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Studies on Persistent Organic Pollutants (Pops) in the Lagos Lagoon 1: Occurrence and Levels of Polycyclic Aromatic Hydrocarbons (Pahs) In Surface Waters of the Lagoon

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Abstract

The Lagos lagoon is one of Africa's largest estuarine ecosystems. Its water receives organic input from a wide variety of sources; atmospheric, industrial, and municipal activities. Environmental impacts from PAHs continue to elicit global concern and attract attention especially in Lagos and its environs. Some PAH compounds which are toxic, mutagenic, or carcinogenic could be taken up and accumulated by aquatic organisms from polluted water, thereby resulting in the modern day diseases in humans. This research aims at assessing the decline of the water quality of Lagos lagoon due to land-based human activities in order to reveal the extent of exposure of humans to PAH risk. The research presents the need for control measures necessary for the minimization or prevention of PAH pollution to ensure a safe environment for our people. All the 16 USEPA priority PAHs were present across all twelve locations in high concentrations irrespective of the distance from the source point. They all exceeded the World Health Organization (WHO) and USEPA recommended maximum contamination levels. The majorities of the PAHs were four to six ring compounds and most predominantly were Benzo (a) pyrene and Pyrene. The pattern was the same for the PAH levels throughout the year. Increase in PAH concentration tracks closely with increase in combustion sources and automobile use. This study evaluated the trends in PAHs, a group of contaminants with multiple urban sources in water samples from twelve points on the Lagos lagoon between February and December 2004.

Keywords: Lagos lagoon; water; polycyclic aromatic hydrocarbons; GC/FID; biochemical oxygen demand.

INTRODUCTION

Persistent Organic Pollutants (POPs) are chemical substances that persist in the environment and can bioaccumulate through the food web and pose a risk of causing adverse effects to human health and the environment. POPs include industrial chemicals like polychlorinated biphenyls (PCBs), combustion by-products like dioxins, Polycyclic Aromatic Hydrocarbons (PAHs) and pesticides like Dichlorodiphenyl - trichloroethane (DDT) among others. In this study, focus is on the PAHs.

Polycyclic Aromatic Hydrocarbons (PAHs) are stable organic chemicals that persist in the environment and bioaccumulate through food chain. Water pollution by PAHs has caused considerable concern worldwide. Coastal and inland water usually act as receptors for these pollutants. PAHs have received considerable attention because of their documented carcinogenicity in experimental animals of several species. Pathways for PAHs to enter surface water include atmospheric fallouts from incomplete combustion, municipal effluents, and oil spillages (Bartle et al 1991 and Anyakora et al 2004).

Davis (2003) has shown that Petrogenic PAHs enter the estuary as a result of spills and leaks of oil and refined oil products. Anthropogenic activities, which distribute PAH contaminants throughout the Lagos lagoon include: industrial production, transportation and waste incineration disposal of municipal and domestic sewages. PAHs are found throughout the environment in the air, water and soil as complex mixtures (Jacob 1994). According to Hellou et al 2002, Delistraty 1997, Schirmer et al 1998a, Schirmer 1998b, and Swartz 1995, PAHs potentially contribute to many of the modern day diseases including cancer, damage to the reproductive system, disrupted endocrine and immune systems, and neurobehavioral effects. There are more than 100 PAHs but the following 16 USEPA priority PAHs are chosen for this study: Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benzo (a) anthracene, Chrysene, Benzo (b) fluoranthene, Benzo (k) fluoranthene, Benzo (a) pyrene, Indeno (1,2,3-cd) pyrene, Dibenzo (a, h) anthracene, and Benzo (g, h, i) pyrelene. Transport to the marine environment occurs both via surface waters and atmosphere; in the water column most PAHs tend to absorb to particles and to be deposited to the underlying sediments. A study by Law et al. (199 showed that transport of PAHs to the marine environment occurs both via surface waters and atmosphere; in the water column most PAHs do not dissolve but rather adsorb onto particles and are then deposited to the bottom sediment.

Their low aqueous solubility, limited volatility and recalcitrance towards degradation allow PAHs to accumulate to levels at which they may exert toxic effect upon the environment (Pavlova et al 2003). From the environmental aspect, Manoli 1996 has found that PAHs are probably the most important analytes because many of these compounds are potential or proven carcinogens.

The Lagos lagoon, the study area, is an urbanized estuarine ecosystem, receiving a load from a number of important large rivers (Yewa, Ogun, Ona and Oshun) draining more than 103, 626 sq km of the country. The lagoon is a great expanse of shallow water with depths ranging as low as 0.9 meters in some points and up to 25meters at the dredged areas (Webb 1958) (See figure 1.1). Much greater seasonal changes occur by the influx of fresh water during the rainy periods. The lagoon borders the forest belt and empties directly into the Atlantic Ocean at the harbor. During the rainy season, large volume of fresh water passes through the harbor into the sea. The estimated area of the Lagos lagoon is 150.56km^2 while the harbor is about 8km long and leads to the wharf area of Apapa where shipping and other oil related activities are concentrated (fig1.1). The lagoon is also impacted by industrial, agricultural and municipal activities that keep increasing with the ever growing population of Lagos, the most highly populated city in the country, presently harboring not less than 15% of the total population (about 150 million) of Nigeria. The above activities contribute highly to the presence of polycyclic aromatic hydrocarbons (PAHs) in the environment. As shown by Meador et al 1995, the highest concentrations of PAHs are generally found around urban areas.

The Lagoon is well exposed to heavy load of pollutants (PAHs inclusive) due to the activities that take place along the shores of the lagoon.
The loading of pollutants into the lagoon also affects the quality of water greatly as shown in figure 2. This was reflected in the colour and appearance of the lagoon water, ranging from oily at some locations to gray, slightly yellow, turbid, and dark at some locations.

The PAH mixtures in Lagos lagoon are expected to be highly variable and complex due to the large number of sources contributing to their discharge. Ajao 1996 showed that the high input of pollutants into the lagoon from land-based sources accounts for about 44% while atmospheric sources contribute 33% of all potential pollutants.

The toxic effects exerted on the environment by PAHs, coupled with the fact that Lagos lagoon is the major source of sea foods to the people of Lagos and its environs have been the motivating factors for this study. This study therefore seeks to assess the chemical availability of and the level of PAHs in Lagos lagoon water.

MATERIALS AND METHODS

Sample Collection and Preparation

Water samples were collected from 12 selected locations on the lagoon in April and May 2004. Water samples (IL volume) were collected from the surface by submerging a pre-cleaned Winchester amber glass bottle by hand. This is essential to prevent decay due to ultraviolet radiation. The sampling apparatus and glassware were precleaned with dichloromethane.

All the reagents used were of pesticide analysis grade (or equivalent). Dichloromethane was purchased from Supelco, USA. Granular anhydrous sodium sulphate (AR grade, Malinkrodt) was muffled at 400ºC for 4hrs prior to use. The boiling chips approximately 10/40 mesh were heated to 400ºC for 30 minutes and then extracted with DCM prior to use. All glassware were washed, rinsed with distilled water, dried at 105ºC for 1hr and then rinsed with DCM prior to use. Previous studies by Lindhardt 1994 and Guerin 1999 have shown that extraction with dichloromethane (DCM) is very effective. Water samples were extracted by Liquid – Liquid method (EPA 1984). About 500ml the samples were extracted using 30ml DCM three times. The first aliquot was used to rinse the sampling bottle so as to include adsorbed material extract. The three solvent extracts were combined over anhydrous sodium sulphate. All samples were spiked with a known volume (100µl i.e. 200ng/ml) of surrogate spiking solution, a mixture of three $_{13}$C-labelled PCBs (13C - PCB 52, 13C - PCB 153 and 13C – PCB 37), prior to extraction. The extracts were then concentrated to about 2ml, using rotary evaporator. Florisil cleanup was carried out on the extracts before GC analysis.

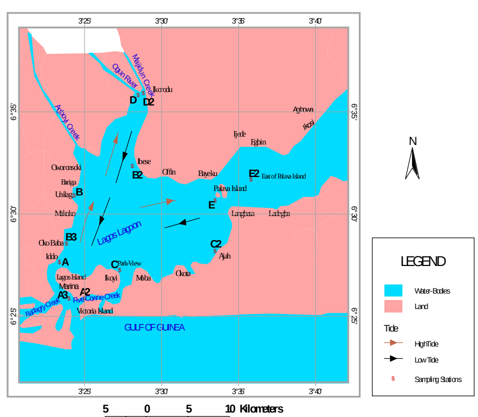

Fig. 1.1: Lagos Lagoon showing Sampling Stations (A-E) and the Direction of the Tide

Sample Analysis: Analysis was run on a Hewlett-Packard (Avondale, PA) Model 5890/5970 Gas Chromatograph with a mass selective detector (quadrupole mass analyzer, 70eV) equipped with a Hewlett-Packard 7673A autosampler and a 30m x 0.25mm. I.D. X 0.10 µm DB-5 film thickness column. 1µl sample was injected using a splitless injection mode at 250ºC injection temperature and GC-MSD interface temperature of 280ºC. A mixture of three 13C-labelled PCBs (13C - PCB 52, 13C - PCB 153 and 13C – PCB 37) was used as surrogate standard. The PAHs were identified and quantified by comparison of retention times and spectra of internal standards. Concentrations were not corrected for the recovery of internal standards. Some of the compounds, for instance, Naphthalene were found in the method blanks but during integration all the concentrations found in the blank were removed from the samples.

Fig. 2: Location B3, Okobaba lagoon front, between Ibadan and Ondo streets

Determination of Physicochemical Parameters

The physico chemical quality of the water was also assessed and correlated to the concentration of PAHs in that location (table 1).

Dissolved Oxygen (DO) and Biochemical Oxygen demand (BOD) determinations were carried out in the water samples from the Lagos lagoon in the Environmental / Analytical laboratory, of the University of Lagos. Azide modification of the Winkler method was used according to APHA et al. (1999).

RESULTS AND DISCUSSIONS

The results show that the PAHs were present across all the 12 locations on the lagoon in high concentrations, irrespective of how far the location was from the source point (fig. 3).

The selected locations include: Lagos harbour (petrogenic source), Okobaba (pyrogenic source), Iddo (other anthropogenic sources), Ikorodu (far from suspected
Sources), Ibese and Offin (far from suspected sources), and East of Palava Island (farthest of the locations from the suspected sources), PAH concentrations were seen to be high at all the locations (table 2).

Table 1: Relationship between DO, BOD, and PAH concentrations from selected locations on the Lagos lagoon in April and June 2004

April 2004						June 2004			
Location	Location description	DO (Mg O_2/L)	BOD (Mg O_2/L)	No. of PAHs	Total PAHs (Mg/L)	DO (Mg O_2/L)	BOD (Mg O_2/L)	No. of PAHs	Total PAHs (Mg/L)
A	Iddo	1.12	56	9	$1.7x10^3$	2.95	25.4	15	$1.78x10^3$
A3	Lagos habour	1.41	187	14	$3.74x10^3$	1.52	55.98	15	$4.04x10^3$
B2	Ibesse & Offin	2.54	96.5	10	$3.22x10^3$	4.06	137.16	15	$2.65x10^3$
B3	Okobaba	1.83	91	13	$7.33x10^3$	2.85	50	13	$2.19x10^3$
D2	Ikorodu	3.76	20	16	$3.6\ x10^3$	6.4	22.35	12	$1.23x10^3$
E2	East of Palava Island	3.86	20	16	$3.69x10^3$	7.21	16.04	13	$4.43x10^3$

The BOD levels of the lagoon water were found to be generally quite high with corresponding low DO values (fig. 3).

The BOD as well as the PAH levels were generally higher in the months of April than in the months of June, while the DO levels were lower in the months of April than in the month of June. The presence of organics (PAHs inclusive) created high oxygen demand. The oxygen depletion occurs in the lagoon as a result of high Biochemical Oxygen Demand (BOD). This was possibly due to increased organic load introduced by indiscriminate discharging of untreated sewage and sludge, emissions from automobile and boats exhausts, surface run-off, waste oil discharge into the lagoon and other shipping and oil related activities in the harbor and along the shores of the lagoon. This was evident by the high levels of the PAHs obtained when the DO levels were low and the BOD levels high.

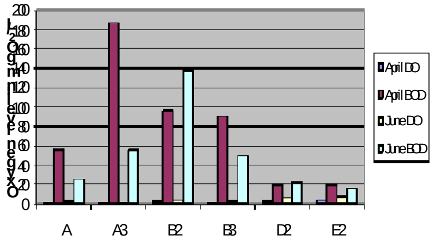

Fig 3: Dissolved Oxygen (DO) and Biochemical Oxygen Demand (BOD) levels of Lagos lagoon water in April 2004

The PAH ranges in Lagos lagoon water samples in 2004 are shown in table 2.

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Table 2: PAH ranges in the water samples of the Lagos lagoon in the year 2004

S/N	PAHs	Range in Lagos lagoon water (µg/L)	MCL in µg/L (EC)	MCL in µg/L (USEPA)
1	Naphthalene	12.42 – 317		
2	Acenaphthylene	21.5 – 363		
3	Acenaphthene	16.60 – 300		
4	Fluorene	9.27 – 210		0.2
5	Phenanthrene	12.30 – 155		0.2
6	Anthracene	7.51 – 139		0.2
7	Fluoranthene	15.02 – 172	0.03	
8	Pyrene	27.70 – 902		0.2
9	Benzo (a) anthracene	83 – 14,400		0.2
10	Chrysene	20.72 – 192		0.2
11	Benzo (b) fluoranthene	96.34 – 1,228	0.03	0.2
12	Benzo (k) fluoranthene	35.58 – 1,610	0.03	0.2
13	Benzo (a) pyrene	28.61 – 70.27	0.01	0.2
14	Indeno(1,2,3-cd)pyrene	13.55 – 692	0.03	0.2
15	Dibenzo(a,h)anthtracene	11.37 – 202		0.2
16	Benzo (g,h,i) perylene	29.65 – 1,300	0.03	

MCL – Maximum contaminant level
EC – European community
USEPA – United States environmental protection agency

Tables 2 confirms that the PAH levels in Lagos lagoon water in 2004 were far above the MCLs set by United States Environmental Protection Agency (USEPA) and the European Community (EC) member states MCLs for drinking water.

The PAHs determined were 2 to 6 - ring compounds. The majority of the PAHs observed were 4 to 6-ring compounds and most predominantly present were Benzo (a) anthracene, Benzo (k) fluoranthene, Benzo(b)fluoranthene, Benzo (g,h,i) perylene and Pyrene. This indicated that the PAHs in Lagos lagoon water were mainly from combustion sources. Uncombusted sources (e.g. oil seeps petroleum spills) contain predominantly two – and three - ringed compounds whereas combustion sources (e.g. automobiles, domestic heating with coal, forest fires etc.) result in predominantly four to six ringed species (Van Metre *et al.*, 2000). Most species of fish will readily assimilate PAHs. However studies (Lu et al, 1977, Varanasi & Gmur 1981) have shown that benzo (a) pyrene (BaP) can be accumulated to potentially hazardous levels in fish and invertebrates. The formation of reacting intermediates, such as diol and phenol epoxides of BaP have been implicated in mutagenesis and carcinogenesis of mammals.

The 2-3-ring compounds were also detected in many of the samples but lower concentrations. Eisler (1987) states that many PAHs such as Naphthalene and Phenanthrene are acutely toxic at whole body concentrations above 50,000ppm and deleterious such that responses occur at concentrations in the range of 100 – 5000ppm. The six-ring aromatic hydrocarbon, BaP, which was present in concentrations much higher USEPA and EC maximum contaminant levels, is among the most toxic PAH compounds, causing deleterious effects at whole body concentrations above 100ppm.

The pattern is the same when comparing the PAH levels in the lagoon water throughout the year.

The results in figures 4 to 14 show that the PAHs were distributed across all the locations on the lagoon. This signified that PAHs can easily be transported from their source points to places far away from such points.

Two to three – benzene ring compounds, Naphthalene, Acenaphthylene, Acenaphthene, etc, were found to be moderately available. These three are not known carcinogens or mutagens.

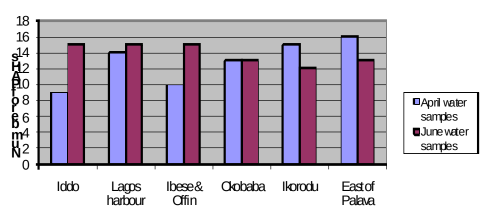

Fig. 4. Number of PAHs in Lagos lagoon water samples from selected locations in April and June 2004

The total PAH levels in the water were found to be very high and this was as a result of the continuous uncontrolled generation of these compounds from the multiple urban sources in the city (fig. 5).

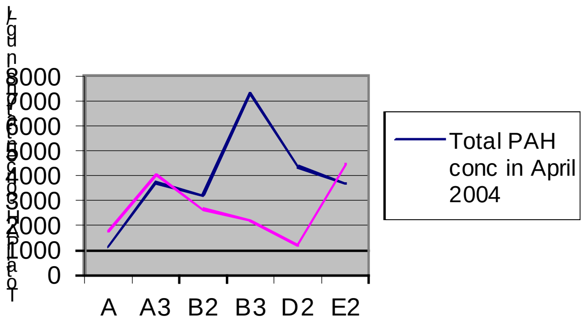

Fig. 5: Total PAH levels in Lagos lagoon water samples of April and June 2004

Total (16) PAH concentrations in Lagos lagoon ranged from 1780µg/L in Iddo (A) to 7200µg/L in Okobaba in April 2004. A decrease was observed in June 2004 where the highest total PAH concentration of 4000µg/L was found in Lagos harbor (A3) where oil related activities take place. Ikorodu sample in the month of April had a total (16) PAH concentration of 4360 µg/L while Iddo in the same month had 1170µg/L. In the month of June, the Ikorodu (D2) water had a total PAH concentration of 1230µg/L; while Iddo had in the same month 1780µg/L. Comparing Lagos lagoon in Nigeria with other large lakes, total PAH levels in Lagos lagoon were found to be several magnitudes higher. This is evident in the report by Klecka et al., (2000), which showed total (4) PAH concentration ranging from 0.000211 to 0.00110ng/mL in Western Mediterranian in Barcelona.

Figs. 6 to 13 show the concentrations of individual PAHs at the six selected locations in April and June 2004. B(a)P and pyrene concentrations were found to be consistently high in the two months at all the six locations. Figs. 6 and 7 show individual PAH concentrations at Iddo in April and June 2004 respectively.

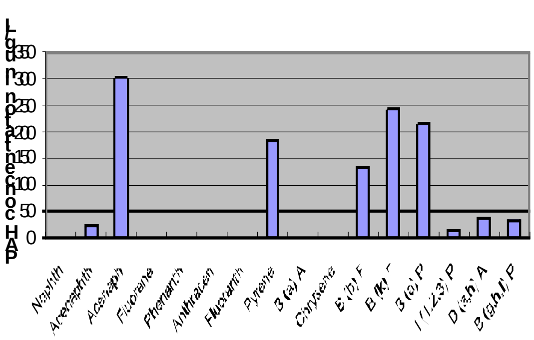

Fig. 6: PAHs in Iddo water sample in April 2004

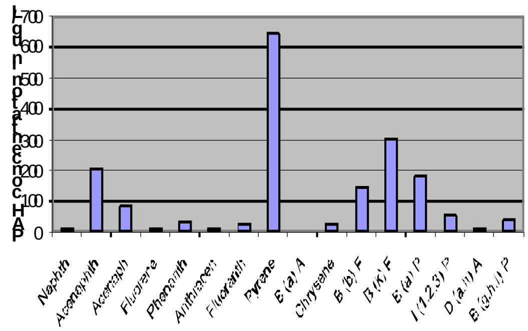

Fig 7: PAHs in Iddo water sample in June 2004

Iddo is one of the busiest of the selected locations. Iddo is highly populated and is full of activities which include, industrial, agricultural, municipal, fishing, and several other activities. This area is both residential and industrial. Here the discharge of sewages and wastes from various sources into the lagoon is very rampant. This location is also very close to the disused power station at Ijora, which functioned between 1921 and 1978. PAH concentrations in Iddo were generally higher in the month of June than in April. The month, June, is found within the center of the rainy season, and the samples for this study were taken during a heavy rain fall. This result reflected the impact of drainage on the lagoon, which acts as a receptor for the wastes and pollutants that are not properly disposed. In Iddo, the PAH with the highest concentration was pyrene (about 640ug/L) as shown in fig. 7.

Figs. 8 and 9 show individual PAH concentrations at the Lagos harbour in April and June 2004 respectively.

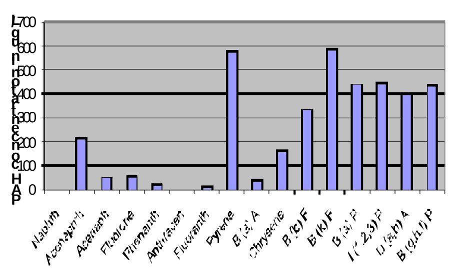

Fig 8: PAHs in Lagos harbour water sample in April 2004

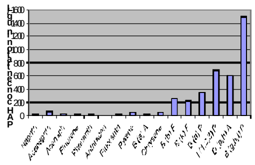

Fig 9 PAHs in Lagos harbour water sample in June 2004

At the harbour, shipping, oil related businesses and other activities take place. The bridge called Outer Marina Bridge passes by the harbour and links Eko Bridge which takes to Iddo. At the Lagos harbour, PAH concentrations were higher in June. The higher PAHs were predominant, with Benzo(g,h,i)perylene concentration (1500ug/L) being the highest as shown in fig. 9.

Figs. 10 and 11 show individual PAH concentrations at Okobaba in April and June 2004. Okobaba (B3) is the location where incessant burning of saw dust takes place. The Third Mainland Bridge, passes the lagoon close to this locations.

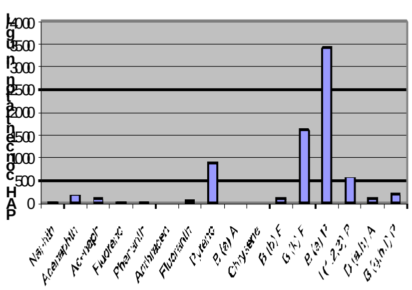

Fig 10 PAHs in Okobaba water sample in April 2004

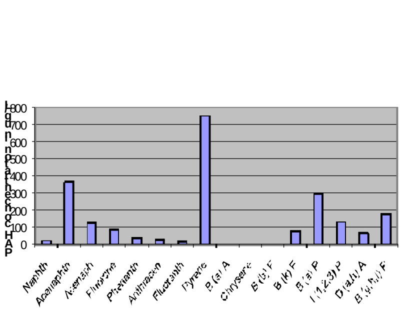

Fig. 11: PAHs in Okobaba water sample in June 2004

PAH concentrations at this location were found to be much higher in the month of April than in June. This was possibly due to greater pollutants movements and distribution from the source point in the lagoon during rainy seasons. The highest PAH concentrations in the lagoon water were found in Okobaba in April, and these were B(a)P (3400ug/L) and pyrene (950ug/L). The concentrations in June were lower (eg. B(a)P was 300ug/L while pyrene was 750ug/L). PAHs such as benzo (a) pyrene can have LC50s as low as a few microgrammes/Litre towards fish when exposed to UV (Varanasi et al., 1981). They can also show appreciable toxicity to sediment dwelling invertebrates. LC50 values of 0.5 to 10mg/kg (concentration in sediment) have been reported for marine amphipods for benzo(a)pyrene, fluoranthene and phenanthrene (Varanasi et al., 1981).

Figs. 12 and 13 show individual PAH concentrations at Ikorodu in April and June. PAH concentrations at Ikorodu were also found to be higher in the month of April than in June.

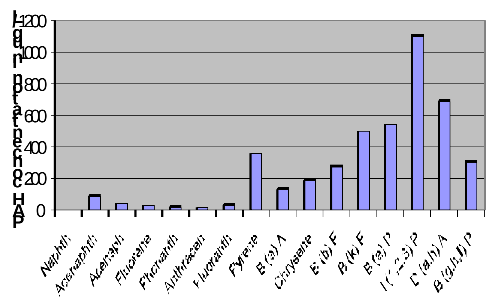

Fig 12 PAHs in Ikorodu water sample in April 2004

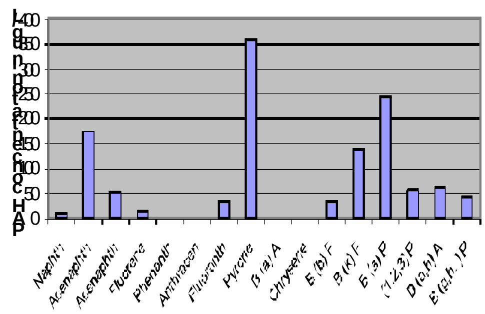

Fig 13 PAHs in Ikorodu water sample in June 2004

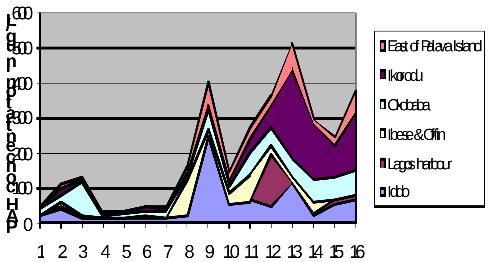

Fig.14: Levels of 16 priority PAHs in water samples from six selected locations of Lagos lagoon in April 2004

In the month of April, Naphthalene was absent in Iddo, Lagos harbour, Ibese & Offin, and Ikorodu. At Okobaba and East of Palava Island it was present in low concentrations (12.42µg/L and 40.24µg/L respectively).

Anthracene was absent in Iddo, Lagos harbour, Ibese & Offin, and Okobaba. Benzo (a) anthracene was present only at the harbour, Ikorodu and East of Palava Island. Phenanthrene and Fluoranthene were absent in Iddo and Ibese but present in the other locations in low concentrations (still much higher than the USEPA and WHO recommended MCLs). Chrysene was absent in Iddo and Okobaba. Benzo (b) fluoranthene, Benzo (k) fluoranthene, Benzo (a) pyrene, Indeno (1,2,3, -cd) pyrene, Dibenzo (a, h) anthracene, and Benzo (g, h, i) perylene (4- 6 ring compounds) were present in all the locations at high concentrations (lowest at Iddo).

Acenaphthylene and Acenaphthene (2-ring compounds) were also present in all the locations in concentrations lower than those of the 4- 6 ring compounds. The PAHs, apart from the long-term impact on the people as potential causes of modern day diseases, also has a short-term effect on the ecosystem as it depletes (along with other organics) the oxygen available for the survival of living organisms in the lagoon.

CONCLUSION

From the results obtained, it can be concluded that the levels of PAHs in the Lagos lagoon are dangerously high as they all exceeded the accepted ranges. It is also concluded that PAHs can easily spread and be transported across the globe via water bodies like the Lagos lagoon and the Atlantic Ocean. Okobaba was identified as having known carcinogens like benzo(a)pyrene and pyrene at levels that posed a risk to the environment. The presence of these potentially toxic substances provide a legitimate basis for initial concern and a more detailed assessment of PAH – related risks to important elements of the Lagos lagoon.

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