Accumulation and distribution of Hg and 210Pb in superficial sediments from a coastal lagoon in the SE Gulf of California associated with urban-industrial and port activities

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ORIGINAL ARTICLE

Accumulation and distribution of Hg and ^{210}Pb in superficial sediments from a coastal lagoon in the SE Gulf of California associated with urban-industrial and port activities

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Abstract The Estero de Urías Lagoon (EUL) is an inner shelf barrier coastal lagoon, located in the Mexican Pacific Coast (SE Gulf of California). It is surrounded by Mazatlán City, one of the most important international tourist areas of Mexico. To provide a comprehensive reassessment of the concentration levels and spatial variability of Hg and ^{210}Pb in the EUL, 40 surface sediment samples were analyzed for several geochemical variables (e.g. grain size distribution, organic matter and reference element concentrations) that could explain the observed variability of Hg and ^{210}Pb. The Hg concentrations ranged from 23 to 214 ng g^{-1}, whereas ^{210}Pb activities varied from 20 to 56 Bq kg^{-1}. No defined distribution pattern was observed for Hg and ^{210}Pb concentrations in the lagoon and no evidence of a common atmospheric delivery route was observed. The sediments from EUL were found contaminated by Hg, and according to international guidelines 48 % of the sampling sites have concentrations that could be harmful to biota.

Keywords Hg contamination · ^{210}Pb in superficial sediments · Estero de Urías coastal lagoon · Gulf of California

Introduction

In the international community there is a considerable concern about Hg contamination in aquatic ecosystems due to its high toxicity at low concentrations and also because it enters easily the biological systems (Porto et al. 2005). In fact, Hg is known to be incorporated into diverse components of trophic webs of marine ecosystems, including marine plants, invertebrates and fish (UNEP 2002); moreover, it can also be potentially toxic to humans through fish consumption (USEPA 2004). Nevertheless, Hg occurs naturally in the environment; its sources include degassing from the earth crust, volcanic activity and forest fire. In addition, there are several human activities that release Hg to the environment which includes metallurgical processes, fossil fuel burning, wood pulping, paint and chlor-alkali industries, battery production and agriculture (Morel et al. 1998). In fact, according to the literature, these anthropogenic activities have increased its global incidence in modern times (Lamborg et al. 2002).

Estuarine systems are particularly important; they are considered among the most productive marine ecosystems in the world, mainly because they serve as areas for migration routes and nursery grounds for many organisms (Rodríguez-Barroso et al. 2009). In recent years the impact by Hg contamination in these aquatic systems has been stronger than in the past decades, especially in estuarine

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systems adjacent to urban, industrial or agricultural areas (e.g. Ram et al. 2003; Ruiz-Fernández et al. 2009a).

The estuaries accumulate Hg and other metals in different environmental reservoirs; however, sediments act as the major reservoir for metals (Caccia et al. 2003). Sediments can accumulate contaminants from direct or diffuse sources; furthermore they can also act as a source of contaminants (Adams et al. 1992; Burton and Scott 1992). The lifetime of Hg in sediments is so long that they can be considered as sinks for this metal, which is mainly bound to organic matter, with a little fraction associated with Mn and/or Fe-oxides near the redox boundary (Canário et al. 2003). Sediment characteristics usually represent the average condition of water quality (Förstner and Wittmann 1983; Salomons and Förstner 1984). In the scientific literature there are several recent studies about Hg contamination of superficial sediment in coastal systems associated with human activities; they demonstrate that this problem exists in different regions in the world (e.g. Ram et al. 2003; Green-Ruiz et al. 2005; Ruelas-Inzunza et al. 2009; Rodriguez-Barroso et al. 2010).

The ^{210}Pb ($T_{1/2} = 22.23 \pm 0.12$ year; DDEP 2010) is a radioactive element of ^{238}U series. The ^{210}Pb occurs naturally in sediments, soils, atmosphere and natural waters. Part of the ^{210}Pb, referred to as base ^{210}Pb (Sánchez-Cabeza et al. 2007) is formed in situ in soils or sediments, in equilibrium with its parent radionuclide ^{226}Ra present in the sediment minerals. Another part of ^{210}Pb is mobilized through the atmosphere (known as excess ^{210}Pb). The atmospheric ^{210}Pb results from the radioactive decay of ^{222}Rn emanating from soils (Turekian et al. 1977); it can attach to atmospheric particulate matter and later it is deposited on the surface of the soil and water bodies as dry and wet deposition. The behavior of ^{210}Pb allows its use in environmental studies, e.g. biogeochemical processes in the oceans (e.g. Nozaki et al. 1991; Wei and Murray 1994), atmospheric deposition and anthropogenic pollution (e.g. Shotyk et al. 2002; Kaste et al. 2003), sedimentary processes (e.g. De Master et al. 1991; Robert et al. 1997) and geochronology of sediments (e.g. Ruiz-Fernández et al. 2009a).

In the coastal zone a part of ^{210}Pb is supplied by atmospheric deposition and in shallow zones it can be the primary source to sediments (El-Daoushy 1988). Additionally, ^{210}Pb can be carried and dispersed in coastal sediments by rain runoff and marine currents. Similarly to Hg and others metals, the ^{210}Pb in sediments can bind to clay minerals, organic matter, oxides and hydroxides of Fe and Mn and other sediment components (Wolfe et al. 2004), which can be vehicles to distribute the ^{210}Pb in coastal sediments, according to the water body hydrodynamic. In relation to this, in the Mexican Pacific it has been demonstrated that ^{210}Pb in the shallow coastal environments is limited by low ^{210}Pb atmospheric fallout, sediment mixing and the abundance of coarse sediments (Ruiz-Fernández and Hillaire-Marcel 2009).

Previous studies have suggested (e.g. Lamborg et al. 2000; Fitzgerald and Engstrom 2005) that Hg and ^{210}Pb can show similar transformational pathways in the atmosphere, in which particle scavenging has a dominant role to remove both elements through rainfall. Hg in the vapor phase is oxidized through a homogenous gas-phase reaction and, after being dispersed in the atmosphere with the wind, it is deposited in superficial soils and sediments; this process is analogous to the gas-phase radioactive decay of ^{222}Rn, the gas-to-particle phase conversion into ^{210}Pb and its subsequent precipitation. In fact, a correlation between Hg and ^{210}Pb in particulate matter and rain has been reported (Lamborg et al. 2000).

In the current study, Hg concentration and ^{210}Pb activities and their spatial distribution were determined in surface sediments collected at the EUL, with the purpose of evaluating if a significant correlation existed between Hg and ^{210}Pb concentrations in the study area. The hypothesis of the study is that both elements were mainly delivered attached to atmospheric particulate material and their accumulation and distribution was related to geochemical variables.

The grain size distribution and the concentrations of calcium carbonate, organic matter and reference elements (Li, Fe, Mn, Ti and Al) were also determined to understand the local processes that influence the Hg and ^{210}Pb distribution in the surface sediments from the EUL.

Materials and methods

Study area

The EUL is located on the Pacific coast (northwest of Mexico), adjacent to the city and harbor of Mazatlán (Fig. 1), which is an industrialized city with 438,434 inhabitants (INEGI 2011) and an urban area of ~103 km^2. The weather in the region is subtropical with an average annual rainfall of 746.9 mm. It has a marked dry season (from November to May) and the maxima rainfall occurring mainly from July to September (CONAGUA 2011). The prevailing winds during the year blow from W and NW with an average speed of 2.6–3.4 m s^{-1}. EUL was classified by Lankford (1977) as an inner shelf barrier type. It is a saline vertically mixed water body (salinity range 25.8–38.4 ‰), with mixed tides of 1.5 m amplitude (Páez-Osuna et al. 1990) with slight diurnal tidal action (Martin and Gutiérrez-Galindo 1989). It is a shallow (<2 m up to ~12 m depth), and narrow (<1 km width) water body; it has 18 km in length and a surface area of ~18 km^2. A

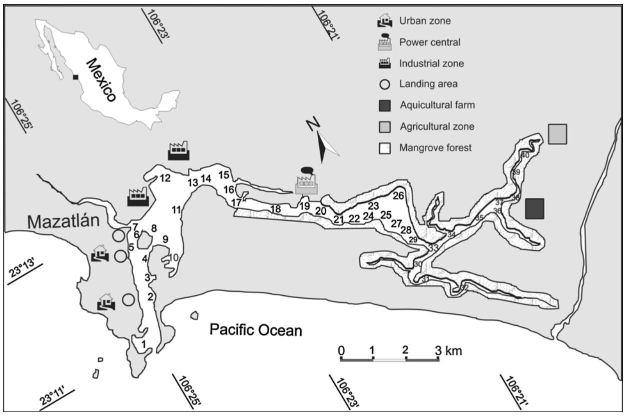


Fig. 1 Estero de Urías lagoon map showing the sampling sites of superficial sediments (*numbers*)

mass accumulation rate of 0.12 g cm^{-2} $year^{-1}$ and a sediment accumulation rate of 0.33 cm $year^{-1}$ have been reported for the intertidal mudflat of EUL by Ruiz-Fernández et al. (2009a). The EUL is surrounded by the shipping terminal and the industrial zone (mostly food processing plants) of Mazatlán City; and ca. 3 km of the proximal section to the sea is regularly dredged to preserve an artificial navigation channel of <12 m depth. It also receives runoff from agriculture wastes (41,700 ha of cultivated areas, with 5,500 ha of irrigated fields) with the main crops being sorghum with 34,000 ha and corn with 2,300 ha (OEIDRUS 2008) although most of the freshwater supply to EUL derives from urban runoff. The inner zone of the lagoon is surrounded by mangrove forests with a small-scale artisanal fishing activity and a semi-intensive shrimp farm (340 ha), which uses and discharges water in the inner part of the lagoon. The harbor activities (e.g. international shipping, transport of petroleum hydrocarbon, shipyards, dredging of navigation channel), industrial and municipal waste discharges are potential prominent sources of contaminants from the urbanized portions of the lagoon. A major fossil fuel electrical generation station uses estuarine water for cooling purposes; it is located approximately 9 km from the lagoon mouth. Previous studies in EUL have reported high concentrations of Cd, Pb, Zn and Cu (Soto-Jiménez and Páez-Osuna 2001), Hg (Jara-Marini et al. 2008; Ruiz-Fernández et al. 2009a) and pesticides and polychlorinated biophenyls (Galindo-Reyes et al. 1999; Reyes et al. 2003) which have been associated with point discharges from municipal, dock and industrial activities.

Sampling

Superficial sediment samples were collected from 40 stations in the lagoon (Fig. 1); using a Van Veen grab made of stainless steel, during the dry season: November and December 2009 (stations 26–40 and 15–25, respectively) and January 2010 (stations 1–14). During the sampling special care was taken to collect just the top 1 cm by using a crystal clear PVC cylinder (10 cm diameter). All the samples were transferred to acid-washed (Moody and Lindstrom 1977) polyethylene recipients and stored at −20 °C until analysis.

Laboratory analysis

Sediment samples were oven dried at 50 °C for 120 h. About 2 g was separated for grain size analysis and 5 g ground to a powder with a porcelain mortar and pestle for the rest of the analyses (aliquots of 0.25 g of dry sediment were used for each analysis). Grain size distribution was evaluated through standard sieve (sand content) and pipette (silt and clay) methods (Galehouse 1971). Organic matter (OM) was determined by Walkey–Black method as described by Loring and Rantala (1992); carbonate content ($CaCO_3$) was analyzed by treating the sample with 1 N HCl and the acid excess was titrated with 0.5 N NaOH (Stuardo and Villarroel 1976). Sediment samples were digested in a mixture of $HNO_3 + HCl + HF$ (Loring and Rantala 1992) by using Teflon PFA containers on a hotplate at 130 °C for 12 h. Metal analyses were determined by atomic absorption spectrophotometry (Fe, Al and Mn by flame, Li and Ti by graphite furnace, and Hg by cold vapor); The accuracy of the metal analyses was evaluated with the reference materials IAEA-356 and IAEA-453 and concentrations obtained fell within the certified value intervals (1σ) for each reference material. The analytical precision was estimated by the analysis of six replicates of a homogenized sample; the variation coefficient for Hg was estimated to be about 6 %, and for Fe, Mn, Al, Ti and Li, 3, 5, 4, 4 and 3 %, respectively.

The total ^{210}Pb activities in the superficial sediments were determined by measuring the activity of its daughter product ^{210}Po (Schell and Nevissi 1983) assuming secular equilibrium between the two radioisotopes. About 0.3 g of sediment samples were spiked with ^{209}Po as a recovery tracer and were acid digested (1:5:4 $HF + HNO_3 + HCl$) in closed containers on a hotplate (130 °C, over-night). The digested material was evaporated to dryness and the digestion residue was converted to a chloride salt by repeated evaporation with HCl concentrate and then dissolved in 0.5 M HCl in presence of ascorbic acid (~2 g) as a reducing agent for Fe^{3+}. ^{210}Po was spontaneously deposited on a spinning Ag disc (Hamilton and Smith 1986) and the activity was measured by α-spectrometry (Ruiz-Fernández et al. 2009a).

In order to evaluate the relationships between the measured variables, a Pearson correlation matrix and Principal component analysis (PCA) were calculated by using the software Stat Soft Statistica 7.0.

Results and discussion

Figure 2 shows the spatial distribution of grain size fractions (sand, silt and clays), the concentrations of OM, $CaCO_3$, reference metals (Fe, Mn, Al, Ti y Li) and Hg, as well as the ^{210}Pb activities, determined in the superficial sediments collected in the EUL.

Grain size, $CaCO_3$ and OM concentration

Surface sediments at EUL were not homogenous in terms of grains size distribution: sand abundance ranged from nil to 100 %, silt from nil to 37 %, and clays from nil to 78 %. Sediments from stations 1 to 26, located between the lagoon mouth and ~13 km inside the lagoon (a zone characterized by a high hydrodynamic energy) were predominantly sandy; excepting the stations 7, 15, 16, 21 and 23 that had a greater predominance of silt and clay sediments, possibly associated with transport of terrigenous materials through runoff. Site 27–40 (located in the inner part of the lagoon) presented a greater predominance of silt–clay sediments (~80 %), most likely associated with the low hydrodynamic energy conditions of this lagoon zone.

The $CaCO_3$ concentrations in sediment were found in the range of 0.2–43.0 %, with the higher values observed in six sampling stations located between the middle and the innermost parts of the coastal lagoon (>30 %). The OM sediment concentrations ranged between 0.9 and 15.9 %; the higher values were mostly observed in the inner part of the lagoon, although a peak value was also found at sampling site 7, in the surroundings of a stream discharge, close to the lagoon mouth. The OM and $CaCO_3$ concentrations found in this study were similar to or higher than the intervals previously reported in other parts in the world and similar to or lower than previous studies reported for the EUL (Table 1).

Reference elements (Al, Fe, Li, Ti and Mn)

The metal concentrations observed in the surface sediments of EUL varied as follows: Fe, from 0.4 to 1.9 %; Mn, from

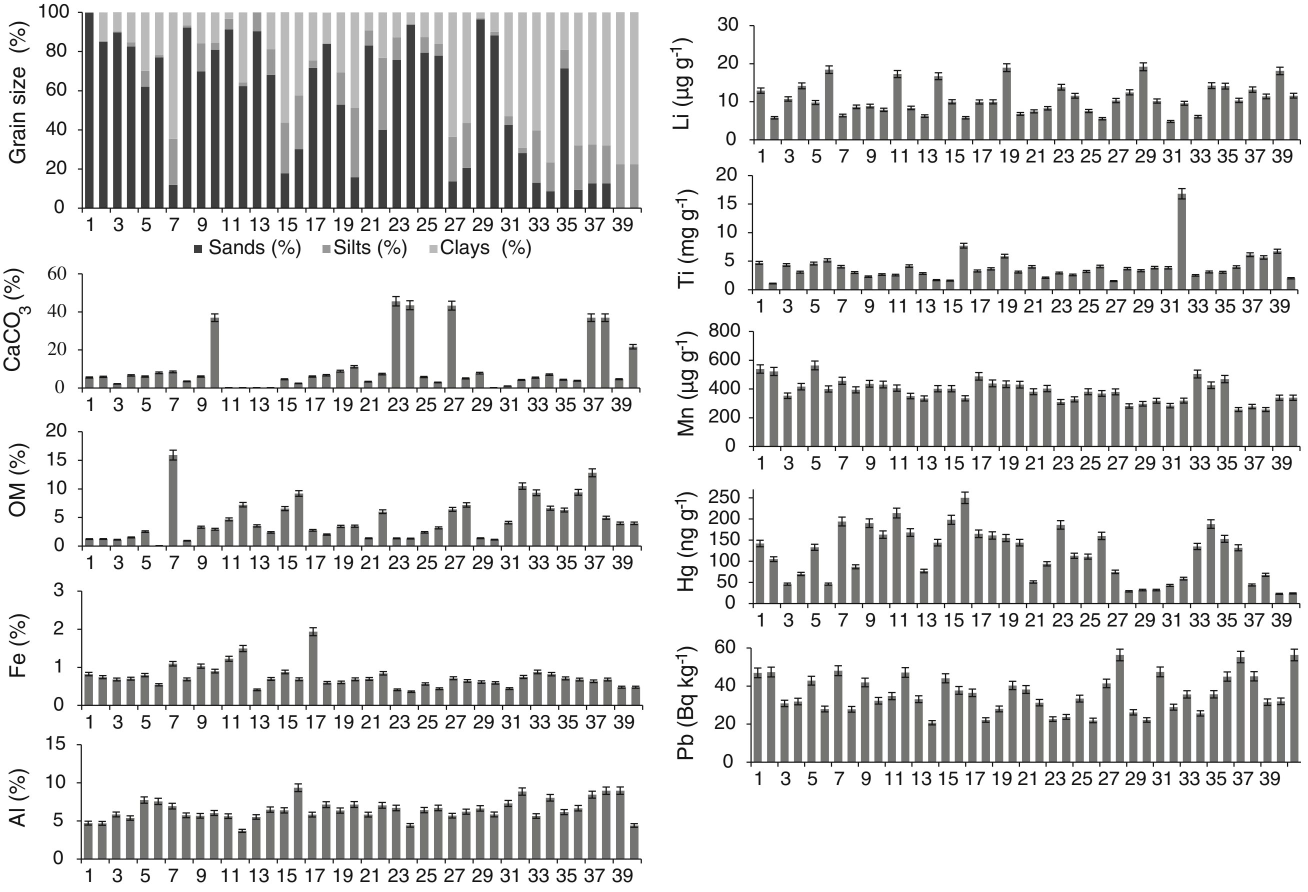


Fig. 2 Grain size content (sands, silts and clays), reference metals concentrations (Fe, Al, Li, Ti and Mn), Hg concentration and ^{210}Pb activity in superficial sediments from Estero de Urías Lagoon

Table 1 Geochemical variables (OM, $CaCO_3$, Al and Fe in %, Ti in mg g^{-1}, Li and Mn in µg g^{-1}), Hg (ng g^{-1}) and ^{210}Pb (Bq kg^{-1}) values for coastal sediments

Study site	OM	$CaCO_3$	Al	Fe	Ti	Li	Mn	Hg	^{210}Pb	Classification	References
Cádiz Bay and Sancti, Spain	6–16	10–35	NA	NA	NA	NA	101–506	100–1,700	NA	NC to C	Rodriguez-Barroso et al. (2010)
Sado estuary, Portugal	<1–10	NA	NA	NA	NA	NA	NA	3–540	NA	NA	Lillebø et al. (2011)
Ulhas estuary, India	3–5	NA	3–11	4–11	NA	NA	500–12,000	50–17,560	54 ± 11	NC to C	Ram et al. (2003)
Sagua estuary, Cuba	NA	NA	3	4	3	NA	NA	1,840 ± 90	62 ± 7	C	Díaz-Asencio et al. (2009)
La Plata estuary, Argentina	NA	NA	NA	NA	NA	NA	NA	NA	265–408	NA	Di Gregorio et al. (2007)
Sepetiba and Ribeira Bay, Brazil	NA	NA	3–10	2–6	3–7	NA	227–872	NA	100–200	NA	Carvalho Gomes et al. (2009)
Coatzacoalcos estuary, Mexico	1–2	<1	5–8	3–5	NA	NA	376–887	NA	20–32	NA	Ontiveros-Cuadras et al. (2012)
Coatzacoalcos estuary, Mexico	<1–5	NA	1–7	NA	NA	NA	NA	10–1,060	NA	NC to MC	Ruelas-Inzunza et al. (2009)
Gulf of Tehuantepec, Mexico	<1	22	4	1	4	11	200	NA	119	NA	Ruiz-Fernández et al. (2009b)
La Paz Lagoon, Mexico	1–2	17–28	7–8	1–2	NA	NA	150–200	18–23	NA	NC	Kot et al. (1999)
Guaymas Bay, Mexico	2–12	NA	3–11	1–4	NA	NA	90–386	300–2,300	NA	MC to C	Green-Ruiz et al. (2005)
Ohira, Mexico	12–27	3–8	4	NA	NA	NA	NA	50–300	2–5	MC	Ruiz-Fernández et al. (2009a)
Chiricahueto, Mexico	3	8	1	NA	NA	NA	NA	200	25	MC to C	Ruiz-Fernández et al. (2009a)
EUL, Mexico	2–23	1–40	2–8	2–8	NA	9–30	150–600	NA	NA	NA	Soto-Jiménez and Páez-Osuna (2001)
EUL, Mexico	4–7	5–14	NA	NA	NA	NA	NA	110–460	NA	NC to C	Jara-Marini et al. (2008)
EUL, Mexico	19	5	1	NA	NA	NA	NA	300	40	C	Ruiz-Fernández et al. (2009a)
EUL, Mexico	<1–16	1–46	4–10	<1–2	1–17	5–19	257–564	23–256	21–56	NC to C	This study
Threshold effect limit (TEL)	NA	NA	NA	NA	NA	NA	NA	130	NA	NA	Buchman (2008)

NC uncontaminated, *MC* moderately contaminated, *C* contaminated, NA not available

257 to 564 µg g^{-1}; Al, from 3.7 to 9.4 %; Ti, from 1 to 17 mg g^{-1} and Li, from 5 to 19 µg g^{-1}. No defined spatial distribution pattern was observed for any of the reference metals analyzed, in agreement with observations from a previous study (Soto-Jiménez and Páez-Osuna 2001). The concentrations of the five reference metals (Al, Fe, Li, Ti and Mn) found in this study were comparable to or lower than to those reported in local studies and other studies in coastal sites (Table 1).

Mercury concentrations and spatial distribution

Hg concentrations in superficial sediments from EUL ranged between 23 and 214 ng g^{-1}. The highest Hg concentrations were found in the proximal section to the sea, comprising the sampling sites in the vicinity of the shipping terminal, the industrial and urban areas and the thermoelectric plant, whereas, the lower Hg concentrations were found in the sampling sites located in the inner part of the lagoon; although it is worth noticing that Hg concentrations in the sampling sites 33–36 (inner part of the lagoon) were comparable to those previously described for the most anthropized section of the lagoon. These high Hg concentrations in sites 33–36 might be associated with the confluence of three potential Hg contamination sources adjacent to the inner part of EUL, i.e. Hg transported by runoff from small extensions of agriculture fields, the shrimp farm effluents (as observed elsewhere by Costa et al. 2013) and the discharge of domestic (raw or partially treated) wastewaters from recent semi-urbanized settlements, entering EUL through the tidal channel located between sampling sites 33 and 34 (Fig. 1). In addition, in the inner part of the EUL there is a reduction of the water flux and thus, an increase of material retention (in comparison with the proximal section of the lagoon to the sea) which has been related to the narrowing of the lagoon shape (Montaño-Ley et al. 2000); this is likely the reason for which the high Hg concentrations in stations 33 and 34 are focused in a small portion of the EUL.

The concentrations of Hg in superficial sediments observed in this study were in the same magnitude to or lower than other coastal sites in Mexico and in other areas of the world, considered as moderately contaminated or contaminated with Hg (Table 1). The Hg concentrations in this study were similar to or lower than previous local studies (Jara-Marini et al. 2008; Ruiz-Fernández et al. 2009a), which could suggest that Hg concentrations in Estero de Urías are stable in recent years; however, it has been demonstrated that Hg has increased in past 20 years, due to local anthropogenic activities (Ruiz-Fernández et al. 2009a).

Hg concentration associated with toxic effects

Forty-eight percent of studied sites at EUL showed Hg concentration above the Threshold Effect Limit ($\mathrm{TEL} = 130\ \mathrm{ng\ g^{-1}}$) which is the concentration above which adverse biological effects might occur (Buchman 2008). Therefore, such Hg levels can be associated with probable unsafe conditions for the biota. Most of these sampling sites are located in the middle part and near to the mouth of the EUL (which corresponds to the most industrialized zone of the lagoon) and the sampling sites 33–36 that are located at the innermost part of the lagoon. The comparison to the TEL value suggested that in 48 % of the analyzed sites there are Hg concentrations that could be harmful to the biota in EUL.

^{210}Pb activities

The ^{210}Pb activities in the surface sediments ranged from 20 to 56 Bq kg^{-1} and showed a heterogeneous spatial distribution along the EUL. Spatial variability of ^{210}Pb activity can result from the combination of several factors such as the hydrodynamics of the water body associated (mainly with tidal and residual currents), bioturbation and anthropogenic activities, such as the port activities, e.g. dredging, displacement of ships, construction and maintenance of docks that may contribute to the redistribution of old (^{210}Pb-depleted) sediment to the surface at specific sites. Some of these factors have been associated with difficulties to achieve geochronology studies with ^{210}Pb in coastal water bodies in the region (Ruiz-Fernández and Hillaire-Marcel 2009).

As shown in Table 1, the ^{210}Pb activities in this study were consistent with other studies previously carried out in the region (e.g. Ensenada del Pabellón, Ruiz-Fernández et al. 2001; and Ohuira Lagoon, Ruiz-Fernández et al. 2007), although relatively higher than those previously reported for a mudflat area in the surroundings of the EUL (Ruiz-Fernández et al. 2009a). The ^{210}Pb activities found in this study were similar to or lower than those reported for surface sediments in other coastal areas (Table 1). Previous studies have suggested that, at the latitudes at which the Gulf of California is located, ^{210}Pb fluxes are very low in comparison to other areas of the world (Liu et al. 2001) and that the dominance of westerlies during most of the year is one of the most important factors to this low ^{210}Pb fluxes (Ruiz-Fernández et al. 2001). In addition, other factors such as the abundance of coarse grain size that characterize the coastal sediments (Ruiz-Fernández and Hillaire-Marcel 2009), the low rainfall regimen at the NW region of Mexico (Ruiz-Fernández et al. 2002) and the scarce contribution from fluvial runoffs to the lagoon, could also contribute to the low ^{210}Pb activities observed in the EUL coastal zone.

Correlation between geochemical parameters, Hg concentrations and ^{210}Pb activities

Table 2 presents correlation coefficients (Pearson product test; $P < 0.05$) among the measured variables in this study. A significant positive correlation was found between OM concentrations and the fine grained fraction (silt + clay) and a negative correlation between OM concentrations and sand percentage. Fe concentrations presented a significant positive correlation with Mn concentrations; whereas Al and Ti correlated significantly to each other, and both elements also correlated with OM concentrations. Al concentration showed a significant positive correlation with fine sediments fractions; as well as a significant negative correlation with the sand percentage.

The significant correlation of OM with grain size has been reported in numerous studies (e.g. Horowitz 1991; Green-Ruiz et al. 2005; Rodriguez-Barroso et al. 2010). OM might be bound to particle surface in the form of a

Table 2 Correlation coefficient matrix (Pearson product, $P < 0.05$) between geochemical variables, Hg concentrations and ^{210}Pb activities, in the superficial sediments from Estero de Urias lagoon

	OM	Sand	Silt	Clay	Fe	Al	Li	Mn	Ti	Hg	^{210}Pb
$CaCO_3$	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
MO	1	−0.71	0.56	0.69	NS	0.35	NS	NS	0.33	NS	0.49
Sand		1	−0.80	−0.97	NS	−0.47	NS	NS	NS	NS	−0.40
Silt			1	0.63	NS	0.34	NS	NS	NS	NS	0.32
Clay				1	NS	0.47	NS	NS	NS	NS	0.39
Fe					1	NS	NS	0.41	NS	0.44	NS
Al						1	NS	NS	0.58	NS	NS
Li							1	NS	NS	NS	NS
Mn								1	NS	0.44	NS
Ti									1	NS	NS
Hg										1	NS

NS not significant correlation ($P < 0.05$)

monolayer that coat the grain surface (Hedges and Keil 1995) or it can be distributed in discrete patches over the mineral surface, in cracks and fissures (Ransom et al. 1997; Mayer 1999). Fine sediments have greater contact surface than coarse sediments, and, therefore, the OM concentrations tend to be higher when associated with fine sediments.

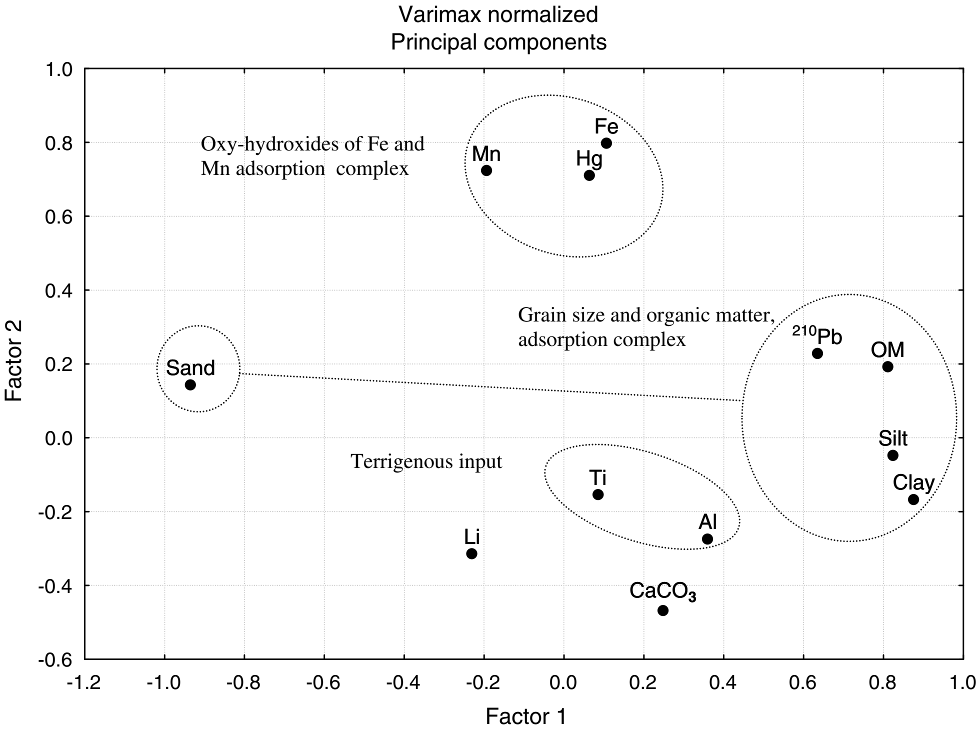
Al and Ti are considered conservative constituents of terrigenous materials (Loring and Rantala 1992) and its common provenance might explain the correlation between these elements; whereas the positive correlation between Al and the fine-grained sediments is most likely due to that Al is one of the most abundant elements in soil, being a component of clays (Edmondson 1991). Finally, the significant positive correlation between OM concentrations and the terrigenous elements Al and Ti might be explained on the basis of a continental contribution of the three components, most likely through runoff and erosion.

Significant positive correlations were also found between the concentrations of Hg with Fe and Mn. The oxyhydroxides of Fe and Mn are associated with the capture of the Hg and other metals (Turner et al. 2004), mostly due to the high capacities of adsorption and the low degree of crystallinity of Fe and Mn oxyhydroxides, caused by their successive cycles of dissolution-precipitation, which facilitates the nonstoichiometric incorporation of metals into the oxides (Chao 1984). The determination coefficients obtained for the relationships between Hg with Fe, and with Mn were rather low ($r^2 = \sim 0.20$), which indicates that <20 % of the Hg variability might be explained due to sequestration by oxyhydroxides of Fe and Mn concentrations present in surface sediments of the EUL, and, therefore, there are other factors that could explain the Hg concentration distribution, such as the local anthropogenic sources that could alter the natural geochemical behavior of Hg.

The ^{210}Pb activities showed significant positive correlations with the contents of fine material (silts and clays) and the OM concentrations, as well as a significant negative correlation with the sand content. The determination coefficient found between ^{210}Pb and fine material content suggested that about 20 % of ^{210}Pb is scavenged by fine grained material in the EUL. The significant correlation between fine sediments content and ^{210}Pb activities has been also reported in other studies where the ^{210}Pb activity may be up to 8.5 times higher in clays than in sands (He and Walling 1996; Ontiveros-Cuadras et al. 2012). The latter reflects the increased adsorption capacity of the fine materials, explained by their high surface area compared with coarser materials (He and Walling 1996). In fact, the limited absorption capacity of sandy sediments can cause a dilution effect of ^{210}Pb activities in sediments (Ontiveros-Cuadras et al. 2012), as demonstrated by the negative correlation between ^{210}Pb activities and the sand content. The correlation between ^{210}Pb and OM has been previously reported in other studies (e.g. Legeleux et al. 1996; Ruiz-Fernández et al. 2004). It has been documented that ^{210}Pb can be sequestrated by the OM binding to the negatively charged functional groups (Wolfe et al. 2004), where the ^{210}Pb can remain stationary (Vile et al. 1999); this means that the capture and distribution of ^{210}Pb through the OM is a relatively stable process. In fact, the concentration of OM in sediments can give an idea of the distribution pattern that ^{210}Pb activity can have in the sediments, as previously explained by Ruiz-Fernández et al. (2012).

No significant correlation was found between Hg concentrations and ^{210}Pb activities in the sediments. Lamborg et al. (2000) have previously reported correlations between Hg and ^{210}Pb in the atmospheric particulate matter and wet deposition; however, coastal lagoon sediments are more complex; they are integrated by a mixture of particles accumulated from a variety of sources. The lack of correlation between ^{210}Pb activities and Hg concentrations might be explained on the basis of the anthropogenic contribution of Hg to the lagoon. The direct discharges of residual waters contaminated with Hg (urban and agriculture wastes) to the lagoon are very likely the main source of

Fig. 3 Scatterplots of PCA loading by individual variables



this metal, whereas the ^{210}Pb activities are mainly the result of atmospheric sources.

Hg sediment contamination at the Estero de Urías Lagoon

To evaluate the degree of Hg contamination in sediments of the EUL, the Enrichment Factor (EF) was calculated according to the formula by Buat-Menard (1979):

$$\mathrm{EF} = [\mathrm{Hg}]_s/[\mathrm{M}]_s/[\mathrm{Hg}]_b/[\mathrm{M}]_b.$$

where $[\mathrm{Hg}]_s$ and $[\mathrm{Hg}]_b$ represent the Hg concentration in the sample (s) and the background or pre-anthropogenic concentration (b), respectively, and [M] is the concentration of the reference metal (normalizing element, in this study was Al).

Enrichment Factor calculations assumed that the chemistry which determines sorption or dissolution of metals is the same through the sediments and generally referred to natural or baseline concentration levels (Ruiz-Fernández et al. 2009a). The choice of Al concentrations as a normalizer of Hg concentrations was based on the significant correlation ($P < 0.05$, $r = 0.5$) observed between Al concentrations and the percent content of clays in the sediments at the lagoon. The background values for Al and Hg (4.9 ± 0.1 % and 28.12 ± 3.61 ng g^{-1}, respectively) used to estimate the EF corresponded to the pre-industrial (~1,900) concentrations of both elements (unpublished data) from a sediment core collected in a mudflat in the surroundings of the EUL. The EF of Hg ranged from 0.4 to 8. According to the pollution categorization based on the enrichment ratio from Sutherland (2000), the surface sediments in EUL show depletion to minimal enrichment of Hg, suggesting no contamination or minimal contamination in 29 % of sites, including sites 3, 6 and 2, as well as most of sampling sites located in the most distal part of the lagoon. In contrast, 58 % of sites in the lagoon showed moderate Hg enrichment, suggesting moderate contamination; and 13 % of sites showed significant Hg enrichment, suggesting a significant contamination signal. The significantly contaminated sites are located between the entrance of the lagoon and up to 5 km inside the navigation channel (corresponding to the port area, in front of the more industrialized and urbanized zone from EUL).

PCA

Factor analysis is a technique commonly used to analyze geochemical matrices by creating one or more factors, each representing a cluster of interrelated variables within the data set (Delvalls et al. 1998). The factor loading is related to the input provided by individual variables to a given factor and the variables that most strongly load a particular factor can be used to assign the physical meaning of this factor (Szefer and Kaliszan 1993). PCA extracted three factors (eigenvalue >1) that accounted for more than 63 % of the total variance from the 12 variables included in the data set (Fig. 3), and they are:

Factor 1 (Grain size and organic matter adsorption complex) explained 31 % of the variance, and includes OM and fine sediments (clay and silt) with positive loadings, as well as sand with a negative load.

Factor 2 (Oxyhydroxides of Fe and Mn adsorption complex) explained almost 20 % of the variance. Fe, Mn and Hg are grouped under this factor indicating that Hg in sediments is most likely scavenged from the water column by the oxyhydroxides of Fe and Mn, and it can explain partly the Hg distribution in superficial sediments of EUL.

Factor 3 (Terrigenous) explained 14 % of the variance and included Ti and Al. These elements have a terrigenous origin and can be associated with eroding soil due to runoffs carried to the superficial sediments from EUL.

Conclusions

The surface sediments of the EUL were found to be polluted by Hg. The Hg concentration in the surface sediments of the lagoon did not show a defined pattern of spatial distribution, and no evidence was found that physical or geochemical sediment factors (e.g. grain size or organic matter content) could unambiguously explain the variability observed in Hg concentrations among sites. The highest concentrations of Hg were found in the intermediate zone of the lagoon, where there is an increased anthropogenic pressure (associated to discharges from urban-industrial areas, the port terminal and the thermoelectric plant) and four sites in the inner part of EUL likely associated with agricultural and domestic wastewater discharges. The results of the correlation analysis and PCA showed that Hg concentrations are associated with Fe and Mn concentrations (most likely, Hg is adsorbed onto Fe and Mn oxyhydroxides) and this association might partially explain the Hg distribution in superficial sediments from EUL. The ^{210}Pb activities found in the surface sediments of the EUL were in the typical range of activities determined in other coastal lagoons of the region (e.g. the coastal lagoons of Chiricahueto and Ohuira) and resulted from atmospheric deposition. The lack of significant correlation observed between the ^{210}Pb activities and the Hg concentrations in sediments of the EUL suggested that there is not a common origin of both elements, and this is most likely related to the contribution of the urban and industrial point sources of Hg to the lagoon. The study has provided evidence that there is a significant Hg contamination in the lagoon, and considering that 48 % of the sampling sites showed Hg concentration that can be unsafe to the biota (and potentially to humans through fish consumption) and therefore, it is advisable to enhance the environmental regulations order to improve the quality of this important aquatic resource.

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