Polycyclic aromatic hydrocarbons in sediments from Sontecomapan Lagoon, Veracruz, Mexico

Laura Georgina Calva B.[1]
y Alfonso V. Botello[2]

[1]Laboratorio de Ecosistemas Costeros. Departamento Hidrobiología. Universidad Autónoma Metropolitana Iztapalapa. México, D.F., c.p. 09340.
[2]Laboratorio de Contaminación Marina. Instituto de Ciencias del Mar y Limnología. Universidad Nacional Autónoma de México. Apdo. Postal 70305, Mexico, D.F. c.p. 04510.

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ABSTRACT

In this study a determination of the concentrations of 15 polycyclic aromatic hydrocarbons (PAH) in surface sediments from Sontecomapan Lagoon, Veracruz was made; these compounds were evaluated by means of gas chromatography using capillary columns of high resolution and flame ionization detection (GC-FID). During the period 1991 to1992, it strengthen that exist differences between dry and rainy season, dry period showed the highest total PAH's concentrations ranged from 25.42 to 49.49 μg/g of dry sediment and in rainy period these ranged was recorded from 10.15 to 21.98 μg/g. In descending order, of the individual compounds of PAH's in dry season were: Chrysene (49.66 μg/g) > Benzo(a)anthracene (14.57 μg/g) > Benzo(b)fluoranthene (8.23 μg/g) > Benzo(k)fluoranthene (5.53 μg/g) > Anthracene (5.44 μg/g) > Pyrene (5.41 μg/g). For rainy season were: Chrysene (27.92 μg/g) > Benzo(a)pyrene (2.71 μg/g) > Benzo(k)fluoranthene (2.62 μg/g) > Benzo(a)anthracene (2.58 μg/g) > Benzo(b)fluoranthene (2.25 μg/g) > Pyrene (2.02 μg/g). The presence of 4, 3 and 5 benzene rings in PAH's suggests that these compounds are primarily of pyrogenic origin, this is the result of surrounding vegetation fires, the lubricants, and gasoline. PAH's goes into lagoon system through river discharges, by tidal action from the coast and from atmospheric transport.

Keywords: pollution, polycyclic aromatic hydrocarbons (PAH's), sediments, Sontecomapan, Veracruz.

RESUMEN

En este estudio se determinaron las concentraciones de 15 hidrocarburos aromáticos policíclicos (HAP) en sedimentos superficiales de la Laguna Sontecomapan, Veracruz por medio de cromatografía de gases de alta resolución empleando columnas capilares y detector de ionización de flama. Durante el período 1991-1992, la época de secas presentó las mayores concentraciones de HAP totales en sedimento seco en un intervalo de 25.42 a 49.49 μg/g y en el período de lluvias su concentración fue de 10.15 a 21.98 μg/g. Respecto al desarrollo jerárquico de los HAP individuales, en época de secas en orden descendiente estuvieron: Criseno (49.66 μg/g) > Benzo(a)antraceno (14.57 μg/g) > Benzo(b)fluoranteno (8.23 μg/g) > Benzo(k)fluoranteno (5.53 μg/g) > Antraceno (5.44 μg/g) > Pireno (5.41 μg/g). Para lluvias: Criseno (27.92 μg/g) > Benzo(a)pireno (2.71 μg/g) > Benzo(k)fluoranteno (2.62 μg/g) > Benzo(a)antraceno (2.58 μg/g) > Benzo(b)fluoranteno (2.25 μg/g) > Pireno (2.02 μg/g). La presencia de HAP conformados por 4, 3 y 5 anillos bencénicos sugiere que estos compuestos son principalmente de origen pirogénico, por la quema de vegetación circundante, gasolina y lubricantes. Los HAP ingresan al sistema a través de las descargas de los ríos, la acción mareal de la costa y por transporte atmosférico.

Palabras clave: contaminación, hidrocarburos aromáticos policíclicos (HAP), sedimentos, Sontecomapan, Veracruz.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are environmental pollutants that constitute a potential risk to public health and to other forms of life (Martel *et al.* 1986). Some of them, such as benzo(a)pyrene, are powerful carcinogens, associated with the etiology of some human cancers. Nowadays there is a great deal of literature about the levels of PAHs in coastal systems around the world (e.g. Abu-Hilal and Khordagui, 1994; Botello *et al.* 1993; Valette, 1993; Zhang *et al.* 1993) and most of these studies have been carried out in areas that receive the direct influence of industrial and municipal discharges.

However, literature specifically about the presence and distribution of PAHs in coastal and estuarine environments of tropical areas, as well as the effects of PAHs on marine biota is less abundant. Such studies are necessary in order to understand the behavior, mechanisms of transport, and bioaccumulation of PAHs in estuarine species. Sontecomapan Lagoon, Veracruz, is situated on the Gulf of Mexico. It is a highly productive ecosystem of mangrove forest, which has been affected during the last two decades by immoderate tree-felling, primarily for agricultural purposes, specially in Los Tuxtlas region (Menendez, 1976; Angeles, 1997), where the most important activity is the tobacco industry, which uses the mangle tree with the aim of building galleries and burn it to dry tobacco leaves. The above mentioned is important because PAH's arise from natural forest and prairie fires (Wakham *et al.* 1980), most of PAH's in the environment are derived during the incomplete combustion of organic matter at high temperatures (Neff,1979).

The sediments of this system is constituted of silt-clay in the south and center of the area, while the northern zone is mainly sand. It is important to mention that polycyclic aromatic hydrocarbons, PAH's, are associated with fine particles in the coastal waters and their range the distribution pattern and extension of the accumulation of PAH's in the estuarine systems are extremely variable (Olsen *et al.*1993). Recent studies indicate that PAH's are not only detected in urban areas, but also in rural and remote areas, due to atmospheric transport (Valette, 1993).

The objective of this study were to evaluate the concentrations, distribution and origin of PAH's with different molecular weight in the sediment samples in order to investigate correlation degree between those PAH's and the percentage of organic carbon, as well as to estimate the preference of accumulation of these compounds in the different season and types of sediments in the lagoon.

MATERIALS AND METHODS

Sontecomapan Lagoon is located in the South of Veracruz State on the Gulf of Mexico, between 18°30'-18°34' N and 94°54' - 95°02' W, it has an area of approximately 8.9 Km^2 and has been an important fishing town (Fig. 1). This study comprises six sampling times between March 1991 up to August 1992 (sampling time was between the 08:00 and 15:00 hr) during the dry and rainy season, respectively. Each one sampling period the temperature was evaluated using a cup thermometer (precision of 0.1 °C), the salinity was recorder using an "American Optical" refractometer (precision of 0.5 o/oo) and the pH was measured with a "Conductronic" field potentiometer (exactitude of 0.1 pH unites).

Surface sediments were taken from 8 sites through lagoon surface by means of a small stainless steel van Veen grab sampler (6 L) and frozen (4°C) in glass jars previously cleaned and rinsed with bidistilled acetone. In the laboratory these were defrosted, and dried through 48

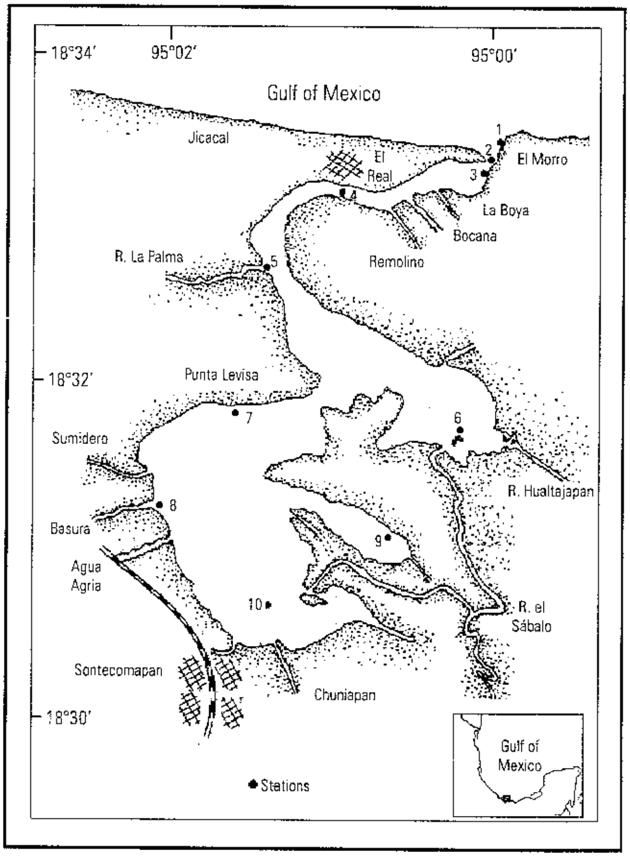

Figura 1. Sampling sites for sediments in Sontecomapan Lagoon, Ver.

hr at 45°C, then the sediments were sifted in a mesh to 0,25 mm.

The analytical procedures for extraction and purification of PAHs were carried out by the method of CARIPOL (1986), according to UNEP (1992). Each set of 3 samples was accompanied by a complete blank and a spiked blank. 10 g of dried sediment were soxhlet extracted with methanol (100 ml) and KOH, and standards additions were added before extraction. The aromatic standard addition contains phenanthrene or fluoranthene 200 µl. The saturated and aromatic fractions were purified by adsorption chromatography using 20 cm long columns, packed with alumina (5% deactivated with water). The solvent elution's for the fraction 1 (saturated) was make with hexane. Fraction 2 contained aromatics was eluted with hexane:methylene chloride (7:3) and with methylene chloride. The extracts were evaporated to near 2 ml in a rotary flask evaporator and analyzed directly by gas chromatography.

Quantification of the aromatic fraction was make by means of a Hewlett Packard gas chromatograph model 5890 equipped with 30 m x 0.25 mm ID x 0.25mm bonded 5%-phenilmethylsilicone, fused silica column (temperature programmed 40 - 300°C at 6 °C/min). Nitrogen was used as carrier gas (flow 1 ml/min). Quantification performed was using a mixture of PAH's with well-known standards of the "Chemical Service" PPH-10M. The limit of detection for individual aromatic compounds was 0.01 µg/g and recovery yields were up to 90%. The accuracy and precision of the method employed is accredit by intercomparison exercise Internatinal Atomic Energy Agency (IAEA-140) for Petroleum Hydrocarbons (1997).

Organic carbon determination was based on method of Gaudette *et al.* (1974) in which exothermic heating and oxidation with K_2CrO_7 and concentrated H_2SO_4 are followed by tritation of the excess dichromate with 0.5 N $Fe(NH_4)_2(SO_4) \times 6H_2O$.

Sediment composition (sand, silt, and clay content) was measured by standard wet sieving and pipette analysis (Folk, 1974).

In order to determinate if there was significant difference in PAH's concentrations between the dates of collection in two years (dry and rains) it was carried out an ANOVA. For multiple comparisons among means a least significant differences (LSD) was used. Statistical analysis and data transformation were conducted using STATISTICA for Windows release 4.5 (STATISTICA 1993). The limit of significance was settle down with an alpha error of 0.05 (Daniel, 1995).

RESULTS AND DISCUSSION

Our results showed that Sontecomapan Lagoon is oligo-polyhaline, according to the Venice classification system with a range since 0-27; with an average salinity of 5.8 in the southern zone and 13.9 in the northern zone. The lagoon connects with the sea by means of a channel approximately 4m in wide. The central area has a range of salinity from 1 to 8, and the southern zone has an average salinity of less than 2.7, manifesting the importance of the fluvial input. The average water temperature (27.1 °C) demonstrates a close relationship with the environment temperature (26.9 °C), due to the shallowness of the system. The average depth is 1.67 m with a water transparency of 0.68 m and pH values were 7.4.

Table 1 shows the total concentration of PAHs, the sediment characteristics, and the percentage of organic carbon present in the samples, taken from 8 different sites in Sontecomapan Lagoon. These samples were collected during the cycle 1991-1992. The percentages for organic carbon remained below 4%, with the highest average of 1.88% in September 1991, and the lowest 1.13% in August 1992; also during March 1991 was found the highest concentrations of total PAHs (49.49 µg/g), decreased in November 1991 (25.93 µg/g), and the lowest concentrations were registered in July (10.46 µg/g), and September, (10.15 µg/g), which corresponds to the rainy season in that area.

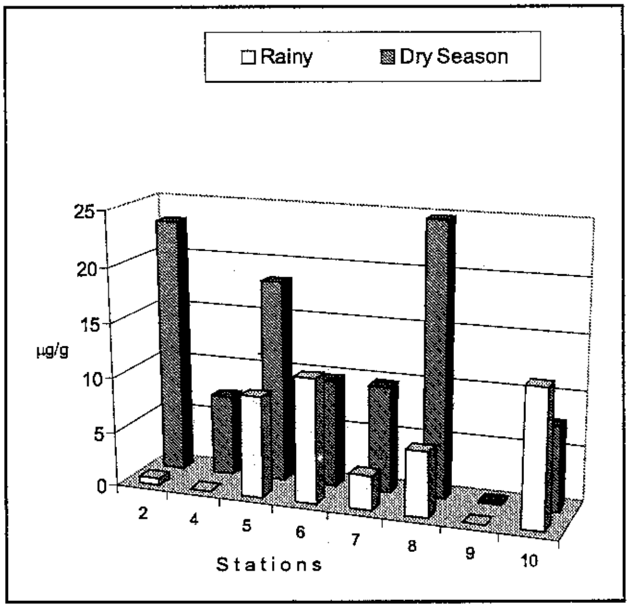

Figura 2. Levels of total PAH's in sediments in dry and rainy season from Sontecomapan Lagoon, Ver. Cycle 91-92.

Table1. Distribution of PAH's (µg/g) in Sediments from Sontecomapan Coastal Lagoon, Ver., Mexico. Cycle 1991-1992.

Station	Type of Sediment	Organic Carbon (%)	March 1991	July 1991	September 1991	November 1991	March 1992	August 1992	Total PAH's
2	Sand	0.24	12.06	<0.01	<0.01	10.22	0.85	0.55	23.68
4	Sand	0.5	3.91	<0.01	<0.01	2.9	0.52	<0.01	7.33
5	Sand	0.76	11.01	5.77	<0.01	7.35	<0.01	3.41	27.54
6	Sand-Clay	1.76	0.66	3.14	4.64	0.26	8.83	3.5	21.04
7	Sand-Clay	0.49	0.43	<0.01	3.18	4.12	5.12	<0.01	12.85
8	Clay-Silt	2.3	13.98	1.55	2.31	0.69	10.1	2.02	30.65
9	Clay-Silt	2.38	0.14	<0.01	<0.01	0.02	<0.01	<0.01	0.16
10	Clay-Silt	3.17	7.3	<0.01	0.02	0.37	<0.01	12.49	20.18
Total PAHs			49.49	10.46	10.15	25.93	25.42	21.98	
Mean PAHs			14.56	6.18	3.17	5.46	8.01	7.86	
Est.Desv.			5.67	2.13	1.86	3.79	4.25	4.2	
O.C.%			1.41	1.28	1.88	1.57	1.68	1.13	

In dry season (March and November, 1991; March, 1992) the sampling stations that showed high concentrations of total PAHs were station 8 (24.77 µg/g) situated in the shared outlet of the Agua Agria river, the De Basura river and the Del Sumidero river; station 2 (23.13 µg/g) located near the mouth of the lagoon; and station 5 (18.36 µg/g) which is influenced by discharges from the La Palma river (Fig. 2).

It is clear that some PAH's are coming from the coast, and get into the lagoon system, this was confirmed in this study by the fact that the greatest average concentrations of dissolved hydrocarbons were detected in the area around the mouth of the lagoon, with 10.17 μg/l, and by the observation of patches of tar all along the external bar of the lagoon. This as consequence of the fact that PEMEX (Petróleos Mexicanos) has oil-drilling platforms in the Gulf of Mexico, and because of the intense traffic of ships and oil-tankers that circulate in the region.

Botello and Villanueva (1992) refer the fact that the petroleum off-shore explotation is particularly important in the Gulf of Mexico, where a large number of off-shore drilling platforms operate. Furthermore, at approximately 80 Km to the south of Sontecomapan Lagoon, the rivers Coatzacoalcos and Tonalá run into the sea, and along their banks are Latin America's biggest petrochemical complexes.

For rainy season (July and September, 1991; August, 1992) the station with the highest concentrations were 10 (12.51 µg/g) which is at the mouth of the Chuniapan river, the Sontecomapan river and the Coscoapan river, and where there is also the main municipal waste outflow that born in Sta. Martha´s mountain range, and station 6 (11.29 µg/g))

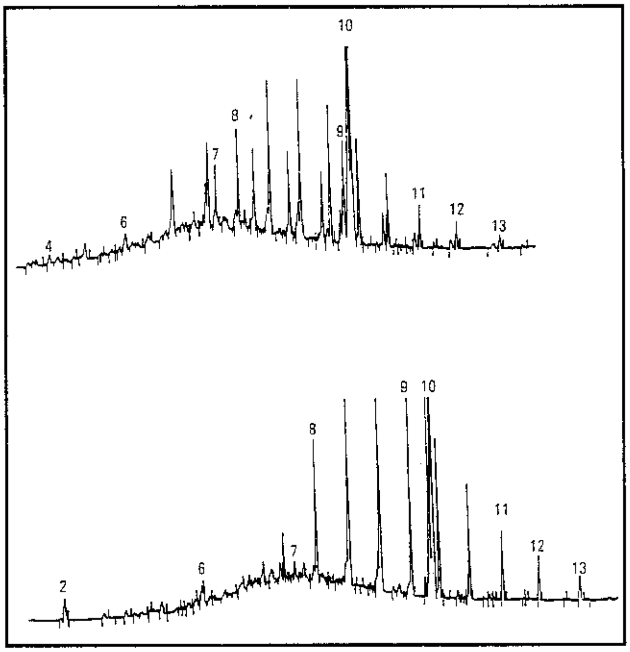

Fig. 3 Chromatograms GC-FID of PAH compounds (µg/g dry wet) in sediments collected in sediments a) Station 8-March, 1991 and b) Station 6-March, 1992 sampled at Sontecomapan Lagoon. Peak identification is as follows: 2, acenaphthylene; 4, fluorene 6, anthracene; 7, fluoranthene; 8, pyrene; 9, benzo(a)anthracene; 10,chrysene; 11, benzo(b)fluoranthene; 12, benzo(k)fluoranthene; 13, benzo(a)pyrene.

in El Cacahuate Estuary that receives the outflow of the Pollos river (Fig. 3), the Hualtajapan river and the Sábalo river and lastly stations 5 (9.18 μg/g)) and 8 (5.88 μg/g). Station 9 registered the lowest concentrations of PAH's (0.16 μg/g)) in dry season, it was located in the Del Fraile Estuary which is influenced by the river of the same name. For rains period both station 9 and 4 stations didn't show PAH's presence and, in the station 2 the level was of 0.55 μg/g), both areas located in the zone of marine influence of the lagoon.

The differences between dry and rain seasons were significant (t-Student's; $P = <0.035$ two-sample assuming unequal variances), the period with highest mean levels of PAH's was dry season 33.61 μg/g (25.42-49.49 mg/g) with respect to rainy season with 14.2 μg/g (10.15-21.98 μg/g).

In general, for the cycle 1991-1992 Stations 2 and 5 registered a clear predominance of chrysene (22.6 and 22.48 μg/g). However, ten to eleven different PAHs made up of from 2 to 5 benzene rings, were detected in stations 6, 8 and 10 indicating differential and fate and transport process may be impacting (Table 2).

Figure 4 shows the major total PAHs identified in the sediments in Sontecomapan Lagoon in dry season their concentrations are as follows: chrysene (49.66 μg/g), benzo(a)anthracene (14.57 μg/g), benzo(b)fluoranthene (8.23 μg/g), benzo(k)fluoranthene (5.53 μg/g), anthracene (5.44 μg/g) and pyrene (5.41 μg/g).

The principal PAH's in rainy season were similar but their concentrations are lowest than dry season: chrysene (27.92 μg/g), benzo(a)pyrene (2.71 μg/g), benzo(k) fluoranthene (2.62 μg/g), benzo(a)anthracene (2.58 μg/g), benzo(b)fluoranthene (2.25 μg/g) and pyrene (2.02 μg/g).

In agreement with the results obtained with ANOVA, it strengthens that exist differences between dry and rain seasons, this could be the result to that in rainy period the lowest levels detected were due to that PAH's were rapidly removed by sedimentation processes coupled with complex phenomena occurring at the fresh water/marine water interface.

LSD test results include the three sampling epoch during dry season nevertheless statistical approaching shows that August results are similar to dry season ones,

In Sontecomapan Lagoon a rapid transport of PAH's through the water column is supported by the high accumulation of PAH in sediments near the river mouths. Kennish (1992) pointed out that even though the pollutants are trapped in the sediments, they are able also to pass

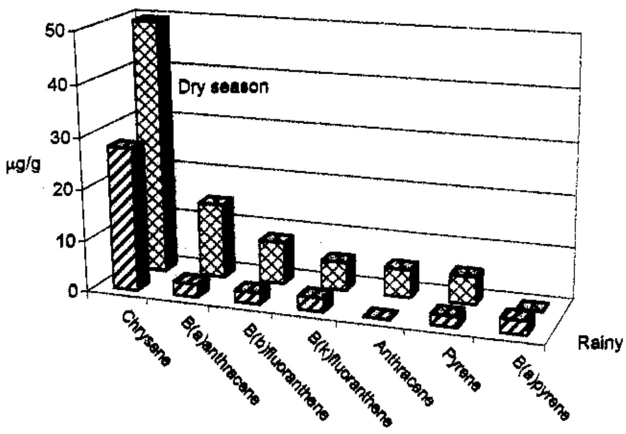

Fig. 4. PAH's distribution in dry and rainy season from Sontecomapan Lagoon, Ver. Cycle 91-92.

back into suspension, which results in a solution of the hydrocarbons in the water column.

In this lagoon 4 benzene rings PAH's represent the highest percentages (84.21%), then were 3 rings compounds (9.32%), 5 rings (4.46%), and finally 2 rings (2.01%). Naphthalene and phenanthrene are relatively rapidly degradable, nevertheless PAH's of high molecular weight are specially resistant and persistent in the environment for long periods of time (Pitter and Chudoba, 1990).

The acquirement of the ratio phenanthrene/anthracene and fluoranthene/pyrene allows us to appreciate the source of contamination. Hence, a ratio of phenanthrene/anthracene <10 and fluoranthene/pyrene >1 indicated that the contamination by PAH's is due to combustion processes (Raoux, 1991). The concentration ratios calculated for phenanthrene/anthracene ranged from 0.12 to 0.6 with a mean concentration ratio of 0.36, and for fluoranthene/ pyrene ranged from 0.32 to 2.5 with a mean concentration ratio of 1.24. This finding suggests a general common origin for PAH's, possibly pyrolysis related.

A featured noted in Table 2 is the dominance of the 4-ring PAH's over 3-ring compounds with the corresponding mean concentration of 21.2 mg/g , which suggest too a pyrolytic origin for the compounds detected in this study.

Steinhauer and Boehm (1992) observed that the grain size in sediments affected considerably the PAH distribution. In muddy and clay sediments, the concentrations are higher than those registered in sandy sediments. In this sense, correlating the sediment grain size data reported by González *et al.* (1994), with the PAHs values registered in this study, no significant values were found in the Sontecomapan Lagoon (gravel r=0.08, $P=0.02$; sand r=0.22, $P=0.14$;

Table 2. Total levels of PAH's (µg/g) in sediments from Sontecomapan coastal lagoon, Ver., Mexico. Cycle 1991 - 1992

Compound	Stations								
	2	4	5	6	7	8	9	10	Total
Naphthalene	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Acenaphthylene	<0.01	<0.01	<0.01	<0.01	<0.01	0.84	<0.01	1.01	1.85
Acenaphthene	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Fluorene	<0.01	<0.01	<0.01	0.5	<0.01	<0.01	<0.01	0.54	1.04
Phenanthrene	<0.01	<0.01	<0.01	0.04	<0.01	0.03	<0.01	0.52	0.59
Anthracene	<0.01	1.39	1.25	1.33	<0.01	0.9	<0.01	1.86	6.73
Fluoranthene	<0.01	1.31	0.77	1.56	0.68	1.42	0.02	0.28	6.04
Pyrene	0.26	1.06	1.34	1.44	0.76	2.1	0.14	0.33	7.43
Benzo(a)anthracene	0.82	2.36	1.06	1.51	1.29	9.06	<0.01	1.05	17.15
Chrysene	22.6	0.4	22.48	7.29	8.05	3.95	<0.01	12.81	77.58
Benzo(b)fluoranthene	<0.01	0.36	0.64	3.06	0.93	4.59	<0.01	0.9	10.48
Benzo(k)fluoranthene	<0.01	0.45	<0.01	2.48	0.7	4.02	<0.01	0.5	8.15
Benzo(a)pyrene	<0.01	<0.01	<0.01	1.83	0.44	3.74	<0.01	0.38	6.39
Indeno(1,2,3-cd)pyrene	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Benzo(ghi)perylene	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Total	23.68	7.33	27.54	21.04	12.85	30.65	0.16	20.18	143.43

mud $r=-0.20$, $P=0.19$; and clay $r=-0.33$, $P=0.06$). The organic carbon results correlated significant with those of the fraction for fine grain sediments (mud $r=0.90$, $P=0.002$; clay, $r=0.91$, $P=0.13$), while the fractions for large grain sediments are negative (gravel $r=-0.49$, $P=0.005$; sand $r=-0.93$, $P=0.00003$). It was also found that the organic carbon content diminishes in relation to an increase in grain size. This demonstrates that there is an association between the organic carbon content and the sediment grain size, but that there is no correlation between the percentage of organic carbon and the PAHs concentration (dry season $r=-0.25$, $P=0.04$, and rainy season $r=-0.38$, $P=0.14$).

Given that PAHs have a low solubility and are hydrophobic in nature, they tend to be enriched in the organic and inorganic particles in the air, and can be carried to all ecosystems by the action of atmospheric agents (Guzzella and De Paolis, 1991).

On the studied area, the results obtained here indicate that exists significant atmospheric transporting of PAHs, given that there are no industries around the lagoon system. In this regard, Valette (1993) states that PAHs are not only to be found in urban areas but also in remote rural areas, as a result of atmospheric transfer. On the other hand, Lipiatou and Saliot (1991) report a PAHs concentration of 0.18 µg/g in areas that do not receive a direct inflow of hydrocarbons, and this finding leads to the conclusion that these compounds are being transported to the Sontecomapan Lagoon from urban and industrial areas, given the high concentrations determined in this present study.

The results obtained are compared to those from other coastal systems along the Gulf of Mexico, the observation show that this lagoon is one of the areas with high PAHs mean values (7.54 µg/g), levels comparable to those of Tampamachoco, Tamps. (7.65 µg/g) in the north of Veracruz, an affected area by discharges from industrial complexes. In descending order, these mean values are followed by those registered for Tamiahua Lagoon, Ver. (6.86 mg/g) and Pueblo Viejo Lagoon, Ver. (6.27 µg/g) (Botello and Calva, 1998).

CONCLUSIONS

From this present investigation the conclusion is drawn that, in spite of not being an urban area nor being surrounded by petrochemical or other industrial complexes, Sontecomapan Lagoon, Veracruz, is receiving inadvertent additions of PAH's. These PAH's are primarily of pyrogenic origin, coming from the inflows of the rivers; from the coast; from the spillage of oils, lubricants and gasoline; from the burning of surrounding vegetation; and from atmospheric transport.

During the period from 1991-1992, it streghten that exist differences between dry and rain season, dry period showed the highest total PAH's concentrations indicating the importance of pluvial and fluvial sources as well as sedimentation processes coupled with complex phenomena occurring at the fresh water/marine water interface. In Sontecomapan Lagoon a rapid transport of PAH's through the water column is supported by the high accumulation of PAH in sediments near the river mouths.

The predominant PAH compounds in sediment samples are, in descending order: Chrysene > Benzo(a)anthracene > Benzo(k)fluoranthene > Pyrene > Anthracene > Benzo(a)pyrene. The presence of compounds made up primarily of 4 and 5 benzene rings, proves the hypothesis that the origin of the PAH's in the lagoon is anthropogenic, justifies from the pyrolysis of organic matter and fossil fuels.

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