Academia Journal of Environmental Sciences 2(7): 108-115, October 2014
DOI: http://dx.doi.org/10.15413/ajes.2014.0112
ISSN: 2315-778X

Academia Publishing

Research Paper

Polycyclic aromatic hydrocarbons (PAHs) pollution in marine waters and sediments at the Tema Harbour, Ghana

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Accepted 12th July, 2014

ABSTRACT

Polycyclic aromatic hydrocarbons as a major pollutant from automobile exhaust, combustion of all forms of fuels and other industrial activities, find their way into water bodies which may have adverse effect on aquatic life. Tema, being an industrial city, has no records of PAHs pollutants hence this study at the Tema Harbour seeks to measure their concentrations which may affect water and sediment quality. This will give baseline guidance to the authorities in the control of pollutants. Samples of surface sediments and water were collected from 10 sites at the Tema Harbour complex in Tema, Ghana, comprising of the canoe landing site, the inner fishing harbour, the outer fishing harbour and the main harbour. The concentrations of the polycyclic aromatic hydrocarbons (PAHs) were analysed by GC with FID detection after Soxhlet and Liquid-liquid extractions were performed on the sediments and water, respectively, using dichloromethane. A broad distribution of the PAHs were identified with the total mean concentration of ∑20 PAHs ranging from 28.6 to 190.3 µg/g w.wt in sediments and from 33.2 – 84.5 µg/L in water. The levels of PAHs in the sediments and water at the canoe landing site were higher than those at the other basins of the harbour complex. Concentrations are however, equally distributed in the inner and outer fishing basins while variations were observed at the different sampling sites within the main harbour. The result from this study provides a valuable range of PAHs concentration that can help in the control of pollutants and in future assessment of water and sediment quality.

Key words: Polycyclic aromatic hydrocarbon (PAH), Tema harbour, marine sediment, pollution, water.

INTRODUCTION

Combustion of coal, oil, gas, wood, garbage and other organic substances are known to release many pollutants including polycyclic aromatic hydrocarbons (PAHs) into the environment (Kuusimaki et al., 2002; Liu et al., 2008). Automobile exhausts, industrial emissions and smoke from burning of wood, charcoal and tobacco contain high levels of these pollutants (Anyakora et al., 2011; Dyaneshwar et al., 2012; Linsey et al., 1999; Miguel et al., 1998; Khillare et al., 2005) which are released into the ambient air, only to be deposited on land, vegetation and in water bodies.

Coastal and inland waters usually act as receptors for pollutants (SEPA, 2010). Watercraft engines are known to release substantial amounts of petroleum products into the surrounding waters contributing to the PAH load of the ecosystem (Oris et al., 1998). Pathways for PAHs to enter surface water include atmospheric fallout, urban run-off, municipal effluents, industrial effluents, and oil spillage (Essumang, 2010; Gilbert, 2006; Bihari et al., 2007; Roozbeh et al., 2012). PAHs have been identified as important class of marine contaminants especially the di- and tri-aromatic compounds that are known to be narcotic

to marine organisms such as the mussel, *Mytilus edulis,* while many high molecular mass PAHs are suspected to be probable human carcinogens (Widdows et al., 1995).

The increase in anthropogenic activities at the harbour may have led to the increase in discharge of PAHs into the environment (Oris et al., 1998; FAO corporate document repository). Human beings are at higher level of food web therefore they accumulate polycyclic aromatic hydrocarbons as they consume marine animals such as fishes (Fisler, 1987). PAHs induce neoplastic effects in aquatic organisms (Bihari et al., 2007). Sediment dwelling organisms are most likely to be adversely affected by these PAHs as the sediment acts as a sink for PAHs (CCME, 1999). Hepatic tumors have been reported in some sediment dwelling fish, although PAH sediment levels are usually lower than those found in these studies (Henry, 2002).

Tema Harbour is one of the largest harbours in West Africa spanning a land area of over 3.9 million square metres. It handles over 80% of all goods imported into Ghana. The harbour serves many landlocked countries in the sub-region and also is a very vibrant fishing region and a canoe landing site for local fishermen (Ghana Ports Authority, 2014). Many industries are located in Tema and these include food and beverages processing companies, oil processing companies, aluminium, steel and cement works and small scale processing industries (Ghana Ports Authority, 2009). The industrial activities coupled with high vehicular traffic and large cargo trucks may release large amounts of pollutant into the environment. The effect of fishing boats, canoes (with outboard motors) and cargo ships on water quality is unknown. There are no records of the levels of PAHs in sea water and sediments at the Tema Harbour, hence the aim of this work was to assess the levels of PAH in waters and sediment of the Tema harbour complex. The results obtained from this investigation would give a direction to the control and regulation of pollutants by the Ghana Ports and Harbours Authority, and other stakeholders and provide a baseline range of PAHs concentrations upon which future assessment of water and sediments quality would be referred to.

MATERIALS AND METHODS

The city of Tema is located on the Greenwich Meridian at the south-eastern part of Ghana in West Africa at 5°38'N and 0°01'E. The Tema Harbour (Figure 1) comprises of a Main Harbour, an Outer Fishing Harbour, an Inner Fishing Harbour and a Canoe Landing Site. It also has 77200 m^2 of pavement area for the storage of containers, steel products and other conventional cargo. The closed storage area, which is about 25,049 m^2, consists of six (6) sheds with a total storage capacity of 50,000 tonnes of cargo (Ghana Ports and Harbours Authority, 2008). There are also goods storage facilities, parks for heavy duty trucks and other vehicles. Cold Stores, Commercial Banks, Offices and other buildings are located in the vicinity to facilitate business activities.

Sampling

Each surface water or sediment sample was collected from each sampling site and stored into a 2.5 L amber Winchester bottle. Selection of sampling sites depended on activities occurring in the harbour (Table 1). Three samples were collected randomly in a particular sampling site within a radius of 10 m and mixed together after which a representative sample was taken for analysis. The pH of the water samples were adjusted to below 2 by addition of 2 M hydrochloric acid (MERCK) to inhibit biological activity (Liapis et al., 2008). Wide mouth sampling (glass) bottles (500 ml), lined with aluminium foil were used to collect sediments from the bottom of the sea, at the same sites where the water samples were collected. The samples were placed in an ice chest containing melting ice to keep them under a temperature not more than 4°C.

Extraction of PAHs

(a) Sediment: About 5 g of the sediment was homogenized with sodium sulphate in a ratio of 1:4 and were air dried. This was done gradually in an agate mortar and pestle until it became loose and free-flowing. The sediment was Soxhlet extracted with dichloromethane (SIGMA-ALDRICH) according to the method described by Kelly et al. (2007), USEPA (2003a); Wolska (2008) and Md Suhaimi (2007).

(b) Water: Sixty millilitres of dichloromethane was added to the water in the sample bottle and shaken to extract the PAH, and the whole mixture was transferred into a 5 L separating funnel. Twenty millilitres of dichloromethane was used to rinse the inner surface of the sample bottle and transferred to the separating funnel. The extraction was done according to the method recommended by Magdalena et al. (2006), USEPA (2003b) and Zakaria and Mahat (2006).

Clean-up

About 0.5 g of activated alumina was added to the concentrated extract and transferred into a conditioned column (30 ×1.5 cm ID). The PAHs was eluted from the column with 25 ml of hexane/dichloromethane mixture in the ratio of 5:1. The volume of the eluate was reduced to 1.00 ml and quantitatively transferred into a GC vial (Magdalena et al., 2006; Zakaria and Mahat, 2006)

GC/FID analysis

The PAH measurements were performed using the Agilent

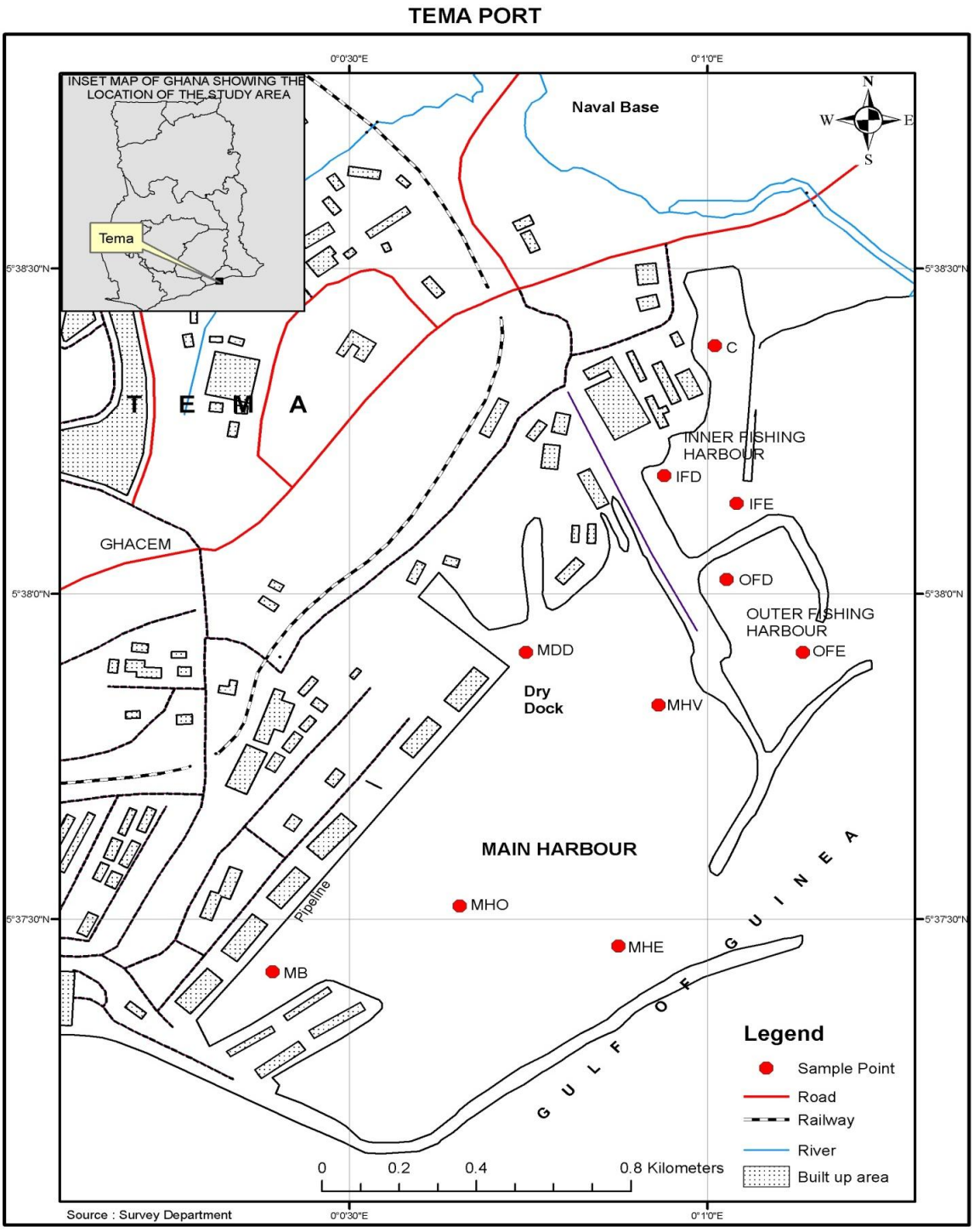

Figure 1. The lay-out of Tema Harbour Complex showing the sampling sites.

Technologies 6890N network gas chromatograph system with flame ionization detector on a SLB-5MS fused silica (5% phenyl methyl polysiloxane) capillary column (30 × 0.25 mm ID, and 0.25 µm film thickness) (SIGMA-ALDRICH). After the system was conditioned, 3 µl of each sample was injected manually into the inlet, in the split mode of ratio 39.6:1 of carrier gas to sample, of the GC at a temperature of 300°C, pressure of 231.2 kPa and flow rate of 173 ml/min.

The oven was programmed as follows: initial temperature was kept at 60°C for 2 min; then heating resumed to 170°C at a rate of 40°C/min and continued to

Table 1. Sampling sites.

Location Name	Location Code	Description
Main Harbour Entrance	MHE	Ships' entering point
Main Harbour Oil berth	MHV	Crude oil &VALCO dock
Main Harbour berths	MB	Cargo ships dock
Main Harbour middle	MHO	Central portion of the main port
Main Harbour dry-dock	MDD	Dry Dock entrance
Outer Fishing Entrance	OFE	Fishing vessels' entering point
Outer Fishing Dock	OFD	Fishing vessels' dock
Inner Fishing Entrance	IFE	Fishing vessels' entering point
Inner Fishing Dock	IFD	Fishing vessels' dock
Canoe Landing Site	MC	Local fishermen landing site

220°C at a rate of 10°C/min. The rate of heating changed to a rate of 10°C/min to 290°C. The system was held at this temperature for 10 min resulting in a total runtime of 33.75 min. The detector temperature was held at 300°C with the flow rate of 55 ml/min for hydrogen, 450 ml/min for air and 15 ml/min for make-up gas, helium (USEPA, 2003c; Braithwaite and Smith, 1999).

Recovery analysis

Recovery analysis was done to verify the extent of PAHs extraction. This involved spiking of the samples (sediments and water) with known concentrations of deuterated *p*-terphenyl standard solution. The recovery test samples were taken through the same extraction, clean-up and instrumentation as done for the main samples. Recovery of samples was between 85 - 98% and this falls within the recommended range of 60 – 120% (USEPA, 2003b; Kelly et al., 2007).

Extraction efficiency analysis

In the absence of a standard reference material for method validation, the extraction efficiency was determined by sampling a different set of sediment and water samples from relatively less polluted area and spiked with known concentration of working deuterated PAH internal standard solution: 1 and 3 µg for water and sediments respectively. This contained Acenaphthene-d_{10}, Chrysene-d_{12}, Naphthlene-d_8, Perylene-d_{12}, and Phenanthrene-d_{10}. It was allowed to rest for about 10 minutes so that there would be adsorption of the PAHs unto the particles before homogenization and extraction.

The spiked sediments were taken through the same analytical method as done for the samples. The solvent was collected after the 6 h of extraction for clean-up and analysis. Fresh solvent was added to the same sample and extraction continued for another 3 h. This fresh solvent was collected and taken through the same analytical method. This was to determine the amount of PAHs that is not extractable after the 6 hours used for the samples.

RESULTS AND DISCUSSION

Twenty polycyclic aromatic hydrocarbons were identified including the sixteen priority ones (USEPA, 2003c) in the sea water and sediments collected from ten different locations in the harbour. The identified polycyclic aromatic hydrocarbons are benzo(a)anthracene B(a)A, benzo(a)pyrene B(a)P, benzo(b)flouranthene, B(b)F, benzo(k)fluoranthene B(k)F, chrysene (CHY), dibenz(a,h)anthracene (DaHa) and indeno(1,2,3-cd)pyrene (IP) which are known human carcinogens. The others are non-carcinogenic PAHs namely: napthalene (NAP), acenapthylene (ACY), acenaphthene (ACE), fluorene (FL), phenanthrene (PHE), anthracene(AN), fluoranthene(FLU), pyrene(PY) and benzo(g,h,i)perylene which make up the 16 priority PAH pollutants. The rest are cyclopenta(c,d)pyrene (CyP), benzo(j)flouranthene B(j)F, benzo(e)pyrene B(e)P and anthanthrene (ANT).

Total mean concentrations of the PAHs in the sea water are generally less than concentrations in sea sediments (Table 2 and Figure 2). Polycyclic aromatic hydrocarbons are non-polar, hydrophobic compounds, which do not ionize. They have a relatively low solubility in water, but are highly lipophilic. Dissolved and colloidal organic fractions also enhance the solubility of PAHs which are incorporated into micelles (Neff, 1997). Due to their hydrophobic nature, PAHs entering the aquatic environment exhibit a high affinity for suspended particulates in the water column. As PAHs tend to adsorb to these particles, they are eventually settled out of the water column onto the bottom sediments. Thus, the PAH concentrations in water are usually quite low relative to the concentrations in the bottom sediments (Moore and Ramamoorthy, 1984).

An ANOVA was done to analyse the concentrations of

Table 2. Total mean concentration of ∑20 PAHs found in sediments (µg/g) and water (µg/L) collected at the Tema Port.

Sampling sites	Sediments	Water
MC	190.3±4.5	53.9±1.5
OFE	56.2±1.7	36.5±1.3
OFD	83.5±1.6	62.0±1.5
IFE	161.2±3.5	33.2±0.3
IFD	144.7±5.7	84.5±1.5
MDD	28.6±0.5	47.1±2.5
MHV	34.9±1.5	65.7±1.9
MHE	102.2±2.2	50.6±0.8
MHO	116.4±5.1	61.4±1.2
MB	63.7±2.4	63.5±1.0

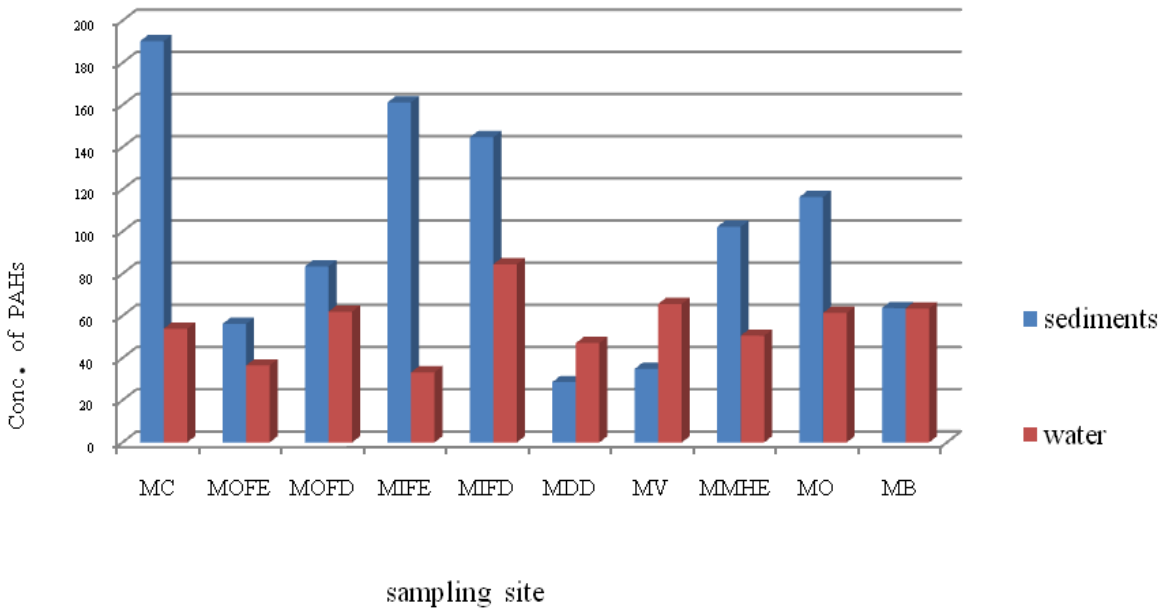

Figure 2. Comparison of total mean concentration of ∑20 PAHs found in sediments (µg/g) and water (µg/L) collected at the Tema Port.

PAHs obtained. The results show that concentrations of PAHs at the canoe landing site (MC) are significantly different and higher than levels in the inner fishing harbour, IFE and IFD, with p-values of 0.003 and 0.018 respectively. This ascertains the fact that the canoe landing site is more polluted than the inner fishing harbour due to the numerous anthropogenic activities that go on there, as a result introducing more PAHs. There is a canal that discharges effluents from the Chemu Lagoon into the canoe landing site. The Chemu Lagoon carries a lot of the industrial waste from the industries in Tema into the sea (Nukpezah, 2010). The canoes normally are propelled by outboard motors with two-stroke engines which are known to release large amounts of petroleum products into the surrounding waters than four-stroke engines (Oris et al., 1998).

Sediments at all these sites are made of fine grains and they are oily probably as a result of oil spillage, changing of dirty oil in the engines of fishing vessels and washing of equipment by fishermen. Some fishmongers also smoke fish at the canoe landing site and this could contribute to the high levels of PAHs. However, there is no significant difference in the concentrations of PAHs at IFE compared to concentrations at IFD since both sampling points are in the same basin therefore sources and conditions of PAHs pollution are similar.

PAHs pollution at the outer fishing harbour is less as compared to the inner fishing harbour such as the canoe landing site and even some locations in the main harbour. There are generally fewer activities here than at the inner

fishing harbour and the canoe landing site. The outer fishing harbour is sandwiched between the main harbour and the inner fishing harbour and for that matter, may not be exposed to other sources of pollutants.

Five sites are located in the main harbour for sampling due to its size and layout so that a representative sample is obtained to show the degree of pollution of water and sediments in the basin. An ANOVA result obtained depicts that the concentrations of PAHs at the sites in the main harbour are significantly different from one another. Concentrations of PAHs at site labeled MDD are significantly different from those at site MHE and MHO with p-value of 0.015 and 0.004, respectively. However, concentrations are almost equal with levels at sites MHV and MB which are all docking sites. Site MHV also shows that concentrations of PAHs are significantly different from levels at site MHE and MHO with respective p-values of 0.026 and 0.007. Site MHE has concentrations of PAHs which are significantly different from concentrations at site MDD and MHV with p-values of 0.015 and 0.026, respectively. On the other hand, they are almost equal to concentrations at site MHO and MB.

Looking at the map (Figure 1), the three sites are on the western part of the harbour which may imply that pollutants are introduced from the same source and are transported by the eastward flowing coastal Guinea Current. There are some canals that discharge industrial effluent directly into the harbour and this could be one of the reasons for the higher concentrations at that part of the port. The mean concentrations of PAHs at site MHO show that levels are significantly different from levels at sites MDD and MHV with p-values of 0.004 and 0.007, respectively. Mean concentrations at site MB show no significant difference with concentrations at all the other sites.

Both particle size and total organic content of sediments have been shown to be important factors in sediment PAH distribution, suggesting a particle size effect due to differences in adsorptive surface area (Ho et al., 1999)

PAH concentrations are found to be greater in muddy sediments from the estuaries of the Rivers Blyth, Tyne, Wear and Tees in NE England. Offshore sites and those with coarser sediment types generally yielded much lower concentrations (Trapido, 1999). In the main harbour, some sites are found to be rocky, consisting of large particle sizes and for that matter lower concentrations are recorded there. For example, there are sandy sediments collected at the MDD site as opposed to muddy sediments at the MB site, which show lower concentrations. Sediments at some of the sites too are more oily than others and these may account for the differences in the concentrations of PAHs in the main harbour.

The total mean concentrations of the ∑20 PAH at the 10 sampling sites range from 28.6 to 190.3 µg/g in sediments. Chanbasha et al. (2003) report that the total concentration of 16 PAH, classified as USEPA (2003c) priority pollutants within Singapore's marine waters that vary between 15.22 and 82.41 µg/g in the northeastern region and between 13.63 and 84.92 µg/g in the southwestern region. In this project work, the ∑16 PAHs was in the range of 25.85 - 155.14 µg/g. These relatively high results, might be due to the fact that the sediments in the port are restricted in movement as compared to the open sea sediments which are highly subject to movement, and for that matter pollutants that are released into the port remain there and may accumulate.

In lakes and oceans pollutants are transported through currents (Kenn, 1992). There are many currents in the oceans, which are wind-driven and this enables a pollutant to travel from one continent to another (Hecht, 1997). We usually count on the ability of the oceans to reduce pollutant concentration, the so-called 'self-cleaning ability' of oceans (ACS, 2013; Lenntech, 2009) but this does not always work, because the movement of the currents in the oceans is not uniform (Segar, 2012). This causes inshore waters to often have substantially higher levels of pollution than the open sea (Zakania and Mahat, 2006). However in a harbour, the marine hydrodynamics are relatively confined, as the tidal exchange is restricted (Segar, 2012). Therefore, the oceanic currents and sedimentation rates facilitate particulate dispersion differently especially in the different basins at the port.

The results show that the total concentrations of the PAHs are comparatively lower than those reported for sediments from Victoria Harbour, Hong Kong which ranged up to 387 µg/g (Srogi, 2007), Kitimat Harbour, Canada, ranged up to 10,000 µg/g (Simpson et al., 1996), Times Beach, New York, U.S.A. ranged up to 480 µg/g (Roper and Chery, 1994), Santander Bay, Spain ranged up to 344.6 µg/g (Viguri et al., 2002) and Harbour of Refuge, Rhode Island Sound, U.S.A. ranged up to 730.0 µg/g (Ho et al., 1999).

Conclusion

Results obtained from this study show that the sea water and sediments at the Tema Harbour are polluted by some amounts of PAHs. The total mean concentrations of the ∑20 PAH at the 10 sampling sites ranged from 28.6 to 190.3 µg/g w.wt, in sediments and from 33.2 – 84.5 µg/L in water. It can also be realized that the canoe landing site is the most polluted basin at the Tema harbour complex. There are significant differences in the mean concentrations of PAHs at the various sampling sites in the main harbour basin. It can be concluded that PAHs at the right side (south-west) of the main port are of higher concentrations than at the left side.

Recommendations

With the increasing rate of population as a result of

economic and industrial activities at Tema, issues about pollution should not be left to get to levels where it becomes uncontrollable. This includes the levels of PAHs in water, biota, soil sediments, food and air in the city. A knowledge of PAH concentrations in air at Tema would help explain the rate of transportation and transformation of PAHs in the sea sediments and water. Efforts should therefore be made to control the activities at the harbour, especially the canoe landing site, to reduce the concentrations of pollutants introduced into the harbour.

ACKNOWLEDGMENT

The authors wish to appreciate the immense support from the Quality Control Laboratory of the Tema Oil Refinery and Port and Harbours Authority which are based in Tema, Ghana.

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Cite this article as:

Gorleku MA, Carboo D, Palm LMN, Quasie WJ, Armah AK (2014). Polycyclic aromatic hydrocarbons (PAHs) pollution in marine waters and sediments at the Tema Harbour, Ghana. Acad. J. Environ. Sci. 2(7): 108-115.

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