2006). ERL and ERM values were derived for nine trace metals (As, Cd, Cr, Cu, Pb, Hg, Ni, Ag, Zn). SQGs should only be applied to soft, aqueous sedimentary deposits of fine-grained material (NOAA 1999).

To evaluate the area of influence of terrestrial discharge, its distribution pattern in the coastal area, and the factors that characterize the sediments adjacent to the mouth of the rivers "Antigua" and "Jamapa", taking into account the adjacent shelf area, a geochemical study was conducted by collecting and analyzing surficial sediments from these areas. The impact of metal concentration was measured using several indicators that can discriminate excess metal contents and thus the impact of anthropogenic inputs.

Study area

The study area is located in 18°53′ to 19°43′ north latitude and 94°55′ to 96°30′ west longitude, in the southwest Gulf of Mexico, across the central basin region of Mexico. The study area covers approximately 1,471.4 km² (Fig. 1). La Antigua and Jamapa rivers discharge their waters into the Gulf of Mexico, which is considered a marginal sea because it is protected from the Atlantic Ocean by the Caribbean Arc.

The state of Veracruz, located in the central part of Mexico in the Gulf of Mexico side, is the third most populated state of Mexico (three of 32) with 7.643 million inhabitants. It ranks first in electrical energy production, and it has the only nuclear plant in the country. Veracruz leads the production of basic chemicals; it ranks second in the manufacture of iron, steel, and candy; and it is fourth in the extraction of oil and gas. It is the first sugar cane producer, citrus (orange, pineapple, lemon), and papaya and is the second coffee producer. It has the highest cattle production in the country (INEGI 2011). Due to the industrial, agricultural, and livestock development of

Fig. 1 Study areas (**a**). Bathymetry and sampling sites location, the continental slope area (**b**), La Antigua area (**c**), and the Jamapa area (**d**)
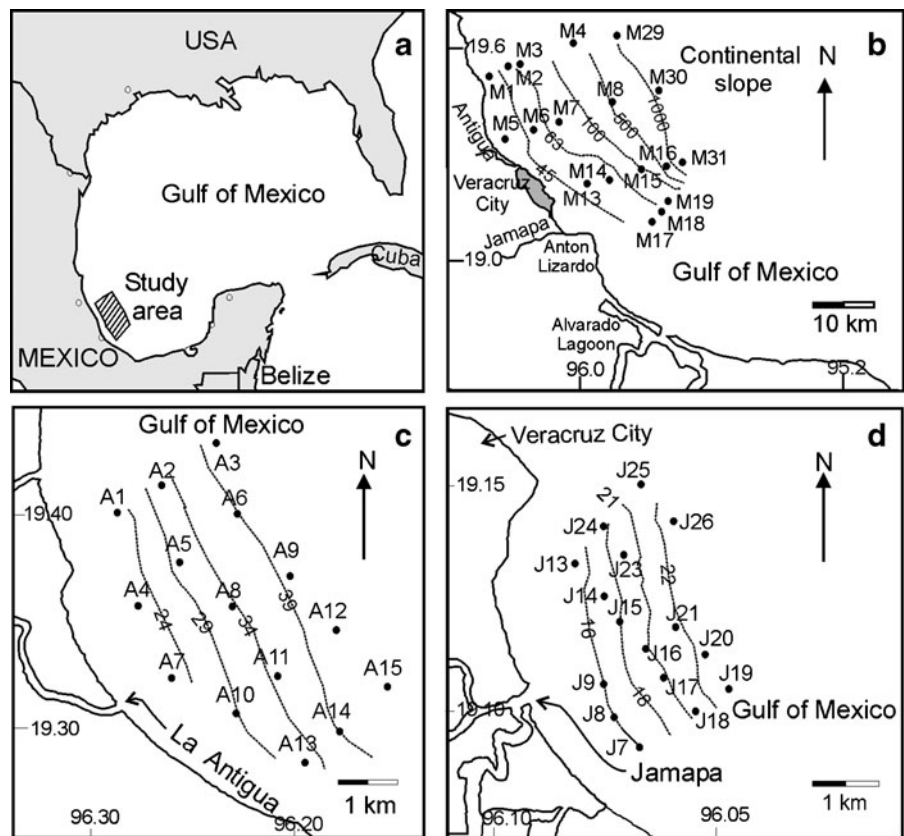
Veracruz area, an increase in human population has taken place, with the associated anthropogenic impacts. The study of the coastal area related to discharges from two of Veracruz's major rivers (Jamapa and La Antigua) may provide information of the different processes that take place in the continental area and which may impact the coastal zone.

The area off the coast of Veracruz experiences hydrocarbon prospection and exploitation; its geological conditions are favorable for the presence of significant volumes of liquid and gaseous hydrocarbon deposits. Several oil platforms exist in the area, which is also experiencing intense prospecting (Barbosa Cano 2007).

La Antigua River originates in the State of Puebla in the Sierra Madre Oriental at 3,350 m above sea level; its drainage basin covers an area of 3,443.9 km^2, with a mean annual flow of $1,789.66 \times 10^6$ m^3 (D.O.F. 2012). Along its watercourse, there are several dams. The river mouth is located 20 km northeast of the Port of Veracruz.

The Jamapa River originates from the slopes of Pico de Orizaba in the State of Puebla at 5,700 m above sea level; its drainage basin covers an area of 3,912 km^2, and it has a mean annual run off of $1,895 \times 10^6$ m^3 (Tamayo 2001). Eleven dams are located along the course of the Jamapa River, which leads to causing a decrease in the river flow and a reduction in the input of sandy sediments to the marine environment, the river mouth is located in the area known as Boca del Rio in the southwestern Gulf of Mexico.

Both basins are dominated by Quaternary soils, sedimentary rocks, mainly conglomerate, limolite, and volcanic igneous rocks (SGM 2005). The study area was divided into three regions (Fig. 1). An equidistant grid was chosen in two of these regions that covered the coastal area close to the discharge mouths of La Antigua River and the Jamapa River, respectively, whereas the last section corresponded to the continental slope area.

Materials and methods

Data and the sampling were carried out during the rainy season on August 2008 using the R/V Justo

Sierra. Fifteen surface sediment samples were collected in each of one the coastal areas located across La Antigua and Jamapa Rivers mouths (Fig. 1c, d). Samples were collected at depths varying from 15 to 44 m, with a gradual increase in depth following isolines parallel to the coastline. Eighteen sediment samples were collected in the continental shelf area in front of the Jamapa and La Antigua River coastal areas, in depths ranging from 30 to 1,608 m (Fig. 1b).

Sediment samples were taken using a Smith MacIntyre dredge. The sediments samples were collected in a plastic bag and stored at 4 °C until analysis. The particle size distribution of the samples was measured using a laser diffraction analyzer Coulter model LS-230. Five grams of homogenized sediment sample was mixed with 50 ml of dispersant solution of sodium hexametaphosphate (100 g/L). The mixture was allowed to stand for 24 h, after which it was stirred and a small portion was taken for analysis. Sediment samples were dried at 55 °C for 48 h and homogenized in a SPEX mill 8000. Organic carbon content was determined by oxidation with $K_2Cr_2O_7$, and a reverse titration was performed to remove excess $K_2Cr_2O_7$ not consumed in the oxidation of the organic matter (Gaudette et al. 1974). The carbonates were evaluated using a technique proposed by Hesse (1971), which uses a reverse titration of an excess of HCl that did not react with the carbonate and a solution of sodium hydroxide. Major elements were analyzed by glass bead preparation of the samples by a 1:1 sample: fusing ($LiBO_2/LiB_4O_7$, ultrapure grade), using a spectrophotometer Siemens SRS 3000 XRF. The accuracy of the method was evaluated using the standard AGV-1 and was 100 ± 2 % for all the oxides, except for MnO and P_2O_5 where values were 93 and 106 %, respectively. The coefficient of variation was between 0.2 and 1.5 % except for P_2O_5 that presented values of 3.79 %. The trace metals analysis was performed by the extraction of 50 mg of sample with a 10:1 ratio of HF/HNO_3 (distilled three times) mixture, in a Teflon vial. The vial is capped and heated at 115 °C for 24 h; after this time, a small amount of $HClO_4$ was added, and the mixture was taken to dryness; the evaporated residue was dissolved with 25 ml of 2 % HNO_3 and let stand for 24 h before the analysis by means of an ICP-MS Agilent 7500ce. The accuracy was evaluated using the standard MESS-3. The percentage of recovery was V 98.7 %, Cr 104.2 %, Co 99.9 %, Ni 106.3 %, Cu 106.1 %, Zn 98.9 %, Cd 41.7 %, Pb

107.5 %, and As 119 %, and the variation coefficient was V 3.0 %, Cr 3.0 %, Co 4.0 %, Ni 6.0 %, Cu 4.0 %, Zn 3.0 %, Cd 3.0 %, Pb 2.0 %, and As 3.0 %. The software used for statistical analysis was Statistic 6.

Results and discussion

The textural characteristics of sediments from the studied area show the highest mud content and the lowest sand content in the continental slope area (Fig. 2). This is associated with the depth of the area, where due to the size of the water column the energy of the sea floor is low and allows the deposition of fine particles. The sediments of the coastal area adjacent to La Antigua River mouth present higher content of mud than sediments near the mouth of the Jamapa River; this is associated with the characteristics of each of these basins, and the coarse sediment retention was due to the dams located in the main course of the rivers. The Dam "Puente Nacional" at 19.342° latitude and −96.4222° longitude is located at 16 km from La Antigua River mouth, retaining coarser sediment particles. The dams in the Jamapa River basin are located in secondary river branches not in the main stream; therefore, the retention of coarse sediments is not so high. When strong winds affect the area, the shallow sediments may receive high wave energy and be
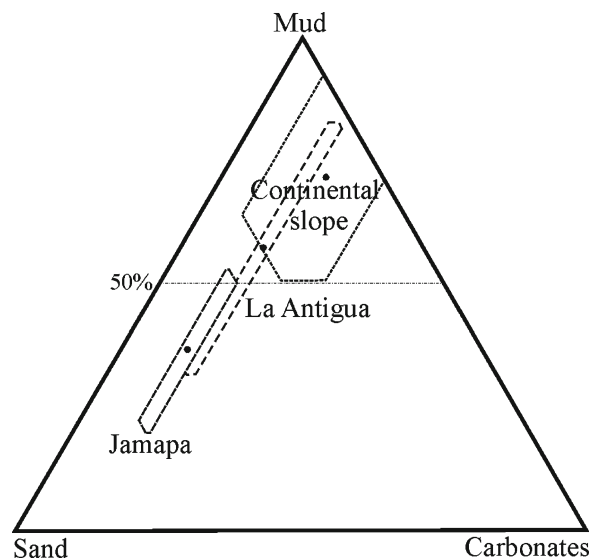

Fig. 2 Triangular diagram with sand, mud, and carbonate poles. The polygons are constructed from the average with one positive and one negative standard deviation

mixed; in consequence, the depth of the studied areas may be also an important factor; in La Antigua, the depths varied from 22 to 43 m; and in Jamapa area, the depth ranged from 15 to 25 m; when strong winds affect the area, the shallow sediments will be mix. This is supported by the distribution pattern of sediments in La Antigua River coastal area (Fig. 1c), where the sediments closest to the shore showed the highest sand content A1 (84.3 %), A4 (86.5 %), A7 (71.2 %), and A13 (77.4 %), compared with the average percentage of sand observed in the area (32.8 %). Differently, station A10 is characterized by a low sand content (11.1 %) and a high percentage of mud (88.8 %), suggesting that this area is influenced by the mud (silt and clay) discharges from La Antigua River. The high values of fine sediment composition at stations A14 and A15 with mud contents of 95.7 and 85.2 %, respectively (Fig. 1c), suggest that the sediments carried by the river move in a southeast direction at this time of the year, which corresponds to the rainy season in the river basins.

The mud content in the Jamapa River coastal area was higher than average (41 %) (Table 1) in stations located in the southern part of the mouth of the River (Fig. 1d) located in stations J9 (72.0 %), J8 (75.2 %), J16 (56 %), J17 (48 %), J18 (53 %), and J19 (54.4 %),

while in the north of the river mouth, the sediment samples were predominantly formed by sands with values higher than the average (55.7 %), located at sampling points J13 (87.9 %), J14 (78 %), J15 (64.8 %), J23 (65.3 %), J24 (81.1 %), and J25 (69.9 %).

Carbonate content in the three areas presented great variations (Fig. 2). The higher average carbonate concentration (29.5 %) was found in the continental slope area; values of 16 to 24 % were observed, except for a few sites with values exceptionally high (38 to 78 %) at M18, M19, M15, M17, and M14. These high concentrations suggest the presence of an ancient carbonate bank, associated with a lower sea level, and to the presence of the VRS (Veracruz Reef System) (Rosales-Hoz et al. 2008). The range of carbonate concentration in La Antigua area varies from 13.48 to 20.28 %. All stations close to the coastline (Fig. 1c) A1, A4, A7, and A13 showed carbonate values below or close to the average value of 17.12 % observed in the area. The carbonate content in the Jamapa area was between 9.54 to 17.14 %; sampling sites with values above the average (13.67 %) were located (Fig. 1d), to the north of the river mouth, with the highest value located at sampling point 25 (17.14 %), which is close to the reef banks present to the north of the area.

Table 1 Range and average concentration of texture and major elements in sediments from the studied areas

Units	La Antigua		Jamapa		Continental slope	
%	Range	Average	Range	Average	Range	Average
Sand	22.00–43.53	33.56[a]	24.74–87.99	59.00[a]	0.14–85.20	13.97[b]
Silt	10.97–72.70	51.85[a]	9.08–60.50	33.82[a]	11.47–75.09	60.80[b]
Clay	2.52–24.04	15.29[a]	2.82–15.06	7.17[a]	3.24–35.23	20.79[b]
Mud	13.49–95.73	67.15[a]	12.00–75.25	40.99[a]	14.70–99.80	81.52[b]
Org. C.	0.35–1.63	0.64[a]	0.36–0.94	0.53[a]	0.08–0.89	0.56[b]
Carb.	13.48–20.28	17.12[a]	9.54–17.14	13.67[a]	16.09–77.82	29.48[b]
Al_2O_3	10.54–13.49	11.58	10.59–14.97	12.85	5.24–13.42	11.02
SiO_2	48.56–55.97	52.86	51.45–56.54	53.26	20.20–53.78	45.76
Fe_2O_3	4.21–6.59	4.97	4.03–6.28	5.40	2.62–5.21	4.26
TiO_2	0.64–1.17	0.78	0.60–1.00	0.83	0.25–0.65	0.56
MnO	0.05–0.10	0.07	0.07–0.08	0.07	0.05–0.31	0.12
CaO	8.39–11.23	10.00	7.59–10.65	9.23	8.79–33.46	13.69
Na_2O	2.42–3.17	2.72	2.58–3.51	3.06	1.47–3.58	2.65
K_2O	1.54–1.98	1.80	1.43–1.79	1.61	0.77–2.28	1.85
MgO	1.80–2.63	2.10	2.28–2.68	2.44	1.78–2.68	2.19
P_2O_5	0.10–0.23	0.17	0.07–0.20	0.16	0.09–0.19	0.14

[a] $n = 15$ sediment samples
[b] $n = 18$ sediment samples

The average values of organic carbon in the three areas were very similar. However, at some specific sites, high values are observed, as in La Antigua, where the high value observed at A10 (1.63 %), and its location in front of the river mouth suggests that this value is associated with La Antigua River discharge. A similar situation was observed in the Jamapa River area; sampling points with values above the average (0.53 %) were located close to the river mouth: J8, J9, and J16. In the continental slope area, higher values of organic carbon were located toward the deepest parts of the area, in station M16 (0.89 %). Very low organic carbon values (0.11 %) were found in the area of the slope where stations 15 and 19 were located, the slope break is very strong, and there is a lack of organic matter deposition in these areas.

The chemical analysis of major elements (Table 1) in the three areas showed the highest average values of Al_2O_3, SiO_2, Fe_2O_3, TiO_2, Na_2O, and MgO in Jamapa; this may be related to the higher sand content present in this area. Sampling points J7, J8, and J19, which are located across the river mouth, present values above the average for these oxides. In La Antigua area, values above the average for Al_2O_3, Fe_2O_3, TiO_2, and MnO_2 were found at station A10, which, based on its low sand content, receives the greatest amount of sediments from La Antigua River discharges. The elements associated with heavy minerals, such as Fe_2O_3, TiO_2, MnO, and MgO, were the highest at station A1, which is located to the north of the river mouth, near the coast. Similar data results were found by Rosales-Hoz et al. (2005) in the Panuco River coastal area, situated 363.6 km north of La Antigua River mouth, where heavy minerals are located near the coastline.

The concentration ranges found for trace metals are presented in Fig. 3. The highest average concentration for V, Co, Cu, Zn, and Cd were found in La Antigua area (Fig. 3a). The highest average concentration for Cr and Ni were found in Jamapa area (Fig. 3b), and the highest average concentration for As and Pb was found in the continental slope area (Fig. 3c). Similar metals ranges were found in La Antigua and Jamapa coastal area, with lower values in the continental slope area. The coastal areas of Jamapa and La Antigua present at some specific sites metal concentrations values above the average.

The data from La Antigua showed that the highest values of V, Cr, and Ni were present in A1 and A7
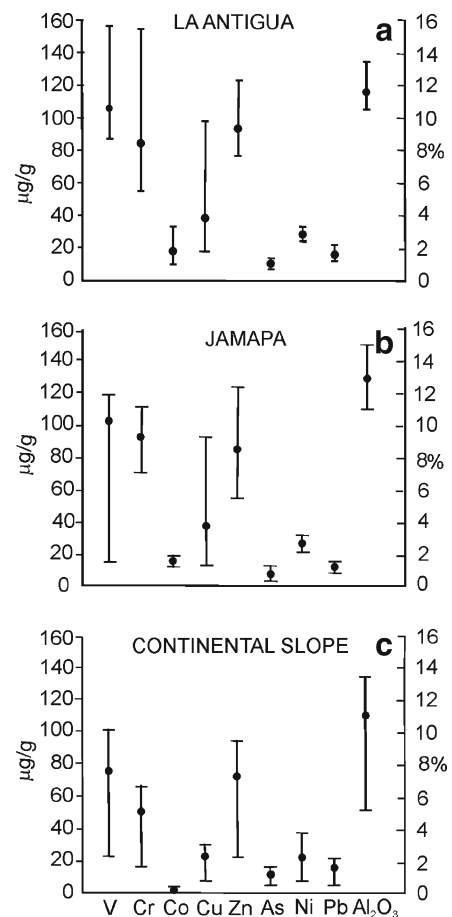

Fig. 3 The average, maximum, and minimum metal concentration (in milligrams per kilogram) and Al_2O_3 (in percent) in surface sediments from: La Antigua area (**a**), the Jamapa area (**b**), and the continental slope area (**c**)

stations, which are the sites where the highest concentrations of Fe_2O_3, TiO_2, MnO, and MgO; this may be due to the influence of source rocks characterized by basic and intermediate volcanic rocks, composed by heavy minerals; this was observed in thin sections. Among them were found magnetite (Fe_3O_4) and ilmenite (Fe, Mg, Mn (TiO_3)) that may also contain Cr, pyroxenes, and olivine (Klein and Hurlbut Jr 1997; Rollinson 1993). Magnetite analyses may show considerable amounts of Mg and Mn substituting for Fe^{2+} and Al, Cr, Mn^{3+}, and Ti^{4+} substituting for Fe^{3+}, which may contain V as an associated element (Rollinson 1993).

High values of Al_2O_3 and Fe_2O_3 were measured in sites M29, M30, and M31 located in the deepest part of the study area, which is formed mainly by mud (Fig. 2). In these sites, high concentrations of V, Ni, Cu, Zn, and

Pb were observed. On the other hand, the site M17 presents the lowest trace metal values and has a high concentration of sand and carbonates, suggesting that carbonates are probably diluting the trace metal content. A comparison of the values obtained in the coastal area in front of the Jamapa River mouth with values from the Jamapa River in the continental area (Rosales-Hoz et al. 2007, 2008) showed an enrichment of carbonates, Ca, Cr, Cu, and Zn in the coastal area.

To analyze how geochemical, textural, and bathymetric characteristics influence the distribution of sediments in the three areas studied, a cluster analysis was performed using the data from all the areas. The cluster obtained showed the presence of four distinct groups (Fig. 4):

Group 1: formed by sites M29, M30, and M31 from the continental slope area (Fig. 5c), their samples present very low sand values (0.51 %) and the highest average concentration of clay (28.6 %), silt (70.9 %), Al_2O_3 (16.2 %), Fe_2O_3 (6.2 %), MnO (0.30 %), Na_2O (4.1 %), V (94.8 mg/kg), Ni (32.7 mg/kg), As (12.3 mg/kg), and Pb (19.9 mg/kg). The stations in this group are the deepest in the studio with a depth range from 276.4 to 1,301.6 m.

Group 2: consisted of stations M17 and M14 from the continental slope area (Fig. 5c). It presents the highest average concentration of carbonates (48.7 %), CaO (37.8 %), and MgO (3.6 %). It had low average content of trace and major elements as Al_2O_3 SiO_2, TiO_2, and Fe_2O_3.

Group 3: formed by stations A3, A10, A6, A12, A9, A15 from La Antigua area (Fig. 5a) and stations M6, M7 M3, M2, M13 (Fig. 5c) from the continental slope area. This group is mainly characterized by intermediate depths (30–77 m), greater amount of fine materials (suggesting that the influence of the winds that move the seabed and water column are low), intermediate carbon organic concentration (0.55 and 0.71 %), carbonates (14 and 23 %), and trace elements. Al_2O_3 and K_2O are above the average suggesting a greater amount of clay minerals.

Group 4: formed by all the samples studied in Jamapa coastal area and four sampling points A1, A4, A7, and A13 from La Antigua area. Samples from La Antigua (Fig. 5a) are characterized by higher sand (71–86 %) and carbonate (15–17 %) concentrations. Samples J13, J24, and J25 from Jamapa characterized by high sand (70–88 %) and carbonate (14–17 %) concentrations are grouped differently from all others samples analyzed from Jamapa (Fig. 4).

A factor analysis was performed to evaluate the main parameters that control the spatial distribution

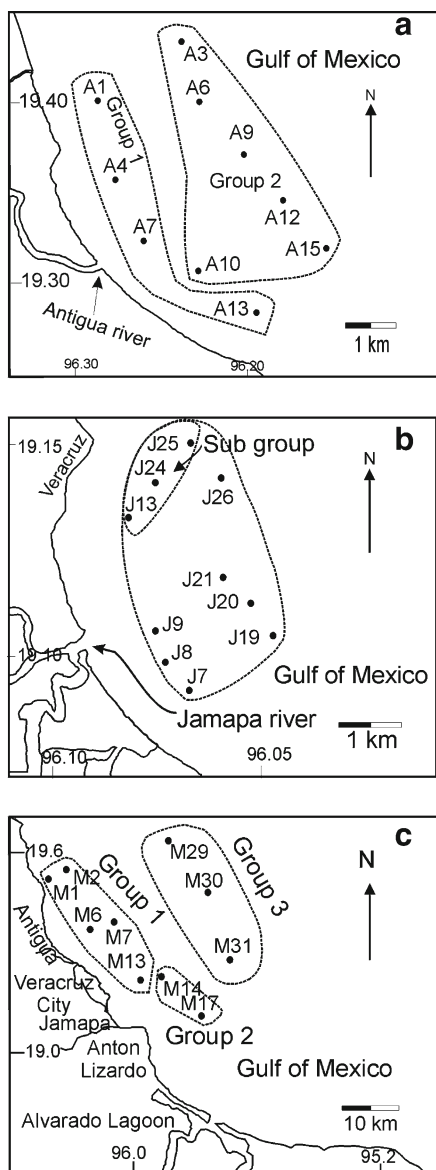**Fig. 4** Cluster analysis from the studied area
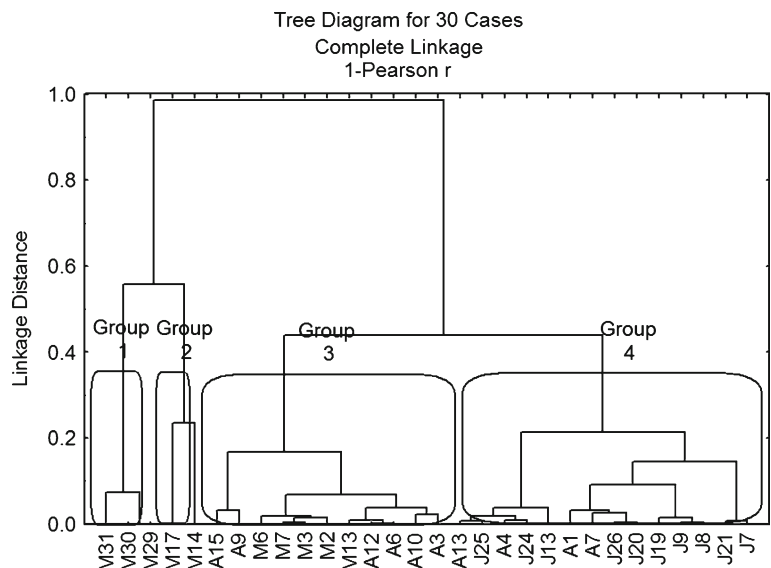
Fig. 5 Distribution of groups according to the Cluster. La Antigua area (**a**), the Jamapa area (**b**), and the Continental slope area (**c**)

of the sediments in order to identify the main causes of variation among the samples (Duan et al. 2010). The factor analysis of the three areas is shown in Table 2. The data show that the system variance can be explained by up to four factors related to sediment texture, clay mineralogy, heavy minerals, carbonate content, and anthropogenic influence.

In La Antigua, Jamapa, and the continental slope area, the grain size is one of the main factors that characterized sediment distribution. The size debris

are associated, among other factors, with the influence of bathymetry, river discharges, and the type of minerals which in turn depends on the source rocks. Depending on the mineral density and the dynamics present in the area, the sediments will be deposited at specific sites. In La Antigua, the predominant type of sediment texture was silty-clays and silty-sands, with a few samples formed mainly by sands. In Jamapa, sands and sandy-silts were predominant, while in the continental slope silty-clays were the dominant sediments.

Fine suspended sediment particles allow for a greater adsorption of metals in the water column to take place (Duan et al. 2010, Zhang et al. 2009, Reddy et al. 2004, Brayan and Langston 1992, Horowitz and Elrick 1987). Sediment texture (factor 1) shows positive interactions with depth, silt, clay, organic carbon, and some trace metals. The sediment texture plays an important role in the variability of trace metals found in these areas. The proportions of fine particles, in addition to the size of some minerals that most likely contain some of the trace elements studied, are very important factors in metal distribution in the sediment samples.

The minerals that constitute the sediments (factor 2) play an important role in the distribution of sediments in the area. Trace metals have a greater or lesser affinity to associate with the crystal structure of the different minerals that make up the sediment. In the three areas, studied mineralogy plays a significant role on sediment distribution.

In La Antigua area, the volcanic lithics and the heavy minerals content (factors 2 and 3) were an important factor in sediment distribution. Meanwhile, in Jamapa and in the continental slope area, the carbonate content of the sediments (factor 3) was a decisive factor in the sediment distribution pattern in the area.

Finally, the absence of significant associations between any of the parameters studied and Cu or Zn would suggest that these metals most likely come from human activities. Therefore, this factor (factor 4) found in La Antigua and Jamapa coastal areas is believed to be associated with anthropogenic influence.

Enrichment factors EFs were evaluated in the areas studied using the metal content in the upper continental crust (McLennan 2001). According to Birth (2003), using the average metal concentration in the crust as a

Table 2 Factor analysis for the different areas studied

Factor 1		Factor 2		Factor 3		Factor 4	
Sed. texture		Clay minerals		Heavy minerals		Anthropic	
Depth	0.94	CO_3	0.78	Fe_2O_3	0.60	Cu	−0.94
Sand	−0.94	Org.C	−0.89	TiO_2	0.60	Zn	−0.68
Silt	0.92	SiO_2	0.73	V	0.86		
Clays	0.95	Al_2O_3	−0.83	Cr	0.60		
TiO_2	−0.76	CaO	0.78	Ni	0.96		
Fe_2O_3	−0.68	Na_2O	−0.74				
MnO	−0.94	P_2O_5	−0.73				
MgO	−0.83						
K_2O	0.97						
Cr	−0.79						
Co	−0.63						
As	0.67						
Cd	0.73					La Antigua	
Pb	0.99						
Exp.Var.	11.49		5.25		3.96		2.50
Prop. total	0.459		0.21		0.15		0.10
Sed. texture		Heavy minerals		Carbonates		Anthropic	
Org. C	0.76	SiO_2	−0.89	Depth	0.82	Cu	0.92
Sand	−0.95	TiO_2	0.79	CO_3	0.78	Zn	0.92
Silt	0.95	Al_2O_3	0.70	Na_2O	−0.75	Cd	0.60
Clays	0.87	Fe_2O_3	0.81	Co	−0.70		
Al_2O_3	0.60	MnO	0.91	As	−0.64		
CaO	−0.60	MgO	0.75				
K_2O	−0.60	P_2O_5	0.69				
V	0.67	V	0.63				
Cr	0.79						
Ni	0.68						
Cd	0.69					Jamapa	
Pb	0.82						
Exp. Var.	8.09		6.28		3.68		3.49
Prop. total	0.324		0.25		0.147		0.14
Heavy minerals		Sed. texture		Carbonates			
Depth	0.85	Org. C	0.74	CO_3	−0.79		
Clay	0.82	Sand	−0.62	SiO_2	0.93		
Al_2O_3	0.78	Silt	0.67	TiO_2	0.79		
Fe_2O_3	0.91	V	0.77	MgO	−0.86		
MnO	0.95	Cr	0.88	CaO	−0.84		
Na_2O	0.91	Co	0.61	As	0.62		
K_2O	0.71	Cu	0.81				
Ni	0.72	Zn	0.81				
		Cd	0.82			Continental slope	
		Pb	0.68				
Exp. Var.	7.90		7.62		6.73		
Prop. total	0.31		0.30		0.26		

reference, the EF values obtained indicate the following conditions: (a) EF ≤1 indicates no enrichment, (b) EF=1–3 indicates minor enrichment, (c) EF=3–5 indicates moderate enrichment, (d) EF=5–10 indicates moderately severe enrichment, (e) EF=10–25 indicates severe enrichment, (f) EF=25–50 indicates very severe enrichment, and (g) EF ≥50 indicates extremely severe enrichment. The use of the upper continental crust as a reference value allows comparison with the values obtained in the studied areas, as well as with the EF reported in other coastal areas. However, the high EFs obtained for As using the upper continental crust as a reference (average 8.67 in La Antigua, 7.05 in Jamapa, and 9.68 in the continental slope area) made necessary the use of background metal concentration in order to clarify the observed As enrichment values obtained.

Metal concentration from 113 sites (Table 3) located in the coastal area of interest were used as background reference; 28 samples were collected at three different times of the year, 3 km off the coast from Veracruz harbor and Jamapa River mouth; 15 samples were collected at two times of the year in Bahia Vergara located north of Veracruz Harbor (Rosales-Hoz et al. 2007; Celis-Hernández 2009; Ceron-Alvarado 2008).

The EF values obtained using the two reference values are similar for the studied metals, except in the case of As, where EFs obtained using background As values were 0.99 in La Antigua, 0.81 in Jamapa, and 1.11 in the continental slope. Using data from the upper continental crust as a reference, EFs As values 8.7 times the value obtained using the background reference were observed in all the areas, which suggests that As enrichment observed is related to the lithology present in the area.

Using metal values from the continental crust as a reference, all the metals from the area present minor enrichments on the average (Fig. 6), except As that presents a moderate severe enrichment. The range of EFs values in La Antigua area (Fig. 6a) is larger than in Jamapa (Fig. 6b) and the continental slope area (Fig. 6c).

All analyzed metals in La Antigua, except for Ni, had EFs average values greater than one (Fig. 6b) that corresponds to a minor enrichment which is most likely associated with natural or anthropogenic sources, according to Hung and Hsu (2004). Arsenic concentration corresponds to a moderate severe enrichment. Some specific sites show higher enrichment, site A15 presents moderate severe enrichment of Cu, and high minor enrichment of Zn and Co. This site is associated with the fine material supplied by La Antigua River (Fig. 1c). Site A1 presents the highest minor enrichment of V, Cr, and Ni. It contains high concentration of TiO_2, Fe_2O_3, MnO, and MgO that were abundant in this area. Site A3 presents the highest minor enrichment in the area of As, Cd, and Pb; the sediments at this site are 98.5 % mud.

Average EFs in Jamapa (Fig. 6a) are lower than in La Antigua. As presents moderate severe enrichment. Cu, Zn, V, Cr, and Co present minor enrichment. Ni, Cd, and Pb show no enrichment. Site J21 shows moderate severe enrichment of Cu and minor enrichment of Zn; this site is associated with fine materials supplied by the river. Site J9 presents the highest minor enrichment of V and Cr; the location of this site (Fig. 1d) suggests that these enrichments are related to river discharges.

The average EFs for the continental slope area (Fig. 6c) were the lowest. Only Pb and As had higher

Table 3 Metal concentration in the continental crust and average metal values from the coastal area (in milligrams per kilogram) used as reference to evaluate contamination indices

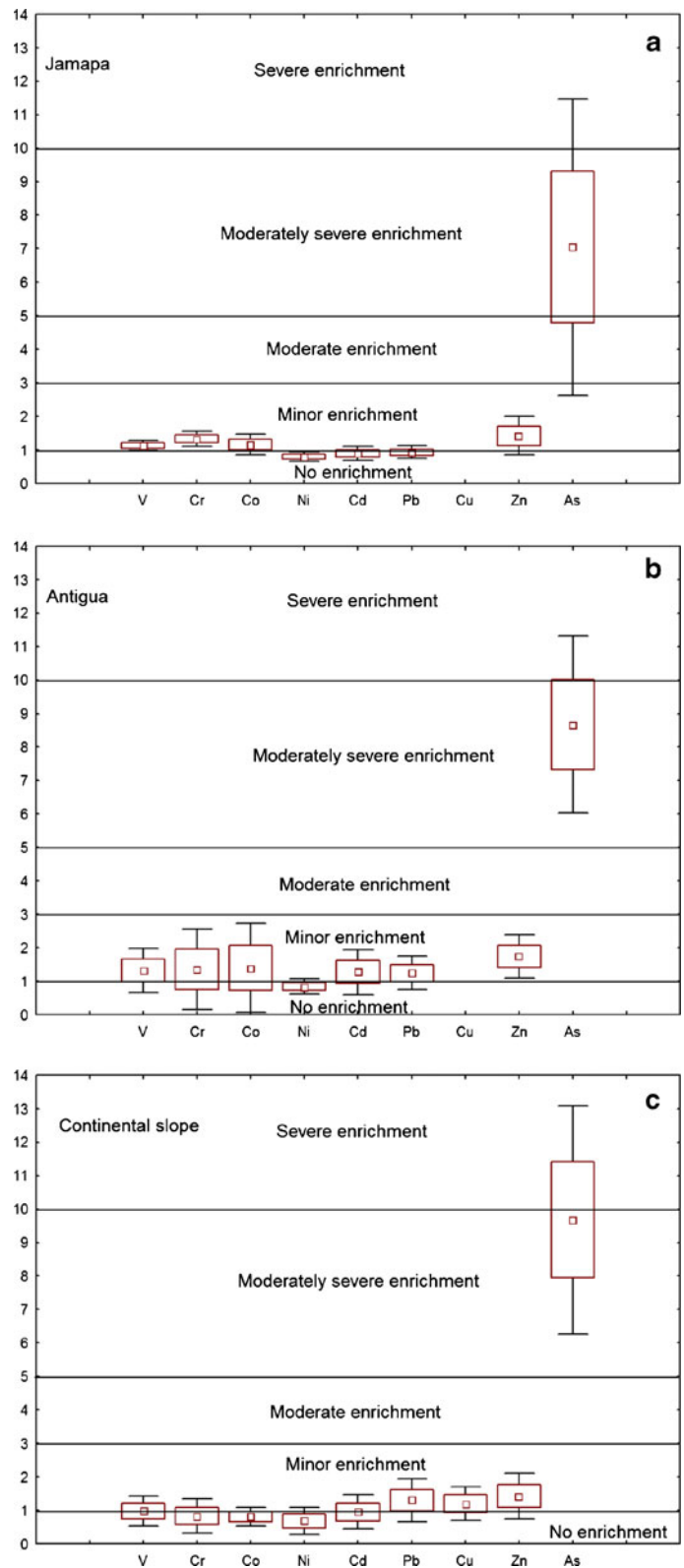|	Al	V	Cr	Co	Ni	Cu	Zn	As	Cd	Pb
Upper cont. crust[a]	80438	107.0	83	17	44	25	71	1.5	0.1	17
Background conc. levels[b,c,d]	40337	101.9	66.6	14.7	30.8	15.6	60.6	9.7	0.1	12.4

[a] McLennan (2001)

[b] Rosales-Hoz et al. 2007

[c] Celis-Hernández 2009

[d] Ceron-Alvarado 2008

Fig. 6 Enrichment factors for metals in each area. La Antigua area (**a**), the Jamapa area (**b**), and the Continental slope area (**c**)

average values than in the other areas studied. Sampling point M13, located in front of the Jamapa River discharge area, presented high concentrations of mud, and it had the highest EFs corresponding to a minor enrichment for Zn, Pb, Cu, Cd, V, and Cr. The location of this site (Fig. 1b) suggests an association to river discharges.

The fact that some of the metals studied were found enriched in the sediments from the three areas suggests anthropogenic inputs from the mainland. It has been reported (Sadiq 1992; Reimann and Caritat 1998) that the use of pesticides, herbicides, and fungicides in farming are most likely responsible for the presence of As, Cu, and Zn. Additionally, Cu and Zn are related to wastewater discharges, which may originate from the city of Veracruz. On the other hand, Zn can be attributed to activity in the port of Veracruz because it is used on boats and port infrastructure to prevent corrosion generated by seawater (Reimann and Caritat 1998). Pb has been associated with incineration of garbage, sewage discharges, coal combustion, traffic, metallurgy, welding, etc. (Brayan and Langston 1992). These activities are carried out in the city of Veracruz and the surrounding areas. V, Co, and Cd have been linked to the burning of coal, geogenic dust, and the metal industry. Cd and Co have been linked to fertilizers, sewage discharges, trash incineration (Reimann and Caritat 1998), contributions from farming, and fuel burning by small boats and power plants.

Degree of contamination (C_d) In the present study, the contamination factor (C_f) values were calculated using the data from ten samples from each area for the studied metals (V, Cr, Co, Ni, Cu, Zn, As, Cd, Pb). C_f was evaluated using the two reference materials from Table 3. The C_f values obtained from the two references were very similar in all the cases, but were higher using metal concentration values from the coastal area as a reference. In order to compare the C_f values obtained in the present work with values from other coastal areas, C_f and C_d values obtained using the upper continental crust are used (Table 4).

C_f values from La Antigua and Jamapa are very similar for V, Cr, Co, and Ni; La Antigua coastal area presents higher C_f values for Cu, Zn, As, Cd, and Pb. The highest values were for As, Cu, and Zn. The continental slope area presents the highest C_f values for Pb (1.49) and As (1.15) from the three studied areas, suggesting a different mechanism of deposition for these metals. The modified degree of contamination (mC_d) in the three studied areas presents values above one, suggesting metal enrichment in the areas relative to the reference material used. Higher values were found in La Antigua coastal area.

The geo-accumulation index (I_{geo}) Seven classes of the geo accumulation index were defined: class 0 ($I_{geo} \leq 0$, unpolluted), class 1 (I_{geo} 0–1, unpolluted to moderately polluted), class 2 (I_{geo} 1–2, moderately polluted), class 3 (I_{geo} 2–3, moderately to extremely polluted), class 4 (I_{geo} 3–4, strongly polluted), class 5 (I_{geo} 4–5, strongly to extremely polluted), and class 6 ($I_{geo} \geq 5$, extremely polluted).

The I_{geo} values obtained in the present study (Table 5) showed that average I_{geo} values are below 1.0 for all metals studied, except for As; only a few specific sites present different degrees of pollution. In La Antigua area sampling site, A1 presents I_{geo} values above one for As (strongly polluted), Cr and V (moderately polluted), and site A15 with values above 2 for Cu (moderately to extremely polluted) and Zn (moderately polluted). Site A3 has values of As (strongly polluted) and Cd (moderately polluted). In Jamapa site, J21 presents I_{geo} values above one for As (strongly polluted), Cu (moderately to extremely polluted), and Zn (moderately polluted) and site J7, As (moderately to extremely polluted), Cu (moderately to extremely polluted), and Zn (moderately polluted).

Sediment quality guidelines Based on the data reported by Long et al. (1995), an analysis of the concentration of metals evaluated in each studied area showed

Table 4 Contamination factors (C_f) and modified degree of contamination (mC_d)

	C_f V	C_f Cr	C_f Co	C_f Ni	C_f Cu	C_f Zn	C_f As	C_f Cd	C_f Pb	mC_d
La Antigua[a]	0.99	1.02	1.05	0.64	1.57	1.32	6.59	0.95	0.95	1.67
Jamapa[a]	0.96	1.12	0.96	0.67	1.47	1.20	5.81	0.75	0.78	1.53
Cont. Slope[a]	0.72	0.59	0.58	0.51	0.87	1.04	6.91	0.68	0.95	1.43

[a] Upper Continental crust

Table 5 Geoaccumulation index (I_{geo})

	V	Cr	Co	Ni	Cu	Zn	As	Cd	Pb
Antigua									
Avg.	0.55	0.51	0.52	−0.07	0.99	0.96	3.29	0.49	0.47
Max.	1.14	1.50	1.50	0.19	2.55	1.37	3.70	1.21	0.92
Min	0.30	0.00	−0.21	−0.22	0.07	0.70	3.04	0.04	0.08
SD	0.28	0.53	0.67	0.14	0.85	0.25	0.21	0.35	0.31
Jamapa									
Avg.	0.51	0.74	0.53	0.01	0.84	0.38	3.10	0.15	0.23
Max.	0.74	1.01	0.63	0.18	2.47	1.38	3.76	−0.29	−0.07
Min	0.06	0.35	0.31	−0.44	−0.35	0.25	2.72	0.43	0.42
SD	0.22	0.20	0.11	0.18	0.91	0.32	0.27	0.25	0.16
Cont. slope									
Avg.	−0.01	−0.27	−0.25	−0.56	0.30	0.53	3.33	−0.05	0.40
Max.	0.50	0.25	0.33	0.36	0.76	1.01	3.96	0.45	0.94
Min	−1.61	−1.69	−1.03	−2.17	−1.03	−1.09	2.43	−1.41	−1.06
SD		0.59	0.37	0.79	0.61	0.67	0.39	0.58	0.65

Metal concentration in the upper continental crust

(Table 6) that the percentage of adverse effects that could be inferred varied between the metals and from one area to the other. It was observed in the three studied areas that the reported concentrations of Cd, Pb, and Zn might rarely be associated with adverse biological effects (Table 6).

In the coastal area adjacent to the mouth of La Antigua River, adverse biological effects might be observed in 100, 100, 50, and 40 % of the sites due to As, Ni, Cu, and Cr concentrations, respectively, and might rarely be observed in 100 % of the sites studied due to the presence of Cd, Pb, and Zn (Fig. 7a).

The sediments of the coastal area adjacent to the mouth of the Jamapa River may cause adverse biological effects occasionally in 90, 80, 60, and 20 % of the sites due to Ni, Cr, As, and Cu concentration, respectively. Adverse biological effects might rarely be observed in 100 % of the sites studied for Cd, Pb, and Zn (Fig. 7b).

This means that the percentage incidence of adverse biological effects that might be observed in sediments

Table 6 Sediment quality guidelines and percent of adverse effect incidence

Metal	Antigua[a]	Jamapa[a] (mg/kg)	Cont. slope[a]	Guidelines[b] (mg/kg)		Percent (ratios)[b]		
						Incidence of effects		
				ERL	ERM	Rarely ≤ERL	Occasionally ≥ERL and ≤ERM	Frequently ≥ERM
As	8.2–13.0	6.6–14.0	5.4–16.0	8.2	70.0	5.0	11.1	63.0
Cd	0.1–0.2	0.05–0.09	0.02–0.1	1.2	9.6	6.6	36.6	65.7
Cr	56.0–156	70.0–111	17.0–66.0	81.0	370.0	2.9	21.1	95.0
Cu	18.0–97.0	13.0–93.0	8.0–28.0	34.0	270.0	9.4	29.1	83.7
Pb	12.0–22.0	11.0–15.0	5.0–22.0	46.7	218.0	8.0	35.8	90.2
Ni	25.0–33.0	22.0–33.0	7.0–38.0	20.9	51.6	1.9	16.7	16.9
Zn	77.0–123	56.0–124	22.0–95.0	150.0	416.0	6.1	47.0	69.8

[a] This work

[b] Long et al. 1995

Fig. 7 Classification of samples according to the sediment quality guidelines for La Antigua area (**a**), the Jamapa area (**b**), and the continental slope area (**c**) (*dark bar* ≤ERL, *open square* ≥ERL, and ≤ERM)
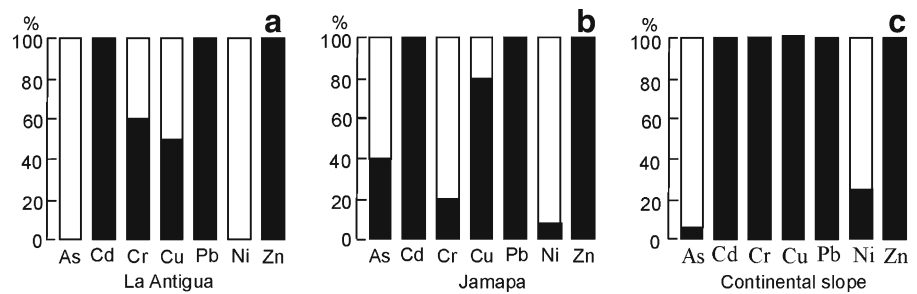
from La Antigua and Jamapa coastal area will be, according to Table 5, of 11.1 % for As, 16.6 % for Ni, 29.1 % for Cu, and 21.1 % for Cr. The concentrations in the continental slope, being lower than ERL, should not cause adverse biological effects for Cd, Cr, Cu, Pb, and Zn; only occasionally benthic fauna could be negatively affected in 90 and 80 % of the sites studied due to arsenic and Ni concentrations, respectively (Fig. 7c).

Ni concentrations evaluated in this study did not show enrichment in relation to Ni found in the continental crust. However, according to SQG index (Long et al. 1995), low Ni concentrations (≤20.9 mg/kg) may have a negative impact on fauna, and Ni concentrations up to 38 mg/kg were found in the area. The opposite could be the case for Zn because the high concentrations (150 mg/kg) needed to produce adverse effects on fauna were not found in this study, even

Table 7 Enrichment factors from different Coastal areas

Area	Cu	Zn	Pb	Ni	Cr	Cd	As
Thermaikos Gulf, Greece[a,b]	0.50–5.30	0.90–4.40	0.60–12.5	–	0.10–2.10	–	–
Offshore SW[c,d] Taiwan	0.92–1.50	0.72–3.31	0.87–1.17	0.67–1.16	0.98–6.96	1.25–11.23	0.70–1.99
Estuaries near[d,e] Bohai Bay China	0.77–2.81	1.01–2.11	0.92–1.93	0.67–1.49	0.89–1.97	0.76–1.98	–
Yangtze River[f,g] Intertidal Zone	0.50–2.40	1.20–2.60	1.3–2.3	1.10–2.80	1.10–5.60	1.70–6.80	–
Kaoping SW[g,h] Taiwan	–	1.18–3.50	1.20–4.94	0.53–2.38	0.85–2.92	0.16–1.65	–
Sundance Harbor Red Sea Cost[g,i]	1.10–5.00	1.00–5.10	0.10–5.20	1.00–5.00	–	–	–
Dongjian[d,j] Harbor Tianjin	0.43–0.89	0.70–1.27	0.62–6.39	–	–	0.02–0.09	0.70–1.59
La Antigua[g,k] Coastal Area	0.92–5.22	1.23–2.31	0.93–1.63	0.68–1.09	0.81–2.71	0.89–1.99	6.92–1.59[g] 0.79–1.27[d]
The Jamapa[g,k] Coastal Area	0.73–4.39	1.10–2.06	0.80–1.07	0.69–0.91	1.11–1.43	0.66–1.02	4.92–12.97[g] 0.56–1.49[d]
The Veracruz Continental slope area[g,k]	0.59–1.51	0.57–1.93	0.58–1.83	0.27–0.98	0.37–1.22	0.45–1.40	7.49–12.67[g] 0.86–1.45[d]

[a] Christophoridis et al. 2009

[b] The average shale content

[c] Selvaraj et al. 2010

[d] Background concentration levels

[e] Feng et al. 2010

[f] Zhang et al. 2009

[g] Metals in the upper continental crust

[h] Hung and Hsu 2004

[i] Idris 2008

[j] Guo et al. 2010

[k] This study

though Zn enrichment can be observed at some sites of the studied area.

The use of different pollution indicators has become more common recently. In Table 7, enrichment factors from different marine areas were shown. By analyzing the data in the table, it is possible to quickly visualize the prevailing conditions in each area. The high values observed in Table 7 for different coastal areas were associated with anthropogenic contributions in all cases. EFs reported in Veracruz's coastal area for As were high compared to data reported by Dongjian Harbor (Guo et al. 2010), where the authors report that As and Zn concentrations were affected seriously by human activities, although the EFs in Dongjian Harbor were calculated using values in unpolluted sediment samples. The EFs reported for Cu at some sites in the present study were as high as the values at some sites in the Thermaikos Gulf, Greece (Christophoridis et al. 2009). EFs for the rest of the metals studied were below the data reported in Table 7. The coastal area near Jamapa River mouth is very close to the Port of Veracruz, where three industrial parks exist; just one of them has 231 different industries (concrete, paints, steel, among others) (INEGI 2011). Additionally, the sewage treatment plant from Veracruz City is located in the coast line, and activities related to ship maintenance are taking place. It has been reported that anti-fouling paint used on the hulls of ships is one of the major sources of Cu pollution in aquatic environments (Idris 2008). The coastal area near La Antigua is not affected directly by industries discharges; however, EFs in La Antigua were higher for Cu, Zn, Pb, Cr, and Cd. The sampling site where the highest EFs were found was associated with high mud content and the higher depths of La Antigua area.

Conclusions

The textural and chemical sediment characteristics allowed understanding the distribution pattern of the sediment supplied by La Antigua and Jamapa rivers toward the studied coastal area of the Gulf of Mexico. The main factors that determine sediment distribution were clay minerals, heavy minerals, carbonates content, and anthropogenic inputs. Trace metal concentrations in the sediment samples spread throughout the coastal area showed significant spatial variation, and higher concentration levels were observed for some metals in localized zones.

Metal enrichment was higher in La Antigua coastal area. Cooper presents the highest EF value at some sites in Jamapa coastal area. Metal enrichment in the Veracruz Continental Slope area was lower than in the coastal areas studied for all metals except for As and Pb. Similar metal enrichments were found through the use of different indexes used to evaluate contamination. EFs gave a bigger number of enriched metals in each area than C_f index and I_{geo} index. The metals with higher concentration showed enrichment with the three different indexes used. In La Antigua area, As, Cu, Zn, Co, Cr, and V; in Jamapa, As, Cu, and Cr; and in the continental slope area, As, Zn, and Pb present enrichment with EFs, C_f, and I_{geo}. The modified degree of contamination (mC_d) allows for an overall view of the conditions of the system, given it was obtained by the sum of all the metals studied. The mC_d data signals La Antigua as the area with higher metal enrichment. According to the sediment quality guidelines, the metals that might produce occasional adverse effects to organisms were Ni, As, Cu, and Cr in the coastal area and As and Ni in the continental slope area.

An integrated approach that includes the geochemical and textural characteristics of the sediments, the metal concentration, and several indexes indicative of pollution was necessary to explain metal concentration levels and distribution patterns related to continental discharges to the coastal environment. The data present in this paper do not show an evidence for contamination from the hydrocarbon industry. However, the data from this work provide a geochemical baseline against which any future contamination caused by expansion of the port activities and the hydrocarbon industry can be assessed.

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