THE CORRELATIONS BETWEEN MERCURY SPECIATION AND DISSOLVED ORGANIC MATTER IN THE SEDIMENT OF THE RED SEA

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ABSTRACT

This study addresses the correlations between Hg and organic matter in recent sediment; samples were collected from the Gulf of Aqaba, Red Sea coasts (oligotrophic regions) during 2010. In the present study TOC analyzer was used to determine Total Organic Carbon (TOC) and Total Nitrogen (TN) concentrations and the total amount of mercury (Hg_T) in sediment samples were analyzed by Hydra-C mercury analyzer. The obtained results indicated that, mercury, TOC and TN average concentrations in the Red Sea were 85.42 ng g^{-1}, 5.10 and 4.45 mg L^{-1}, respectively. The results show that the Total Organic Carbon (TOC) in sediment represents the sum of various organic compounds, which may play a completely different role in the distribution and accumulation of Hg. slightly correlations between the TOC and the concentration of Hg in the studied sediment arise mainly from the labile portion of organic matter released. These compounds primarily consist of easily degradable algal-derived lipids and various pigments, which are petrographically described as a soluble Organic Matter (OM). The preserved OM in sediment is commonly entrapped within the cell walls of phytoplankton and also appears as a surface coating on sediment particles. The strong affinity between Hg and OM is due not only to its chemical reactivity, but also to the physical characteristic of these labile compounds, which plays the most important role in the distribution of Hg in sediment.

Keywords: Total organic carbon, total nitrogen, mercury, sediment, gulf of Aqaba-Jordan

1. INTRODUCTION

Anthropogenic mercury in the aquatic environment has been an increased awareness due to their toxicity for the life so it is important to understand the fate and reactivity with such environment. Paraquetti *et al.* (2004) showed that the trace metal are reacted and sorbed to the suspended particulate matter. In an aquatic environment mercury occurs in different forms including elemental mercury, Ionic mercury and methyl mercury (Berthon, 1995). Methyl mercury is one of the most toxic forms of mercury species; it tends to bioaccumulate and biomagnify in the fatty tissue of fish more than other forms. Watras and Huckabee, (1994) prove that the most mercury in the most fish species occurs as methyl mercury which is transferred to human via the food chain. Mercury

like other metals speciation is affected by reaction with organic and inorganic compounds that present in water.

Dissolved Organic Matter (DOM), which is ubiquitous in aquatic environments, is bind to trace metals which affect their speciation, solubility, mobility and toxicity (Buffle *et al.*, 1988). There is increasing evidence that DOM interacts with mercury as well which is altering its speciation and bioavailability in aquatic environments (Loux, 1998). In coastal waters, more than 60% of the dissolved mercury is associated with organic matters or suspended particles (Fitzgerald and Lyons, 1973). Whereas Dissolved Organic Carbon (DOC) appears to be the most important species which control the bioavailability of mercury due to high binding capacity (Barkay *et al.*, 1997, Cai *et al.*, 1999). The

previous studies showed a strong interactions between Hg and DOM have also been indicated by a positive correlation between their concentrations in many natural waters (Andren and Harriss, 1975; Lindberg and Harriss, 1974; Mierle and Ingram, 1991; Meili *et al.*, 1991; Driscoll *et al.*, 1995; Hurley *et al.*, 1995; Watras *et al.*, 1995; Baeyens *et al.*, 1996; Kolka *et al.*, 1999; Shanley *et al.*, 2002).

Strong ionic binding is one of the most important reactions between mercury and reduced sulfur sites in soil and DOM which facilitates the mobility of mercury across water column and sediment (Wallschlager *et al.*, 1996) and enhanced results in increased water column's mercury concentrations and help in increased sequestration of mercury in sediments (Cossa and Gobeil, 2000). The pore water's mercury that adsorbed to sediment particles may affect the biota. Generally, the highest mercury concentrations in sediments (maximum 35 mg g^{-1}) were found at 30-40 cm depth, corresponding1 to the period of maximum industrial production (Pereira *et al.*, 1998a). However, particulate mercury escapes when the surface sediments are resuspended and transported by the tide to the rest of the lagoon (Pereira *et al.*, 1998b). The contribution of the dissolved mercury was not evaluated well due to the low concentrations found and reported in several studies (Lucas *et al.*, 1986; Pereira *et al.*, 1998a; 1998b; Ramalhosa *et al.*, 2001).

This study presents is the first study dealing with the distributions of reactive mercury in sediments, as well as dissolved organic matter, in Red Sea-Aqaba and contributes basic information required for regional balances of carbon and nitrogen fluxes, budgets and cycles associated with mercury.

2. MATERIALS AND METHODS

Study area: The study area is located at the northern end of the Gulf of Aqaba (**Fig. 1**), which is the northward extension of the desert-enclosed Red Sea. The maximum depth of the gulf is 1830 m; it is 180 km long and 5-26 km wide. Oligotrophic conditions prevail in the gulf waters and evaporation (350 cm y^{-1}) greatly exceeds precipitation (3 cm y^{-1}) (Reiss and Hottinger, 1984). Surface sediment samples were collected by SCUBA divers from the upper layer of the sediment (0-2 cm) using cylindrical plastic tubes for the surface samples and using the cores for the core samples (0-17 cm) , after drying; samples were sieved through 63um sieve and preserved in the freezer until analysis.
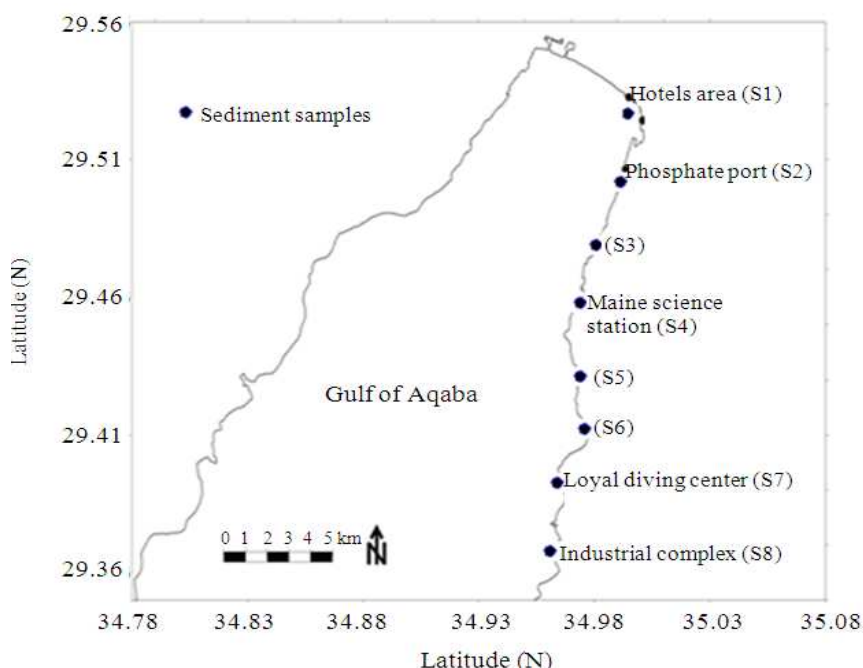

Fig. 1. Sampling site the Gulf of Aqaba (S1; Hotel Area, S2; Phosphate Port, S3; Clink, S4; Marine Station, S5; Station 5, S6; Station6, S7; Royal Diving Center, S8, Industrial Port)

Table 1. Correlation with the certified values

Sample	Certified value Hg_T(µg/g)	measured	% recoveries	Certified value MeHg(µg/g)	measured Range	% recoveries
ERM-CC580	132	134	102	0.075	0.051-0.066	68-88

2.1. Dissolved Organic Matter Extractions

Sediment DOM was extracted by adding 10.0 mL of deionized H_2O to 1.00 g of sediment l in a 15 mL centrifuge tube. The suspensions were shaken on an orbital shaker for 30 min at room temperature (22-18C). Centrifuged at 900× g for 30 min and filtered through 0.45 mm Acrodisk syringe filters. The extraction period was selected to minimize microbial DOM alteration during extraction (Zhou and Wong, 2000).

The concentration of Total soluble Organic Carbon (TOC) in the extracts was determined using a TOC analyzer (Shimadzu's TOC-V Series/Oldenburg University, Germany). Absorbs at 240 nm are obtained using a diode - array detector All DOM solutions were diluted with DI-H2O to set absorbance at 240-0.10 nm to minimize inner filtration effects. Fluorescence measurements were obtained using a spectrofluorometer with the excitation range set from 240-400 nm and the emission range set from 300-500 nm in 3 nm increments. Instrumental parameters were excitation and emission slits, 5 nm; response time, 8 s; and scan speed, 240 nm min^{-1} (Zhou and Wong, 2000).

2.2. Mercury Determination

The total amount of mercury (Hg_T) in sediment samples was determined by taking exact sample weight (200-300 mg) and the samples were analyzed by Hydra-C mercury analyzer, this machine is capable to analyze solid and liquid samples without pre-treatments. For methyl mercury (MeHg) determination, a 0.5-1.0 g sample was extracted using 15 mL (HCl: CH3OH, 1:1 (v/v) for 2 h. After the extraction step, the mixture was centrifuged and the supernatant was separated from the sediment by decantation. Multi extracts were mixed and the pH was adjusted to 4.5 with the addition of acetate buffer, followed by derivatization with the addition of 1 ml of 1% sodium tetraphenylborate. The derivative, phenyl methyl mercury (MeHg Ph) was extracted with hexane and directly analyzed by GC-MS after concentration to about 1mL; solvent exchange to ethyl acetate also was used for the analysis by HPLC-fluorescence technique.

In parallel to this procedure, a derivatization with KBr is now being used for other more samples. For mercury determinations, the US EPA Method 7473 was adopted; aliquots of homogenized samples were weighed (0.1- 0.3 g) in pre-cleaned nickel boats before placement on the Hydra C auto sampler. Samples were analyzed based on the principle of thermal decomposition, amalgamation and atomic absorption spectrophotometry detection. The tuna fish (BCR-463) reference material was analyzed to evaluate the accuracy of the analytical method, the result (Hg: 2.75 µg g^{-1}) was in excellent agreement with the certified value (Hg: 2.85 µg g^{-1}).

Two Certified Reference Materials (CRM's) were used to evaluate the accuracy of the analytical methods; the tuna fish BCR-463 and the estuarine sediment ERM-CC580. The results presented in **Table 1** show excellent correlation with the certified values. Recoveries in the range of (95-102) % and (68-80) % was achieved for Hg_T and MeHg, respectively.

3. RESULTS

3.1. Total Organic Carbon (TOC)

The TOC values range from 7.10-2.05 mg L^{-1} with a mean value of 5.10 mg L^{-1} and a standard deviation of 1.59 (**Fig. 2**) for the all sampling locations in a depth interval of 0-14 cm. These values are not linearly related to depth. The values were relatively low in the continental slope sediments and increased within the continental rise (**Fig. 3**).

3.2. Total Nitrogen (TN)

The TN concentration ranges from 4.23-4.54 mg L^{-1} in the superficial sediments (**Fig. 2**), with a mean value of 4.45 and standard deviation of 0.15 in the all stations sampled at depths from 0-17 cm. The TN concentration in superficial sediments displays a polynomial pattern with increasing depth (**Fig. 4**).

3.3. C/N Ratio

TOC shows a direct relation with TN in the superficial sediments of the deep Gulf of Aqaba and adjusts to a linear regression with a correlation coefficient that explains of the cases (r2 = 0.89; **Fig. 5**). The carbon-nitrogen ratio (C/N) for the samples, from depths of 0-15 cm, has a mean value of 1.14 with a standard deviation of 0.34. C/N does slightly change with increasing depth. The lowest C/N ratios are observed (C/N = 0.48) and the highest (C/N = 1.57).
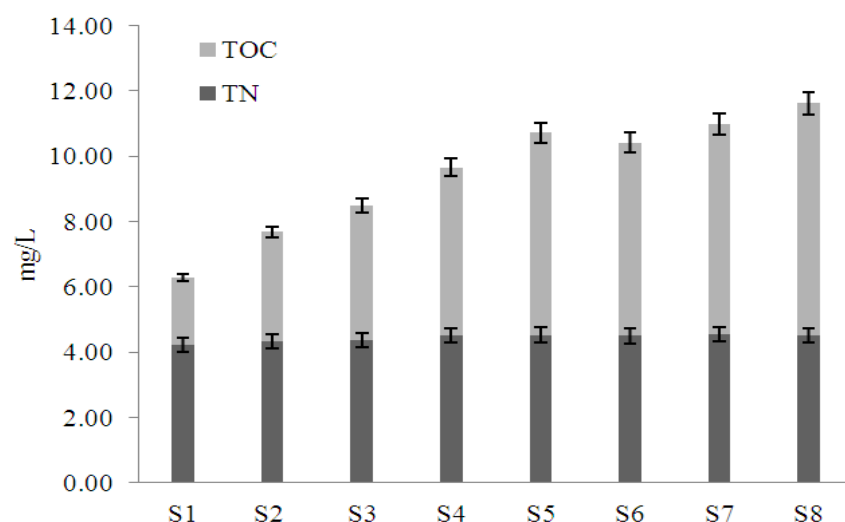
Fig. 2. The total concentrations (mg/L) of Total Organic Carbon (TOC) and Total Nitrogen (TN)
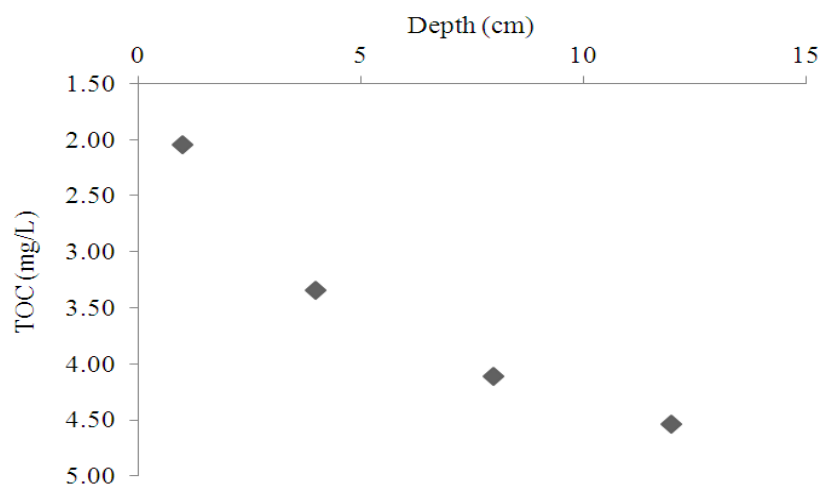

Fig. 3. The total concentrations (mg/L) of TOC with depth (cm)

3.4. Total Mercury

As shown in **Fig. 6**, the concentration of total mercury (all forms of mercury, including inorganic and methylmercury) in the top centimeters of Gulf of Aqaba sediments ranges from approximately 28.9-176.60 ng of mercury per gram of dry sediment and (StD 4.25, 18.0 respectively). While sediment mercury concentration generally correlated with organic matter content, the sharp peak in sediment mercury we observed in the top centimeters does correspond to a high organic matter content. It is instead due to the point source of mercury.

Comparisons between variations of organic matter throughout the sediment cores and the concentrations of Hg were carried out using a leaner regression correlation. The correlation coefficients between the TOC, TN and the concentrations of Hg in the core are shows a significant value (**Fig. 7a and b**). The relationship between Hg and organic compounds throughout the Aqaba sediment core is also shown as a regression plot reiterating a poor relationship between Hg, TOC. However, the linear relationship between Hg and TN indicates that the distribution of Hg in the Aqaba sediment samples mainly relates to the labile compounds.
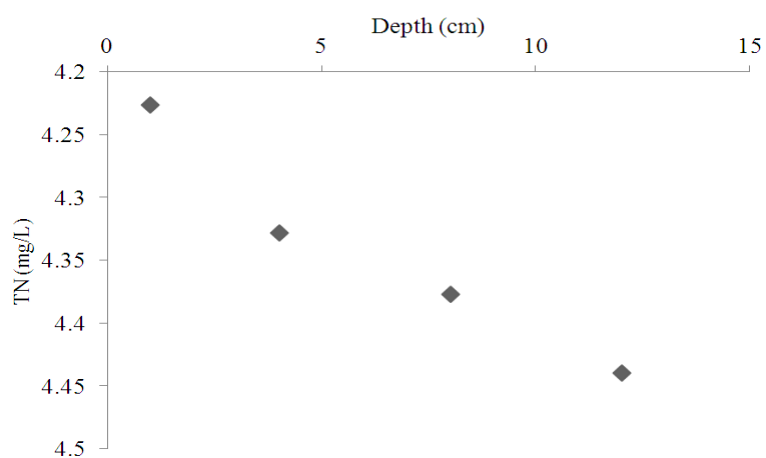
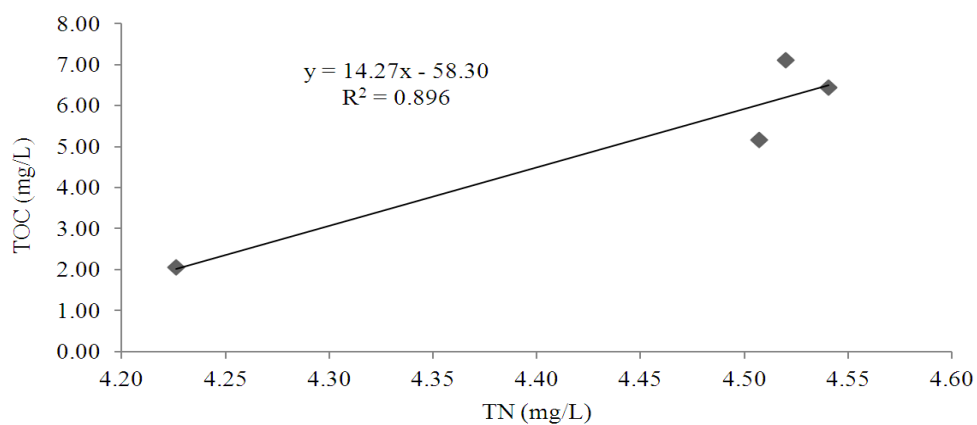**Fig. 4.** The total concentrations (mg/L) of TN with depth (cm)

$$y = 14.27x - 58.30$$
$$R^2 = 0.896$$

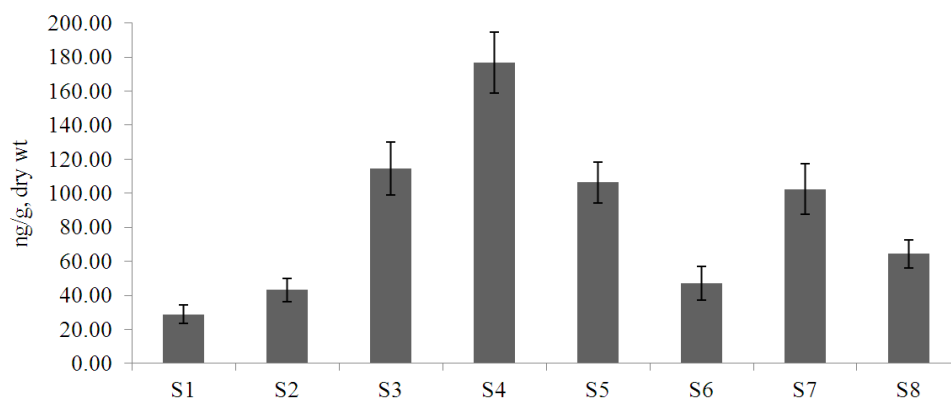**Fig. 5.** Relationships between TOC and TN

Fig. 6. The concentration of total mercury (all forms of mercury, including inorganic and methylmercury (in the top centimeters of Gulf of Aqaba sediments
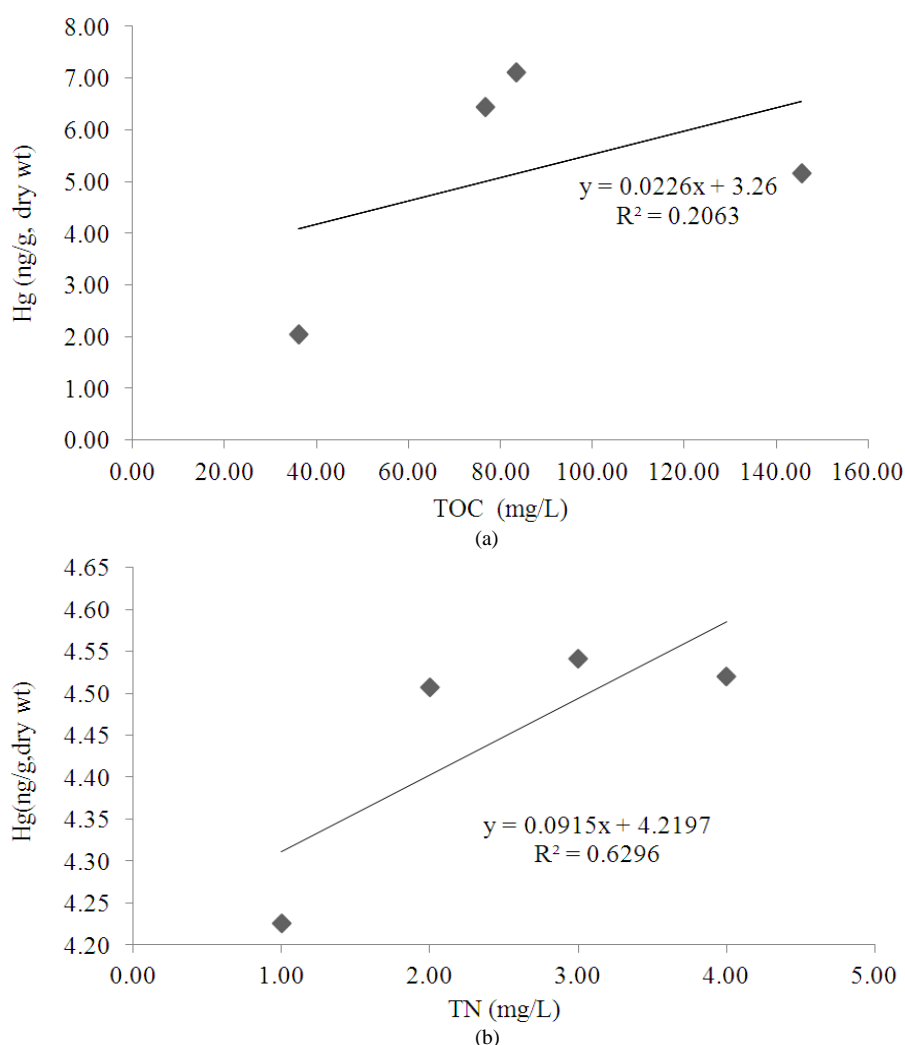
$$y = 0.0226x + 3.26$$
$$R^2 = 0.2063$$

(a)

$$y = 0.0915x + 4.2197$$
$$R^2 = 0.6296$$

(b)

Fig. 7. (a and b) Relationship between Hg and organic matter

4. DICUSSION

Seiter *et al.* (2004) demonstrated TOC values for sediments under well oxygenated conditions are <0.5%. The mean values recorded in this study are slightly higher than expected (1.02 ± 0.3%, range 0.41-1.42%,), but lower than those recorded in the continental margins (>1.5% in Seiter *et al.*, 2004; 4-16% in Arthur *et al.*, 1998). The TOC values are similar to those recorded in the northern Gulf of Mexico (0.34-1.59% in Goni *et al.*, 1997; 1998; Gordon and Goni, 2004; 0.37-1.3% in Morse and Beazley, 2008) and those from other basins at similar latitudes, i.e.,: The Argentina Basin (0.26-1.72% in Stevenson and Cheng, 1972) and the Arabian Sea (0.04-

1.5% in Grandel *et al.*, 2000). The highest value of this study is in agreement with the values described by Seiter *et al.* (2004) for tropical regions, including the continental margin of western Africa.

The TN values span over a narrow range (TN = 0.85-0.91%) which are higher than those expected for the northern sector of the Gulf of Mexico (TN = 0.08-0.17% in Goni *et al.*, 1998; Gordon and Goni, 2004). This is explained by the broadest geographical coverage of this study. The obtained TN values are higher for those from the Argentina Basin (TN = 0.031-0.167% in Stevenson and Cheng, 1972) and than those recorded in the continental margin of the Arabian Sea (0.1-0.4% in Suthhof *et al.*, 2000) and than those recorded in the China Sea (0.02-0.19% in Kao *et al.*, 2003).

The organic material of photoautotrophic origin mixes with continental organic matter near the coast and is usually transported by turbidity currents to the abyssal plain (Suthhof *et al*., 2000; Mari *et al*., 2001; Epping *et al*., 2002; Gordon and Goni, 2004). Values of Carbon-Nitrogen atomic ratio (C/N) suggests an organic matter input derived from vascular plants (Meyers, 1994), characterized by limiting nitrate concentration (Mari *et al*., 2001). The mean value of our study (C/N = 1.41 ± 0.34) is lower the Redfield ratio (C/N = 5.7; Gordon and Goni, 2004; Weston and Joye, 2005). The TN of the sediments avoids being remineralized into the water column by adsorption to the mineral clay surface, hence recording smaller C/N ratios (Stevenson and Cheng, 1972; Suthhof *et al*., 2000) that are explained by selective degradation of the organic compounds and by immobilization of N by microorganisms during early diagenesis (Meyers, 1994). Other factors that can indicate the origin of organic matter in the sediments, but not performed in this study, include the Hydrogen index, microscopic observations and carbon stable isotopic composition (Stein, 1990; 1991). The stable isotope composition of particulate organic matter in deep seawater samples from the same stations in the Gulf of Mexico ($\delta13C$ -25.3 to -22.70 ‰ in the abyssal plain and $\delta13C$ -24.11 to -22.12 ‰ in the continental slope) indicate a photoautotrophic origin, with depleted values caused by the time of residence in the water column with a contribution of suspended material from the sediment (González-Ocampo, 2005; González-Ocampo *et al*., 2007).

The surface sediments collected in this study presented mercury concentrations comparable to the values recorded in a previous study (Pereira *et al*., 1998a). In the present study, the highest mercury contents were found in the area near the industrial discharge and were positively correlated to both the organic carbon and total nitrogen. The organic carbon present sediments may result from industrial effluents and also from the decomposition of extensive algae coverage and plankton inside the gulf, as stated by Monterroso *et al*. (2003).

The previous studies of Schuster *et al*. (2008) and Selvendiran *et al*. (2008a) showed that there is a connection of wetlands and riparian areas to streams during hydrologic events can flush stored organic matter and Hg to downstream ecosystems. Both dissolved and particulate C plays an essential role in the transport of Hg fractions. The result of the this study is in agreement with other studies in the region that report DOC as an important transport mechanism for THg (Driscoll *et al*., 1995; Selvendiran *et al*., 2008b; Shanley *et al*., 2008).

There was a consistent relationship between Hg and DOC across our sites with, which is higher than the studied reported by Grigal (2002) for sites across the Northern Hemisphere. The similar slopes between our study and those reported by Grigal (2002) for the THg DOC relationship are likely a function of strong correlation between FTHg and the export of humic material associated with hydrophobic organic matter (Mierle and Ingram, 1991; Grigal, 2002).

5. CONCLUSION

Sediments can be a source of dissolved mercury and organic matter because of the activity of microorganisms. As the deposited mercury is buried beneath new layers of sediment, organic matter in the sediment is consumed by certain types, by bacteria living a few millimeters below the sediment water interface, beyond the reach of oxygen. These bacteria get their energy through chemical reactions that involve sulfur and iron and produce toxic methylmercury as a byproduct.

6. ACKNOWLEGMENT

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