Available online at http://ajol.info/index.php/ijbcs

Int. J. Biol. Chem. Sci. 9(2): 1120-1129, April 2015

ISSN 1997-342X (Online), ISSN 1991-8631 (Print)

International Journal of Biological and Chemical Sciences

Original Paper http://indexmedicus.afro.who.int

Distribution of Polycyclic Aromatic Hydrocarbons (PAHs) in a tropical coastal lagoon (Grand-Lahou lagoon, Côte d'Ivoire)

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ABSTRACT

The distribution of 8 polycyclic aromatic hydrocarbons (PAHs) in the surface sediments of the Grand-Lahou lagoon (Côte d'Ivoire) was investigated using a high performance liquid chromatography (HPLC). The total concentrations of PAHs were between 1.55 and 437.52 µg/g in the dry season. Pyrene, benzo (b) fluoranthene and fluoranthene have the highest concentrations. In the rainy season, the total PAHs concentrations varied between 46.35 and 1222.73 µg/g. Pyrene, benzo (a) pyrene and benzo (a) anthracene were the most present in the sediments of the lagoon of Grand-Lahou during the wet season. Possible sources of PAHs in the lagoon of Grand-Lahou are oil and fuel spills from ships and fishing boats, combustion of waste, domestic waste disposal and especially the production of charcoal on the Grand-Lahou lagoon shores. A clear dominance of high molecular weight (HMW) PAHs (pyrene, Benzo (k) fluoranthene, Benzo (a) pyrene, indeno (1,2,3-cd) pyrilène, Benzo (g, h, i) pyrilene, Benzo (a) anthracene , Benzo (b) fluoranthene) was observed compared to low molecular weight (LMW) PAHs (fluoranthene).

Keywords: Polycyclic Aromatic Hydrocarbon (PAH), coastal lagoon, pollution, West Africa.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are neutral nonpolar organic compounds, consisting of at least two aromatic rings containing only carbon and hydrogen atoms. They are mainly formed during the incomplete combustion of organic matter (pyrolitic sources) (Eisler, 1987; Baek et al., 1991) and during the slow maturation of the organic matter accumulated in deep sedimentary environments (petrogenic sources) (Wang et al., 2004). These two sources have distinct formation mechanisms that are realized with different kinetics. PAHs from pyrolitic and petrogenic sources exhibit different chemical behaviours and distribution in sediments. In particular, PAHs from pyrolitic processes are more strongly associated to sediments and much more resistant to microbial degradation than PAHs of petrogenic origin. PAHs may also be formed by micro-organisms from biogenic precursors such as di- and tri-terpenes, steroids, pigments or quinones in sediments or recent soils (Wakeham et al., 1980); these precursors may come from terrestrial or

DOI : http://dx.doi.org/10.4314/ijbcs.v9i2.47

aquatic biological tissues (plants, animals, bacteria, macro- and micro-algae) (Wang et al., 2004).

PAHs are ubiquitous compounds present in all environmental compartments (atmosphere, water column, biota, sediment, soil). In the aquatic environments, due to their hydrophobic nature, PAHs are mostly adsorbed on the particulate material and are associated to organic matter due to their physicochemical properties. They reach then the aquatic bottom by decantation of particles; thus the sedimentary mass constitutes the major reservoir of hydrophobic compounds (Neff, 1979). For this reason, the sediments are of great interest in the environmental assessment of aquatic ecosystems.

Polycyclic aromatic hydrocarbons are among the most monitored pollutants particularly in shallow coastal areas subject to anthropogenic inputs and natural emissions (Li et al., 1998). Accumulation and persistence of PAHs present a risk to human health and the environment, firstly due to their characteristics and also because of the many sources of exposure. PAHs have toxic, mutagenic and carcinogenic properties (Juhsz and Naidu, 2000; Semlali et al., 2012). However, very few studies have been conducted in African tropical coastal lagoons (Soclo et al., 2000; Nyarko et al., 2011; Semlali et al., 2012).

In Côte d'Ivoire, the coastal region is characterized by the presence of many lagoons of which the most important are, from west to east, the Fresco, the Grand Lahou, the Ebrié and the Aby lagoons. These coastal ecosystems play an important role in the reproduction of fish and shrimp, and are irreplaceable habitats for many species of migratory and local birds (Kouassi, 2005). They are also important for the Ivorian economy since they provide suitable areas for fishing, aquaculture, tourism, transport activities, etc. Unfortunately, these lagoons undergo intense human activities (production of charcoal, fisheries, transport ...) susceptible of causing PAHs pollution.

This work aims at studying the distribution of PAHs in superficial sediments of the Grand Lahou lagoon (Côte d'Ivoire).

MATERIALS AND METHODS

Study site

The Grand-Lahou ecosystem (Figure 1), with an area of about 190 km², is located between 5°25'W and 5°10'N. It is actually a complex of four small lagoons (Figure 1) comprising:

- The Tagba lagoon located at the east side, with a mean depth of 3 m. It covers an area of approximately 57 km². However, there are some areas (close to the inlet) as deep as 8 m. It communicates with the Atlantic Ocean and receives the Bandama River;
- The Mackey lagoon, with up to 2 m depth, is the shallowest. It receives waters from the Gô and Boubo Rivers, has an area of about 28 km² and is connected to the Tagba and Tadjo lagoons;
- The Tadjo lagoon with an area of 90 km^2, is the largest. It is strongly influenced by the Boubo river;
- The Niouzoumou lagoon directly communicates with the Tadjo lagoon. It is parallel to the barrier beach. It has an area of 15 km^2 and a maximum depth of 3 m. It connects to the Tagba lagoon by an artificial channel called Groguida canal.

The Grand-Lahou lagoon is fed by the Bandama and the Boubo Rivers. The Bandama River has a total length of 1,050 km (Laé, 1982), a total catchment area of 97,000 km² and an annual average rate of about 172 m^3/s (Girard et al., 1971). According to Durand and Chantraine (1982), the Bandama River alone contributes to 95% of the continental inputs. The Boubo is characterized by two flood seasons with an average flow of 29.10 m^3/s in June and about 16 m^3/s in November. Between the two flood periods, there is an inter-flood which usually occurred during the month of August with an average flow of 13.01 m^3/s. The Grand-Lahou lagoon also directly receives rainfall which represents 5% of the continental inputs (Durand and Chantraine, 1982). It is connected to the Atlantic Ocean by a channel (Hie, 1986) whose width is between 150 and 200 m (Abe et al., 1993). The oceanic waters volume penetrating in the lagoon has not been assessed.

The Grand-Lahou lagoon system is supplied with sediment from rivers, streams and runoff. The sediment is composed of sand and silt grain from 1.6 to 45.10^{-6} m. The channels, with depths between 3 and 5 m are covered with vases and clay to fine grains less than median 5.10^{-6} m (Abe et al., 1993). The water content of these vases is high: 200 to 700% by weight of dry sediment. Minerals are found in decreasing order of importance, kaolinite, illite and montmorillonite swelling illite inter-layered; the calcite content of 0 to 15% depending on the presence of shells and mollusks. Pyrite is present in vases rich in organic matter. The organic matter is relatively abundant in fine sediments found in channels and ditches where its accumulation is related to the hydrodynamics, and in areas with high inputs of continental plant or planktonic debris. The concentrations are between 0 and 1% in sandy sediments and exceed 20% in vases (Abe et al., 1993).

Sampling

The sediment sampling campaigns were conducted during the dry and wet season in ten stations (Figure 1) spreaded along the Grand-Lahou lagoon system. Sediment samples were collected using a metal tipper, packaged in aluminum foil and transported in an ice box to the laboratory. At the laboratory, the sediment samples were dried on aluminum foil at room temperature and stored in a refrigerator at 4 °C for extraction.

Extraction and purification

25 g of sediment previously dried in ambient air, crushed in a mortar and sieved were weighed and introduced in a flask. 50 ml of dichloromethane are then added to the sediment and mixed vigorously overnight with a mechanical stirrer. The mixture was filtered through a filter paper and placed in a round bottom flask. The filtrate was then evaporated to dryness on a rotary evaporator with a water bath to a temperature below 30 °C to minimize loss of volatile components. The substrate is recovered with 10 ml of methanol for the purification phase.

The extracts are purified by conditioning on a C18 cartridge which is previously activated with 10 ml methanol. The cartridge must not be dried for more than 30 seconds before the next step. All of the prepared sample is passed through the cartridge and is left dripped. The eluate is collected in a vial for analysis by HPLC.

HAP analysis

Determination of PAHs was carried out using a Shimadzu liquid chromatograph LC-20AT Model equipped with a getter Model DGU-20A5, a SHIMADZU UV/Visible detector, model SPD-20A and a SIL-20A injector model.

The device allows an ultraviolet detection at variable wavelengths and it allows to obtain gradients of solvent using a pump. Also, with software (LC - solution), it can control the whole system and ensure the data acquisition. The separation was performed on an analytical column type VP-ODS, 154 mm length and 4.6 mm inner diameter with a degree of pre-column of 10 x 4.6 mm.

The mobile phase consists of a mixture of acetonitrile (A) and ultrapure water (E) (acetonitrile/water (80/20)) with a flow rate of 0.5 mL/min and the column temperature was 40 °C. The injections were performed with an automatic injector; the injection volume was 20 µl and the analysis time was 10 min.

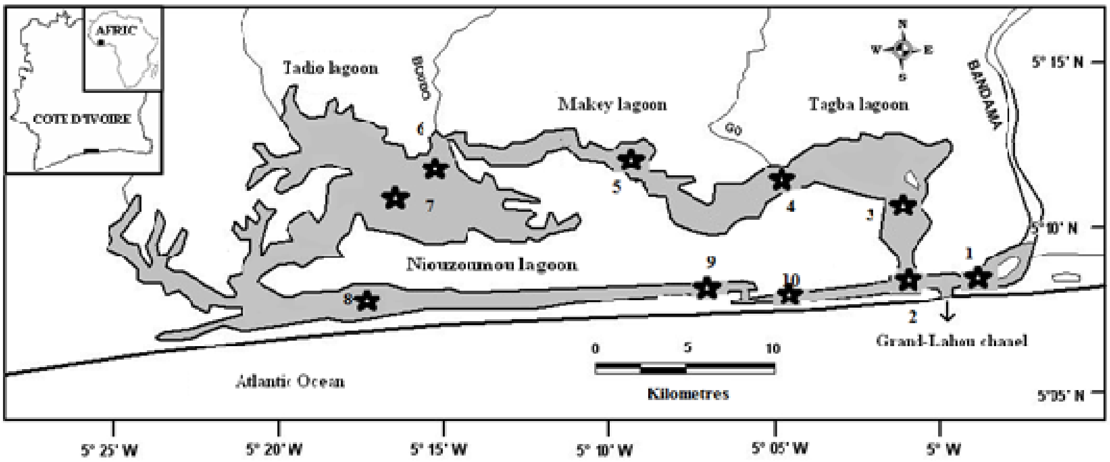

Figure 1: Sampling stations in the Grand-Lahou lagoon.

RESULTS AND DISCUSSION

Concentrations of polycyclic aromatic hydrocarbons are displayed in Tables 1 and 2. A total of eight (8) PAHs compounds were identified in the sediments of the Grand Lahou ecosystem. These are Fluoranthene, Pyrene, Benzo (k) fluoranthene, Benzo (a) pyrene, indeno (1,2,3-cd) pyrilene, Benzo (g, h, i) pyrilene, Benzo (a) anthracene and benzo (b) fluoranthene.

The average concentrations of PAHs vary from LD to 105 µg/g for fluoranthene; from LD to 381.08 µg/g; for pyrene; from LD to 162.87 µg/g for benzo (k) fluoranthene; from LD to 18.70 µg/g for benzo (a) pyrene, from LD to 6.35 µg/g for indeno (1,2,3-cd) pyrilene; from LD to 1.27 µg/g for benzo (g, h, i) pyrilene; from LD to 5.18 µg/g for benzo (a) anthracene and from LD to 10.69 µg/g for benzo (b) fluoranthene.

The sediments of the Grand-Lahou lagoon have relatively high concentrations in PAHs than those of the African lagoons of Chemu, Korle and Kpeshie (Essumang et al., 2009), Cotonou (Soclo et al., 2000) and Bizerte (Trabelsi and Driss, 2005) (Table 2). Data obtained from these works are also significantly higher than other coastal ecosystems from other parts of the world (Table 3).

The total concentration of PAHs was between 1.55 and 437.52 µg/g in the dry season. Pyrene, benzo (b) fluoranthene and fluoranthene have the highest concentrations. In the rainy season, the total PAHs concentrations varied between 46.35 and 1222.73 µg/g. Pyrene, benzo (a) pyrene and benzo (a) anthracene were the most present in the sediments of the lagoon of Grand-Lahou during the wet season.

Apart from the stations 9 and 10 which have high concentrations of benzo (a) anthracene, stations 1, 2, 3, 4 and 5 are more contaminated with PAHs during the two hydroclimatic seasons (Figures 2 and 3). These stations are located in the area of the lagoon characterized by the influence of the freshwater (the Boubo and the Bandama rivers) inputs and by intense agricultural activities leading to deforestation and charcoal production. In Station 1, is regularly discharged the ferry engine motor oil that carries the local populations from one bank to another. On the contrary, stations 6, 7, 8, 9 and 10 are less contaminated and are located in the sector of the lagoon under the oceanic influence.

Table 1: Concentrations of PAHs (µg/g) in sediments samples collected in the Grand-Lahou lagoon during the dry season.

	St 1	St 2	St 3	St 4	St 5	St 6	St 7	St 8	St 9	St 10
					Concentration (µg/g)					
Fluoranthene	LD	0,92	9,52	31,39	105,84	LD.	LD.	LD	LD	LD
Pyrene	LD	25,04	LD	381,08	LD	6,21	2,02	2,19	LD	LD
Benzo(k) fluoranthene	LD	LD	162,87	LD	LD	LD	LD.	0,69	LD	0,46
Benzo(a)pyrene	0,96	13,72	LD	18,70	12,10	LD	0,73	LD	1,05	LD
Indéno(1,2,3-cd) pyrilene	LD	LD	LD	6,35	LD	LD	0,36	LD	2,83	LD
Benzo(g,h,i)pyrilene	0,48	1,27	LD	LD	LD	LD	LD	LD	LD	LD
Benzo(a) anthracene	LD	3,66	LD	LD	LD	0,25	LD	LD	LD	5,18
Benzo(b) fluoranthene	0,11	4,48	10,69	LD	LD	0,02	0,16	0,12	LD	LD
Total HAPs	**1,55**	**49,09**	**183,08**	**437,52**	**117,94**	**6,48**	**3,27**	**3,00**	**3,88**	**5,64**

Table 2: Concentrations of PAHs (µg/g) in sediments samples collected in the Grand-Lahou lagoon during the wet season.

	St 1	St 2	St 3	St 4	St 5	St 6	St 7	St 8	St 9	St 10
					Concentration (µg/g)					
Fluoranthene	LD	LD	LD	LD	LD	LD	LD	LD	LD	LD
Pyrene	227,86	249,40	33,06	87,15	1021,22	46,35	57,35	192,36	LD	LD
Benzo(k)fluoranthène	57,33	LD	LD	0,09	LD	LD	LD	LD	LD	30,05
Benzo(a)pyrene	226,33	24,55	LD	13,16	153,36	LD	LD	4,59	LD	LD
Indeno(1,2,3-cd)pyrilene	LD	LD	LD	LD	LD	LD	LD	LD	LD	12,44
Benzo(g,h,i)pyrilene	LD	LD	LD	LD	LD	LD	LD	LD	LD	LD
Benzo(a)anthracene	124,47	7,60	LD	12,37	48,15	LD	LD	11,55	784,05	121,28
Benzo(b)fluoranthene	LD	LD	LD	LD	LD	LD	LD	LD	LD	LD
Total HAPs	**635,99**	**281,55**	**33,06**	**112,77**	**1222,73**	**46,35**	**57,35**	**208,50**	**784,05**	**163,78**

LD: Limit detection; St: Station;

Table 3: Comparison of sediment PAH concentrations (µg/g dry wt) measured in this study compared with those in other countries.

Location	Number of PAH compounds analyzed	Mean PAHs values	References
Gironde Estuary (France)	17	0.0185 – 4.88	Budzinski et al., 1997
Arcachon Bay (France)	16		Baumard et al., 1998b
Barcelona Harbour (Spain)	16	1.74 – 8.42	Baumard et al., 1998a
Orbetello lagoon (Italy)	16	0.0006 – 0.203	Specchiulli et al., 2010
Vietnamese lagoons (Vietnam)	18	0.112 – 0.628	Giuliani et al., 2008
Mediterranean coast (Morocco)	16	0.01 – 0.55	Pavoni et al., 2001
Abu Qir Bay (Egypt)	16	Nd – 2.66	Khairy et al., 2009
Bizerte lagoon (Tunisia)	16	0.083 - 0.447	Trabelsi and Driss (2005)
Cotonou lagoon (Benin)	14	0.025 – 1.45	Soclo et al., 2000
Chemu (Ghana)	16	0.0078 – 0.011	Essumang et al., 2009
Korle (Ghana),	16	0.00 – 0.009	Essumang et al., 2009
Kpeshie (Ghana),	16	0.00162 – 0.0047	Essumang et al., 2009

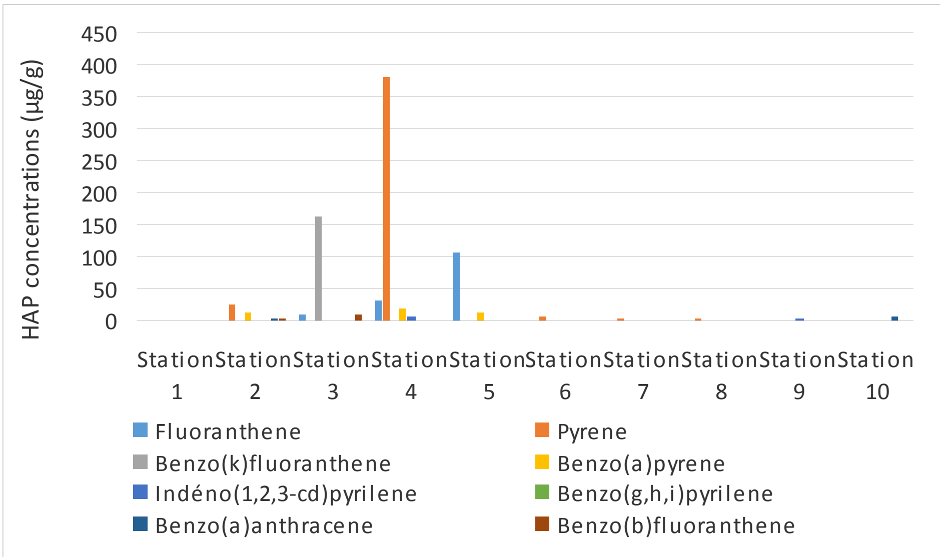

Figure 2: Distribution of PAHs concentrations in surface sediments collected in the Grand-Lahou ecosystem during the dry season.

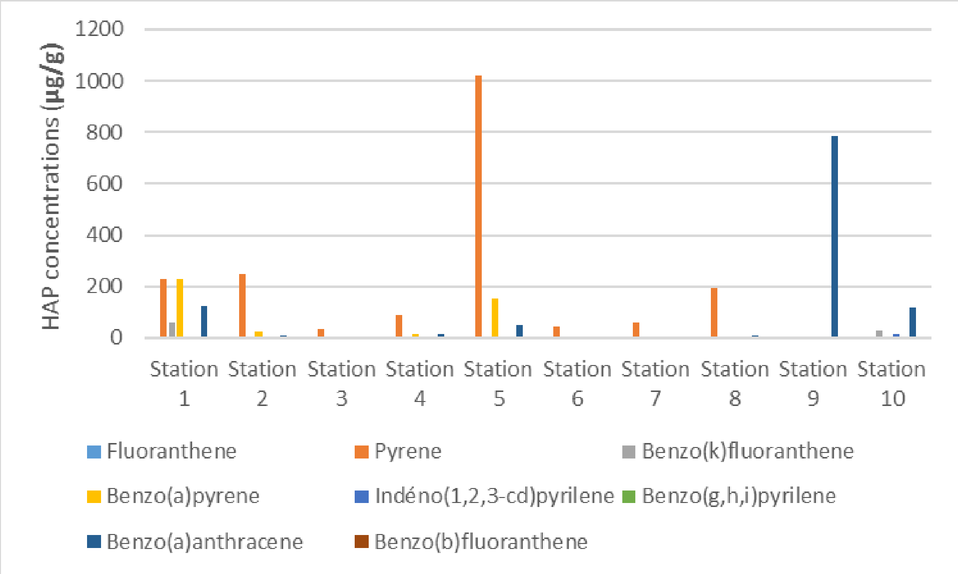

Figure 3: Distribution of PAHs concentrations in surface sediments collected in the Grand-Lahou ecosystem during the wet season.

The presence of PAHs in sediments is an indication of the contamination of the Grand-Lahou lagoon. The sediment compartment is the ultimate repository of hydrophobic contaminants introduced into aquatic environments. It is often referred to as a well or trap of these organic compounds. Thus the sediment provides an essential link in the study of the contamination of the environment by PAHs since it can be used as a record of contamination. It is also necessary to determine the level of PAH in this compartment because it is not only a well of PAHs but it can be a source of the water column contamination in certain circumstances or conditions. Desorption and remobilization of surface material to the water column due to current events or natural/anthropogenic disturbance may increase the bioavailability of PAHs to the biota, mainly benthic biota.

Possible sources of PAHs in the Grand-Lahou lagoon are oil and fuel spills from ships and fishing boats, waste combustion, domestic wastes disposal and especially the production of charcoal on the Grand-Lahou lagoon shores. High concentrations observed during the rainy season can be attributed to the contribution due to leaching of soil contaminated with PAHs. According Karichkoff et al. (1979), the spatial variations in the PAHs concentrations can also be explained by the heterogeneity of the particle sediment. Indeed, the hydrophobic organic contaminants (COH) are generally associated with the fine fraction of sediments.

A clear dominance of high molecular weight (HMW) PAHs (pyrene, Benzo (k) fluoranthene, Benzo (a) pyrene, indeno (1, 2, 3-cd) pyrilène, Benzo (g, h, i) pyrilene, Benzo (a) anthracene, Benzo (b) fluoranthene) is observed. Low molecular weight (LMW) PAHs is only represented by fluoranthene. LMW PAHs are different from HMW PAHs by their number of aromatic rings, and therefore by their molecular weight.

These differences play on their physicochemical and toxicological properties that differ. However, these molecules are poorly soluble, hydrophobic and very slowly degraded; these characteristics are especially true that the compound has a high cycle number. Thus, PAHs tend to adsorb to suspended solids and accumulate in sediments as well (specifically on fine particles) in living organisms. It should be noted that the abundance of heavy compounds could indicate contamination from domestic origin, among others, the production of charcoal on the banks of the lagoon. The low presence of LMW hydrocarbons could also be attributed to the high microbial activity in the lagoon environment. According to Lei et al. (2007), microbial activity contributes significantly to the reduction of LMW PAHs in sediments since the PAHs with a small number of aromatic rings are more easily degraded by bacteria in the water column, which reduces the number of molecules that reaches the bottom. Finally, one can note that LMW PAHs being more soluble, they are subject to evaporation and dilution in the water body (Khim et al., 2001).

Molecular ratios, such as phenathrene/anthracene(Phe/An), Fluoranthene/Pyrene (FL/Pyr), Chrysene/ Benzo(a)anthracene (Chrys/B[a]A) have been proposed for interpreting PAH composition and identifying possible sources (Soclo et al., 2000; Yunker, 2002; Doong and Lin, 2004). According to Yunker (2002), when the ratio of An /An+Phe < 0.1, the PAHs were mainly from petroleum contamination, while it is > 0.1, PAHs were from combustion source. PAHs with Fl/Fl+Pyr < 0.40 were from petroleum contamination, while a PAHs with 0.4 <Fl/Fl+Pyr < 0.5 were mainly from oil combustion, and PAHs with Fl/Fl+Pyr > 0.5 were from combustion of coal, wood and grassed matter (Yunker et al., 2002). Bicego et al. (2006) used the following four PAHs isomer pair ratios to identify possible sources in sediments of the Santos and Sao Vicente Estuary of Sao Paulo (Brazil): An/An+Phe; Fl/Fl+Pyr; B[a]A/B[a]A+chry; IP/IP+BP (IP: Indeno (123-cd) Pyrene; BP: Benzo (e) pyrene). Regarding the B[a]A/B[a]A+chry ratio, PAHs with 0.2 < B[a]A/B[a]A+chry < 0.35 could be related to a mixed origin. PAHs with B[a]A/B[a]A+chry < 0.2 were mainly from petroleum contamination, while PAHs with B[a]A/ B[a]A+chry > 0.35 were from combustion source. Finally, PAHs with IP/IP+BP < 0.2 were from petroleum origin input, while PAHs with 0.2 < IP/IP+BP < 0.5 were mainly from oil combustion, and IP/IP+BP > 0.5 were typical of the combustion of coal, wood and grassed matter. In this work, none of these ratios can be applied. PAH components such as Fluoranthene, Indo Pyrene have not been found in the coastal lagoon of Grand-Lahou.

The presence of benzo (a) anthracene and benzo (b) fluoranthene in the sediment is a public health problem especially since these pollutants are considered by International Agency for Research on Cancer (IARC) and Environmental Protection Agency (EPA-USA) as a potential carcinogen to humans (Agency for Toxic Substances and Disease Registry, 1995). This is risk to local residents who use the lagoon for several activities (fishing, recreation, transportation...). Pyrene is found in large quantities in the lagoon at any season. This chemical compound is known to be very hydrophobic, low volatility, more recalcitrant to biodegradation than LMWPAHs and therefore persistent in soils and contaminated sediments. It is mutagenic but it is not considered as a carcinogen. Its effects on the ecosystems are relatively poorly known.

The analysis of the presence of HAPs in the Grand-Lahou coastal lagoon showed a clear dominance of high molecular weight (HMW) PAHs (pyrene, Benzo (k) fluoranthene, Benzo (a) pyrene, indeno (1,2,3-cd) pyrilène, Benzo (g, h, i) pyrilène, Benzo (a) anthracene , Benzo (b) fluoranthene). Low molecular weight (LMW) PAHs was only represented by fluoranthene. The total concentrations of PAHs were between 1.55 and 437.52 µg/g in the dry season. In the rainy season, the total PAHs concentrations varied between 46.35 and 1222.73 µg/g. The possible sources of PAHs in the Grand-Lahou lagoon are oil and fuel spills from ships and fishing boats, waste combustion, domestic wastes disposal and especially the production of charcoal on the Grand-Lahou lagoon shores.

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