UNIVERSITY OF CAPE COAST

DISTRIBUTION, TOXICOLOGICAL EFFECTS AND REMEDIATION OF POLYCYCLIC AROMATIC HYDROCARBONS IN SELECTED LAGOONS IN GAHANA

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BY

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Thesis submitted to the Department of Fisheries and Aquatic Sciences of the School of Biological Sciences, College of Agriculture and Natural Sciences, University of Cape Coast, in partial fulfilment of the requirements for the award of a Doctor of Philosophy (Ph.D.) degree in Oceanography and Limnology

NOVEMBER 2020

DECLARATION

Candidate's Declaration

I hereby declare that this thesis is the result of own original research and that no part of it has been presented for another degree in this university or elsewhere.

Candidate's signature …………………………. Date …………………………

Name: RAHMAT QUAIGRANE DUKER

Supervisors' Declaration

We hereby declare that the preparation and presentation of the thesis were supervised in accordance with the guidelines on supervision of thesis laid down by the University of Cape Coast.

Principal supervisor's signature ……………………...Date ……………………

Name: DR. NOBLE K. ASARE

Co-supervisor's signature ……………………... Date ……………………….

Name: PROF. EDWARD A. OBODAI

ABSTRACT

Polycyclic Aromatic Hydrocarbons (PAHs) are ubiquitous and persistent organic pollutants. Their dynamics in aquatic environments have been extensively studied, but studies mainly focused on their distribution in single compartment of the aquatic environment. A study on 16 PAHs were analysed in water, sediments, and fish tissues from Benya, Brenu and Fosu Lagoons in Ghana, using GC-MS. The potential application of activated charcoal for the remediation of PAH-contaminated sediment was also investigated. PAHs were extracted using SPE cartridges and QUECHERS, whilst diagnostic ratios were used to determine the sources of the PAHs. The concentrations of 16 PAHs ranged from 32.37 (fillet) to 2050.47 (visceral) µg/kg/w.w, 20.81 to 24801.38 µg/kg/d.w and 1.17 to 3.39 µg/L for fish tissues, sediment and water respectively. The total PAHs recorded in the sediments demonstrated both spatial and seasonal variations in the distribution of PAHs. Environmental factors such as temperature and pH, as well as the total organic carbon {F (4, 292) = 18.19, p <0.001, R^2 = 0.20} were also noted to influence the dynamics of PAHs in these lagoons. Indiscriminate burning of wastes, shipping activity, vehicular emissions and sewage were among the main sources of PAHs. More than 50 % of individual PAHs in the sediments exhibited a moderate level of ecological risk for aquatic life. The calculated ILCR for sediments (varied from 2.5×10^{-6} to 6.0×10^{-5}) and fish tissues (from 2.0×10^{-5} to 4.6×10^{-5}) were above the USEPA safety limit (1×10^{-6}); an indication of cancer-related effects to humans who are exposed to the sediments and consumption of *Sarotherodon melanotheron* in these lagoons. Activated charcoal is recommended as an effective carbonaceous material to remediate the sediments of these lagoons.

KEY WORDS

Polycyclic Aromatic Hydrocarbons (PAHs)

Coastal lagoons

Sarotherodon melanotheron

Ecological risk

Human health risk

Remediation

LIST OF ACRONYMS

AC	Activated charcoal
Ace	Acenaphthylene
Acp	Acenaphthene
Ant	Anthracene
BaA	Benz[a]anthracene
BaP	Benzo[a]pyrene
BbF	Benzo[b]fluoranthene
BC	Black carbon
BFRs	Brominated flame retardants
BkF	Benzo[k]fluoranthene
BP	Benzo[g,h,i]perylene
Chr	Chrysene
DahA	Dibenz[a,h]anthracene
ERL	Effect range-low
ERM	Effect range-median
F	Fluorene
Flu	Fluoranthene
GC-MS	Gas chromatography-mass spectrometry
HMW	High molecular weight

HOCs	Hydrophobic organic compounds
ILCR	Incremental Lifetime Cancer Risk
IP	Indeno[123-cd]pyrene
LMW	Low molecular weight
mERM-Q	mean effect range-median quotient
MCL	Maximum Contaminant Level
MPCs	Maximum permissible concentrations
Naph	Naphthalene
NCs	Negligible concentrations
PAHs	Polycyclic aromatic hydrocarbons
PCBs	Polychlorinated biphenyls
PEC	Potency equivalent concentration
PFCs	Polyfluorinated compounds
Phe	Phenanthrene
POPs	Persistent organic pollutants
Pyr	Pyrene
QUECHERS	QUick, Easy, CHeap, Effective, Rugged and Safe
RQ	Risk quotient
Solid Phase Extraction	SPE
SQG	Sediment quality guidelines

TEF	Toxicity equivalency factors
TEQ	Toxic equivalent quotient
USEPA	United States environmental protection agency
WHO	World Health Organisation

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DEDICATION

To people who are doing their best in their field of work for the sake of humanity. Know the impact is worth it, so keep moving!!!

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CHAPTER ONE

INTRODUCTION

Coastal lagoons are vital ecosystems that provide various ecological, socioeconomic, and cultural services (Gray, 1997; Harris & Vinobaba, 2013; Newton et al., 2018; Small & Nicholls, 2003). These services include providing food and water, erosion control, nutrient cycling, human settlements, tourism, spiritual and recreational benefits. However, due to the increase in anthropogenic activities, such ecosystems have become exposed to higher levels of polycyclic aromatic hydrocarbons (PAHs). Given PAHs' toxicological effects, the knowledge about their sources and fate in the aquatic environment is imperative. PAHs are persistent, poorly degradable, bioaccumulative, and can be transported over long distances (Ekere, Yakubu, & Ihedioha, 2017; Francisco, 2016; Sojinu, Sonibare, & Zeng, 2013). They can undergo partitioning between water, sediments, and biota in the aqueous environment, depending on the contamination level. Thus, it is imperative to periodically assess the ecological health of lagoons exposed to PAHs contamination. Most studies have focused on a single medium investigation (i.e., either water, sediment, or biota), with fewer studies looking at the ecosystem's comprehensive media. It is essential to employ a more comprehensive approach to identify the possible synergistic or antagonistic behaviours influencing the concentration of PAHs in different compartments (media) in such ecosystems. For this purpose, this work investigated the water, sediment, and biota (fish) of three coastal lagoons in Ghana to assess the distribution and adverse effect of PAHs in the ecosystems selected.

Sediment is the main sink of PAHs when released into an aquatic ecosystem. Hence, when these pollutants are above the permissible limit, the sediment is being remediated to restore the ecosystem's health and other vital uses like fisheries and recreation (Kupryianchyk et al., 2011). Currently, the use of activated carbon, as opposed to the traditional method, dredging, is the upcoming approach for remediating contaminated sediment. Therefore, this study's goal was not only to give a broader insight into the distribution of PAHs in aquatic environments but also, to consider remedial action for management purposes.

1.1 Background to the Study

The 550 km coastline of Ghana, which forms part of the Gulf of Guinea, has more than 90 reported lagoons (Armah, Luginaah, & Kuitunen, 2012; Nixon et al., 2007; Yankson & Obodai, 1999) spanning its entire stretch. These lagoons are categorised mainly into two types: the “open” and the “closed” lagoons. The former is always opened to the sea and connected with rivers that flow all year round. An open lagoon is partly under tidal influence, while a closed lagoon remains closed to the sea for most of the year by a barrier such as a sand bar (Biney, 1990). In this study, both types were considered. Lagoons play a vital role in almost all biogeochemical processes that sustain the biosphere, as well as offer different services and functions that have contributed significantly to the human growth and development (Gaglio et al., 2019; Inácio et al., 2018; Newton et al., 2018; Sousa, Lillebø, Gooch, Soares, & Alves, 2013; Velasco, Pérez-Ruzafa, Martínez-Paz, & Marcos, 2018). For instance, coastal lagoons play a crucial role in the hydrological cycle by storing, regulating, and recharging both surface and groundwater supplies. Coastal lagoons also absorb, filter, process, and reduce the presence of nutrients, pollutants, and wastes. They are a tank for primary production, providing nutrients for aquatic organisms, and suitable breeding and nursery grounds for freshwater and marine species (Newton et al., 2018). In the global carbon cycle, coastal lagoons act as sinks for carbon, thereby reducing its emissions. Additionally, coastal lagoons provide protection from natural hazards by acting as a protection belt on the coast, mitigating the vulnerability of coastal areas to sea-level rise and erosion (Gaglio et al., 2019; Inácio et al., 2018; Newton et al., 2018; Sousa et al., 2013; Velasco et al., 2018). However, due to the increase in anthropogenic activities, such ecosystems have become exposed to intense human pressures, affecting their ecological status.

Most inland activities from agriculture, human settlements, and industries generate pollution and land erosion (causing siltation), all of which impact coastal water bodies (Armah, Luginaah, et al., 2012; Secco, Pellizzato, Sfriso, & Pavoni, 2005). Nutrients (nitrogen and phosphorus), agricultural chemicals (pesticides, insecticides, herbicides, and fungicides), heavy metals, and PAHs are some of the contaminants released into the coastal water bodies (FAO, 2019; Gilbert, Dodoo, Okai-Sam, Essumang, & Quagraine, 2006; Nixon et al., 2007; Odjer-Bio, Belford, & Ansong, 2015). Most studies in Ghana have focused on contaminants such as nutrients levels and heavy metals (e.g., Akoto, Bismark Eshun, Darko, & Adei, 2014; Ansa-Asare, Mensah, Entsua-Mensah, & Biney, 2009; Armah, Luginaah, et al., 2012; Clottey, 2018; Gilbert et al., 2006; Nixon et al., 2007; Obodai et al., 2011; Odjer-Bio et al., 2015), with limited data on the levels and distribution of PAHs in the coastal lagoons in Ghana. Industrial activities, fossil fuel combustion, vehicular emissions, sewage drainages, defecating grounds, animal husbandry, vehicle washing, incineration of wastes, among others, are known anthropogenic sources of PAHs (Essumang, 2010; Gilbert et al., 2006). Almost all of these activities currently occur in the vicinity of these studied coastal lagoons. Therefore, it is essential to periodically assess the physical, biological, and chemical relationships that may determine lagoons' ecological health under the influence of PAHs contamination. This current study seeks to achieve that by investigating the distribution and the effects of PAHs in selected brackish water systems using water, sediment, and fish samples.

1.1.1 Polycyclic aromatic hydrocarbons

PAHs are organic compounds consisting of conjoined aromatic rings with or without heteroatoms (Manoli, Samara, Konstantinou, & Albanis, 2000; Neff, Stout, & Gunster, 2005; Schwarzenbach, Gschwend, & Imboden, 2003). Thousands of PAHs and other organic pollutants belonging to about 660 parent PAH compounds can be found in the environment. (Sander & Wise, 1997). Parent PAHs are those without alkyl groups and are exclusively fused rings of carbon and hydrogen conjoined to each other. They range from the monocyclic molecule of benzene (molecular weight, MW = 78) up to nine-ringed structures (MW up to 478). PAHs consisting of one or more alkyl groups are known as alkyl PAHs. In this study, the focus was on sixteen PAH species (parent compounds) recognised by the USEPA and EU as priority pollutants due to their higher potential of adverse effects in humans and aquatic species than the rest of the groups (Figure 1). The 16 priority PAHs include: Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benz[a]anthracene, Chrysene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[a]pyrene, Indeno[1,2,3-cd]pyrene, Dibenz[a,h]anthracene, Benzo[g,h,i]perylene (Wenzl, Simon, Anklam, & Kleiner, 2006).

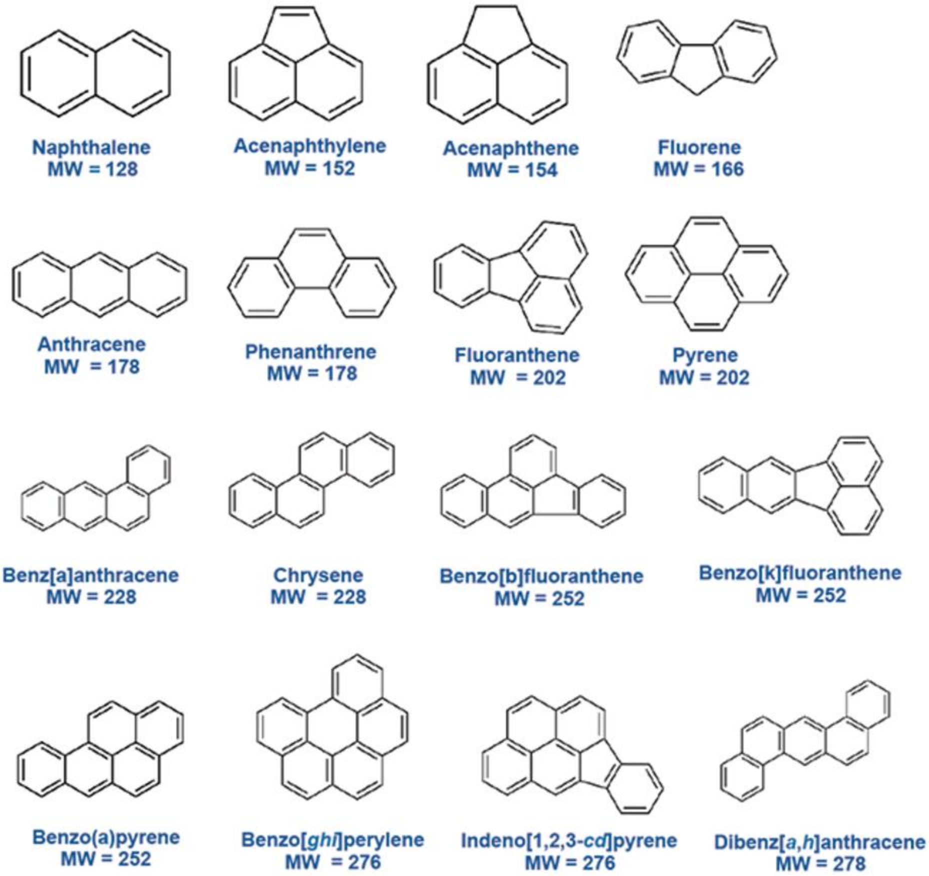

Figure 1: Nomenclature, chemical structure, and molecular weight for 16 -priority PAHs. Source (Advion, 2017).

Generally, PAHs are classified into three groups based on the temperature at which they form, or their origin (Boehm, Neff, & Page, 2007; Mitra, Dellapenna, & Dickhut, 1999). These are: pyrogenic, petrogenic and biogenic or diagenetic origin. It has been realised that the distribution of PAHs varied among different PAH sources (Grimmer, Jacob, & Naujack (1981), Laflamme & Hites (1978), and Youngblood & Blumer (1975). These findings have led to ongoing efforts to discover the appropriate molecular indices of PAH distribution that would allow source characterization of contaminated areas. These indices aid in identifying ubiquitous PAH markers, PAH abundance, modes of distribution, and whether they are of pyrogenic or petrogenic sources. (Stogiannidis & Laane, 2013). According to Yunker et al. (2002), Santos, Brehm, Filippe, Reichert, & Azevedo (2017), and Ashayeri et al. (2018), these molecular indices are possible due to the differences in their thermodynamic properties, distinguishing the most stable isomer from the least stable, making it a judgemental tool to differentiate their sources. One challenging situation in identifying PAHs' origin is the possible coexistence of various contamination sources and transformation processes that PAHs may undergo before final deposition in the analysed media. Nevertheless, compounds might display comparable pathways that might be applied to identify their source.

PAHs are not synthesized chemically for industrial purposes. Yet, there are some commercial uses for many PAHs. They are commonly used as intermediaries in pharmaceutical manufacturing (by acting as catalysts for broader and more complex pharmaceutical products), agricultural products, photographic products, thermosetting plastics, lubricating materials, and other chemical industries (Abdel-Shafy & Mansour, 2016; Timbrell, 1981). Some other specific uses of some PAHs are:

- *Acenaphthene:* manufacture of pigments, dyes, plastics, pesticides, and pharmaceuticals.
- *Anthracene:* diluent for wood preservatives and manufacture of dyes and pigments.
- *Fluoranthene:* manufacture of agrochemicals, dyes, and pharmaceuticals.

- *Fluorene:* manufacture of pharmaceuticals, pigments, dyes, pesticides, and thermoset plastic.
- *Phenanthrene:* manufacture of resins and pesticides.
- *Pyrene:* manufacture of pigments.

Other PAHs may be contained in asphalt and roofing tar (Abdel-Shafy & Mansour, 2016).

PAHs are also grouped as low molecular weight (LMW) and high molecular weight (HMW) compounds depending on their molecular structure. The LMW PAHs are those having two or three benzene ring structures (e.g., naphthalene), while HMW PAHs have four or more benzene ring structures (e.g., Benzo[a]pyrene). The HMW PAHs are less water-soluble, less volatile, and more lipophilic than LMW PAHs (Edokpayi, Odiyo, Popoola, & Msagati, 2016; Wania & Mackay, 1996). Due to their high hydrophobicity, PAHs tend to bind to organic particles and undergo sedimentary adsorption when introduced into aquatic environments. Therefore, they end up in depositional areas such as the substrate of aquatic systems or bio-accumulate organisms such as fish. In fish, this could occur directly through ingestion of contaminated food at lower trophic levels (Logan, 2007; Meador, Stein, Reichert, & Varanasi, 1995), adsorption of dissolved contaminants through diffusion from water across their gills and skin (Abdel-Shafy & Mansour, 2016; Gobas, Wilcockson, Russell, & Haffner, 1999) or indirectly through HOC release after re-suspension or bioturbation. Fishes can, therefore, serve as bioindicators for PAHs in aquatic environments (Abdel-Shafy & Mansour, 2016; Pointet & Milliet, 2000).

Bioaccumulation of PAHs in fish and accumulation of PAHs in the sediment of an aquatic ecosystem can negatively affect biota's health and productivity. Fish occupies a key position in aquatic and terrestrial food webs. Humans consume fish, and many species, including other fish, piscivorous birds, and several scavengers also preyed on fish. Hence, changes in fish populations as a result of the effects of PAHs can lead to changes in other populations relying on them for food (Logan, 2007). Some of the adverse effects of PAHs on fish species include growth reduction (Christiansen & George, 1995; Nyarko, Botwe, & Klubi, 2011), endocrine alteration (Meador et al., 1995), malformations of embryo and larvae (Camus & Olsen, 2008; Carls et al., 2008) and DNA damage (Caliani et al., 2009).

1.1.2 Black-chinned tilapia (Sarotherodon melanotheron)

In Ghana, wetland ecosystems form about 10% of the country’s total land surface area, with lagoons and estuaries being the most valuable wetlands (Okyere, Aheto, & Aggrey-fynn, 2011). They are associated with salt marshes, mangroves swamps, tidal flats, macro-benthic fauna, and sensitive habitats housing many fish and wildlife species that support the country’s economy (Attuquayefio & Gbogbo, 2009). Ghana's lagoons have historically produced high fish yields, predominantly *Sarotherodon melanotheron* (Nixon et al., 2007; Odjer-Bio et al., 2015; Pauly, 1976). With such knowledge, the current study selected *S. melanotheron* for the PAHs assessment in fish. With the abundance of *S. melanotheron* in several lagoons in Ghana, this study's results would provide requisite monitoring information on the level of PAHs in Ghana's coastal lagoons.

Sarotherodon melanotheron, commonly called black-chinned tilapia, is an ecologically engaging bio-indicator that exhibits exceptional adaptive capacities, especially its tolerance to salinity. It is a cichlid, and normally populates freshwater and brackish water ecosystems. They can reproduce at salinities between 0 to 120 ppt (Panfili et al., 2004; Panfili, Thior, Ecoutin, Ndiaye, & Albaret, 2006). This species is endemic to West Africa and geographically, ranging from Senegal to Zaire (Mireku, Blay, & Yankson, 2016). It represents one of the key fish species caught in most coastal lagoons in Ghana. Its abundance makes the species a vital resource for subsistent fisheries in the brackish water ecosystems. The species is predominantly planktivorous, feeding mainly on phytoplankton and zooplankton. It is a paternal mouthbrooder which is known to exhibit parental care (Eyeson, 1979; Mireku et al., 2016; Ndimele, Kumolu-Johnson, Aladetohun, & Ayorinde, 2010; Pauly, 1976; Zuh, Abobi, & Campion, 2019). The males pick up the fertilized eggs and incubate them until they are released as free-swimming fry (Avarre et al., 2014). *S. melanotheron* is one of the primary sources of protein diet for the communities surrounding these lagoons (Obodai et al., 2011).

It is known that fish rapidly metabolise PAHs (Hellou & Payne, 1987; Zelinkova & Wenzl, 2015) to intermediates, which either bind to liver DNA or form conjugates, which then pass into the bile. For example, the composition and concentrations of PAH metabolites in the fish's visceral are expected to indicate the extent of their exposure to PAHs (Pointet & Milliet, 2000). With this understanding, the visceral of the fish was of interest in this current study. Fish fillet may also be exposed to dissolved contaminants by diffusion of contaminants from water across their skin (Bandowe et al., 2014; Cheung, Leung, Kong, & Wong, 2007). Therefore, the fish fillet was also considered in this current study. With background knowledge that direct diffusion of contaminants occurs between the gills and the surrounding water during respiration in black-chinned tilapia, PAHs' concentration in the fish head was also considered in this present study.

1.1.3 Contaminated sediment remediation

When contaminants are released into aquatic environments the hydrophobic contaminants like PAHs rapidly adsorb onto suspended material, biota, and sediment (Arias, Spetter, Freije, & Marcovecchio, 2009; Liang, Tse, Young, & Wong, 2007; Montuori et al., 2016; Santos et al., 2017; Zheng, Wang, Lei, & Nan, 2016). Due to the presence of organic matter and other domains in the sediment (such as black carbon soot), PAHs principally adsorb onto the sediment (Bucheli & Gustafsson, 2000; Cornelissen et al., 2005; Gustafsson, Haghseta, Chan, Macfarlane, & Gschwend, 1997; Koelmans et al., 2006). Therefore, the sediment is the main sink of many of these persistent organic pollutants found in the aquatic environment.

Due to the eco-toxicological effects associated with the presence of PAHs in an aquatic environment, researchers are coming out with innovative and cost-effective technologies that will effectively remove these pollutants without destruction to the ecosystem. Also, because sediment is the primary storage and sinks for PAHs, the contaminated sediment is treated or remediated. Such treatment or remediation aid to restore the ecosystem and other essential uses like fisheries and recreation (National Research Council, 2007). Before considering any of the existing remediation methods for treating contaminated sediment, the goal is to eliminate or control contaminant sources. Contaminated sediment management strategies seek to reduce contaminant adverse effects on human health, the environment, as well as cost effective techniques. Another objective is to reduce the adverse effects of using the remediation technique itself, such as habitat destruction or modification (Zeller & Cushing, 2006). Generally, PAH-contaminated sediment is either dredged (Ghosh, Luthy, Cornelissen, Werner, & Menzie, 2011; Mudge, 2007; National Research Council, 2007), or capped with clean material such as sand, silt, or clay (Murphy, Marquette, Reible, & Lowry, 2006; Samuelsson, Hedman, Elmquist Kruså, Gunnarsson, & Cornelissen, 2015). They may also be bioremediated (Dhar, Subashchandrabose, Venkateswarlu, Krishnan, & Megharaj, 2019; Hale, Meynet, Davenport, Martin Jones, & Werner, 2010) or carbonaceous materials such as activated carbon is use in amending the contaminated sediment. Among the above listed approaches, the use of activated carbon as an amendment is the upcoming technique known to be a promising mechanism for remediating organic and inorganic pollutants deposited in sediment (Bes & Mench, 2008).

In the past century, the rise in biomass burning and fossil fuel usage has tremendously increased black carbon (BC) contribution to the environment (Griffin & Goldberg, 1983; Koelmans et al., 2006). BC is formed from weathering graphitic carbon in rocks (Dickens, Gélinas, Masiello, Wakeham, & Hedges, 2004) or a product of incomplete combustion of fossil fuels and vegetation (Cochrane, 2003; Griffin & Goldberg, 1983). Black carbon is known to be ubiquitous, including the atmosphere (Cochrane, 2003; Laborde et al., 2012), soils (Cheng, Lehmann, & Engelhard, 2008; Liang et al., 2008), ice (Hansen & Nazarenko, 2004; Ming et al., 2009), aquatic sediments (Gustafsson et al., 2001; Schmidt & Noack, 2000), among others. Due to BC's adverse impacts on the environment and human beings, it is relevant to policymakers. For example, it has been reported that BC can catalyse several chemical reactions in the atmosphere, such as nitrous acid formation, which contributes to photochemical ozone and smog formation (Cochrane, 2003). Also, as a component of aerosols, BC can absorb solar radiation, thereby adversely impacting the climate (Ramanathan, Crutzen, Kiehl, & Rosenfeld, 2001). Additionally, it has also been reported that BC can cause various respiratory and cardiovascular diseases in humans (Armstrong, Hutchinson, Unwin, & Fletcher, 2004).

Contrary to these adverse effects, it has been discovered recently that BC also exhibits a beneficial role in the environment (Koelmans et al., 2006). The naturally occurring carbonaceous materials in sediments, such as soot and charcoal, often referred to as BC, can effectively bind organic pollutants (Cornelissen et al., 2005; Koelmans et al., 2006). This binding is parallel to clean manufactured carbonaceous material like activated carbon, used in water-cleaning technologies and as poison control for many years (Karanfil & Kilduff, 1999; Karanfil, Kitis, Kilduff, & Wigton, 1999; Karapanagioti, 2007). These knowledge have led to studies investigating whether intentionally adding types of activated carbon to polluted sediments can reduce the risk towards aquatic organisms as well as the risk of transport in leaching of contaminants (Ghosh et al., 2011; Hilber & Bucheli, 2010; Rakowska, Kupryianchyk, Harmsen, Grotenhuis, & Koelmans, 2012). Rather than excavation and relocation of contaminated material, the use of activated carbon sorbent amendments could directly reduce chemical exposure on-site, thereby saving costs compared to traditional methods (Hilber & Bucheli, 2010; Jonker, Hoenderboom, &

Koelmans, 2004; Rakowska et al., 2012). In this work, the carbonaceous material considered is activated charcoal.

Activated Charcoal (AC) is a porous material with a high surface area and affinity for adsorbing organic chemicals (Accardi-Dey & Gschwend, 2002; Ghosh, Talley, & Luthy, 2001). The presence of activated charcoal in contaminated sediment reduces pore water concentrations by sequestration of the chemicals through partitioning or adsorption to the surface within its pore structure (Accardi-Dey & Gschwend, 2002; Ghosh et al., 2001; Ghosh, Zimmerman, & Luthy, 2003). AC has slow kinetics of contaminant desorption, which implies that hydrophobic organic compounds' fluxes to aqueous phase are low, limiting contaminant mobility in the aquatic environment, thus, making it an effective material for sediment remediation (Ghosh et al., 2001).

In Ghana, dredging and bioremediation (natural attenuation) are the traditional ways considered when sediment remediation is needed. This current study looked at the possibility of activated charcoal (a form of activated carbon) as a remediating material for PAH-contaminated sediment.

1.2 Statement of the problem

PAHs have been comprehensively studied in the coastal environment in many parts of the world (Haftka, 2009; Niu et al., 2018; Perugini et al., 2007; Specchiulli et al., 2011; Zhang, Liang, Wang, Guan, & Zeng, 2012; Zhu et al., 2004), including their adverse effects on the aquatic environment and human health. However, significant data gaps exist on its sources and levels in Ghana’s coastal waters, where most studies (conducted almost a decade ago) on PAHs focused mainly on sediments and water (e.g., Armah, Luginaah, et al., 2012;

Essumang, 2010; Gilbert et al., 2006), with no data on the distribution and eco-toxicological effects of PAHs on fish from these ecosystems. In Ghana, knowledge on PAHs when considering fish has mainly been on smoked fish and marine fishes. There is little or no information on fishes from brackish water, not to talk of PAHs residual levels in different target organs of the fish.

Most studies have also focused on a single medium, rather than different compartments of the aquatic ecosystems. With the tendency of PAHs partitioning into different compartments of an aquatic system, it is essential to survey and monitor the PAHs levels in these compartments to effectively understand the mobility of PAHs in an aquatic ecosystem. From such surveys, fishes, and their relationships with sediments and the water column can serve as a tool for risk assessment of PAHs in the aquatic environments. In the present study areas, there is a gap in a comprehensive survey on PAHs where the sediment, water, and fish are considered coherently. This present study could be considered as the first effort to coherently analyse PAHs in water, sediments, and fish of these studied lagoons.

PAHs are also on the limelight for environmental chemists, toxicologists, as well as regulatory agencies. First, they are contaminants that are released into the environment from diverse sources; principally, from industrial, petro or agricultural chemicals (Arias et al., 2009; Boehm et al., 2007; Mitra et al., 1999). Unlike other human-induced pollutants, PAHs are not usually released in their pure chemical state. They are associated with burning processes, therefore affecting their resilient and bioavailability in the environment. Thirdly, the number of PAHs found in the environment far outweigh the few number of other persistent organic pollutants. There exist more than 10,000 individual PAHs (Arias et al., 2009; Logan, 2007). Most of these persistent organic pollutants have been banned or have some restrictions in their releases into the environment. These restrictions are not effective with PAHs due to their ever present in the environment; especially during their formation through combustion of fossil fuels, petroleum recovery and transport (Arias et al., 2009; Culotta, De Stefano, Gianguzza, Mannino, & Orecchio, 2006). The continuous use of fossil fuels worldwide is a surety evident of release of PAHs into the environment. Lastly, PAHs have been reported to affect the nutritional value and health of human populations who rely mainly on fish for their animal proteins. It is worth to note that PAHs are one of the first and largest sets of compounds known to be strongly mutagenic to laboratory animals and man (Nyarko et al., 2012).

1.3 Purpose of the study

In Ghana, industrial, agricultural, domestic activities, automobile exhaust, and open-air incineration of waste are the major sources of PAH contamination (Nyarko et al., 2012). However, in scaling these to the plausible sources of PAHs contamination to coastal waters, the main sources include municipal and domestic wastes, high rates of indiscriminate dumping and burning of municipal waste within the peripheries of the water bodies, extensive use of coal tar to coat dugout canoes for artisanal fishing activities, poor conditions of two-stroke engines of outboard motors, and from automotive repair shops. These coastal waters are frequently used for domestic and recreational purposes, and fish species caught from such lagoons serve as a source of animal protein for the indigenous people. They are also of significance to the people's traditional heritage and provide a livelihood for fishers, especially in such communities. Therefore, the purpose of this study is to contribute to existing knowledge on the current trend of PAH distributions in such aquatic ecosystems and their potential human health risks. PAHs were chosen over other types of pollutants because of their tendency to partition to sediments and biota, eventually, harmful to both the aquatic environment and humans that depend on such ecosystems.

Sediment is an important reservoir for PAHs; hence, in treating pollutants in the aquatic environment, the sediment is mostly the focal point. For this reason, activated charcoal was considered as a remediating material to remediate PAH-polluted sediment. Such an experiment was to test the effectiveness of activated charcoal in remediating contaminated sediment. Its outcome will provide a mechanistic understanding of the amount of activated charcoal that significantly reduces PAHs' bioavailability through laboratory experiments.

The outcome of this present study will be used to assess the ecosystem quality, one of the thematic areas of the USAID/UCC capacity building project, “Research on Biodiversity and Ecosystems Health Monitoring” in Central and Western Regions of Ghana.

1.4 Research objectives

This work's main objective was to evaluate the dynamics of PAHs in coastal lagoons and investigate the potential remediation of PAH-contaminated sediment using activated charcoal under laboratory conditions.

The specific research objectives were to:

1. Assess the concentration levels and distribution of PAHs in the water, sediment, and fish tissues of three coastal lagoons.
2. Identify possible contamination sources for mitigation purposes.
3. Determine the influence of Total Organic Carbon (TOC), temperature, pH and salinity on PAH levels in the lagoons.
4. Assess the ecological risk associated with PAH contamination levels in the lagoons.
5. Evaluate the potential health risk implications of PAHs in the consumption of fish from lagoons.
6. Determine the influence of activated charcoal on the freely dissolved concentration of PAHs in PAH-contaminated sediment.

1.5 Significance of the study

Ghana is one of the developing countries with emerging industries in West Africa. In 2010, Ghana started offshore oil production in commercial quantities. Oil exploration is known to be a key contributing factor to PAHs in the environment. Currently, not much has been put in place when monitoring PAHs in coastal environments in Ghana. Therefore, knowledge obtained from this current study could be fed into an evaluation and monitoring projects that seek to regulate PAH levels in the country.

Most of Ghana’s coastal waters are becoming a matter of crucial concern, given their present state due to pollution. Several studies have confirmed elevated levels of organic pollutants in these coastal waters. There is, therefore, the need for remediation actions to be taken by policy and regulatory agencies for implementation by environmental managers. Findings from this current work seek to provide vital information for environmental managers regarding alleged remedial actions for lagoon management.

1.6 Delimitations of the study

For easy accessibility, the Central Region of Ghana, one of the thematic areas of the USAID/UCC capacity building project, was chosen for this study. Two closed lagoons and one open lagoon were selected for this work. The distribution of PAHs based on lagoon types was explored. Fosu, a closed lagoon, was chosen to give current information on PAHs' distributions in the lagoon. The recent data on PAHs from Fosu Lagoon was almost a decade ago, and the focus was mainly on the sediment medium. Brenu Lagoon was another closed lagoon that was selected. This lagoon is a vital resource for the people of Ampenyi and Brenu Akyenim. This current study was the first to record levels of PAHs in any of the three media selected (i.e., water, sediment, and fish). Benya Lagoon, on the other hand, was the open lagoon chosen for this study. The selection was based on the fact that this lagoon experiences intense fishing activities. The recent data on PAHs from this lagoon was also on levels in the water column, assessed more than a decade ago.

1.7 Limitations of the study

To effectively address the possibility of these lagoons being contaminated with PAHs, sampling was done purposively within the catchment area of potential PAHs source (e.g., near a burning waste, domestic waste, effluents, etc.). However, sampling was done with an approximate distance from the source for all the three lagoons understudy to avoid biases.

Fish samples were not dried before they were blended to ascertain residual levels per wet weight. This approach could have introduced errors to the data as a powdery mixture was not obtained. However, errors were avoided as much as possible by making sure samples were well homogenised by blending.

1.8 Organisation of study

This thesis contains five chapters. Each chapter addressed the objectives of the work in diverse ways. These are outlined below:

Chapter 1 describes the background information on the compounds studied and their prevalence in the environment. This chapter also explained the use of activated charcoal for sediment remediation.

Chapter 2 presented on works that have been done in the area of PAHs including their distribution and eco-toxicological effects on the environment, as well as the theoretical framework on how activated charcoal can be a remediating material to reduce these contaminants in the environment. This approach has led to reducing bioaccumulation of these contaminants. The chapter identified the knowledge gaps that made this work even more compelling.

Chapter 3 elaborates on the various materials and methods needed to achieve each objective effectively. In this chapter, PAHs concentrations in fish, sediment, and water were quantified and assessed to determine their related ecological and human health risks.

Chapter 4 discusses the statistical results achieved during the study of the PAHs' distribution and toxicological effects in the environment.

Furthermore, the inferences that could be drawn from such results were also discussed in this chapter.

Chapter 5 summarized the research, its implications, and pointed out the areas in which further study can emerge.

1.9 Chapter summary

The underlining rationale for the study has been presented in this chapter. The justification as well as benefits to be derived by policy makers in regulating the inputs of PAHs into coastal waters is also considered. The objectives have been clearly outlined and terms have been defined to guide readers.

CHAPTER TWO

LITERATURE REVIEW

The awareness on hydrophobic organic compounds has gained rapid attention in the last decades; especially, the polycyclic aromatic hydrocarbons (PAHs). These are distinctive group of persistent organic pollutants (POPs) that find their way into the aquatic ecosystems, thereby contaminating these ecosystems. At coastal sites, PAHs are released into the aquatic ecosystems mainly through urban runoffs (Gilbert et al., 2006; Liang et al., 2007; Obanya, Omoarukhe, Amaeze, & Okoroafor, 2019; Zhang et al., 2016), industrial processes (Dong, Chen, & Chen, 2012; Gilbert et al., 2006; Sojinu et al., 2013; Sun, Littlejohn, & Gibson, 1998; Viguri, Verde, & Irabien, 2002), vehicle exhausts and spillage of fossil fuels (Abdel-Shafy & Mansour, 2016; Nguyen et al., 2014; Niu et al., 2018; Zakaria et al., 2002; Zhang et al., 2012). Concentrations of PAHs in the aquatic environment are of toxicological importance, especially to the organisms that thrive in these environments. With the rapid increase in industrialization, motorization, and urbanization in Ghana due to population growth, the potential elevation of PAHs loads is of much concern.

In this section, the nature of PAHs, their sources, their concentrations and distribution in the aquatic environments, as well as their toxicological effects on the aquatic lives and humans have been reviewed to provide more insights about the mobility of PAHs in these ecosystems. Most of the works done in the present studied areas focussed mainly on the levels of PAHs, without any clear technique in removing such contaminants from the ecosystems. With such background, the use of activated charcoal was employed to enhance the mechanistic understanding of its use on PAH-contaminated sediment through laboratory experiments. Similar works have also been reviewed in this section to elaborate on the impacts of carbonaceous materials as amendment for contaminated sediments to ensure healthy aquatic ecosystems.

2.1 Properties of PAHs

Over the past decades, copious trace contaminants have been produced and released into the environment through anthropogenic activities such as industry, agriculture and transport. PAHs, example of such trace contaminants, are of particular concern because they are very persistent, poorly degradable, bio-accumulative and are continuously added to the environment (Ekere et al., 2017; Francisco, 2016; Halsall, Sweetman, Barrie, & Jones, 2001; Sojinu et al., 2013). They are a class of organic compounds that comprise fused benzene rings. Each benzene ring is connected to at least one other benzene ring through a shared carbon-carbon double bond (Baird & Cann, 2008; Francisco, 2016; Li & Lee, 2001; Nguyen et al., 2014; Rengarajan et al., 2015). They consist of conjoined aromatic rings with or without heteroatoms (Manoli et al., 2000; Neff et al., 2005; Schwarzenbach et al., 2003). They are found anywhere and everywhere so, they are described as ubiquitous or multimedia contaminants. They can be found in diverse environmental compartments such as: atmosphere, inland and sea waters, sediments, soils, vegetation and some food products (Abdel-Shafy & Mansour, 2016; Bordajandi et al., 2004; Bruce-Vanderpuije et al., 2019; Essumang, Dodoo, & Adjei, 2012; Inomata et al., 2012; Nguyen et al., 2014). They are usually found in the environment as mixtures, rather than single compounds due to their formation (Edokpayi et al., 2016; Logan, 2007; Zelinkova & Wenzl, 2015). For this reason, they are often regarded as a mixture and discussions normally revolve around the total concentration of their mixture to interpret their distribution. In this work, however, both individual and the total PAHs of the mixture were reported.

The behaviour, transport, and fate of PAHs in the environment, principally hinge on their physical and chemical properties. PAHs as pollutants exhibit these three main characteristics (bioavailability, persistence and bioaccumulation), and all these are greatly influenced by these physicochemical properties (Abdel-Shafy & Mansour, 2016; Gehle, 2009; Skupinska, Misiewicz, & Kasprzycka-Guttman, 2004). They are generally described as solids with low volatility at room temperature; are either insoluble or have relatively low solubility in water (hydrophobic) as molecular weight increases, lipid-soluble (lipophilic), have relatively high molecular weights, soluble in many organic solvents and most can be photo-oxidized and degraded to simpler substances (Bergman, 2016; Gehle, 2009; Ohiozebau et al., 2017; Ololade et al., 2012; Ţigănuş, Coatu, Lazăr, Oros, & Spînu, 2013; USEPA, 2000; Viguri et al., 2002).

The production of PAHs usually occurs during the incomplete combustion of carbon-based compounds including, wood, coal, or oil. The makeup of a PAHs pool released from a combusted source relies on the properties of the combusting carbonaceous material and combustion temperature (Dhar et al., 2019). Stogiannidis & Laane (2015) reported that generally, when lesser heat is generated under low amount of oxygen, the rate of incomplete combustion of these organic materials intensifies, thereby producing more PAHs. Due to population density, transportation and proximity of industry, many urban and rural areas have increasing levels of PAHs in their air, water and soil (Cavalcante, Sousa, Nascimento, Silveira, & Viana, 2012;

Foster, Roberts, Gruessner, & Velinsky, 2000; Francisco, 2016; Wang et al., 2013). This is because PAHs are naturally produced from petroleum seeps, volcanoes, forest and bush fires, bacterial and algal synthesis and are as well characterised with long transport potential (Abdel-Shafy & Mansour, 2016; Rengarajan et al., 2015; Wcisłło, 1998). Therefore, they have the ability to also travel to rural areas where anthropogenic activities are minimised. They can be described as "semi-volatile" i.e, under environmental conditions they are able to travel between the atmosphere and the earth's surface in recurrent, temperature-driven cycles of deposition and volatilisation (Cavalcante et al., 2012; He et al., 2014; Wania & Mackay, 1996). This makes them liable to dispersion on a global scale. With such understanding, Brenu Lagoon which is located in one of the rural areas in the Central Region of Ghana was chosen for this current study. This was to establish the influence of long range transport of PAHs on a rural-based lagoon.

In a comprehensive review on anaerobic microbial degradation of PAHs by Dhar et al. (2019), vapour pressure was mentioned as one of the vital element that influences dispersion, transport, and fate of PAHs. Figure 2 gives a general overview of fate of PAHs in the environment. Low molecular weight (LMW) PAHs have higher vapour pressure and are mostly released into the environment in gas phase. The high molecular weight (HMW) PAHs, due to their lower vapour pressure, are released primarily in particulate form. Another physicochemical characteristic that was mentioned in the same article was the octanol-water partition coefficient (log K_{ow}). This characteristic is able to influence the compartmentalisation of PAHs in the environment. Both physicochemical properties (i.e. vapour pressure and log K_{ow}) have been agreed by other authors (Bandowe et al., 2014; Bidleman, 1988; Hawthorne et al., 2011; Meador et al., 1995; Muangchinda et al., 2017; Ololade et al., 2017; Shahbazi et al., 2010) about their tendency to affect behaviour, transport and fate of PAHs in the environment.

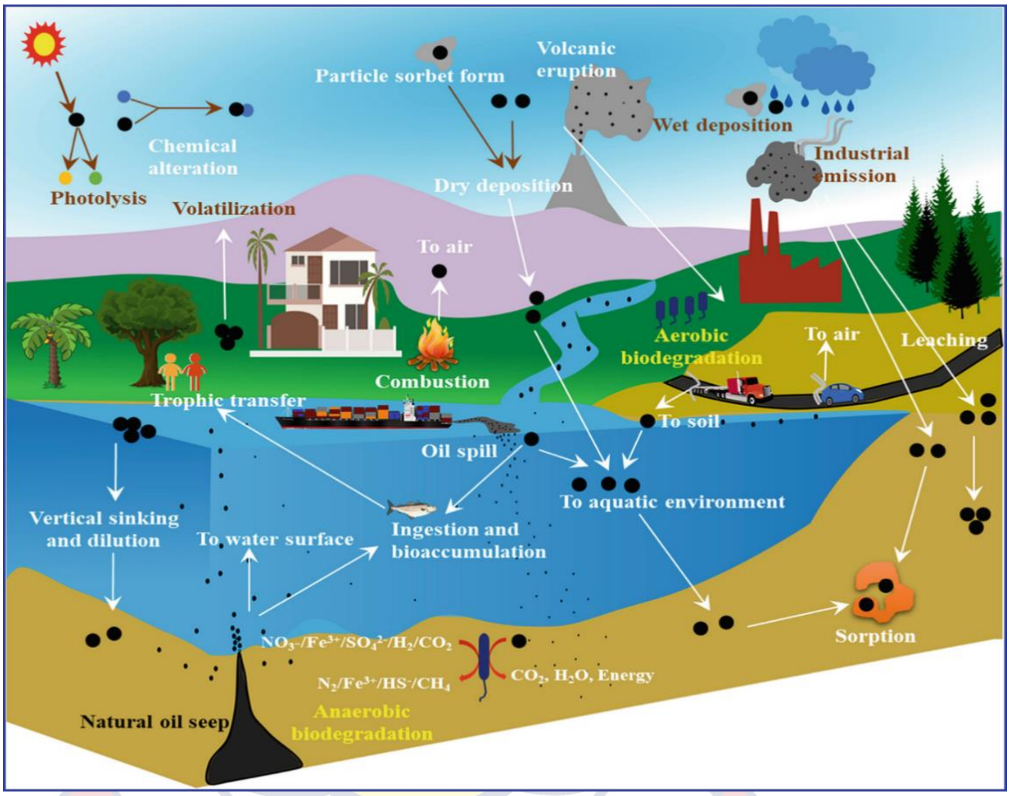

Figure 2: Fates of PAHs (in black dots) in the environment. The most common ways of origin and transfer (Dhar et al., 2019).

Some physiochemical properties of the 16 selected priority PAHs has been presented in Table 1.

Table 1 - *The Physiochemical Properties of 16 Selected Priority PAHs*

PAH	Abbrev	RN	P (Pa)	K_{ow}	bp (˚C)	S (mg/l)
Naphthalene	Naph	2	11	3.37	218^a	32
Acenaphthylene	Ace	3	9.0×10^{-1}	4.1	280	3.9
Acenaphthene	Acp	3	3.0×10^{-2}	3.9	279	3.9
Fluorene	F	3	9.0×10^{-2}	4.18	295	1.9^b
Anthracene	Ant	3	1.0×10^{-2}	4.54	342	0.05^b
Phenanthrene	Phe	3	2.0×10^{-2}	4.57	340	1.1^b
Fluoranthene	Flu	4	1.2×10^{-3}	5.22	375	0.26
Pyrene	Pyr	4	6.0×10^{-4}	5.18	393	0.13^b
*Chrysene	Chr	4	1.4×10^{-6}	5.86	448	0.002
*Benz[a] anthracene	BaA	4	2.8×10^{-5}	5.6	400	0.009-0.014
*Benzo[k] Fluoranthene	BkF	5	5.2×10^{-8}	6.0	480	0.0007-0.008
*Benzo[b] fluoranthene	BbF	5	6.7×10^{-5}	5.8	481	0.0014
*Benzo[a] Pyrene	BaP	5	7.0×10^{-7}	6.0	496	0.003^b
*Dibenz[a,h]anthracene	DahA	5	3.7×10^{-8}	6.5	524	0.0005
*Indeno [123-cd]pyrene	IP	6	1.3×10^{-8}	6.6	536	0.00019^d
Benzo[g,h,i] perylene	BP	6	$1.4\times10^{-8,c}$	7.1	550^c	0.00026

MW = molecular weight, RN = ring number, S = aqueous solubility (25 °C), P = vapor pressure (25 °C), K_{ow} = the logarithm of the octanol-water partition coefficient, bp = boiling point. Unless otherwise specified, all solubility and K_{ow} data are from (Irwin, Van Mouwerik, Stevens, Seese, & Basham, 1997), and all vapour pressure and bp data are from (European Commission, 2001). *Priority PAH pollutant considered carcinogenic. All 16 pollutants are of US EPA and EU priority pollutants.

[a] (Lide, 1992)

[b] Haftka (2009)

[c] Irwin et al. (1997)

[d] Mackay, Shiu, Ma, & Lee (2006)

2.2 Sources of polycyclic aromatic hydrocarbons

Pyrogenic, petrogenic, and biological (biogenesis or diagenesis) are the main sources of PAHs to the environment (Boehm et al., 2007; Mitra et al., 1999). Pyrolysis is the process in which pyrogenic PAHs are produced. The production of pyrogenic PAHs happens when organic substances are yielded to high temperatures (≈ 350 °C to ≥ 1200 °C) under low or no oxygen. There are intentional and unintentional release of pyrogenic PAHs. For instance, during the destructive distillation of coal to coal tar and coke, or during the thermal cracking of petroleum residuals into lower forms of hydrocarbons. These processes produce intentional pyrogenic PAHs. However, unintentional pyrogenic PAHs are produced during incomplete burning of wood, incomplete burning of vehicular fuels, incomplete burning of fuel oils in heating systems, etc. (Abdel-Shafy & Mansour, 2016; Chen, Hsieh, & Chiu, 2003; Gnandi et al., 2011; Zhu et al., 2004).

PAHs are also produced at low temperatures. For example, crude oils contain PAHs that have been formed over decades of centuries at low temperatures of 100–150 °C. The PAHs that are formed as crude oils evolve, and other related processes are known as the petrogenic PAHs. Examples of activities that release petrogenic PAHs to the environment are oil spills, underground and above ground storage tank leaks, small discharges of gasoline, motor oil, and other similar substances linked to transportation (Abdel-Shafy & Mansour, 2016; Stogiannidis & Laane, 2013). Petrogenic PAHs are mainly from petroleum-related substances.

PAHs can also be formed through natural or biological means. They can be synthesized by some flora and fauna or being produced during the degradation of vegetative matter. Other natural sources include forest and brush fires, volcanoes, petroleum seeps, sedimentary rocks having petroleum hydrocarbons, etc. (Baek et al., 1991; Gnandi et al., 2011; Kielhorn & Boehncke, 1998; Manoli et al., 2000; Tolosa, Bayona, & Albaigés, 1996). The incomplete combustion of natural or humanly derived, is known to be the distinct largest contributor of PAHs into the environment.

Researchers have also established some chemical fingerprinting techniques, serving as tools to aid distinguish between groups of PAH (such as, coal-based, wood-based, or oil-based sources) by identifying specific chemical indicators. Chemical fingerprinting technique also aids in identifying non-point sources of PAHs to the environment (Wang et al., 2011). In source apportionment, indices such as PAH diagnostic ratios (also known as molecular isomeric ratios), multivariate analysis (e.g. principal component analysis, cluster analysis) are some of the chemical fingerprinting techniques being used (Liu, Lang, Xue, Liao, & Zhou, 2009). According to Yunker et al. (2002), Santos et al., (2017) and Ashayeri et al. (2018), diagnostic ratios are possible due to the differences in their thermodynamic properties, distinguishing the most stable isomer from the least stable, making it a judgemental tool to differentiate their sources. One challenging situation in identifying origin of PAHs is the possible coexistence of a variety of contamination sources. Not to mention the transformation processes that PAHs may undergo before final deposition in the analysed media. Nevertheless, compounds may display comparable pathways that could be applied to identify their source. Sediment has an added advantage as a valuable long-term record of both natural and anthropogenic origin of PAH contamination. These ratios have been extensively used by many researchers (e.g. Ashayeri et al., 2018; Cardellicchio et al., 2007; Culotta et al., 2006; Santos et al., 2017; Souza et al., 2018; Tay & Biney, 2013; Yunker et al., 2002), as a substantial tool in identifying possible sources of PAHs in an aquatic environment.

In this current study, to identify possible PAHs sources in the samples (i.e., fish, sediments and water), ratios between PAHs isomers were employed.

Bivariate plots of the diagnostic ratios used in this study involved the combination of the following: Ant/(Ant + Phe) and Flu/(Flu + Pyr), Flu/Pyr and Phe/Ant (Ashayeri et al., 2018; Santos et al., 2017; Sun, Zhang, Ma, Chen, & Ju, 2017). Other ratios such as ∑LMW/∑HMW, BaA/(BaA + Chr), InP/(InP + BghiP), BaP/BP and ∑PAHs(4 rings)/∑PAHs(5-6 rings) (Ashayeri et al., 2018; Edokpayi et al., 2016; Souza et al., 2018; Tay & Biney, 2013; Yunker et al., 2002) were also employed in this current study.

2.3 Fate and transport of PAHs in aquatic environments

As ubiquitous contaminants, PAHs can be found in the marine environment, especially in stressed areas like harbours, lagoons, estuaries and other shallow coastal zones under anthropogenic inputs (Baumard, Budzinski, Garrigues, Dizer, & Hansen, 1999; Baumard et al., 1998). Lagoons are recognised as one of the most important ecosystems globally (Gray, 1997; Harris & Vinobaba, 2013; Odjer-Bio, Belford, & Ansong, 2015). They provide lots of ecosystem services and functions, such as habitat and nursery sites for fish species, reference stations for a variety of migratory birds, and food and water sources for some human communities (Gray, 1997; Harris & Vinobaba, 2013). Their continual existence is therefore paramount to ensure continual survival of many organisms, including humans. However, because coastal lagoons lack appropriate watershed management strategies, most of them are prone to pollution. Among the lagoons that exist along the coastline of Ghana, Chemu, Fosu and Korle lagoons have been added to the list of water bodies with ‘dead zones’ (Adjei, Adokoh, Bentum, Hadzi, & Ameho, 2017; Akoto et al., 2014; Armah, Luginaah, et al., 2012). This is worrying as some individuals depend on these lagoons for their livelihood.

Various anthropogenic actions in coastal lands and other catchment areas of coastal waters have contributed to PAH contamination in these aquatic environments (Ashayeri et al., 2018; Gilbert et al., 2006; Jaward, Alegria, Galindo Reyes, & Hoare, 2012; Nyarko, Botwe, & Klubi, 2011). PAHs are released into the aquatic environment, via wastewater effluents (Edokpayi et al., 2016; Haddaoui, Mahjoub, Mahjoub, Boujelben, & Di Bella, 2016), from coke and petroleum refinery industries (Barakat, Mostafa, Wade, Sweet, & El Sayed, 2011), accidental oil spills and leakages (Specchiulli et al., 2011), runoffs (Zakaria et al., 2002; Zhang et al., 2012), vehicular emissions (Armah, Luginaah, et al., 2012; Gilbert et al., 2006), incineration of domestic waste within the vicinity of coastal waters (Gilbert et al., 2006; Nyarko et al., 2011), among others. Residues of these pollutants enter coastal ecosystems via continental runoff, atmospheric deposition, municipal and industrial effluents, and often by direct discharge. Most of these activities can also be identified in the present study areas, thus, the need for monitoring the current levels of PAHs in these coastal lagoons.

When PAHs enter coastal lagoons either through runoffs, direct discharge, wet or dry deposition, they become associated with particulate matter, sediment, or biota. In the water column, PAHs sorb to particulate matter due to their low solubility in aqueous medium and their high ocatanol-water partition coefficient (Balk et al., 2011; Van der Oost, Beyer, & Vermeulen, 2003). The levels of PAH in the sediments are mostly in several orders of magnitude higher than aqueous phase (water column) levels, due to their hydrophobicity. The increased levels of PAHs in the sediment can also be related to low oxygen, making degradation of PAHs very slow. The sediments and aquatic organisms are known to be the main recipient or eventual sinks of PAHs in the aquatic environment (Armah, Luginaah, et al., 2012; Bruno, 2019; Gilbert et al., 2006; Layshock, Wilson, & Anderson, 2010; Odjer-Bio et al., 2015). However, the residues of PAHs in molluscs are generally higher than in the fishes, because fishes have greater ability to metabolise PAHs (Hellou & Warren, 1997; Inomata et al., 2012; Tudoran & Putz, 2012; Zelinkova & Wenzl, 2015).

With the tendency of PAHs partitioning into different compartments of an aquatic system, it is essential to survey and monitor the PAHs levels in these compartments to effectively understand the mobility of PAHs in an aquatic ecosystem. From such surveys, fishes, and their relationships with sediments and the water column can serve as a tool for risk assessment of PAHs in the aquatic environments. It is with these backgrounds that the current study was conducted. Below are some similar works that assess the concentration and distribution of PAHs in different compartments of the aquatic environment.

2.3.1 PAHs in the water column (aqueous medium)

Disposing of solid waste in open dumps and burning of such solid waste, is the most common way of handling solid waste disposal in many developing countries including Ghana. Some of these activities are found even within the vicinity of coastal lagoons. In the process of burning these solid wastes, certain harmful and corrosive chemicals such as PAHs, PCBs, hydrogen chloride, etc. are emitted into these water bodies (Essumang, Adokoh, Afriyie, & Mensah, 2009). With such background, Essumang et al. (2009) conducted a study on the levels of PAHs in leachates from a solid waste disposal site and an effluent from an oil refinery in some water bodies around Accra, Ghana. Sixteen PAHs were analysed and the results demonstrated that there were elevated levels of PAHs in the water sample of the Densu River, Chemu, Korle and Kpeshi lagoons. Concentration of PAHs ranged from below detection level to 14.587 µg/L and acenaphthene, anthracene, Benzo[a]anthracene, Benzo[b]fluoranthene, Chrysene and Phenanthrene recorded at the various sites were above the recommended level set by WHO (0.05µg/L; Kielhorn & Boehncke, 1998). It was also noted from their results that the seven carcinogenic PAHs were detected in different concentrations at the various sites. From their correlation analyses, a significant coefficient of 0.708 at 0.01 level depicted how water samples from Oblogo solid waste site and Weija Dam derive their source mainly from burning of biomass/coal. This demonstrated how open dumping and burning procedure used in Ghana can pollute the environment. It was concluded from their study that inhabitant who swim and bath in the downstream of River Densu are likely exposed to these PAHs and may be at risk of their harmful effects. Combustion of biomass/coal was detected from the diagnostic ratios as the dominant source of PAHs in the study sites (Essumang et al., 2009).

In order to understand the dynamics of PAHs in the water column of Pearl River estuary (China), the seasonal variations of PAHs' concentrations and compositions were investigated (Niu et al., 2018). They observed that the PAH total concentrations and profiles exhibited significant differences when comparing the dry season (summer) with the rainy season (winter). An observation also noted in the work of Santos et al. (2018). In both works, they linked the seasonal patterns to environmental factors such as temperature. Unlike the work of Santos et al. (2018), Niu et al. (2018) separated the water samples into two groups: dissolved and suspended particulate matter sample.

The total PAHs of the 16 priority PAHs varied from 0.012 to 0.16 µg/L in dissolved water and from 2.82 to 112.32 µg/g in the suspended particulate matter. The LMW PAHs dominated in the water samples. However, the HMW PAHs were higher in the suspended particulate matter than in the dissolved water samples. Also, vehicular emissions and combustion of biomass/coal were the potential sources of PAHs into the estuary when diagnostic ratios were employed (Niu et al., 2018).

Marine culture is flourishing in China and signifies a major contributor to the regional economy in coastal zones. Hitherto, studies on the environmental quality of these areas are limited (Zhou & Maskaoui, 2003). So, Zhou & Maskaoui investigated the quality status of Daya Bay, a key aquaculture area in China. The levels of the 16 priority PAHs were analysed in water samples of the bay and the total concentration ranged from 4.228 to 29.325 µg/L. In comparison to other water bodies, the PAH levels in Daya Bay waters were relatively high. A concentration sufficiently high (> 10 µg/L) to cause acute toxicity to some organisms (Kalf, Crommentuijn, & van de Plassche, 1997; Long & MacDonald, 1998). The PAH distribution profile in the bay indicated potential source dependence, as the levels were generally higher in the vicinity of intense anthropogenic activities. From their study, Zhou & Maskaoui (2003) saw the urgent need to establish a robust monitoring programme for PAHs, and not only in water and sediment, but also in organisms as well. According to them, such development will ensure a comprehensive understanding of PAHs concentrations that may be above the recommended environmental quality standards for water bodies. This will aid in taking the proper policy measures to curb PAH contamination in water bodies.

Some of the classifications used in determining PAH contamination in water samples include that used by Ashayeri et al. (2018) and Li et al. (2015) in similar studies. In this classification, PAHs contamination is grouped into 4 categories: low polluted, 0.01–0.05 µg/L; slightly polluted, 0.050–0.25 µg/L; moderately polluted, 0.25–1 µg/L; and highly polluted, above 1 µg/L. This classification was also used to describe the degree of PAH contamination in this current study.

Due to the hydrophobicity nature of PAHs, they are not expected to be readily available in the water column. Once PAHs are incorporated into sediments they are somewhat immobile because their non-polar structures inhibit them from dissolving in water. Nevertheless, PAHs are not entirely insoluble, particularly the LMW PAHs. Hence, small amounts of PAHs do dissolve and become part of the pore water where they are bioavailable to organisms especially living in the water column like pelagic fishes. (Abdel-Shafy & Mansour, 2016; Dong et al., 2012). The dominance of LMW PAHs in the water column is agreed by several studies (Ashayeri et al., 2018; Essumang, 2010; Hu et al., 2010; Jaward et al., 2012; Omayma, Sawsan, & Nady, 2016; Santos et al., 2017).

The HMW congeners on the other hand, are relatively low in water column. This is expected due to their relative high affinity to sorb to particulate matter due and their high partition coefficients (Ashayeri et al., 2018; Bidleman, 1988; Jaward et al., 2012). The presence of pore water organic colloids become an added advantage to the increase levels of PAHs especially, the HMW compounds, beyond their aqueous solubility because PAHs will be sorbet onto these organic colloids. Santos et al. (2017) observed appreciable concentrations of HMW congeners in the water they sampled from upper Iguassu watershed at Parana, Brazil. They found out that surface water analysis (dissolved pollutants) showed predominance of compounds containing more of the LMW compounds while in suspended particulate matter there was a predominance of compounds containing 4, 5 and 6 rings (HMW compounds). They also realised that dominance of LMW PAHs may demonstrate existence of a local origin or a recent generated of LMW PAHs, including atmospheric deposition, sewage discharge, oil spills and fuel from boats. The reason was that LMW PAHs are less stable, and so they degrade and evaporate more rapidly than HMW PAHs (Ashayeri et al., 2018; Hu et al., 2010; Omayma et al., 2016).

2.3.2 PAHs in sediment

Sediments are deposited material consisting of organic matter in various stages of decomposition, particulate mineral matter and inorganic material of biogenic origin (Viguri et al., 2002). The dynamic of PAH in the ecosystem can be determined by the compound’s physicochemical parameters (e.g. vapour pressure, log Kow and solubility) (Bergman, 2016; Gehle, 2009; Ahmed et al., 2017; Ohiozebau et al., 2017; Ololade et al., 2012; Ţigănuş et al., 2013; USEPA, 2000; Viguri et al., 2002). Nevertheless, some environmental characteristics should be considered; such as pH, grain size (%sand and %mud (silt + clay), temperature, organic matter content and salinity. They play vital role in organic compounds adsorption and distribution in the ecosystem (Nguyen et al., 2014; Sojinu et al., 2013; Viguri et al., 2002). In effect, sediment retention capacity to PAHs may be related to these environmental properties as well. With such understanding, some of these environmental characteristics were considered in this current study. The effect of TOC, pH, temperature and salinity on partition of total PAHs in sediments were therefore looked at.

Some studies have demonstrated that the organic matter is a very important variable related to PAHs pollution of sediments; and to correct for factors controlling the partition of hydrocarbons in sediments, hydrocarbon concentrations have been normalised for measured total organic carbon (TOC) in the sediments (Baumard et al., 1999; Knezovich, Harrison, & Wilhelm, 1987; Montuori et al., 2016; Ololade et al., 2017; Viguri et al., 2002; Witt & Trost, 1999; Xue et al., 2016). Therefore, TOC was used to evaluate the organic matter contents of the sediments sampled in this present study. Some studies on the other hand have reported on the possibility that there exist no significant correlation between partitioning of PAHs to sediment and TOC. A regression analysis was carried out to investigate the correlation between the total PAHs concentrations and TOC (Sojinu et al., 2013). It was apparent that there was no significant correlation between the two parameters ($R^2 = 0.21$). Their result was consistent with reports on sediments from the Bahia Blanca estuary, Argentina (Arias et al., 2010) and Shadegan wetland, Iran (Ashayeri et al., 2018). They concluded that lack of positive linear relationship between the PAHs concentrations and TOC values in the sediments indicated that the PAHs were recently generated and, therefore, were yet to fully partition into organic matter in the sediments. Ashayeri et al. (2018) further added that a weaker correlation could also mean concentration of PAHs in sediments are related to the sources of contamination more than the organic matter content.

Studies on PAHs concentration in the aquatic sediment have received much recognition by environmental chemists, toxicologists, and regulatory agencies because of their toxic effect to both aquatic organisms and humans. The sediments are also the most receptive media to PAHs in an aquatic environment. Generally, organic compounds are known to readily adsorb to sediment particles. Hence, sediments can be recognised as a pollution reservoir and origin from which toxic substances may once again be released into the aquatic environment (Francisco, 2016; Specchiulli et al., 2011). Furthermore, sediments are economically effective media in environmental assessment of aquatic systems, and so can be a representative and a useful media for monitoring inputs of PAHs in coastal areas (Ashayeri et al., 2018; Ololade et al., 2017; Specchiulli et al., 2011). For these reasons, most premier studies of lagoons consider the PAH levels in the sediment.

Armah, Luginaah, et al., (2012) and Gilbert et al. (2006) conducted studies on PAH contamination in Fosu Lagoon; a lagoon this present study also looked at. The work that was reported by Gilbert et al. (2006) was the first results ever obtained on PAHs concentrations in the sediment of Fosu Lagoon. The main basis upon which this study was conducted was due to increase in the careless disposal of waste directly into this lagoon. Fifteen PAHs were analysed from the sediment samples. Total PAHs varied from 254,000 to 558,000 μg/kg. Two distinct locations were identified to be a major source of anthropogenic load of PAH; the mechanical shop in the north-eastern sector and the residential area in the northern sector of the lagoon. They established a significant spatial variation, while temporal variations at individual sites were lacking. With the temporal variation, they ascribed the outcome to the fact that sampling was done within the same season (dry season). For spatial variation, they ascribed the outcome to the distinct anthropogenic activities that occurred at different sampling points.

There are other studies that also realised a significant temporal variations while statistically, there was no significant variation at spatial level (Jaward et al., 2012). Gilbert et al. (2006) further demonstrated that these chemicals were released into the lagoon mainly by combustion of wood or coal (pyrogenic sources). They recommended that further studies be done to verify the existence of naturally occurring PAHs in the lagoon. They realised that perylene (5-ring compound) exhibited inverse correlation with the other PAH compounds. Indicating that some fraction of the other compounds may form from the biodegradation of perylene by natural occurring population of sediment microorganisms. Therefore, the need to embark on other studies that would focus on evaluating the degradation capability of the indigenous micro-flora and fauna in this PAH-contaminated sediment. They also added that there is the need for development of clean up or remediation technologies for PAHs.

Armah, Luginaah, et al. (2012) also assessed the levels, distribution and human health risk of PAHs in sediments from the Fosu Lagoon. It was coupled with environmental fate modelling that examined biodegradation behaviour of PAHs in sediment. The observed PAH contamination demonstrated potential anthropogenic impact, mainly from burning. This burning was linked to vehicular emissions and tyre burning which is widespread at the mechanical workshops close by. The total PAHs recorded ranged from 3,000 to 14,000 µg/kg. Unlike the report from Gilbert et al. (2006) who detected 4 carcinogenic compounds in this lagoon, Armah, Luginaah, et al. (2012) measured only one carcinogenic PAH, and that was Benzo[b]fluoranthene. The levels of PAHs in sediments were quite high, and from the results obtained in the PAH-based risk calculations, the non-cancer and cancer risk to children were greater than that of the adult residents. This was attributed to the fact that children are considered to be more vulnerable to chemical-induced risk than their adult counterparts. The fate modelling on the other hand depicted that the PAHs analysed tend to sorb onto sediment in the environment and are not easily biodegradable. They therefore suggested the need to implement long term monitoring of these compounds in order to sustain this fragile ecosystem.

PAHs are also known to be present at especially, stressed areas like harbours, lagoons and other shallow coastal zones exposed to intense human activities (Baumard et al., 1999; Bouloubassi, Fillaux, & Saliot, 2001; Specchiulli et al., 2011). Specchiulli et al. (2011) set out to identify and quantify the 16 priority PAHs (cited by USEPA) in order to develop a framework of environmental quality control for four Italian lagoons. To achieve this, both the source and distribution of PAHs were analysed in sediments sampled from Lesina, Varano, Orbetello and Santa Giusta lagoons. Their results portrayed that concentrations of total PAHs in sediments differed significantly among lagoons and sampling stations, and were as well considerably lower than those reported for other urbanised and/or industrial Mediterranean areas. The lowest total concentration of the 16 PAHs in the sediments was recorded in Varano lagoon (25 µg/kg d.w.), while the highest was recorded in Orbetello lagoon (99 µg/kg d.w.). This output was ascribed to intensive industrialisation and urbanisation within the surrounding of Orbetello lagoon. In relation to the sources of PAHs in these lagoons, diagnostic ratios were used. PAHs were of both pyrogenic and petrogenic origins. Industrial, urban and aquaculture waste, agricultural run-off, oil spillage and traffic of boats were the main contributing factors to PAH contamination in these lagoons. The 4-rings PAHs showed predominance, indicating that HMW compounds readily adsorb to sediment particles. Several studies have demonstrated that HMW PAHs have a high tendency to accumulate in sediments (Ashayeri et al., 2018; Edokpayi et al., 2016; Jaward et al., 2012; Zhang et al., 2016). Their predominance in the sediments could also be due to their low solubility, high hydrophobicity, stable molecular structures, less predisposed to degradation and higher partitioning coefficients in aqueous environments in comparison to the LMW PAHs (Ashayeri et al., 2018; Zakaria & Mahat, 2006; Zhang et al., 2016). Hence, HMW PAHs prefer to adsorb unto sediment or biota in aquatic ecosystems. For this reason, higher percentage of HMW to LMW is expected in sediment samples.

Another study reviewed in this present study was conducted in Ologe lagoon, located in Lagos state, Nigeria. This lagoon is a source of water for domestic, transportation, and fishing activities. Unfortunately, it also serves as a drainage channel for some rivers that are recipients of untreated or partially treated effluents from neighbouring industries and communities (Obanya et al., 2019; Sojinu et al., 2013). These effluents may elevate the concentrations of anthropogenic contaminants (including PAHs) in the lagoon, thereby creating stressors for aquatic biota. With such a background, Obanya et al. (2019) conducted a research to evaluate the occurrence of PAHs in Ologe lagoon. From their results, 11 of the 16 priority PAHs were detected in the sediment, with HMW compounds dominating over the LMW PAHs. The level of ∑(LMW PAHs) was 8.91 μg/kg, while that of ∑(HMW PAHs) was 377.98 μg/kg. Such a distribution of PAHs in Ologe lagoon indicated a pyrogenic source of PAH pollution. They ascribed the pyrogenic PAHs to incomplete combustion of fossil fuel in the generators used by surrounding factories including, food and beverage industries, pharmaceutical plants, breweries, metal finishing, chemical plants, and pulp and paper companies. Benzo[b]fluoranthene was the most predominant in the lagoon and they concluded that such level of Benzo[b]fluoranthene may be chronically toxic to aquatic organisms, in terms of oxidative stress and hepatotoxicity.

Another study by Ashayeri et al. (2018) saw the need to evaluate impact from local sources that may have contributed to significant risk of contamination by PAHs. In view of this, they investigated pollution characteristics and ecological risks of PAHs by sampling contaminated sediments from the Shadegan wetland. Shadegan wetland located at Iran, is known by its distinctive biodiversity and joins the Jarahi River and Persian Gulf waters (Nasirian et al., 2013). For the past few years, this wetland has received diverse contamination from sources (such as: petrochemical complexes, oil pressure boosting station, oil pipelines and petroleum refineries) due to its closeness to an oil field. Such contamination has devastated the wetland's natural resources and pose threat to human health (Chaharlang, Bakhtiari, Mohammadi, & Farshchi, 2016). From their investigations, they found concentrations of total PAHs in sediment samples varying from 10 to 317 μg/kg. Source apportionment of PAHs by molecular isomeric ratios suggested that impact of potential anthropogenic PAH sources included oil spill, bulrush combustion, vehicular exhaust and fishing boat emissions. From the calculated risk quotient (RQ), it portrayed relatively insignificant toxic impact on aquatic ecosystem so far as most individual compounds were concern. Also, the toxic equivalent quotient (TEQ) value of PAHs (1.9 µg/kg) was lower than the safe level (600 µg/kg; Yu, Liu, Xu, & Shen, 2015). They also detected carcinogenic PAHs at all stations hence, recommended that such compounds should be preferentially targeted in next surveys.

Globally, PAHs concentrations in sediments vary between 1 to 760,000 µg/kg, with modal concentrations of 1,000 to 10,000 µg/kg reported for rivers, estuaries, harbours, and coastal areas (Zakaria et al., 2002). The ∑PAHs contamination in sediment has been categorized into 4 classes according to He, Meng, Xu, Zhang, & Guo (2016): low; 0–100 µg/kg, moderate; 100–1,000 µg/kg, high; >1,000 µg/kg. This classification has been used in several studies including: Sundarban mangrove wetland in India (132 to 2,938 µg/kg; Domínguez et al. (2010)), sediments from Lake Maryut, Alexandria, Egypt (106 to 57,800 µg/kg; Zhang, Liu, Chen, & Hou (2014)), Stagnone coastal lagoon of Marsala (72 to 18381 µg/kg; Culotta et al. (2006)) and in the sediment of Shadegan wetland, Iran (10 to 317 µg/kg; Ashayeri et al. (2018)). Such classification was also used in this present study.

2.3.3 PAHs in fishes

Fish can serve as natural bio indicators of PAH contamination in the aquatic ecosystem (Copat et al., 2012; Kammann et al., 2017; Logan, 2007; Pointet & Milliet, 2000). Due to their commercial use and recreational importance, fish are usually the most visible organisms of the aquatic communities. Hence, defects on the skin, barbels, fins, and eyes due to an exposure to PAH-contaminated sediments and water can be observed with the naked eye. It is as a result of these that instigated in some of the earliest investigations of the effects of PAHs on fish (e.g., Baumann, Mac, Smith, &

Harshbarger, 1991; Murchelano & Wolke, 1985; Neff, 1895), and continues to attract attention.

Exposure of fish to PAHs can occur mainly through the organic gill (respiration), gut (ingestion), or integument (dermal contact). For this purpose, fillet, including the skin, visceral and head of fish were analysed in this present study in order to capture all these possible routes of PAHs to *Sarotherodon melanotheron*. Once exposed to these membranes, the PAHs can be transferred to other parts of the fish via the bloodstream (Gobas et al., 1999; Logan, 2007; Pointet & Milliet, 2000). The degree of exposure and accumulation of PAH by fish depends on the bioavailability as well as the solubility of the compounds. The more water-soluble PAHs (for example, LMW compounds), the likelihood of uptake by the fish via the gills. The HMW compounds on the other hand are mostly found in the fish (gut) (Bandowe et al., 2014; Baumard et al., 1999; Logan, 2007; Meador et al., 1995). In as much as accumulation of PAHs depends on the concentrations of the pollutants and their solubility characteristics, it also takes into account the feeding mode or the physiology of the fish (Baumard et al., 1999; Gobas et al., 1999; Logan, 2007). Throughout the life cycle (from laying of eggs through to larvae stage and adulthood), fish maybe exposed to PAHs in sediments and in the water column. For instance, some species dig nests and others have demersal eggs, which expose the eggs to direct contact with sediments. Some larvae may inhabit near to the bottom, making them exposed to sediment contamination. Other eggs and larvae can also be found close to the surface micro layer where elevated PAH concentrations can occur. Some adult fish are benthic, such as flatfish and catfish, and come into direct contact with sediments, and many eat benthic invertebrates that accumulate PAHs. Some are bentho-pelagic (like the fish sampled for this current study; *Sarotherodon melanotheron*), and so are exposed to both the sediment and the water column (Al-khion et al., 2016; Lazartigues, Thomas, Brun-Bellut, & Feidt, 2010; Logan, 2007; Shi et al., 2016). All fish have gills which comprised of thin membranes, distributing oxygen from the water to other tissues. Fish actively transport large quantities of water, and thus the gill tissues can be exposed to PAHs present in the water especially, the contaminated suspended particulate matter (Baumard et al., 1999; Gobas et al., 1999; Logan, 2007).

In addition, bioaccumulation or bio-concentration of PAHs in fish is influenced by the rate of uptake and elimination of these pollutants (Aas, Baussant, Balk, Liewenborg, & Andersen, 2000; Abdel-Shafy & Mansour, 2016). Upon entering an organism, PAHs tend to partition to lipophilic tissues, such as the liver and bile of fish (Bandowe et al., 2014; Meador et al., 1995). Elimination on the other hand, is influenced by both passive diffusion and active bio- transformation in the liver followed by excretion with the bile. A lot of fish species and higher organisms in the food chain are known to metabolise PAHs fairly rapidly. They possess enzymatic systems that metabolize and detoxify PAHs (Inomata et al., 2012; Tudoran & Putz, 2012), thereby eliminating different PAHs at different times (Logan, 2007). The enzyme cytochrome P450 mixed function oxidase system (commonly called CYP1A) is involved in metabolizing PAHs and other organic contaminants, as well as measures of its activity are used as biomarkers. Basically, it catalyses oxidation of PAHs for example, into a more soluble metabolites that can be further conjugated by Phase II enzymes and excreted (Abdel-Shafy & Mansour, 2016; Arinç, Sen, &

Bozcaarmutlu, 2000). However, in a well polluted system, metabolism of PAHs become ineffective as organisms are continually exposed to the contaminants, thereby leading to several adverse effects.

Since it remains hard to accurately predict bioaccumulation in fish, even with highly sophisticated models, analyses of tissue levels are required (Van der Oost et al., 2003) and so in this study, fillet, head and visceral parts were analysed for PAH contamination in black-chinned tilapia. It should be noted that this study is the first to analyse PAHs levels in fish tissues from these lagoons. The concentrations of PAHs detected in fish and shellfish are expected to be much higher than in the environment from which they were taken (Abdel-Shafy & Mansour, 2016).

A work done by Bandowe et al. (2014) came out with the concentrations of 28 PAHs in muscle and gut + gill tissues of 3 demersal fishes (*Drapane africana*, *Cynoglossus senegalensis* and *Pomadasys peroteti*). These were sampled from three locations along the coast of the Gulf of Guinea, Ghana. The mean concentration of the total PAHs in gut + gill tissues was 340 µg/kg, and this was significantly higher than that in muscles (192 µg/kg). From their results, although all fish tissues had benzo[a]pyrene concentrations lower than the EU limit for food safety (2 µg/kg;- EFSA, 2008; Shi et al., 2016), the excess cancer risk from consumption of some fish was higher than the guideline value of 1×10^{-6}. Also, the log-transformed levels of PAHs in the tissues significantly ($p < 0.05$) correlated with their octanol-water partitioning coefficients, indicating that equilibrium partitioning from water or sediment into the fish tissues was the main mechanism of bioaccumulation. They conclude that the health risk arising from the consumption of the studied fish (due to their PAHs content) was generally low.

PAHs were also analysed in five fish species; *Euthynnus alleferatus, Scomberomorus commerson, Sphyraena Sphyraena, Diplodus vulgaris* and *Alepes djedaba,* collected from the western coast of Alexandria, Egypt (Said, 2007). Total PAHs varied from 3,862 to 35,746 µg/kg wet weight, with a mean concentration of 12,112 µg/kg. Benzo[a]pyrene was identified as the dominant PAH congener with a mean concentration of 9,465 µg/kg wet weight in all fish species. They also indicated that the concentrations of PAHs in the different fish species may be attributed to fish habits as well as their ecological niches. The species that generally had the highest concentrations of PAHs was the reef-dwelling carnivorous fish, *Alepes djedaba*. This suggested that near-shore zones of the Alexandria coast were more polluted than offshore zones, undoubtedly portraying land-based sources of pollution.

Another work was conducted to evaluate the risk of PAHs to fish inhabiting in chronically polluted environment; the Bay of Algeciras, Spain (Rojo-Nieto, Oliva, Sales, & Perales, 2014). The concentrations of the total PAHs identified in different target organs (muscle, liver and gills), varied from below detection level to 358.44 µg/kg. Basically, the dominant compounds were Phenanthrene and Pyrene. To evaluate the potential toxicity for fish of PAHs associated with sediments, potency equivalent concentrations (PEC) and screening values (SVs), the biota-to-sediment accumulation factors (BSAFs) and the Toxic equivalent quotients (TEQ) of sediment for fish were calculated. Their results suggested that concentrations in target organs and the TEQs of sediment calculated for fish in relation to the SVs for fish consumption were of concern. They concluded that feral finfish can be useful in PAH risk assessment for chronically polluted environments. Such an assessment could be a productive tool for complementing environmental diagnoses and improving their accuracy.

Zhao, Zhang, Cai, & Chen (2014) also conducted a research on the residual levels of PAHs in different tissues of fish from the largest lake in China, the Poyang Lake. Distribution of PAHs in bighead carp (*Aristichthys nobilis*) and silver carp (*Hypophthalmichthys molitrix*) were analysed. Total PAH concentrations varied from 105 to 513 µg/kg for the several tissues of bighead carp and from 53.9 to 401 µg/kg in that of the silver carp. The main compounds that dominated in the samples were the LMW, indicating the gill-water transfer could be the main exposure pathway for PAHs in the studied fish species. For tissue distribution, the hepatobiliary system accumulated elevated concentrations of PAHs when compared with the extrahepatic tissues, with bile as the dominating tissue for both species. The origin of these PAHs were ascribed to both petrogenic and pyrogenic inputs, with gasoline combustion being the main source. From their results, they concluded that exposure to humans through the consumption of these fishes could induce potential carcinogenic effects. They suggested that further research should be conducted, especially in the area of effects of PAH metabolites.

Several studies have indicated a connection between PAHs exposure to fish and incidences of immune toxicities and cancers (Falcó et al., 2003; Harris et al., 2009; McCallister et al., 2008; Pratt et al., 2011; Šimko, 2002). Many countries as well as the World Health Organisation (WHO) have therefore set guidelines in terms of the maximum concentration limits of PAHs in fish above which the health of humans is at risk (Balk et al., 2011; Logan, 2007; Wenzl et al., 2006). Health risks resulting from dietary intake of PAHs are usually assessed using Toxic Equivalent Quotient (TEQ), Toxicity Equivalency Factor (TEF) relative to Benzo(a)pyrene (BaP) (Ashayeri et al., 2018; Chien et al., 2002; Storelli, 2008; USEPA, 2000; Yi, Yang, & Zhang, 2011; Yoon, Park, Lee, Yang, & Lee, 2007a).

2.4 Ecological effects of PAHs in the aquatic environment

From the 1960s, researchers started to report on PAH contamination in the marine environments. Such reports triggered other studies to evaluate PAHs concentrations that may adversely affect aquatic biota around the world (Arias et al., 2009). As PAHs get into the aquatic system, they partition into fine-grained sediments and suspended particles, after that, they remobilized in the aquatic system, then become bioavailable to organisms (Wetzel & Van Vleet, 2004). They bioaccumulate in biota with high rates of metabolising PAHs (e.g., fish; Hellou & Payne, 1987; Van der Oost et al., 2003; Zelinkova & Wenzl, 2015) or those incapable of efficiently metabolising especially, the parent compounds of PAHs (e.g. some invertebrates, mussels and other bivalves; Arias et al., 2009; Liu et al., 2009). The concentration of PAHs in fish may not significantly exhibit effect of exposure in fish tissue, nevertheless, such concentration could be used as baseline data that can be interpreted using different toxicity criteria. For example, the fish gills is damaged when exposed to anthracene (≈20 μg/kg) (McCloskey & Oris, 1993). Regarding toxicity, compounds with two and three aromatic rings exhibit a significant acute toxicity (Netto et al., 2000) and compounds with a larger number of aromatic rings present a considerable carcinogenic, mutagenic and teratogenic potential (Cavalcante et al., 2007; Hussain, Balachandran, & Hoque, 2015; Witt, 1995; Zheng et al., 2016). PAHs may induce growth inhibition, degradation of the lymphatic system, and interference in hematopoietic functions (Ruiz et al., 2011; Zaghden, Kallel, Elleuch, Oudot, & Saliot, 2007).

PAH-contaminated sediment have also been reported to cause narcotic effects in benthic organisms, tumours in bottom feeding fish, infertility or immune deficiency in many organisms such as oysters (Liu et al., 2009). PAHs accumulated in sediments could also be used by emergent macrophytes (Ning et al., 2010), such as cattail and reed, and can find their way into the aquatic food webs (Liu et al., 2009). If not monitored to control their levels in the sediment, PAHs can pose a potential risk for aquatic lives. Therefore, conducting ecological risk of PAHs is vital for protecting the health of the aquatic environments. Ecological risk assessment has been shown to be a useful tool to characterize the risk of PAHs to organisms and ecosystems (Wu et al., 2011). Interpretive tools are used to determine if sediment-associated contaminants are present at concentrations which could, potentially, impair the designated uses of the aquatic environment. In this respect, the Sediment Quality Guidelines (SQGs), Toxicity Equivalency Factor (TEF), Toxicity Equivalent Quotient (TEQ), mean effect range-median quotient (mERM-Q), and Risk Quotient (RQ) provide scientifically defensible bases for evaluating the potential effects of sediment-associated contaminants on aquatic organisms (Ashayeri et al., 2018; Cao et al., 2010; Cardellicchio et al., 2007; Kalf et al., 1997). The SQGs and TEFs for the 16 priority PAHs have been provided in Table 2. The simultaneous and long-term exposure to a mixture of PAH compounds can cause various adverse biological effects on organisms and aquatic systems, which is not reflected by SQGs. Rather, mERM-Q and TEQ were used to assess the likely combined toxicity or biological risk for exposure to mixed PAHs in sediments (Ashayeri et al., 2018; Cardellicchio et al., 2007; Long & MacDonald, 1998; Long, Macdonald, Smith, & Calder, 1995).

Table 2 - *International Standards for SQGs and TEF for the 16 PAHs of Toxic Potential*

Compound name	SQG-ERL (µg/kg) [a]	SQG-ERM (µg/kg) [a]	TEF values [b]
Naphthalene	160	2100	0.001
Acenaphthylene	44	640	0.001
Acenaphthene	16	500	0.001
Fluorene	19	540	0.001
Phenanthrene	240	1500	0.001
Anthracene	85	1100	0.01
Fluoranthene	600	5100	0.001
Pyrene	665	2600	0.001
Benz[a]anthracene	261	1600	0.1
Chrysene	384	2800	0.01
Benzo[b]fluoranthene	320	1880	0.1
Benzo[k]fluoranthene	280	1620	0.1
Benz[a]pyrene	430	1600	1.00
Indeno[1,2,3-ed]pyrene	240	1400	0.1
Dibenz[a,h]anthracene	63	260	1.00
Benzo[g,h,i]perylene	430	1600	0.01

[a] (Ashayeri et al., 2018; Cardellicchio et al., 2007)
[b] (Ashayeri et al., 2018; Nisbet & LaGoy, 1992)

PAHs present in the water column are not left out either. They also have the potential to bio-accumulate especially in fishes or other organisms that freely live in the water column, thereby posing a potential risk for aquatic ecosystem. TEQ and RQ were applied in this current study to evaluate the ecological risk of aquatic biota's exposure to PAH in the water column.

2.5 Effects of PAHs on human health

The 16 selected PAHs of this current study are among those identified as posing adverse effects on humans. Seven of these compounds are classified by the International Agency for Research on Cancer as probably carcinogenic to humans. These are Chrysene, Benz[a]anthracene, Benzo[k]fluoranthene, Benzo[b]fluoranthene, Benzo[a]pyrene, Dibenz[a,h]anthracene, and Indeno[123-cd]pyrene (IARC Working Group on the Evaluation of Carcinogenic Risks to Humans, 2010).

The main pathways of PAHs exposure to humans is through breathing ambient and indoor air polluted with PAHs, eating PAH-contaminated food, smoking cigarettes, or breathing smoke from open fireplaces (Dahlstrom & Bloomhuff, 2014). In the aquatic environment, PAHs partition into sediments as well as bioaccumulate in mussels and other aquatic invertebrates due to their hydrophobicity and lipophilicity. Through uptake by invertebrates, PAHs can find their way in the aquatic food webs, thereby posing risk to human health through consumption of seafood. Also, ingestion and dermal contact with the water column and sediments are other pathways through which humans are exposed to PAHs in the aquatic environments. It is therefore essential to monitor these contaminants in the aquatic environment to protect human health. In humans, PAHs can caused acute and chronic effects, carcinogenicity, adverse effects on the immune system, teratogenicity, among others.

The effects of PAHs on the health of humans depend on duration and pathway of exposure, the exposure concentration, and the relative toxicity of the exposed PAHs (Dahlstrom & Bloomhuff, 2014). Other contributing factors such as already existing health issues and age can influence the effects of PAHs on a person. Acute toxicity of PAHs on humans is not clear, however, symptoms such as eye irritation, nausea, vomiting, and diarrhoea due to occupational exposures to elevated levels of PAHs have been reported by Unwin, Cocker, Scobbie, & Chambers (2006). Laboratory studies on animals exposed to some PAHs over long time developed lung cancer from inhalation, ingesting and having dermal contact with PAHs (USEPA, 2008). Benzo[a]pyrene is known to be the first carcinogenic compound to be discovered, and usually, noted to cause cancer in animals the most. The PAH metabolites are noted to be the main PAHs that cause immune-toxic effects, and not the parent PAHs. Most of the works published, indicate that immune-suppression is usually the effect after exposure to PAH (Burchiel & Luster, 2001). Also, laboratory studies on mice showed that ingestion of Benzo[a]pyrene caused some embryotoxic effects and low body weight in the offspring of mice (Kristensen et al., 1995). Such effects have not yet been reported in humans, however, a work by Perera, Tang, Whyatt, Lederman, & Jedrychowski (2005) observed that exposure to PAH pollution during pregnancy can cause adverse birth consequences such as, low birth weight, premature delivery, and heart malformations.

Generally, with long-term exposure, PAHs may decreased immune, kidney and liver functions, cause asthma-like symptoms, and lung function abnormalities (Gehle, 2009). All these adverse effects and many more which were not covered in this section establish the need to monitor and control PAHs that are released into the aquatic environments.

2.6 Remediation

Predicting the bioavailability of PAHs in sediments is largely determined by their molecular weight and hydrophobicity. The lipid content of the receptor (organism), the organic carbon content of the sediments and Kow (Meador et al., 1995; USEPA, 2004), are all underlying determinants of PAHs' bioavailability in sediments. Some organic compounds such as black carbon soot, charcoal and coal are known to have strong sorption capacities to hydrocarbons, thereby decreasing the bioavailability of PAHs in sediments (Beckingham & Ghosh, 2011; EPA, 2004; Fadaei, Watson, Place, Connolly, & Ghosh, 2015; Jonker, Suijkerbuijk, Schmitt, & Sinnige, 2009; Samuelsson et al., 2015). Since the sediments become one of the main sink for these contaminants, such sediments can be ingested by biota, contaminants can desorb from the sediment, hence exposing the ecosystem at large. Due to this, several management practices are put in place to ensure a healthy sediment. Some of these include minimising emissions, assessment on monitoring in order to group polluted sediments based on potential risk for humans and ecosystems, and in situ and ex situ remediation approaches for polluted sediments (Kupryianchyk, Rakowska, Grotenhuis, & Koelmans, 2012; Kupryianchyk et al., 2011; National Research Council, 2007).

Conventionally, dredging or in situ capping with clean materials including sand, silt, or clay has been used in treating contaminated sediments in order to achieve sediment quality criteria. Researchers are coming out with new remediation techniques that either supplement or offer less laborious, less expensive, less destructive alternatives to existing methods. Also, making sure that such remediation methods are able to reduce human and ecosystem exposure to contaminants. Hence, investigations on the potential use of carbonaceous materials (CM) to polluted sediment as a means of reducing pore water hydrophobic organic compounds (HOCs) concentrations are ongoing.

2.6.1 Types of remediation

Dredging is a remediation technique where polluted sediment is removed and treated *ex situ* (National Research Council, 2007). It has the advantage of removing almost all contaminants in that specific area. However, in cases where this is not achieved, it can lead to residual contaminants in the sediment and pore water, even exceeding permissible levels. It puts the sediment structure in a deteriorating state, and such a method is very costly. Dredging can also lead to re-suspension of underlying deep sediment particles, temporarily increasing pore water concentrations of contaminants (Clemente, Almela, & Bernal, 2006; National Research Council, 2007). Since treatment is done *ex-situ*, one other disadvantage of this method is the possibility of contaminants leaching from storage facilities. It partially or wholly destroys benthic habitats and communities during excavation (National Research Council, 2007). Nonetheless, dredging has been applied in lakes, rivers, and harbours in the United States of America and other countries because it permanently gets rid of the contaminated sediment (Perelo, 2010).

For *in situ* capping, its purpose is to reduce exposure by creating a 30 – 100 cm defensive barrier made of clean material such as clay, silt, or sand. This clay, silt, or sand is placed on the contaminated sediment bed to isolate the clean material from the contaminated sediment (Murphy et al., 2006; National

Research Council, 2007). Using caps on contaminated sediment is typically less expensive than dredging, and its costs depend mainly on cap design. However, capping does not always effectively achieve the purpose of reducing contaminant transport because of the permeability of capping materials, dynamic topography of the affected area, or erosion of the cap (Bokuniewicz, 1992; Lohse, Epping, Helder, & Van Raaphorst, 1996; National Research Council, 2007; Perelo, 2010). This technique also has a disadvantage of partially or wholly destroying benthic habitats and communities. Also, during the capping material introduction, contaminated sediments can be mixed with clean material or resuspended, making contaminants readily available in the system (National Research Council, 2007; Perelo, 2010). Researchers' goal is to investigate the design of different reactive caps effective to minimise the risk associated with cap permeability (Zimmerman, Ghosh, Millward, Bridges, & Luthy, 2004).

Another process that can remove PAHs from contaminated sediment is biodegradation. Biodegradation of PAHs by organisms like bacteria, fungi, algae, and plants is considered the main detoxification and removal (Perelo, 2010; Samanta, Singh, & Jain, 2002). This process can occur either in an aerobic or anaerobic condition. Bioremediation is, therefore, the term used to explain the process in which contaminants in an environment are degraded into less harmful products through biological means, using the metabolic potential of microorganisms (Scragg, 2005). Bioremediation in sediment is the least disruptive, so far as the ecosystem is a concern and the least expensive restorative approach. Besides being cost-effective, bioremediation is a long-lasting solution, resulting in the complete mineralization of the pollutant.

However, some major disadvantages, such as the processes involved, are time-consuming and less predictable than conventional methods like dredging (National Research Council, 2007; Perelo, 2010).

Different bioremediation strategies include monitored natural recovery (or natural attenuation), biostimulation, bioaugmentation, and phytoremediation (Perelo, 2010). Monitored natural recovery is the method in which the ecosystem undergoes “self-healing.” In such a case, the natural capabilities of the native microbial population and the naturally occurring physical and chemical processes breakdown PAHs into minerals that are not harmful to the existing ecosystem (Perelo, 2010). The main processes involved in this natural recovery method are allowing the deposition of naturally occurring clean particles and new organic matter to be formed. Such processes result in new and clean layers covering the sediment surface and bury the contaminants in the process, thereby disconnecting them from the bed surface layer, the overlying water, and aquatic organisms. In time, other factors such as biological and chemical degradation, sequestration, and bed consolidation contribute to less exposure to contaminants and facilitate natural recovery (Thibodeaux & Bierman, 2003). While no action is needed to start or continue the process, natural recovery is the output of a deliberate, thoughtful decision ensuing after detailed site assessment and characterization; hence, the name “monitored” natural recovery (National Research Council, 1997).

Biostimulation boosts the native population by influencing the parameters that play a role in microbial growth needed for contaminant degradation (Perelo, 2010). Both organic (e.g., sewage sludge) and inorganic nutrients (such as NPK fertilizers) (Atagana, 2008; Chorom, Sharifi, &

Motamedi, 2010; Louati et al., 2014) can be used as stimulating nutrients for bioremediation. Another strategy used in bioremediation is bioaugmentation. Bioaugmentation is the addition of appropriate microorganisms with explicit catabolic potentials into the contaminated environment to achieve the goal of increasing the native population to speed up or aid the degradation process (Adams, Fufeyin, Okoro, & Ehinomen, 2015; Louati et al., 2014; Perelo, 2010). Bioaugmentation has been demonstrated to be successful for remediation of PAH-contaminated sediments with poor or lacking inherent degradation capability (e.g., Juhasz & Naidu, 2000), while other research works have shown that bioaugmentation did not improve biodegradation significantly as compared to natural attenuation (e.g., Tam & Wong, 2008). Some of the major issues to be concerned with bioaugmentation is the survival and activity of the introduced species in the environment of interest, pH, concentration, and bioavailability of contaminants (Thompson, Van Der Gast, Ciric, & Singer, 2005).

Last but not the least strategy of bioremediation is phytoremediation. This method focuses on using plants and algae in the degradation and removal of contaminants from the environment (Peng, Song, Yuan, Cui, & Qiu, 2009; Perelo, 2010; Zarull, Hartig, & Maynard, 1999). Phytoremediation of organic pollutants such as PAHs has the likelihood to mineralize or change the pollutant into the less- or non-toxic form including, carbon dioxide, nitrate, chlorine, and ammonia (Peng et al., 2009). The application of phytoremediation has been recommended for *in situ* sediment remediation of shallow waters (Perelo, 2010; Zarull et al., 1999), including lagoons. Plants can remediate pollutants in sediment through adsorption, transport, and translocation, and by transformation and mineralization by plant enzymes (Meagher, 2000), or algae.

In a study by Yamamoto et al. (2008), benthic microalgae were used for the first time to remediate organically enriched sediments.

Bioremediation approaches promise to eliminate dredging and in situ capping drawbacks. However, their low predictability and sometimes long degradation times, which involve extensive monitoring activities, make this remediation technique stand to be at a disadvantage. Not to mention that many bioremediation approaches are laboratory-based and work differently under different parameters under field conditions. With these backgrounds, researchers are coming out with new remediation techniques that either supplement or offer less laborious, less expensive, less destructive alternatives to existing methods, which can reduce human and ecosystem exposure.

Recently, scientists have recommended an *in situ* technique where carbonaceous materials such as activated carbon were added to contaminated sediments as an alternative to dredging for sediment remediation. (Antrim & Kohn, 2000; Kohn & Evans, 2004; Kupryianchyk et al., 2011). There has been much progress over the past years to comprehend how these carbonaceous materials affect the bioavailability and toxicity of HOCs. However, in Ghana, such data is limited. To the best of the author's knowledge, this current study is the first to explore activated charcoal effectiveness in remediating Ghanaian lagoon sediments contaminated with PAHs. This part of the research is meant to enhance the mechanistic understanding of activated charcoal application on PAHs contaminated sediment through laboratory experiments.

During the late 1960s, organic matter (OM) in soils and sediments was discovered to be the major binding site for HOCs (Lambert, 1968). In the 1970s, the model of normalizing the solid–water distribution coefficient to total organic carbon (TOC) was also found (Chiou, Peters, & Freed, 1979; Karickhoff, Brown, & Scott, 1979). Years on, this theory was tested and many findings reported that there are other domains in the sediments where sorption takes place. Such findings included, elevated sediment–water partition coefficients, nonlinear sorption isotherms, multiphasic desorption kinetics and presence of soot phases (Allen-King, Grathwohl, & Ball, 2002; Cornelissen et al., 2005; Gustafsson et al., 1997; Pignatello & Xing, 1996). These outputs indicated that naturally present CM, like soot and chars, also called black carbon (BC), readily bind HOCs, making them immobile (Ghosh et al., 2003; Gustafsson et al., 1997; Jonker & Smedes, 2000; Koelmans et al., 2006; Luthy et al., 1997; Weber, Huang, & LeBoeuf, 1999). Other developing works now focused on studying the properties of BC (Cornelissen, Kukulska, Kalaitzidis, Christanis, & Gustafsson, 2004; Koelmans et al., 2006), developing ways for its quantification (Cornelissen et al., 2004; Gustafsson et al., 1997; Koelmans et al., 2006) and coming out with a mechanistic understanding of sorption to BC (Accardi-Dey & Gschwend, 2002; Gustafsson et al., 1997; Jonker & Koelmans, 2002). From the diverse studies, commercial activated carbon used in water treatment was suggested by researchers, and so in this current study, activated charcoal (similar to manufactured activated carbon in sorption properties) was used for the remediation experiment.

In situ treatment of HOCs such as PAHs, by either addition of CM to PAH-contaminated sediments, or for ex situ application such as mixing CM in sediment slurries during dredging, transportation, or processing has yielded very good results in terms of its high effectiveness and low cost (Beckingham

& Ghosh, 2011; Grossman & Ghosh, 2009; Jonker et al., 2009; Kupryianchyk et al., 2011; Nybom et al., 2016). The science behind this field of CM technique seeks to answer 3 main research questions (Rakowska et al., 2012). One of them is the mechanistic and technical aspects of CM application. This takes into account CM type, dosage, and mixing procedure. In effect, how CM should be applied to obtain an efficient and cost-effective outcome. The second is the ecological and eco-toxicological effects of CM application. This aspects investigates on improving the sediment quality since HOCs become immobile, as well as possible adverse effects of CM on aquatic biota. This research area studies the effects on species, community or the ecosystem level in order to achieve a required concentrations of HOCs in the aquatic environment. Last but not the least is modelling the effects of CM amendments for various scenarios.

The premier tests with CM addition were laboratory tests aiming mainly on in situ applications for PCB- and PAH-contaminated sediments. Later on, the first pilot-scale field experiments (*ex situ*) were conducted for in situ stabilization of HOCs using activated carbon (Beckingham & Ghosh, 2011; Cho et al., 2009, 2007; Cho, Werner, Choi, & Luthy, 2012; Cornelissen et al., 2011). Since this current study is the first to report on activated charcoal as a remediating material for PAH contaminated sediment of coastal lagoons in Ghana, a laboratory test was conducted.

Zimmerman et al. (2004) investigated on the effect of addition of activated carbon to reduce PAH bioavailability in marine sediments from Hunters Point Naval Shipyard, San Francisco Bay. This was done for the periods of 1 and 6 months. Sediment treated with 3.4 % activated carbon exhibited 84% reductions of PAHs in the aqueous medium and 83% reductions in PAH uptake by semipermeable membrane devices. They ascribed the effectiveness of the activated carbon to its large specific surface area and a pore structure effective for binding the PAHs. The outcome of their results yielded the following conclusions: that adding activated carbon to contaminated field sediment reduces the presence or concentrations of hydrophobic contaminants available to the aqueous medium, the advantage is seen relatively quickly due to optimum contact conditions and the contact time, and lastly, that activated carbon application is a potentially plausible method for in-situ, non-removal treatment of marine sediment contaminated with HOCs.

In another study, activated carbon was introduced into a creosote-contaminated soil and an urban soil with total PAHs of 5,500 mg/kg and 38 mg/kg respectively (Brändli, Hartnik, Henriksen, & Cornelissen, 2008). The total PAHs consisted of the 16-EPA priority PAHs without naphthalene. An amount of 2% powdered and granulated activated carbon were applied to the two contaminated soils. The results for urban soil indicated a 99% and a 64% reduction of freely dissolved aqueous concentration of PAH in soil and water suspensions when powdered and granulated activated carbon were applied respectively. For the creosote-contaminated soil, the reduction for the powdered activated carbon was 63%, while that of the granulated activated carbon was 4%. Comparatively, the general low reduction in the creosote-contaminated soil to the urban soil was attributed to saturation of activated carbon sorption sites by the high PAH concentrations and/or blockage of sorption sites and pores by oil. They concluded that there exist a high chemical effectiveness of activated carbon amendment in soil. However, further research questions are to be answered in terms of how these results translates to possible field application.

Especially, where continuous mixing may not be effective and diffusive PAH transfer through the pore gas and/or pore water is needed to attain a redistribution of contaminants from soil particles to activated carbon. They also recommended that further research is needed to study on the likelihood of secondary effects of activated carbon amendment on soil microbiology, soil nutrients, soil ecology and soil fertility.

With the purpose to know whether there are differences between pollutant binding to biomass and coal-based activated carbon, Amstaetter, Eek, & Cornelissen (2012) performed adsorption test in contaminated marine sediment containing phenanthrene, anthracene, fluorene, pyrene, Benzo(a)pyrene, chrysene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)anthracene, indeno(1,2,3cd)pyrene, Benzo(g,h,i)perylene. Upon introduction of 2% activated carbon, it was evidenced that the present of coal-based activated carbon exhibited more extensive sorption than biomass-based activated carbon. Also, the effectiveness of activated carbon amendment appeared to decrease with compound hydrophobicity. The reason they ascribed to this was that the larger molecules had difficulty in accessing clogged and narrow pores. They also hypothesised that the results obtained could be basis for other polluted sediments because many contaminated sediments exhibit similar sorption properties (strong binding due to the presence of black carbon).

Similarly, Oleszczuk, Hale, Lehmann, & Cornelissen (2012) investigated on the removal efficiency of freely dissolved PAHs (phenanthrene, fluoranthene, pyrene, Benzo(a)pyrene, Benzo(g,h,i)perylene) from sewage sludge using biochars and activated carbon. They reported a significant reduction of freely dissolved PAHs concentration when sewage sludge was amended with biochars and activated carbons. Based on the dose, the reduction varied from 56% to 95% (activated carbons) and from 0% to 57% (biochars). They also observed that it was only for the biochars experiment that there existed a significant difference between short 7-d and long 30/60-d mixing time. The key conclusion from Oleszczuk et al. (2012) was that biomass-based activated carbon might be the optimal sorbent for sewage sludge, because it portrayed a relatively high pollutant sorption effect compared to bitumen-based activated carbon and even much higher than unactivated biochars. Although one may argue of the carbon sequestration effect that is lost in the energy intensive activation process, a previous study on a complete life-cycle assessment of sediment remediation by different amendment projected that biomass-based activated carbon was the optimal material in a global perspective (Sparrevik et al., 2011).

Also, a review by Rakowska et al. (2012) looked at the modern state-of-the-art use of CM as an extensive means for sediment remediation, considering both technical and ecological aspects. They looked at how factors such as CM type, particle size and dosage, sediment characteristics, and properties of contaminants could influence the effectiveness of CM amendment to immobilize HOCs in aquatic sediments. The also discussed the extent to which CM may reduce bioaccumulation and toxicity of HOCs, as well as whether CM itself has negative effects on benthic species and communities. The review was based on literature and datasets from laboratory and field trials with CM amendments. From their reports, presence of domains such as natural black carbon, oil, or organic matter in the sediment decreases the effectiveness of CM amendments. They also indicated that introduction of CM seem to improve the habitat quality for benthic organisms by way of decreasing bioavailable HOC concentrations and toxicity in sediment. Should there be any adverse effect of CM itself on benthic species, it was reported to be insignificant. Thus, the positive effects of decreasing toxicity at low CM concentrations most probably outweigh the insignificant adverse effects detected at higher CM concentrations.

In Ghana, the use of CM as a remediating technique in lagoon sediment is an upcoming research. This current study will be the first mechanistic approach to evaluate the possibility of activated charcoal (sorbent for organic pollutants with similar strong sorption properties as activated carbon) as a remediating material in PAH-contaminated lagoon sediments.

2.7 Chapter summary

In this section, the nature of PAHs, their sources, and their distribution in the aquatic environments, as well as their toxicological effects on the aquatic lives and humans have been reviewed to provide more insights about the mobility of PAHs in these ecosystems. Works on the use of carbonaceous materials as amendment for contaminated sediments to ensure healthy aquatic ecosystems have also been reviewed in this section.

CHAPTER THREE

MATERIALS AND METHODS

3.1 Description of the Study Areas

In this study, three lagoons located in the Central Region of Ghana were selected (Figure 3). This region receives two wet seasons in a year; the major one from April to July and a minor season from September to November, and a dry season from December to March (Gilbert et al., 2006; Logah, Obuobie, Ofori, & Kankam-Yeboah, 2013). One of the lagoons that was considered in this current study was Brenu Lagoon. It is located at Brenu Akyenim and lies within latitude 5°4′0"N–5°4′30"N and longitude 1°25′30"W–1°26′0"W. It is a classical closed lagoon (Yankson & Obodai, 1999) located about 13 km west of Cape Coast in the Central Region. It is fed by rainfall and other five small streams: Esuaka, Obuohu, Asenkye, Asosi, and Burabui (Yankson & Obodai, 1999; Zuh et al., 2019). The lagoon covers an area of 0.82 km^2 (82 ha) and it is separated from the adjacent sea by a sand bar for the greater parts of the year (Zuh et al., 2019). The lagoon has contact with the sea either naturally (overflow of its banks due to precipitation) or artificially by the indigenous people. The latter is normally done to avert flooding of nearby villages and farms. The Brenu Lagoon is bordered by large strands of mangroves, mostly *Avicennia sp.* and small patches of marsh (Zuh et al., 2019). The main economic activities in this area are fishing and farming. Fishing by the inhabitants of Brenu Akyenim and Ampenyi-Ayensudo in this area is seasonal, and this usually occurs during the dry season when farming activities are minimal. One other key characteristic noted within this lagoon's catchment is a road connecting the two towns (Brenu Akyenim and Ampenyi-Ayensudo).

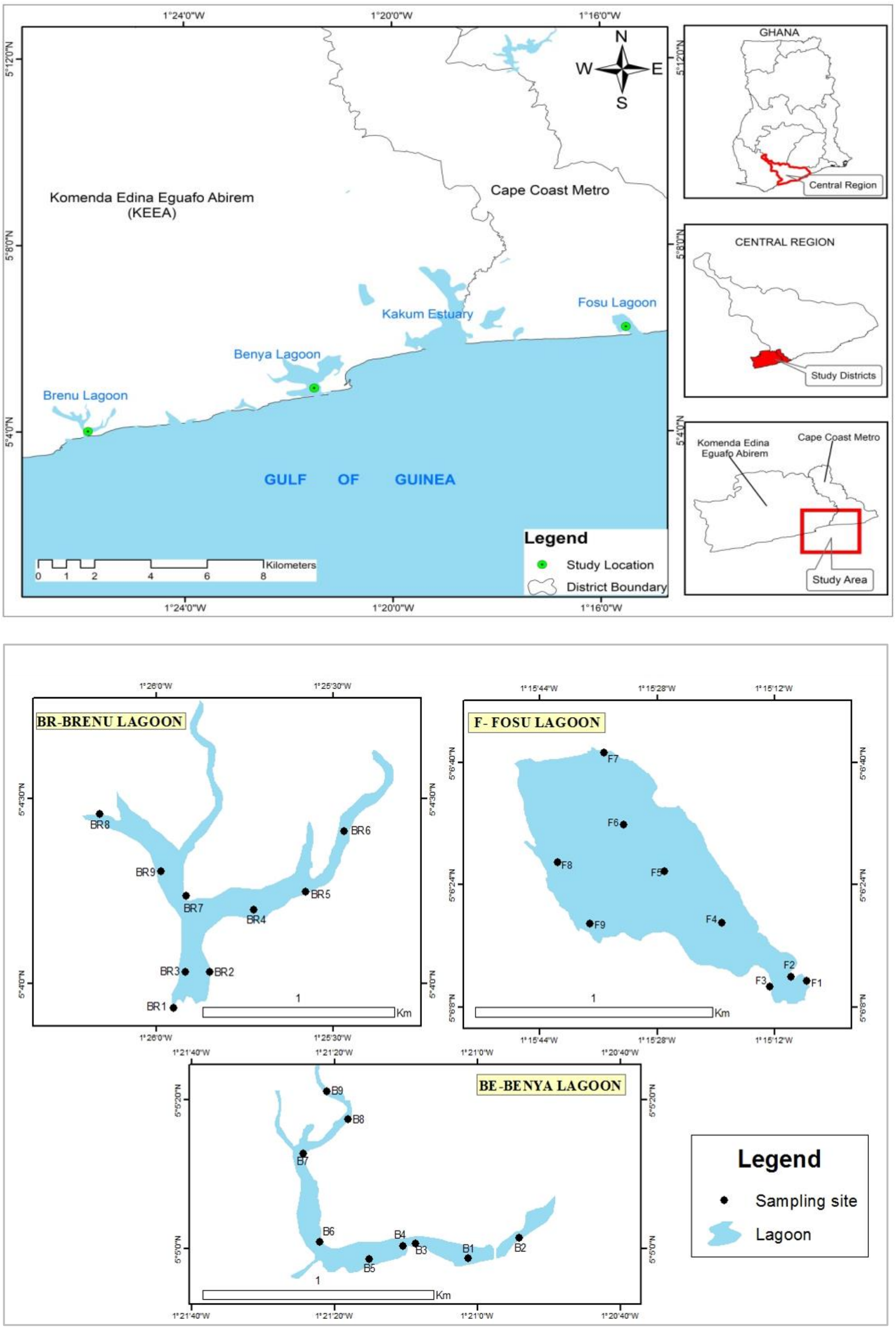

Figure 3: Maps showing the study locations and sampling sites

Another closed lagoon selected for this study was Fosu Lagoon. It is located within the Cape Coast Metropolis and lies between latitude 5°6′40″N–5°6′8″N and longitude 1°15′12″W–1°15′44″W. It is a shallow brackish water body which is separated from the Gulf of Guinea by a sand bar and having an area of approximately 0.61 km^2 (61 ha) with an average water depth of 1.5 m (Blay & Asabere-Ameyaw, 1993; Dankwa, Quarcoopome, Owiredu, & Amedorme, 2016). The sand bar is removed by heavy rainfall or by the local people as part of a series of annual rituals. In addition to its significance as a traditional heritage of the people, it serves as a livelihood for fishers especially, in that area (Armah, Ason, et al., 2012; Dankwa et al., 2016). Other economic activities that can be located in the settlements along the lagoon's bank are automobile servicing garages and metal fabrication.

The lagoon exhibits a wide range of irregular water level fluctuations, a situation that sometimes leads to near dryness during the lean season. The lagoon serves as a reservoir for both untreated liquid and solid wastes from various activities carried out along its banks and even distant surroundings. The source of these waste are directly and mainly from St. Augustine's College, Robert Mensah stadium, the Cape Coast Municipal hospital, the nurses training college, Siwdo mechanic shops, and some households within the buffer zone of the lagoon. Wastes from these human activities flow directly into the lagoon without any prior treatment. These human induced developments along the peripherals of the lagoon have also caused an outgrowth of marshes, which consequently, have adversely affected the physiological changes of the lagoon. This has also led to a decrease in species diversity. The lagoon is covered with mats of mixed vegetation consisting of shrubs of about 1.5 m high, grasses and a few scattered trees. The mangroves have also depleted considerably leaving a few around the shrine area. These activities in the catchment and buffer zone of the lagoon have led to relatively high level of sedimentation causing a siltation of the lagoon (Baffour-Awuah, 2014).

The primary fresh water source to this lagoon is direct rainfall, which is seasonal, and so, even during high rainfall, the lagoon is not able to store enough water due to high siltation. Whenever there is a splash of seawater over the sand bar due to flooding; either naturally (through rainfall) or artificially, larvae of crustaceans such as crabs enter the lagoon. However, these marine species are short-lived due to the quality of the lagoon. The dominant fish species in this lagoon is *Sarotherodon melanotheron* (Dankwa et al., 2016). Fosu Lagoon is the third most polluted lagoon in Ghana, after the Korle Lagoon in Accra and the Chemu Lagoon in Tema (Akoto et al., 2014; Baffour-Awuah, 2014).

The third selected lagoon for this study was Benya Lagoon, an open lagoon. It is located in the Komenda-Edina-Eguafo-Abrem Municipality in the Central Region of Ghana. It lies between latitude 5°5’0’’N–5°5’20’’N and longitude 1°20’40’’W–1°21’20’’W, with a depth which ranges between 0.5 m and 2 m (Armah, Ason, et al., 2012). It exchanges water with the sea throughout the year and partly under the tidal influence (Armah, Ason, et al., 2012; Joiris, Azokwu, Otchere, & Ali, 1998). The Benya Lagoon’s catchment area is one of the few locations in Ghana where salt is produced commercially. The main occupation of the inhabitants, however, is fishing. One key observation with this lagoon, is that it serves as a fish landing quay (the third largest in Ghana) and a berth for numerous canoes and semi-industrial fishing vessels (Obodai et al., 2011).

The selection of these lagoons was based on their geomorphological orientations (closed or open), which largely determine their level of interaction with the sea and the types of activities that prevail within the lagoons' catchment. Open lagoons are always in contact with the sea; hence, they are continuously under tidal influence. In contrast, closed lagoons interact intermittently with the sea through natural or anthropogenic means. Fosu and Benya Lagoons are located in urban areas and have become a reservoir of many industrial effluents and untreated domestic wastewater. The Benya Lagoon also serves as a landing quay for many fishing vessels due to accessibility to the sea almost daily. Brenu Lagoon, on the other hand, seems to experience less anthropogenic activities. However, refuse are incinerated around the lagoon's mouth with a road connecting Brenu Akyenim (where the lagoon is located) and Ampenyi Township.

3.2 Sample collection and preparation

3.2.1 Water and sediment samples

Nine sampling sites (see Figure 3) with a distance of about 100 m to 300 m between them were selected for each lagoon. The choice of the sampling sites was based on their accessibility and nearness to anthropogenic activities that are potential source of pollutants. The nine sampling sites represented three main regions of the lagoon: a proximal, middle, and distal regions (with reference to distance to sea). For the sediment samples, the three sampling sites of each region represented the triplicate for that region. However, for water samples, the three sampling sites of each region represented a replicate. Ekman grab was used to sample sediment after which the samples were kept in aluminium packs. For the water samples, a pre-cleaned (with acetone) 250 mL amber bottles were used. Both sediment and water samples were kept on ice (at 4 ºC) to maintain cells' metabolic activities. Samples were then sent to the Fisheries and Coastal Research Laboratory at the Department of Fisheries and Aquatic Sciences, University of Cape Coast (UCC) for further analysis. At the laboratory, samples (for both water and sediment) that constituted a sampling region were thoroughly mixed to serve as a composite sample. This mixing was done to help minimize within-site variability and ensured that both water and sediment samples collected were genuinely representative of each region (Koh et al., 2004). Suspended particulate matter for PAH analysis was not separated from the dissolved pollutants present in the water samples collected. Hence, water samples were not filtered before extraction.

Debris, pebbles, and other unwanted materials were removed from the sediment samples. The samples were air-dried (average temperature of 30 °C) to constant weight. Composite samples were ground and homogenized in a porcelain mortar and sieved with a 2 mm mesh. Three replicates, each of 10 g, were then weighed from the composite sample for the PAH extraction. Ten grams (10 g) of the same composite were measured for organic matter analysis. For the water samples, 500 mL of water from Benya and Brenu were used for the PAH extraction. However, 250 mL of water sampled from Fosu was used for the PAH extraction. Such volume was used because algae clogged the bed of the Solid Phase Extraction (SPE) cartridges used for the extraction. Sediment samples were collected monthly from December 2017 to February 2019, while the water samples were collected in November 2018 and January 2019. Extraction of PAHs in water samples were done only in the stated two months because that was when needed equipment and materials were available.

3.2.2 Fish samples

S. melanotheron (fish samples) were purchased from commercial fishers who fish on the day of sampling. Samples were collected every other month from January 2018 to January 2019 in each lagoon to minimise cost for PAHs extraction. However, these sampling periods did not disrupt the objectives of the study. Fish samples were wrapped with aluminium foil and kept under ice (at 4°C) to maintain cells' metabolic activities. Samples were then taken to the Fisheries and Coastal Research Laboratory, UCC. Thirty individual fishes of average size; 7.54 ± 0.15 cm, 7.34 ± 0.10 cm, and 7.66 ± 0.11 cm were selected from fishes caught from Benya, Brenu, and Fosu respectively. Twenty-five fish specimens were used for the PAH extraction procedure, while five were used for wet and dry weight analysis. The scales of the fish samples for PAH analysis were removed and then rinsed with distilled water. With the aid of a dissecting kit (pre-sterilised with acetone), each fish specimen was dissected into three main parts: the head, the visceral (gut), and the muscles (including skin). Each part was homogenised, and three replicates (for each fish part) weighing 10 g were used for the PAH extraction. Samples were then stored at -20 °C until extraction was done. All the weights were measured using a Sartorius analytical balance.

The following were used to characterise the lagoons: water quality parameters, water and lipid contents of the fish, and the organic matter of the sediment.

3.3 Water quality parameters

Conductivity (mS/cm), pH, salinity (ppt), temperature (ºC), total dissolved solids (TDS; mg/L), and turbidity (NTU) of each water body was measured *in situ* using a multi-parametric portable instrument (Horiba U-50 series).

3.4 Water content analysis of fish

The water content was calculated as the difference between the wet weight of fish samples measured and the dry weight. The wet weight was measured after external water elimination with a paper tissue. The dry weight was also measured after keeping the specimens in an oven (Genlab) at 105 °C for 24 hours (Federation & APHA, 2005; Sun et al., 2020).

3.5 Lipid content analysis

Soxhlet extraction is usually recommended by the Association of Official Analytical Chemists (AOAC) as the standard method for crude fat analysis (Essumang et al., 2012). The fish fillet was dried in an oven at 105 °C until a constant weight was achieved. About 10 g of the dried finely ground fish fillet (representing S in equation 1) was weighed into a 50 ×10 mm Soxhlet extraction thimble. The sample was then transferred into a 50 mL capacity Soxhlet extractor. A clean, dry 250 mL round bottom flask was pre-weighed (representing W1 in equation 1). About 150 ml of petroleum ether (40–60 ˚C, Lot: 15L100518, CAS number: 64742-49-0; purchased from BDH PROLAB UK) was added to the 250 mL round bottom and connected to the Soxhlet extractor. Extraction was then performed for 4 hours using a heating mantle as a source of heat. After 4 hours, the round bottom flask was placed in an oven at 60 °C for a further 2 hours to allow the petroleum ether to evaporate. The round bottom flask was removed, cooled in a desiccator, and weighed (W2 in equation 1). The percentage of lipid was calculated following the approach described by Thiex (2009) and Essumang et al. (2012):

$$\boldsymbol{lipid\ content}\ (\%) = \left(\frac{\boldsymbol{W2-W1}}{\boldsymbol{S}}\right) \times \mathbf{100}$$ ……………………Equation 1

W1 is the initial weight of the round bottom flask. W2 is the weight of the flask with fat and S is the weight of sample.

3.6 Organic matter content analysis

Loss on Ignition (LOI) was the method adapted for this analysis (Ashayeri et al., 2018; Rather, 1918). Ten grams of sediment was weighed from the composite samples and dried at 105 °C to a constant weight. Upon further drying, the organic matter (OM) was determined when samples were heated to a temperature of 550 °C (in the Cole-Parmer furnace) for 3 hours. The total organic carbon (TOC) content was computed by a correction factor of 1.724 (i.e., OM = TOC × 1.724) (Schumacher, 2002).

3.7 Bioaccumulation models

To adequately comprehend the extent of transfer of contaminants such as PAHs in the trophic chain, bioaccumulation factors models are useful (Froehner, Rizzi, Vieira, & Sanez, 2018; Kwok et al., 2013; Liang et al., 2007; Rojo-Nieto et al., 2014). For this reason, the bioaccumulation factor (BAF) and biota-sediment accumulation factor (BSAF) (Kwok et al., 2013) were employed in this current study. The BAF was estimated using the equation below:

$$BAF = \frac{C_f}{C_s}$$ ---Equation 2

Where C_f is the concentration in fish tissues (in µg/kg wet weight) and C_s the concentration in sediments (in µg/kg dry weight). The extension of bioaccumulation was also measured, taking into account the lipid content of fish tissues. In this case, a BSAF model was estimated by:

$$BSAF = \frac{C_f/L}{C_s/TOC} \text{ ……………………………………. Equation 3}$$

Where C_f is the PAH concentration in fish tissue (μg/kg wet weight), L is the lipid content in the fish tissue in %; Cs is the PAH concentration in sediment (μg/kg dry weight), and TOC is the organic carbon in the sediment in %.

3.8 Sediment remediation experimental setup

With the aid of an Ekman grab replicate sediment samples were collected from the Fosu Lagoon and stored in the dark place until used. Sediment weighting 350 g was measured into 1 L beakers, in replicates of twelve. The total PAHs in the sediment was 2 ppm (2 mg/kg). A 28-day AC experiment was carried out with the collected sediment. Sediment samples were treated with powdered AC (AC; purchased from Central Drug House Limited, India, CAS number: 7440-44-0, particle size passed through 300 mesh, pH: 6.0 – 7.5) at different percentages of 1%, 3%, and 6% under laboratory conditions. These percentages were selected based on literature report that a dosage of 1 - 30% AC resulted in a significant reduction of hydrophobic organic contaminant concentrations in the aqueous phase (Brändli et al., 2008; Kupryianchyk et al., 2011). A fourth sediment sample with no AC treatment (0%) served as a control experiment.

All sediment treatments were then thoroughly homogenised and compacted. Distilled water of volume, 550 ml was gently added to each treatment. The water was allowed to settle for 48 hours prior to the start of the experiment. The overlying water was aerated continuously over the 28 days. Also, weekly measurements of temperature (ºC), pH, salinity (ppt), conductivity (mS/cm), dissolved oxygen (mg/L), and total dissolved solids (mg/L) were carried out using a Hanna instrument 9829 over the experimental period. At the end of the experiment, pore water (freely dissolved) concentrations of PAH were measured using polyoxymethylene (POM) samplers.

3.9 Extraction and analysis of PAHs

QUick, Easy, CHeap, Effective, Rugged and Safe (QUECHERS) method (Albinet, Tomaz, & Lestremau, 2013; Angioni, Porcu, Secci, & Addis, 2012; Ramalhosa, Paíga, Morais, Delerue-Matos, & Oliveira, 2009) was adapted for the extraction of PAHs in the sediment and fish samples. For the water samples, Solid Phase Extraction (SPE) was employed for the extraction. QUECHERS has been extensively used for pesticides and PAHs extractions. It is known to be advantageous over those involving intensive treatments since accurate results can be achieved with minimal sample preparation in a short time (Bragança, Plácido, Paíga, Domingues, & Delerue-Matos, 2012; Ramalhosa et al., 2009). It is a method suitable for laboratories that engage in daily routine analyses of large numbers of samples. PAHs in water samples were quantified with reference to USEPA method 8310 (Li & Lee, 2001; USEPA, 1986).

3.9.1 Extraction of PAHs from water samples

3.9.1.1 Cartridge conditioning

The SPE cartridges (HyperSep C18, Thermo Scientific Rockwood, USA) were attached to a manifold connected to a vacuum pump. A volume of 3 mL dichloromethane (HPLC grade, 99.8% purity, Lot: 18D254014, CAS number: 75-09-2) was added to the cartridge and drawn under vacuum in succession. The procedure was repeated four times (3 × 4 ml aliquots of dichloromethane). Similarly, 4 mL of methanol (100%, grade: analytical reagent, Lot: 17J314014, CAS number: 67-56-1), was added, and this was also repeated three times. Lastly, 4 mL of distilled water was added. This was done in 3 successions. About 1 cm of distilled water was retained above the column bed.

3.9.1.2 Sample extraction

For Benya and Brenu water samples, 500 mL of the composite was used for the PAH extraction, while 250 mL was used with samples from Fosu Lagoon. The samples were steadily added to the cartridges and vacuumed under a constant rate of about 15 – 20 mL per minute. The sample beakers were then rinsed with distilled water and added to the cartridge before drawing under a full vacuum for 10 minutes to dry the column.

3.9.1.3 Sample elution and drying

Analytes were eluted with 3 × 4 mL aliquots of dichloromethane under gravity. The extract was collected in a 15 mL Teflon tube before drying the column under a full vacuum. The samples were then reduced to near dryness at 40 °C (for dichloromethane to evaporate) and re-concentrated to 3 mL with ethyl acetate (analytical grade, Lot: 18H014006, CAS number: 141-78-6). An amount of 150 mg of anhydrous $MgSO_4$ (analytical reagent, 99.4% purity) was then added to the sample and vortex-mixed for a minute (with Vortex-Genie 2). Samples were then centrifuged at 4500 rpm for 5 minutes (Eppendorf 5430). Approximately, 1.5 mL of the extract was filtered through a 2-micron (2E-6 m) syringe filter into a 2 mL vial. The final extract was then stored at 4 °C until PAH analysis by the GC-MS.

3.9.2 Extraction of PAHs from fish and sediment samples

Ten grams of fish fillet, head, visceral tissues, and sediment was weighed separately for the extraction process. Each was transferred into a 50 mL Teflon centrifuge tube. Thereafter, 20 mL of acetonitrile (HiPerSolv HPLC grade, Lot: 18L141668, CAS number: 75-05-8) was added to the sample and allowed to settle for 15 minutes. The samples were then vortex-mixed for 2 minutes. Four grams of anhydrous $MgSO_4$ and 1 g of NaCl (analytical reagent, 99.9% purity) were added to the samples. The samples were then vortex-mixed for 1 minute and centrifuged for 5 minutes at 3500 rpm. Thereafter, 5 mL of the extract was transferred into a 15 mL Teflon tube for clean-up. 50 mg of zirconium Z-Sep sorbent (Supel QuE Z-Sep, Lot: 95115) combined with 150 mg anhydrous $MgSO_4$ was added to the extract for clean-up purposes. The samples were then vortex-mixed for a 1 minute and centrifuged for 5 minutes at 3500 rpm. Afterwards, 1.5 mL of the extract was filtered through a 2-micron syringe filter into a 2 ml vial. The final extract was then stored at 4 °C until the analysis of PAH by the GC-MS.

3.10 Preparation of standard solution for calibration

Concentrations of 1.0, 10.0, 20.0, 30.0, 50.0, 10.0 and 200.0 ppb of the native standards were prepared from 1000 ppm stock solution of 8270 megamix (#31850 (e.a), Restek) to prepare the calibration curve. Deuterated internal standards of 1.0 ppm were also prepared from a stock solution of 2000 ppm SV internal standard mix. The standard mix contained acenaphthene d-10, naphthalene d-8, chrysene d-12, pyrene d-12, Phenanthrene d-10. The surrogate used for recovery was p-terphenyl d-14 (B/N surrogate mix, 4/89 SOW). This method was adapted from Essumang et al. (2012).

3.11 GC/MS analysis of samples

The USEPA method 8270 Selected Ion Monitoring (SIM) (USEPA, 1996) was adopted for the GC/MS analysis. A Shimadzu GCMS QP2020 system, having an Automatic Operation Controller (AOC) 20i auto-injector, was used for the analysis. The dimension of the capillary column was 30.0 m (length) × 0.25 mm (ID) × 0.25 µm (thickness) Rtx-5ms fused capillary column, with helium (purity: 99.9995) as the carrier gas.

3.11.1 GC operation conditions

The injection port temperature was set at 265.0 °C and the column oven temperature was initially set at 70.0 °C. Temperature programming was used for GC operations. Here the temperature was initially set at 70 °C and held for 2 minutes. It was then ramped at a rate of 20 °C/min to 90 °C and ramped again at 10 °C/min to 250 °C. The temperature was further ramped at 5.0 °C/min to 300 °C and held for 3 minutes. A total program time of 32 minutes was used and the injection volume was 1.0 µL. The linear velocity flow (42.3 cm/sec for a column flow of 1.33 mL/min, and a total flow of 8.7 mL/min) control mode was used.

3.11.2 MS operation conditions

The electron impact ionization source was used and quantitative data were collected using (SIM) mode with ≥ 2 ions monitored for each compound. The temperatures of the ion source and the interface were set at 230 °C and 280 °C respectively.

3.12 Analytical quality control

Method reagent blank spiked with internal standard (ISTD) and surrogates were first analysed for each batch of sample analysis. Using the GC/MS tuning mixture, manual tuning was conducted every 12 h in conformity to 8270 E/D criteria. Standards and samples were analysed in triplicate (n=3)

Recovery was done in two levels; using the surrogate for instrumental analysis recovery and native standard for method recovery. About 20% of the samples were spiked with 1.0 ppm native standards for the method efficiency (recovery) test. Equation 4 was used to calculate recovery.

$$Recovery = \left(\frac{spiked - unspked}{spiked}\right) \times 100\% \quad \text{..............Equation 4}$$

3.13 Statistical analysis and risk assessment

Parametric (independent t-test and one-way analysis of variance (ANOVA)) and nonparametric (Mann-Whitney U and Kruskal-Wallis tests) tests were computed at a significance level of 0.05. The Bonferroni correction for multiple tests was used to calculate the p values of the Kruskal-Wallis test.

These statistical tests were done to assess the level of significance of PAHs concentrations within the selected three coastal lagoons. Such evaluation aided in interpreting the levels, distributions, ecological, and health effects of PAHs, taking into account the seasonal and spatial variations. The concentrations of PAHs that were less than the detection limit (DL) were considered as DL/√2 (Ashayeri et al., 2018; Hornung & Reed, 1990) for the calculations. Ecological and behavioural studies often contain data that violate parametric tests' statistical assumptions (Kikvidze & Moya-Laraño, 2008; Peres-Neto & Olden, 2001). Therefore, differences in PAHs concentrations among sampling sites, seasons, and fish parts were evaluated to determine if the data met the assumptions of normality and homogeneity of variances. Shapiro–Wilk test for normality, with a significance level of 0.05 (Elie et al., 2015; Razali & Wah, 2011) and the Levene's test for equality of variances, with a significance level of 0.01 (Elie et al., 2015; Gastwirth, Gel, & Miao, 2009) were computed. Among the three media (water, sediment, and fish tissues), only the water samples from Brenu and Fosu Lagoons upon log conformed to the assumptions of normality and homogeneity of variances of parametric test.

Water, sediment, and tissues of fish were compared both within and across lagoons. For both Brenu and Fosu Lagoons, an independent t-test was used to compare the two months in which water samples were taken. However, for water samples from Benya Lagoon, a nonparametric test, Mann-Whitney U was used. In each test, the sampling period was the factor, defined as November 2018 and January 2019. Among the three coastal waters, the levels of the different compounds observed in the water were compared using Kruskal-Wallis test with the factors being type of PAHs, sampling period, and type of lagoon. The concentrations of PAHs in the water samples were expressed in µg/L.

Also, for the tissues of fish and sediment samples, Mann-Whitney U test was used to compute seasonal variations. Kruskal-Wallis test on the other hand, was used to establish spatial variations (for sediment) and the concentrations in different parts of the fish. This was computed for each lagoon. A similar statistical approach was also used to compare PAHs concentrations in sediment and fish tissues across the three lagoons. The factors were: type of PAH, sampling period, type of lagoons, and sections of lagoon (for the fish, the fourth factor was the parts of fish). The concentrations of PAHs in sediment and fish samples were expressed in µg/kg/dry weight (µg/kg/d.w.) and µg/kg/wet weight (µg/kg/w.w.) respectively.

The descriptive statistics for all the 16 USEPA priority PAH concentrations in each media were computed for all the lagoons under study. In considering each lagoon's coherent situation, where all the three media (water, sediment, and fish) were looked at, the robust tests of equality of means (Welch and Brown-Forsythe) were used at a significance value of 0.01. The SPSS statistics software (version 26) was applied for the statistical data analysis.

To identify possible PAHs sources in the samples (i.e., fish, sediments, and water), ratios between PAHs isomers were employed. The following isomeric ratios were used in this study: Ant/(Ant + Phe), Flu/(Flu + Pyr), Flu/Pyr, Phe/Ant, BaA/(BaA + Chr), InP/(InP + BghiP), ∑LMW/∑HMW, BaP/BP and ∑PAHs(4 rings)/∑PAHs(5-6 rings) (Ashayeri et al., 2018; Santos et al., 2017; Sun et al., 2017). Also, Ant/(Ant + Phe) and Flu/(Flu + Pyr), as well as Flu/Pyr and Phe/Ant were used for bivariate plots (Ashayeri et al., 2018; Santos et al., 2017; Sun et al., 2017; Souza, 2018, Tay & Biney). These ratios were used for only sediment and fish and not the water samples due to their affinity to PAHs. Sediment has an added advantage as a valuable long-term record of both natural and anthropogenic origin of PAH contamination, hence results obtained will aid in recognising likely PAHs origins in the studied lagoons.

In determining the total PAHs levels in the sediment of lagoons to water temperature, total organic carbon (TOC), pH, and salinity, descriptive statistics and linear regression models were computed to establish any relationship. Multiple linear regressions were computed to analyse four predictors (%TOC, temperature, pH, and salinity) on total PAHs in lagoon sediments. In this case, %TOC, temperature, pH, and salinity were the four independent variables, while total PAHs was the dependent variable. Such analysis was done for each lagoon.

3.13.1 Ecological risk assessment

Sediment Quality Guidelines (SQGs) were used to evaluate the potential ecological risk of PAHs in sediment samples. These are internationally recognised guidelines based on a set of large toxicity data for aquatic ecosystems (Li et al., 2015). The SQGs opted for in this study were the Effect Range-Low and Effect Range-Median values (ERL/ERM) and the Threshold Effect Levels or Probable Effect Levels (TEL/PEL). Effect of PAH on organisms is categorised based on PAH concentrations and their comparison with two benchmarks ERL (or TEL) and ERM (or PEL). In this case, when concentrations are below ERL or TEL, it is expected not to be dangerous to organisms. When above ERM or PEL, it is expected to regularly portray adverse effects. When concentrations are within the two benchmarks, they cause harm occasionally (He et al., 2014; Long et al., 1995; Sun et al., 2017; Zhang et al., 2016). Since SQGs do not account for mixture toxicity effects of environmental contaminants, mean ERM quotient (mERM-Q) was used to estimate the ecological risk of multiple toxic components (Long & MacDonald, 1998; Long et al., 1995). The coefficient is calculated according to:

$$\mathrm{mERM-Q} = \frac{\sum(\mathrm{Ci/ERMi})}{\mathrm{n}} \quad \text{……………………..Equation 5}$$

C*i* is the concentration of individual compounds, ERM*i* is the corresponding ERM value for compound (*i*), and n is the total number of compounds. According to Li et al. (2015) and Long, MacDonald, Severn, & Hong (2000), the four classification of mERM-Q varied from: ≤ 0.10 for no harmful biological effect; 0.11 – 0.50 for a potential adverse effect; 0.51 – 1.50 for a moderate adverse effect; > 1.50 for a significant harmful effect.

In determining the potential carcinogenic toxicity effect, the risk evaluation approach used was the Toxicity Equivalency Factors (TEFs) (see Table 2). These were used to estimate the toxic equivalent quotient (TEQ) with respect to BaP as carcinogenic PAHs equivalent (Ashayeri et al., 2018; Nisbet & LaGoy, 1992). This was done for fish, sediment, and water samples. The following equation thus computed the TEQ in the samples:

$$TEQ = \sum_i (Ci \times TEFi) \qquad \text{……………………Equation 6}$$

Where TEQ = toxic equivalent quotient, C*i* is concentration of individual PAHs, TEF*i* toxicity equivalency factor of compound (*i*) relative to BaP.

Also, the levels of potential ecological risk indicated by specific PAHs on aquatic biota were characterised by a risk quotient (RQ), which was also calculated as:

$$RQ_{NC_s} = \frac{C_{PAHs}}{NC_s} \qquad \text{………………………………Equation 7}$$

$$RQ_{MPC_s} = \frac{C_{PAHs}}{MPC_s} \qquad \text{……………………………….Equation 8}$$

Where the maximum permissible concentrations (MPCs) and the negligible concentrations (NCs) of PAHs in sediments and water were applied as first suggested by Kalf et al. (1997). NCs are 100 times MPCs (i.e. NCs =

MPCs/100). The NCs and MPCs of individual PAHs in the sediments were listed in Table 3. The RQ value of individual PAH was compared with reference quality values (Ashayeri et al., 2018; Liu, Liu, Chen, Wang, & Cao, 2013; Yu et al., 2015): $RQ_{NCs} < 1.0$ shows that individual PAHs might be of negligible risk; $RQ_{NCs} > 1.0$ shows moderate risk; $RQ_{MPCs} < 1.0$ shows moderate risk; $RQ_{MPCs} > 1.0$ shows significant risk.

In considering $RQ_{\sum PAHs}$, $RQ_{\sum PAHs(NCs)}$ and $RQ_{\sum PAHs(MPCs)}$ were used for the evaluation (Ashayeri et al., 2018; Kalf et al., 1997; Liu et al., 2013):

$$RQ_{\sum PAHs(NCs)} = \sum_{i=1}^{n=16} RQ_{i(NCs)} \quad \text{…………….Equation 9}$$

$$RQ_{\sum PAHs(MPCs)} = \sum_{i=1}^{n=16} RQ_{i(MPCs)} \quad \text{………….Equation 10}$$

The ecological risk classification for $RQ_{\sum PAHs}$ were as follows: $RQ_{\sum PAHs(NCs)} < 1$ shows that the sum of the PAHs exhibit negligible risk; $RQ_{\sum PAHs(NCs)} \geq 1$; < 800 shows moderate risk; $RQ_{\sum PAHs(MPCs)} < 1$ shows low to moderate risk; $RQ_{\sum PAHs(NCs)} \geq 800$ shows high risk; $RQ_{\sum PAHs(MPCs)} \geq 1$ shows high risk.

Table 3 - *NCs and MPCs of individual PAHs in sediments and water*

PAHs	Sediment (µg/kg)		Water (µg/L)	
	NCs	MPCs	NCs	MPCs
Naph	1.4	140	12.0	1200
Ace	1.2	120	0.7	70
Acp	1.2	120	0.7	70
F	1.2	120	0.7	70
Ant	1.2	120	0.7	70
Phe	5.1	510	3.0	300
Flu	26.0	2600	3.0	300
Pyr	1.2	120	0.7	70
Chr	107.0	10700	3.4	340
BaA	3.6	360	0.1	10
BkF	24.0	2400	0.4	40
BbF	3.6	360	0.1	10
BaP	27.0	2700	0.5	50
DahA	27.0	2700	0.5	50
IP	59.0	5900	0.4	40
BP	75.0	7500	0.3	30

3.13.2 Human health risk assessment

In estimating the carcinogenic risk from exposure to PAHs in water, sediment, and fish, the USEPA guidelines, as used by Cheung et al. (2007), was used in this work. By this method, BaP was used as a marker for the occurrence and effect of carcinogenic PAHs. The overall carcinogenic health risk from the measured PAHs was estimated based on the TEFs values (see Table 2) (Nisbet & LaGoy, 1992). In this work, Potency Equivalent Concentration (PEC) equals to the Toxic Equivalent Quantity (TEQ) of PAHs (Shi et al., 2016). The PEC (µg-TEQ/kg) of the 16 total PAHs in each fish sample was calculated with the following equation:

$PEC = \sum_{i=1}^{n}(C_i \times TEF_i)$ ……………………..Equation 11

Where C_i (µg/kg) is the concentration of an individual PAH compound, *i* in fish tissue and TEF*i* is the toxicity equivalence factor of this compound *i* relative to that of BaP. All TEF values were taken from Nisbet & LaGoy (1992).

To evaluate PAHs' potential risk for inhabitants' health, the Incremental Lifetime Cancer Risk (ILCR) model was used to calculate the risk of children and adults, mostly fishers exposed to PAHs through their interactions with water, sediment, and fish in these studied lagoons. They are exposed to these components through two main pathways: (1) ingestion and (2) dermal contact. Some people in these areas use the water for domestic activities and consume fish as part of their protein diet. The exposure dosage through ingestion or consumption (water and fish) and dermal (water and sediment) pathways were calculated using the following equations (USEPA, 2004):

$ILCR_{ingestion} = \frac{CS \times \left(CSF_{ingestion} \times \sqrt[3]{(BW/70)} \times IR_{water} \times EF \times ED\right)}{BW \times AT \times cf}$ ………Equation 12

$$ILCR_{ingestion} = \frac{CS\times\left(CSF_{ingestion}\times\sqrt[3]{(BW/70)}\times IR_{fish}\times EF\times ED\right)}{BW\times AT\times cf}$$ ………Equation 13

$$ILCR_{dermal} = \frac{CS\times\left(CSF_{dermal}\times\sqrt[3]{(BW/70)}\times SA\times AF\times ABS\times EF\times ED\right)}{BW\times AT\times cf}$$ ………Equation 14

Where: $ILCR_{ingestion}$ and $ILCR_{dermal}$ are the Incremental Lifetime Cancer Risk from ingestion and dermal contacts respectively; Carcinogenic state (CS - ng/g) is the TEQ from equation 6; For carcinogenic slope factor (CSF – mg/kg/day), this was based on the cancer ability of BaP: 7.3 and 25 for CSF ingestion and dermal respectively (Knafla, Phillipps, Brecher, Petrovic, & Richardson, 2006; Liu et al., 2007); Body weight (BW – kg) was set at an average of 70 for adults and 15 for children (Ashayeri et al., 2018; USEPA, 2004; Yu et al., 2015); Intake rate of water or fish (IR_{water} or IR_{fish}) was set at 0.0001 kg/day (adults) and 0.00005 kg/day for children (Yu et al., 2015) and 78 g/capita/day (adults) and 39 g/capita/day for children (Bandowe et al., 2014) respectively; SA was the dermal surface area exposure (5000 cm^2/day – adults) and (1700 cm^2/day – children); Average life span (AT) was set at 70 years; Exposure frequency (EF) and exposure duration (ED) were set at 170 days/year and 30 years respectively; cf is the conversion factor which was 10^6; AF is the dermal adherence factor: 0.00001 kg/cm (adults) and 0.00003 kg/cm (children); dermal adsorption fraction (ABS – unit less) was 0.1 (Essumang et al., 2009).

The total risks were the sum of ILCRs of individual compounds in terms of direct ingestion and dermal contact. When an ILCR is between 10^{-6} and 10^{-4} indicates a potential risk (Essumang et al., 2009).

3.13.3 Sediment remediation

Analysis of Variance (ANOVA; One-Way and Two-Way using Minitab 19 Statistical Software) was used to deduce statistical differences between sample groups at 95% confidence level (α = 0.05). Tukey's Pairwise and Simultaneous Comparisons were used to identify specific differences among means. Anderson-Darling's test was used for normality testing, while and homogeneity of variances was subjected to Cochran's test. Data was Log_{10} transformed as it did not conform to normality. The factors were: type of charcoal amendment (0%, 1%, 3%, and 6%) and media (water and sediment). The response was PAHs concentrations.

3.14 Chapter Summary

The materials and methods used to execute the research, as well as the study locations have been described in detail in this Chapter. Statistical analytical tools and software employed to make inferences have also been indicated.

CHAPTER FOUR

RESULTS

The purpose of this study was to provide the current trend of PAH distributions in such aquatic ecosystems and their potential human health risks as the available data on PAHs in some of these coastal waters are almost a decade old (Armah, Luginaah, et al., 2012; Essumang, 2010; Gilbert et al., 2006). The results presented in this section focussed on residual levels of PAHs in water, sediment and fish (*Sarotherodon melanotheron*).

4.1 Quality control results

The six-point calibration curve was linear between 1.0 to 500.0 ppb and the coefficient of determination, $R^2 > 0.99$. These results are indications of very good linearity in an analytical response of the method used. The RF percent relative standard deviation (RF %RSD) was < 15% (Table 4). The recovery of the results was between 77 - 110% for the native spiked samples and that of the surrogate was between 83 - 101%. The recovery results for both instrumental and method efficiency were within the acceptable range for the method 8270 used. The LOD and LOQ of the sixteen detected PAHs are presented in Table 4. The selected ion monitoring chromatogram showing the analysed PAHs have been presented in Appendix A1.

Table 4 - *Quality assurance/ quality control*

Compound name	*Ret. time/min	LOD/ µg/kg	LOQ/ µg/kg	RF %RSD	R^2	Curve
Naphthalene	5.82	0.025	0.082	7.77	0.999	Linear
Acenaphthylene	9.44	0.010	0.034	14.29	0.997	Linear
Acenaphthene	9.86	0.006	0.018	11.54	0.999	Linear
Fluorene	11.11	0.001	0.004	7.66	0.998	Linear
Anthracene	13.52	0.049	0.162	9.43	1.000	Linear
Phenanthrene	13.36	0.044	0.146	14.87	0.998	Quadratic
Fluoranthene	16.20	0.054	0.181	7.92	0.998	Linear
Pyrene	16.71	0.019	0.063	11.27	0.999	Quadratic
Chrysene	19.69	0.041	0.138	6.25	1.000	Linear
Benz[a] anthracene	19.61	0.025	0.084	10.71	0.999	Linear
Benzo[k] Fluoranthene	22.08	0.010	0.034	12.26	0.998	Linear
Benzo[b] fluoranthene	22.02	0.008	0.026	6.80	1.000	Linear
Benzo[a] Pyrene	22.68	0.015	0.050	12.93	0.999	Linear
Dibenz[a,h]anthracene	25.30	0.046	0.155	8.17	0.999	Linear
Indeno [123-cd]pyrene	24.74	0.015	0.049	14.72	1.000	Linear
Benzo[g,h,i] perylene	25.76	0.012	0.041	11.91	1.000	Linear

*Ret. time; retention time
*LOD; Limit of detection
*LOQ; Limit of quantification

4.2 PAHs concentrations and distribution

4.2.1 Concentrations and distribution of PAHs in the Benya Lagoon

4.2.1.1 PAHs levels in water and sediment

All 16 PAHs under investigation (i.e. Naphthalene, Naph; Acenaphthylene, Ace; Acenaphthene, Acp; Fluorene, F; Phenanthrene, Phe; Anthracene, Ant; Fluoranthene, Flu; Pyrene, Pyr; Benz[a]anthracene, BaA; Chrysene, Chr; Benzo[b]fluoranthene, BbF; Benzo[k]fluoranthene, BkF; Benzo[a]pyrene, BaP; Indeno[1,2,3-cd]pyrene, IP; Dibenz[a,h]anthracene, DahA and Benzo[g,h,i]perylene, BP) were detected in the water samples, with the exception of Flu, DahA and BP (Table 5). Total PAHs (∑PAHs) ranged from 1.26 µg/L to 3.11 µg/L, with a mean concentration of 1.61 ± 0.8 µg/L. Naph and BkF recorded the highest and lowest concentrations measured respectively in the 2 months in which sampling was done. In November, the concentrations of Naph and BkF were 0.46 µg/L and 0.03 µg/L respectively, while in January, their respective concentrations were 0.18 µg/L and 0.02 µg/L (Figure 4). Using a nonparametric (Mann-Whitney U test) test, there was no significant difference for the recorded concentrations for both months. High molecular weight (HMW) PAHs, which include potential carcinogens such as BaA, Chr, BbF, BkF, BaP and IP (USEPA), ranged from 0.02 µg/L (BkF) to 0.12 µg/L (IP) in concentrations. The predominant PAHs encountered in the water samples were of low molecular weight (LMW) with the 3-ring structures dominating. Three-ring (3-ring) structures accounted for ≈44% of the total PAHs detected and the 6-ring component had the lowest percentage of ≈7 (Figure 5).

Table 5 - *Basic descriptive statistics for the PAHs concentrations in water and sediment (in dry weight) of Benya Lagoon*

	Concentration in water (µg/L)						Concentration in sediment (µg/kg)					
Compound	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis
Naph	0.144	0.870	0.319	0.275	2.271	5.303	1.555	65.970	18.481	11.484	1.481	2.475
Ace	0.089	0.181	0.106	0.037	2.442	5.971	0.061	28.943	12.094	9.097	0.063	-1.132
Acp	0.085	0.313	0.145	0.084	2.297	5.473	0.050	26.520	9.677	7.085	0.659	0.211
F	0.148	0.301	0.175	0.062	2.447	5.990	0.157	37.035	19.264	12.717	-0.232	-1.479
Ant	0.101	0.243	0.135	0.054	2.310	5.467	BDL	38.440	14.637	8.183	-0.038	-0.274
Phe	0.082	0.169	0.102	0.034	2.372	5.708	BDL	26.445	11.769	7.345	-0.265	-1.260
Flu	BDL	BDL	BDL	-	-	-	0.550	145.390	18.832	20.524	2.957	12.845
Pyr	0.087	0.180	0.105	0.037	2.425	5.903	1.451	122.900	26.121	17.196	2.304	8.842
Chr	0.087	0.178	0.103	0.036	2.447	5.991	3.215	69.840	27.625	15.665	0.906	0.147
BaA	0.046	0.093	0.054	0.019	2.446	5.988	0.800	591.650	37.032	81.732	5.321	30.071
BkF	0.046	0.098	0.057	0.020	2.367	5.682	2.985	68.895	21.855	15.309	1.308	1.111
BbF	0.021	0.044	0.025	0.009	2.440	5.964	0.080	82.580	16.600	16.458	1.662	3.045
BaP	0.082	0.168	0.109	0.039	1.049	-1.331	2.155	73.900	24.392	14.319	1.470	2.341
DahA	BDL	BDL	BDL	-	-	-	BDL	18.242	3.088	5.096	1.442	0.770
IP	0.087	0.177	0.103	0.036	2.443	5.974	BDL	52.890	16.883	9.700	0.591	1.449
BP	BDL	BDL	BDL	-	-	-	BDL	50.005	6.282	11.393	1.988	3.195
Total PAHs	1.260	3.110	1.607	0.737	2.418	5.879	35.820	824.460	284.633	146.239	1.583	3.244

*BDL- below detection limit

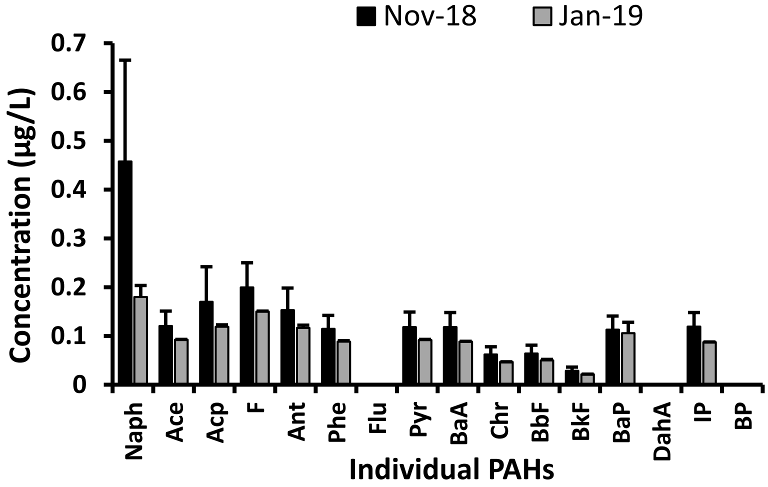

Figure 4: Concentrations of individual PAHs in water samples of Benya Lagoon.

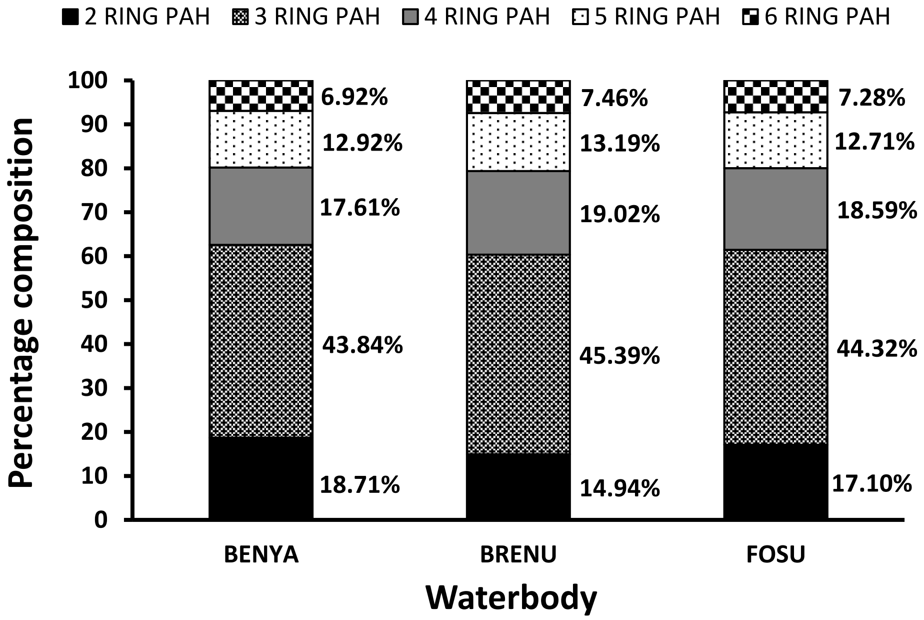

Figure 5: Percentage compositions of 2, 3, 4, 5 and 6 ringed PAH to total PAHs in water.

Unlike the water samples, there were significant variations of PAHs concentrations in sediment samples. Sediment sampled from Benya Lagoon had a ∑16PAHs varying from 35.82 to 824.46 µg/kg, with a mean concentration of 284.63±146.24 µg/kg. BaA was the highest mean concentration of 37.03±81.73

µg/kg (Table 5). Appendix A2 provides the descriptive statistics of the 3 different stations considered in Benya Lagoon. Stations 1 (BST1), 2 (BST2), and 3 (BST3) had a ∑16PAHs varying from 35.82 to 824.46 µg/kg, 89.43 to 807.53 µg/kg and 43.56 to 501.21 µg/kg respectively. Their mean concentrations were 282.62±149.53 µg/kg, 339.89±169.44 µg/kg, and 231.40±90.39 µg/kg respectively. Among the individual compounds, Chr was detected in all the stations to be one of the highest detected compounds. The mean concentrations of Chr were 25.01±13.11, 32.92±18.43, and 24.94±13.94 µg/kg at BST1, BST2 and BST3 respectively. PAHs recorded in sediment samples were predominantly HMW compounds. The 4-ring structures dominated both BST1 and BST2, while 3-ring structures dominated BST3 (Figure 6).

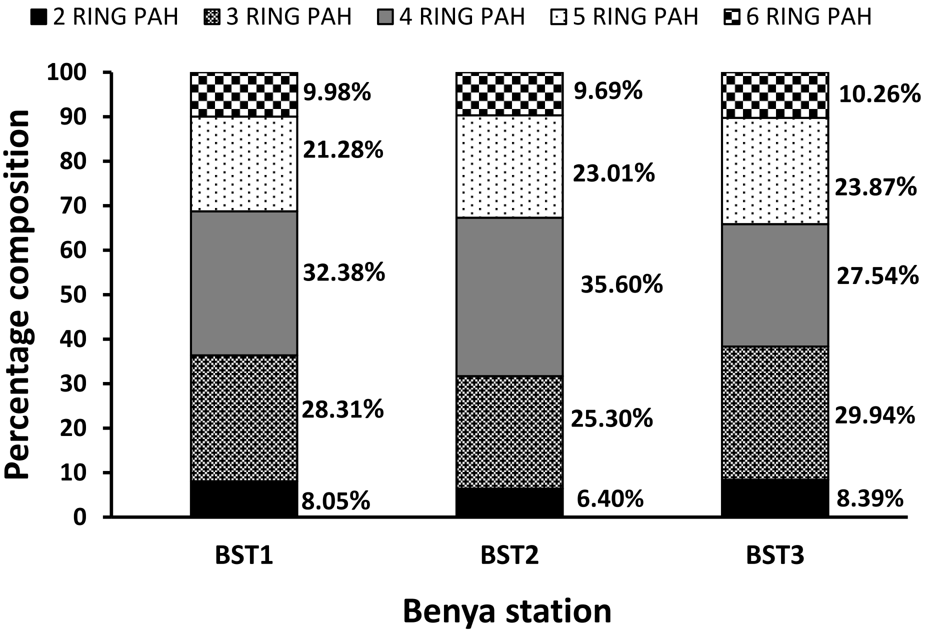

Figure 6: Percentage compositions of 2, 3, 4, 5 and 6 ringed structures to total PAHs in Benya sediment.

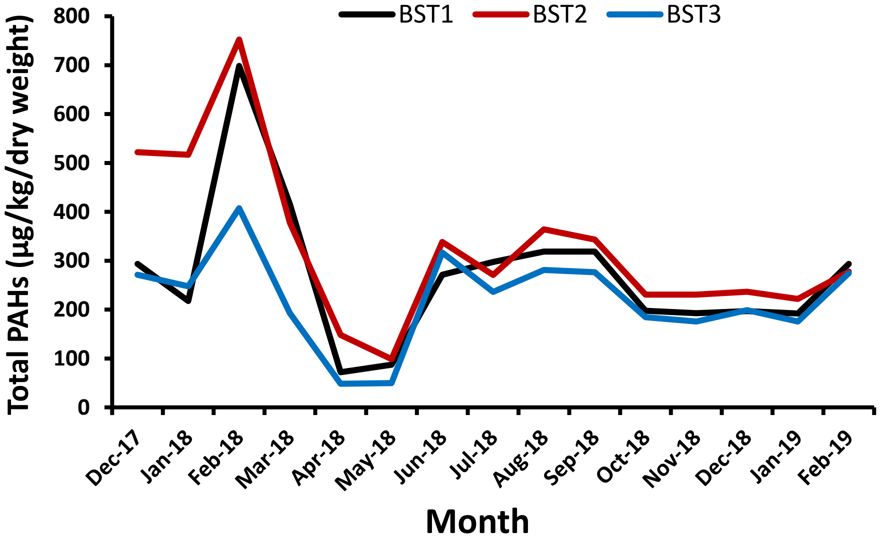

Figure 7: Monthly variations in total PAHs in the sediment of Benya Lagoon.

Temporal trends for total PAH concentrations in the sediment were similar at all the sampling sites (Figure 7). The highest total PAH concentrations were recorded in February 2018 at all three stations (BST1, BST2 and BST3) as 698.50, 752.43 and 407.35 µg/kg respectively. However, the lowest concentrations were observed in April at BST1 and BST3 (71.89 and 48.54 µg/kg respectively), and in May for BST2 (98.93 µg/kg). Figure 8 on the other hand, depicted the monthly distribution of individual PAHs in the sediment for the dry season, while Figure 9 represented that of the wet season.

In applying the Mann-Whitney U test, the total PAHs and individual PAHs (except for Pyr) were significantly different ($p < 0.05$) when the two seasons were compared (Appendix B1). It was noted that the mean rank for the LMW compounds (Naph, Ace, Acp, F, Ant and Phe) were significantly lower ($p < 0.05$) in the dry season than in the wet season and vice versa. The total PAHs were however significantly higher ($p < 0.05$) in the dry season than in the wet season. The mean concentrations for the dry season ranged from 9.19±7.62

µg/kg to 57.46±107.43 µg/kg, and that of the wet season ranged from 0.22±1.06 µg/kg to 24.89±9.56 µg/kg for the individual PAHs. For the total PAHs, the mean concentrations for the dry and wet seasons were 332.49±166.91 and 228.80±91.18 µg/kg respectively. Also, the Kruskal-Wallis test was computed to estimate the differences in PAH concentrations in the sediment at the 3 sampling stations. The total PAHs, together with Flu, Pyr, Chr and BbF were noted to be significantly different ($p < 0.05$) among the 3 stations (Table 6). The concentrations of Flu and Pyr at BST3 were significantly lower ($p < 0.01$) than their concentrations at both BST1 and BST2. For Chr, BbF and ∑PAHs, their concentrations at BST3 was significantly lower ($p < 0.05$) than their concentrations at BST2.

Table 6 - *Kruskal–Wallis nonparametric test showing differences within sampling points*

Compound	Comparison	Test Statistic	P-value [a]
Flu	BST3-BST1	-29.923	0.000
	BST3-BST2	-41.462	0.000
	BST1-BST2	11.538	0.399
Pyr	BST3-BST1	-27.564	0.001
	BST3-BST2	-38.282	0.000
	BST1-BST2	-10.718	0.489
Chr	BST3-BST1	-5.205	1.000
	BST3-BST2	-18.795	0.043
	BST1-BST2	-13.590	0.231
BbF	BST3-BST1	-5.282	1.000
	BST3-BST2	-23.333	0.007
	BST1-BST2	-18.051	0.056
Total PAHs	BST3-BST1	-11.769	0.376
	BST3-BST2	-26.308	0.002
	BST1-BST2	-14.538	0.175

*[a] p values have been adjusted by the Bonferroni correction for multiple tests.

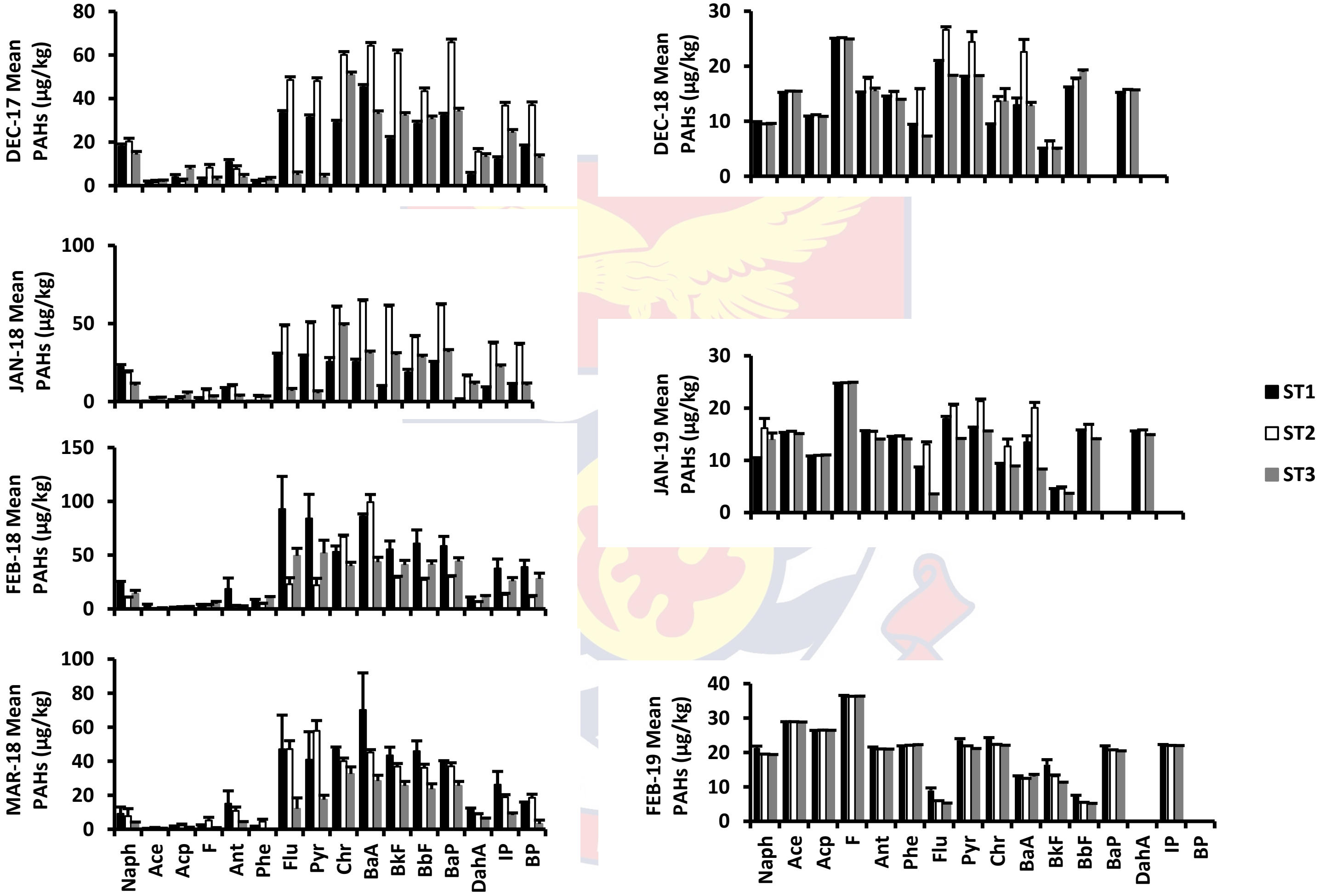

Figure 8: Monthly mean concentrations of individual PAHs in the sediment of Benya Lagoon for the dry season.

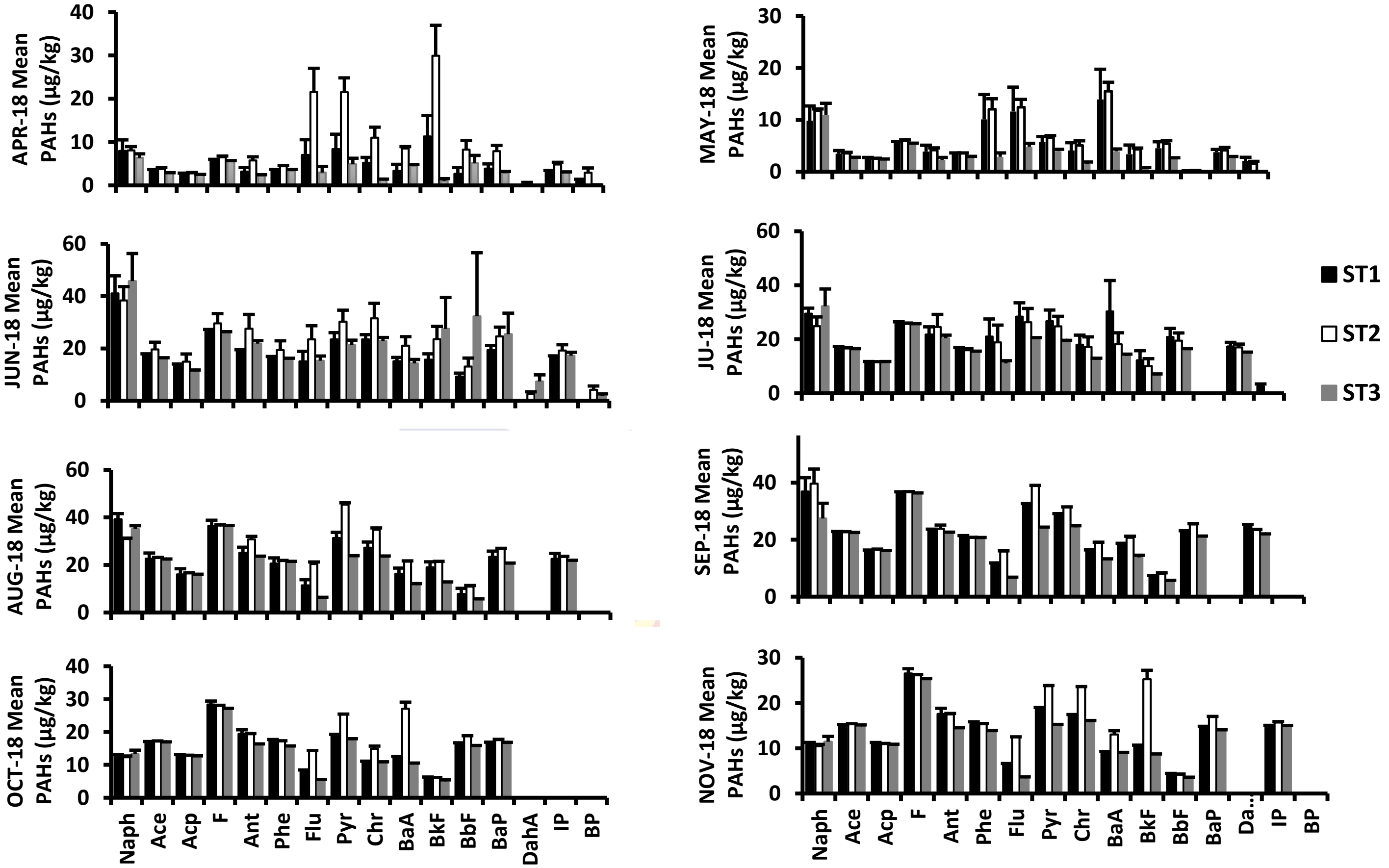

Figure 9: Monthly mean concentrations of individual PAHs in the sediment of Benya Lagoon for the wet season.

4.2.2 PAHs levels in fish parts

Descriptive statistics for the concentrations of individual PAHs in fish parts have been elaborated in Appendix A3. From the results, there were significant variations of PAHs concentrations in fish samples. Benya Fish Fillet (BFF), Benya Fish Visceral (BFV) and Benya Fish Head (BFH) respectively had a ∑16PAHs ranging from 173.04 to 433.29 µg/kg, 300.80 to 3726.16 µg/kg, and 178.41 to 553.61 µg/kg. The ∑16PAHs in the visceral was about twice the concentration in the head and thrice that of the fillet. The mean concentrations were 277.49±57.29 µg/kg (BFF), 745.41±957.22 µg/kg (BFV), and 385.04±94.74 µg/kg (BFH). Pyr (in both BFV and BFH) and F (in BFF) were the highest compounds detected. Their mean concentrations were 30.10±15.45 µg/kg (F), 160.42±392.49 µg/kg (Pyr), and 78.15±56.22 µg/kg (Pyr) for BFF, BFV and BFH respectively. All the 16 PAH compounds were detected in all the fish parts. Out of these, approximately 23%, 27% and 41% contributed to the presence of carcinogenic compounds considering the visceral, head and fillet respectively. In all the fish parts, BP recorded the least concentration of all measured compounds with mean concentrations of 0.51±1.08, 1.37±2.32 and 0.11±0.49 µg/kg in BFF, BFV and BFH respectively.

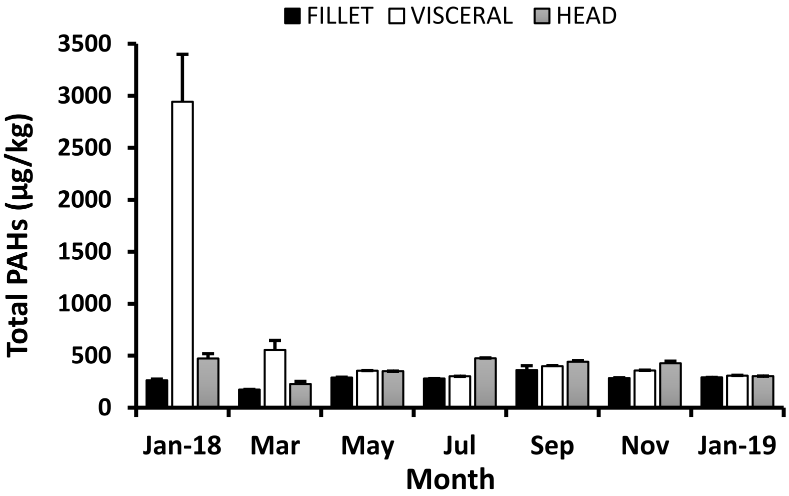

Figure 10: Monthly variations of total PAHs in fish parts from Benya Lagoon.

January, July and September 2018 were the months in which the highest total PAH were recorded in BFV (2942.82 µg/kg), BFH (473.75 µg/kg) and BFF (361.27 µg/kg) were obtained respectively (Figure 10). Furthermore, monthly mean concentrations for individual PAHs were analysed (Figure 11). From Figure 11, the mean concentrations of Pyr (1032.19±264.28 µg/kg), Phe (206.41±64.41 µg/kg) and F (38.91±0.27 µg/kg) were the highest recorded in the visceral part of fish samples during the dry season. These were recorded in January 2018, March 2018 and January 2019 respectively. January 2018 recorded the minimum number of compounds. During the wet season, Flu and Pyr were the dominating compounds and were detected in the fish head. Flu dominated in both May and July 2018 while Pyr dominated in both September and November 2018. In May and July, Flu concentrations in the fish head were 60.85±1.87 µg/kg and 135.11±3.26 µg/kg respectively. For September and November, Pyr concentrations in the fish head were 170.90±9.09 µk/kg and 131.04±2.09 µg/kg respectively.

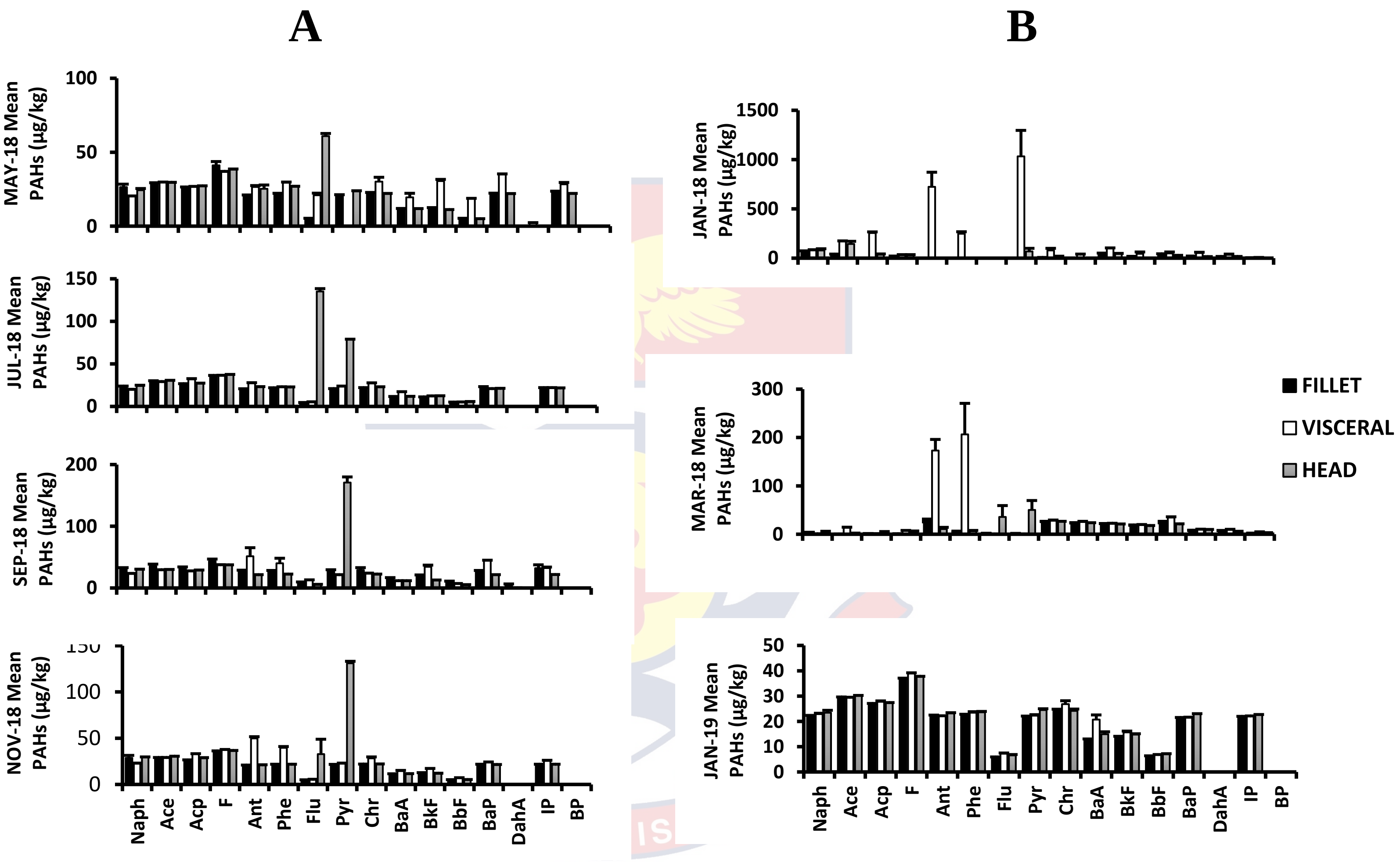

Figure 11: Seasonal variations of mean concentrations of individual PAHs in the fish parts of Benya Lagoon: **A** composed of wet season months and **B** composed of dry season months.

Based on the Mann-Whitney U test, there were significant seasonal variations ($p < 0.05$) in the concentrations of F, Flu, Chr, BbF, BkF, DahA, IP, and BP for the fish samples (Appendix B2). For Chr, BbF, BkF, DahA, and BP the mean ranks were significantly higher ($p < 0.05$) in the dry season than in the wet season. These were in the range of 39.30 µg/kg (BkF) to 44.00 µg/kg (BbF). F, Flu and DahA were also significantly higher ($p < 0.05$) in the wet season than the dry season. The mean ranks were 37.72 µg/kg (IP) to 39.58 µg/kg (Flu). The Kruskal-Wallis test was portrayed significant differences in concentrations of the fish parts. Except for Naph, Ace, F, DahA, and BP, both total PAHs and the other 11 PAH compounds had a significant difference ($p < 0.05$) in concentrations in at least one of the fish parts (Appendix B3). Among these, concentrations of total PAHs, Acp, Ant, Phe, Flu, Pyr, BaA, Chr, BbF, and BkF in BFF were significantly lower than their concentrations in BFV and BFH. The total non-carcinogenic PAHs (∑PAHs non-car) and total carcinogenic PAHs (∑PAHs car) were significantly higher in BFV ($p < 0.01$) than in BFH. It was also in BFV that the highest concentrations for ∑PAHs non-car (572.73 µg/kg), ∑PAHs car (171.28 µg/kg), and ∑16PAHs (745.41 µg/kg) were detected (Figure 12). The percentage compositions of the aromatic rings were as follows: HMW (4 to 6 rings) compounds dominated BFF (51%) and BFH (62%), while the LMW (2 to 3 rings) compounds dominated BFV (55%) as seen in Figure 13.

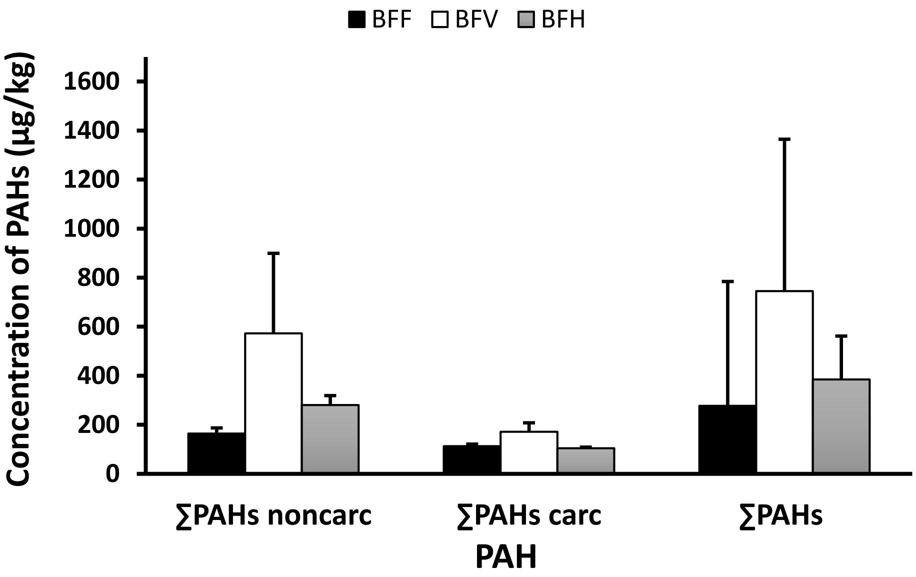

Figure 12: Non-carcinogenic, carcinogenic and total PAHs in fillet, visceral, and head of fish from Benya Lagoon.

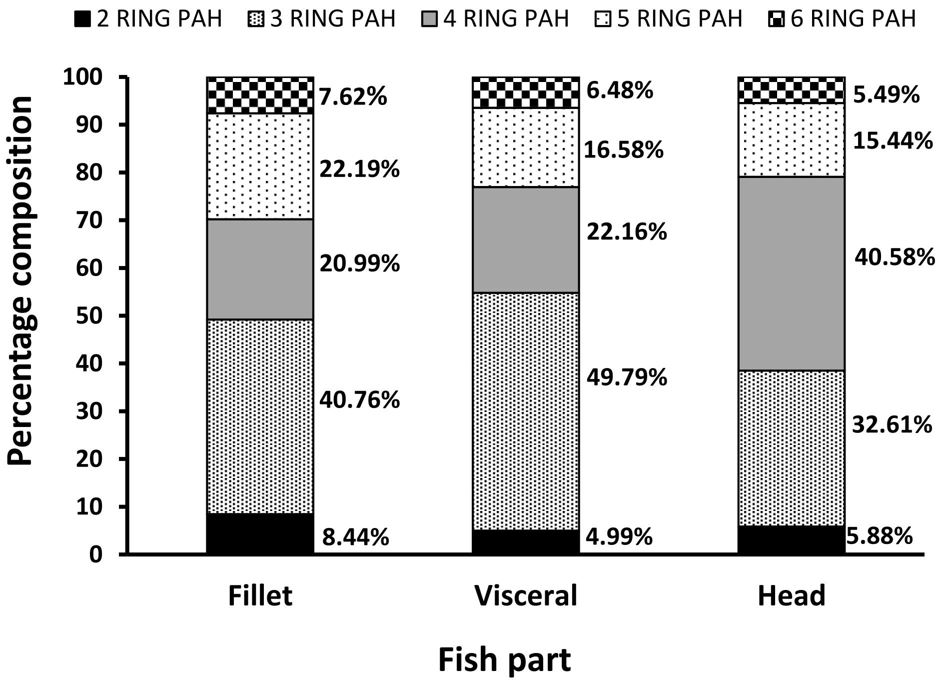

Figure 13: Percentage compositions of 2, 3, 4, 5, and 6-rings PAHs in fish parts.

Furthermore, the total PAHs in all the media (water, sediment and fish) were considered coherently (considering November 2018 and January 2019 samples). It was observed that the highest level of total PAHs was measured in the head of the fish (365.02 µg/kg), with water samples (1.54 µg/L) recording the least concentration. Generally, there was a significant decrease (p <0.01) in concentration in the water samples compared to the other two components (i.e. sediment and fish samples). Also, concentrations recorded in the fish head were significantly higher (p <0.01) than that in sediment and fish fillet (Figure 14).

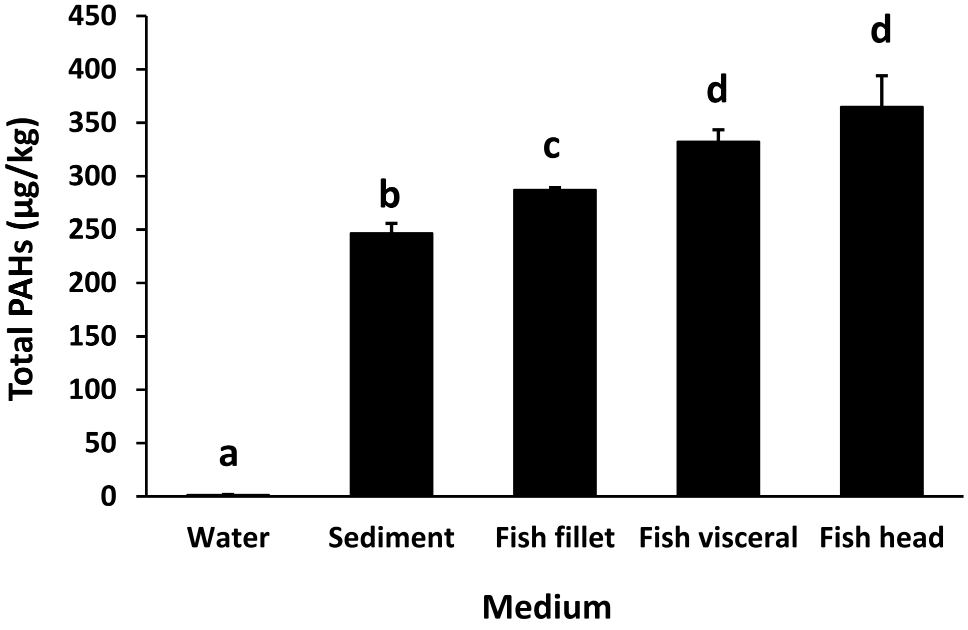

Figure 14: Total PAHs in different media of Benya Lagoon. Statistical differences among media (p< 0.01) are indicated by different letters.

4.2.2 Concentrations and distribution of PAHs in the Brenu Lagoon

4.2.2.1 PAHs levels in water and sediment

As observed in Benya Lagoon, all the 16 PAHs under investigation were recorded in all the water samples except for Flu, DaAh and BP. Results of descriptive statistics have been presented in Table 7, with total PAHs ranging from 1.17 µg/L to 1.43 µg/L and a mean concentration of 1.247± 0.10 µg/L. From Figure 15, Naph (0.23±0.06 µg/L) and F (0.15±0.001 µg/L) were the maximum concentrations for November 2018 and January 2019 respectively. However, BkF was the compound with the lowest concentration for both months. The mean concentrations of BkF in November 2018 and January 2019 were 0.02±0.001 µg/L and 0.03±0.01 µg/L respectively. The water samples were also dominated by LMW compounds. The 3-ring structures dominated with approximately 45% (Figure 5). In conducting an independent t-test, the concentrations of F, Pyr, and IP in November 2018 were significantly higher ($p < 0.05$) than their concentrations in January 2019 (Appendix B4).

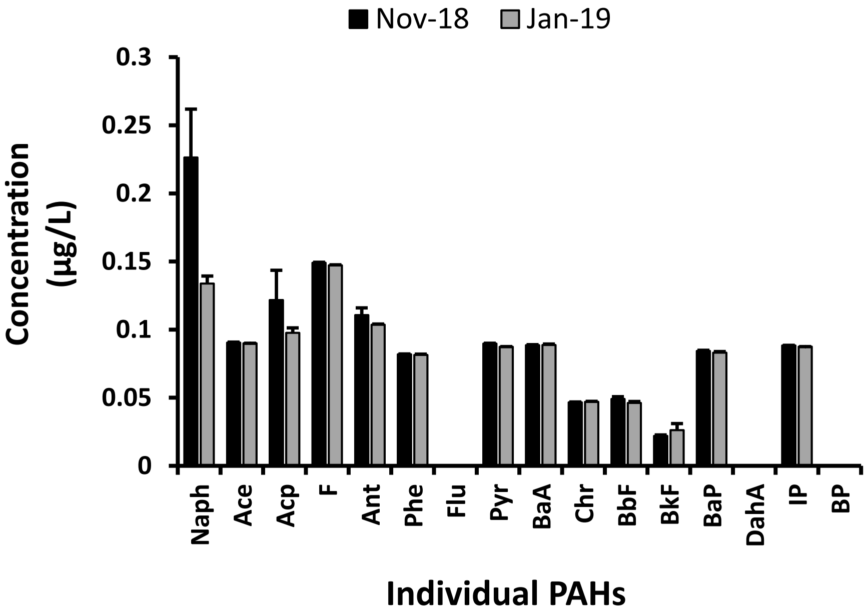

Figure 15: Concentrations of individual PAHs in water samples of Brenu Lagoon.

Table 7 - *Descriptive statistics for PAH concentration levels in water and sediment (dry weight) samples from Brenu Lagoon*

	Concentration in water (µg/L)						Concentration in sediment (µg/kg/dry weight)					
Compound	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis
Naph	0.123	0.295	0.180	0.064	1.397	1.762	BDL	59.670	10.811	11.170	2.177	6.483
Ace	0.090	0.091	0.090	0.001	0.811	-1.798	BDL	29.220	12.309	8.986	0.148	-1.163
Acp	0.083	0.159	0.110	0.028	1.369	1.684	BDL	26.745	8.985	8.173	0.539	-0.683
F	0.146	0.150	0.148	0.001	-0.485	-0.655	0.001	38.805	17.352	14.060	-0.017	-1.570
Ant	0.101	0.119	0.107	0.007	1.377	1.104	BDL	45.300	10.813	9.530	0.413	-0.143
Phe	0.080	0.082	0.082	0.001	-1.381	2.485	BDL	70.350	11.450	11.680	2.074	8.109
Flu	BDL	BDL	BDL	-	-	-	BDL	375.203	8.341	38.629	8.427	75.211
Pyr	0.087	0.090	0.089	0.001	0.069	-2.840	BDL	168.923	12.662	18.568	5.936	46.040
Chr	0.088	0.090	0.089	0.001	0.953	0.374	BDL	28.200	15.052	8.770	-0.561	-1.047
BaA	0.046	0.047	0.047	0.001	-0.219	-1.660	0.029	23.370	9.079	7.093	0.479	-0.546
BkF	0.044	0.052	0.048	0.003	0.857	2.560	BDL	37.500	11.297	7.061	1.456	2.208
BbF	0.020	0.036	0.024	0.006	2.243	5.211	BDL	19.530	5.775	6.132	1.257	0.301
BaP	0.082	0.085	0.084	0.001	-0.088	-1.460	BDL	22.950	13.615	7.530	-0.596	-1.187
DahA	BDL	BDL	BDL	-	-	-	BDL	56.400	3.157	10.289	4.786	22.538
IP	0.087	0.089	0.088	0.001	0.175	-2.491	BDL	55.650	13.470	10.715	1.762	5.224
BP	BDL	BDL	BDL	-	-	-	BDL	16.800	0.839	2.997	4.625	21.050
Total PAHs	1.17	1.430	1.247	0.101	1.613	2.273	20.810	796.110	165.010	116.652	1.688	6.933

*BDL- below detection limit

Sediment samples from Brenu Lagoon had total PAHs concentrations varying from 20.81 to 796.11 µg/kg, with a mean concentration of 165.01±116.65 µg/kg. Fluorene (F) was detected as the highest recorded compound with a mean concentration of 17.35±14.06 µg/kg (Table 7). Appendix A4 also elaborated on the descriptive statistics of the 3 different stations considered in this lagoon. Brenu station 1 (BRST1), Brenu station 2 (BRST2) and Brenu station 3 (BRST3) had a ∑16PAHs varying from 22.06 to 474.50 µg/kg, 21.52 to 283.80 µg/kg and 20.81 to 796.11 µg/kg respectively. Their mean concentrations were 166.06±106.73 µg/kg (BRST1), 158.61±88.68 µg/kg (BRST2) and 170.36±149.04 µg/kg (BRST3). The highest concentration of the compound detected at all the 3 stations was F, while BP was the lowest compound detected at all the stations. Also, the sediment was dominated by HMW compounds constituting about 57%, 60% and 58% for BRST1, BRST2 and BRST3 respectively (Figure 16).

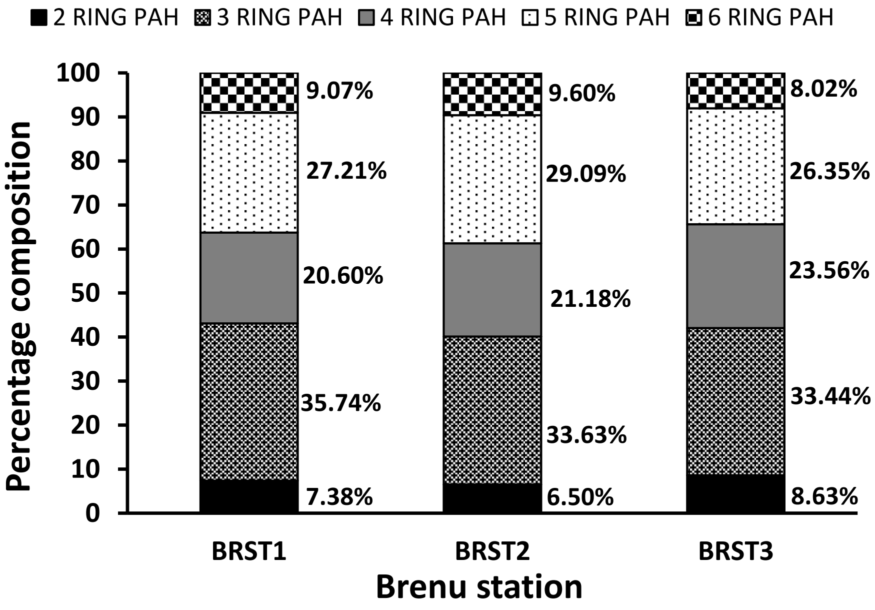

Figure 16: Percentage compositions of 2, 3, 4, 5 and 6 rings to total PAHs in Brenu sediment.

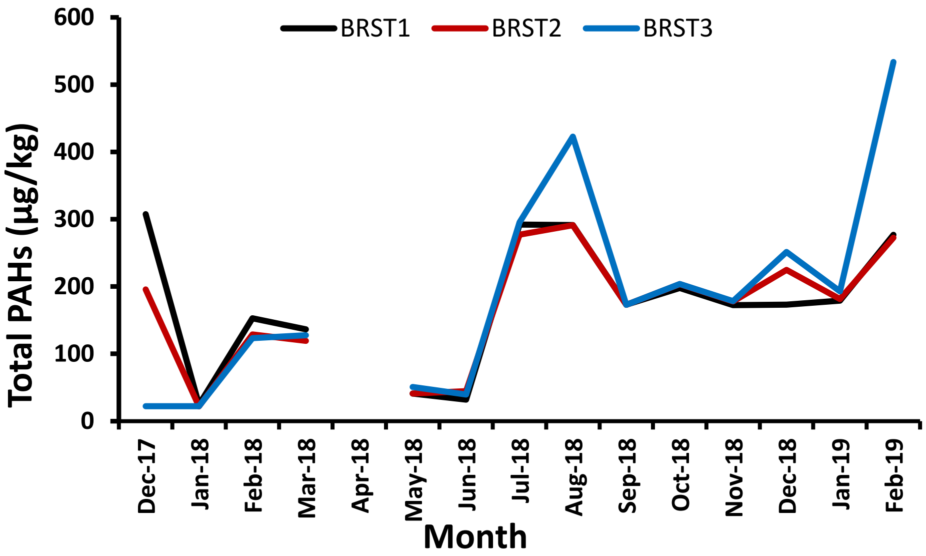

Figure 17: Monthly variations in total PAHs in the sediment of Brenu Lagoon.

Total PAH concentrations from all the three stations generally showed similar monthly trends (Figure 17). The highest concentrations for total PAHs at BRST1, BRST2 and BRST3 were respectively 307.563 µg/kg (in December 2017), 291.060 µg/kg (in August 2018), and 533.565 µg/kg (in February 2019). Besides, the dominant total PAH concentrations for both the wet and dry season were observed at BRST3. All the three stations had their lowest total PAHs recorded in January 2018. Furthermore, the monthly distribution of individual PAHs in the sediment have been presented in Figures 18 and 19. In Figure 18, one distinguishing observation was that unlike December 2017 to March 2018, all the measured compounds were found at all the 3 stations from December 2018 to February 2019. Generally, about 31% of the 16 PAHs were detected in all the 3 stations of the dry season. The highest mean concentration for the dry season was 189.88±106.99 µg/kg (Flu) in February 2019. Concerning the wet season (Figure 19), one similarity that was observed was either Flu or BbF were the least recorded compounds. Their concentrations ranged from 0.73 to 8.36 µg/kg and 0.16 to 7.29 µg/kg for Flu and BbF respectively.

The results from the Mann-Whitney U test indicated significant differences ($p < 0.05$) between the seasons for some PAHs in the sediment (Appendix B5). This included Naph, F, Ant, Phe, Flu, Pyr, BaA, BbF, BaP, DahA, and BP. All the LMW compounds (Naph, F, Ant, Phe), together with Flu and Pyr were significantly higher in the wet season than in the dry season. However, BaA, BbF, BaP, DahA, and BP were significantly higher in the dry season than in the wet season. In using the Kruskal-Wallis test there was no significant difference in the concentrations of the sediment considering both the individual compounds and the total PAHs.

4.2.2.2 PAHs levels in fish parts

All the 16 priority PAHs were detected in the 3 different fish parts, ranging from below detection limit (BDL) to 97.14 µg/kg (F), BDL to 411.57 µg/kg (BaA), and BDL to 322.99 µg/kg (Pyr) in the fillet, visceral, and head respectively (Appendix A5). The ∑16PAHs also ranged from 32.37 to 457.02 µg/kg, 176.80 to 577.37 µg/kg, and 65.38 to 845.85 µg/kg for Brenu Fish Fillet (BRFF), Brenu Fish Visceral (BRFV) and Brenu Fish Head (BRFH) respectively. Their mean concentrations were 284.60±116.50 µg/kg (BRFF), 309.79±90.21 µg/kg (BRFV), and 451.99±208.15 µg/kg BRFH). BP was the compound with the least concentration detected in both BRFF and BRFH. However, DahA was the least detected compound in BRFV. Their mean concentrations were 1.149±2.90 µg/kg (BRFF), 5.162±18.67 µg/kg (BRFH), and 2.392±3.90 µg/kg (BRFV). Among the carcinogenic compounds, BaA recorded the highest mean concentrations in all the fish parts with concentrations of 22.12±9.94 µg/kg (in fillet), 67.55±116.98 µg/kg (in visceral) and 22.84±10.63 µg/kg (in the head).

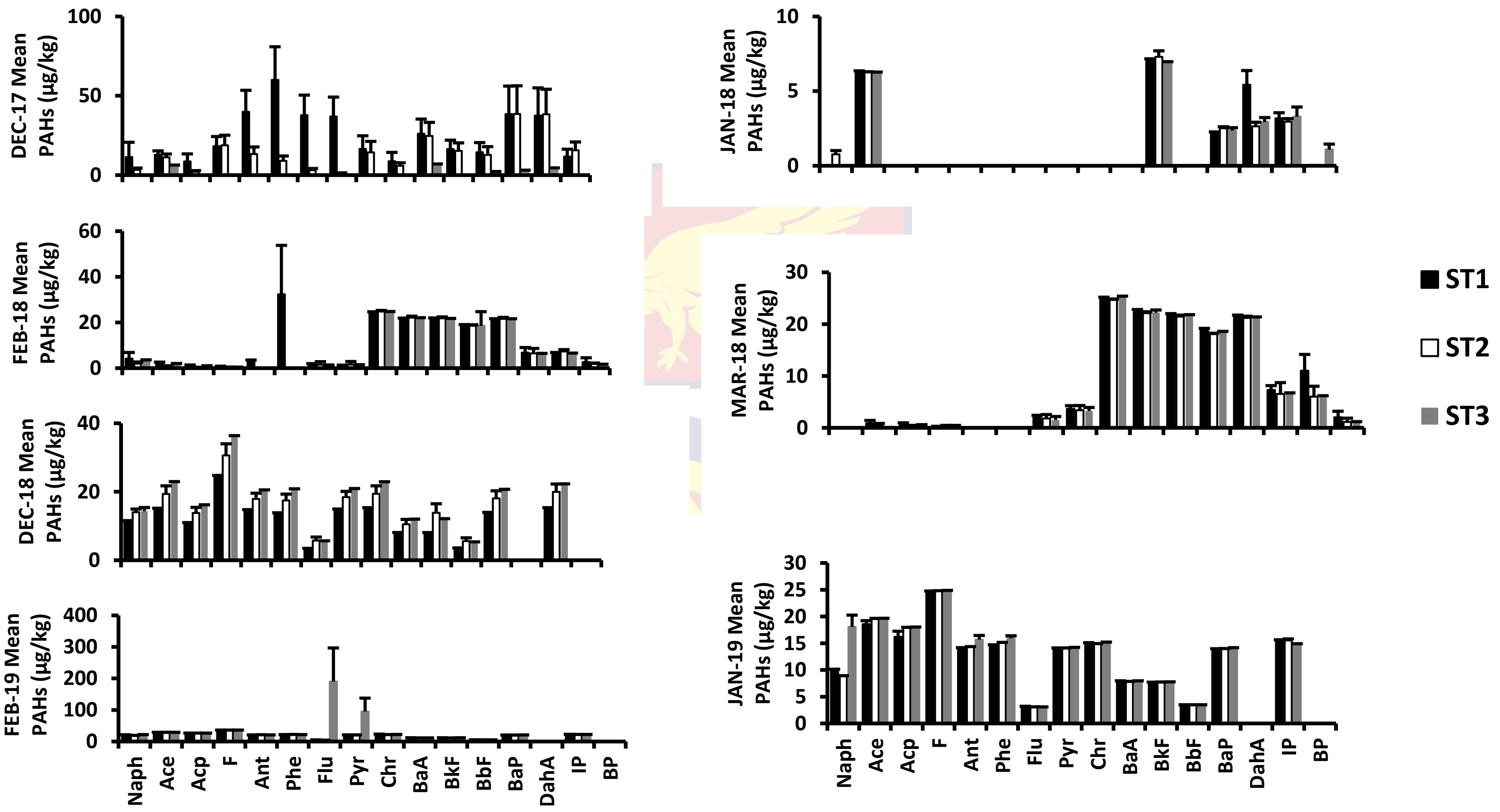

Figure 18: Monthly mean concentrations of individual PAHs in the sediment of Brenu Lagoon for the dry season.

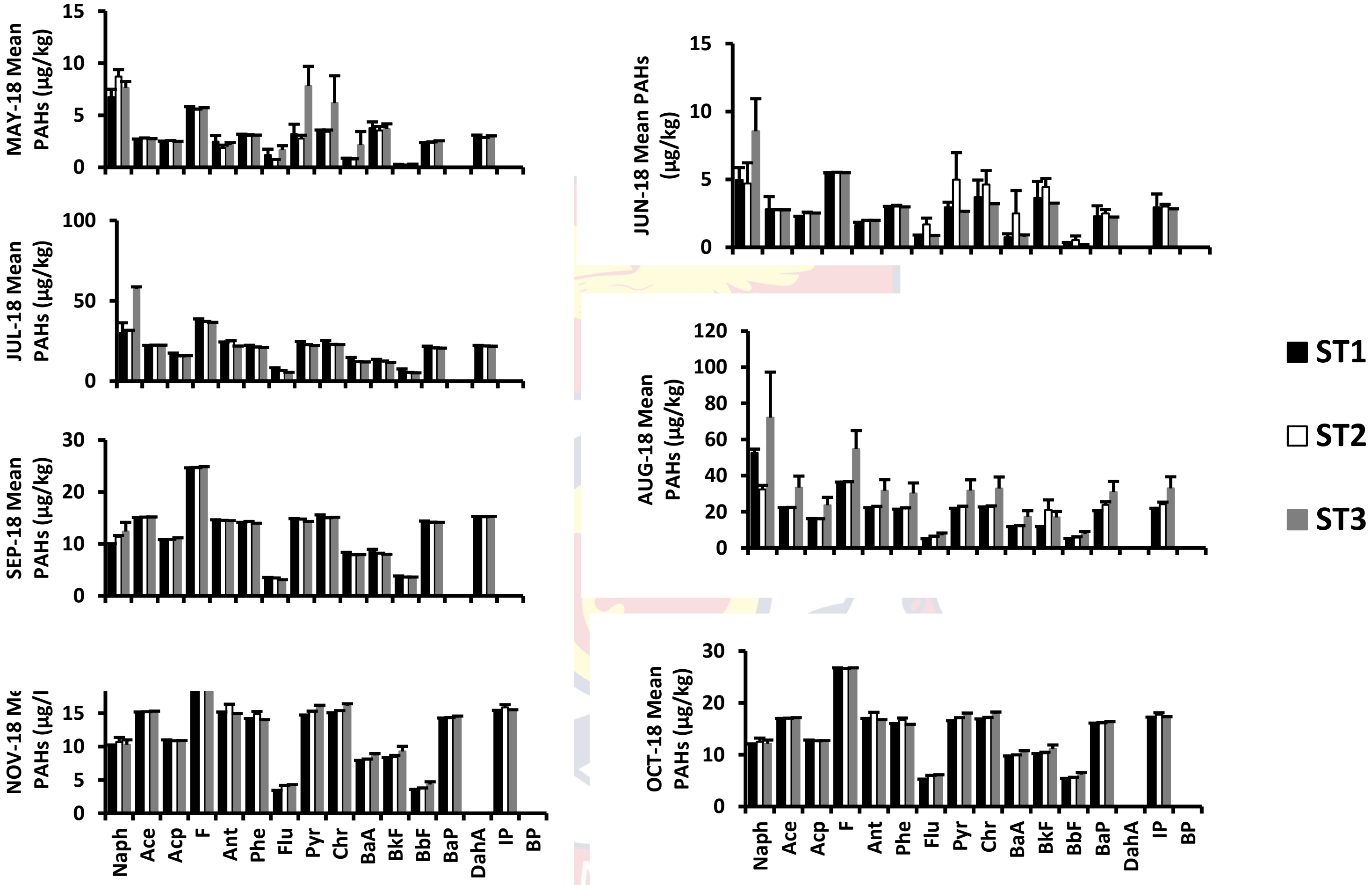

Figure 19: Monthly mean concentrations of individual PAHs in the sediment of Brenu Lagoon for the wet season.

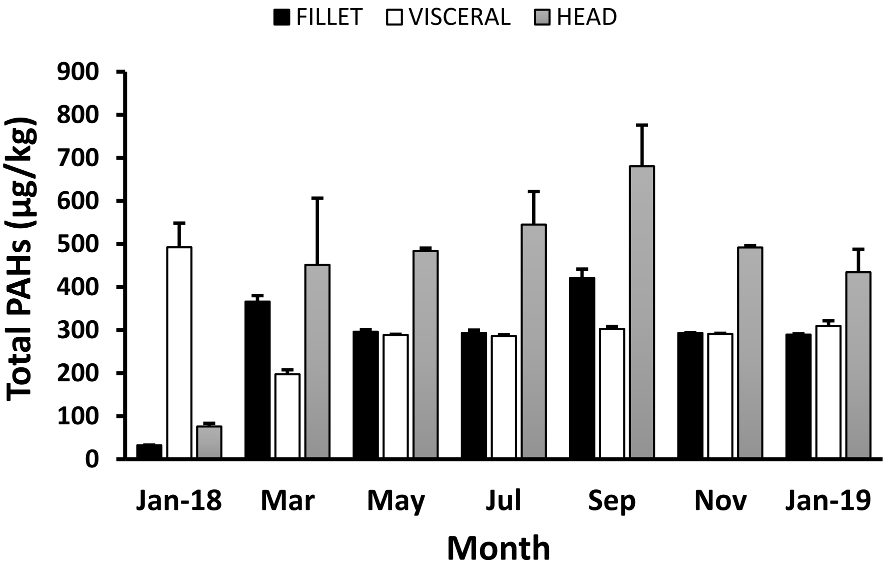

Figure 20: Monthly variations of total PAHs in fish parts from Brenu Lagoon.

From Figure 20, fish head recorded the highest monthly ∑PAH throughout the study period, except in January 2018 where the highest concentration was observed in the visceral part of the fish. Likewise, in the monthly distribution of the individual PAHs, the highest recorded concentrations were generally detected in the fish head (Figure 21). The highest mean concentration in the dry season was 337.75±54.28 µg/kg (Chr) in January 2018, while the highest mean concentration for the wet season was 119.34±2.05 µg/kg (Flu) in May 2018. BkF, BaP, DahA, and IP were the carcinogenic compounds detected in all the fish parts throughout the study period.

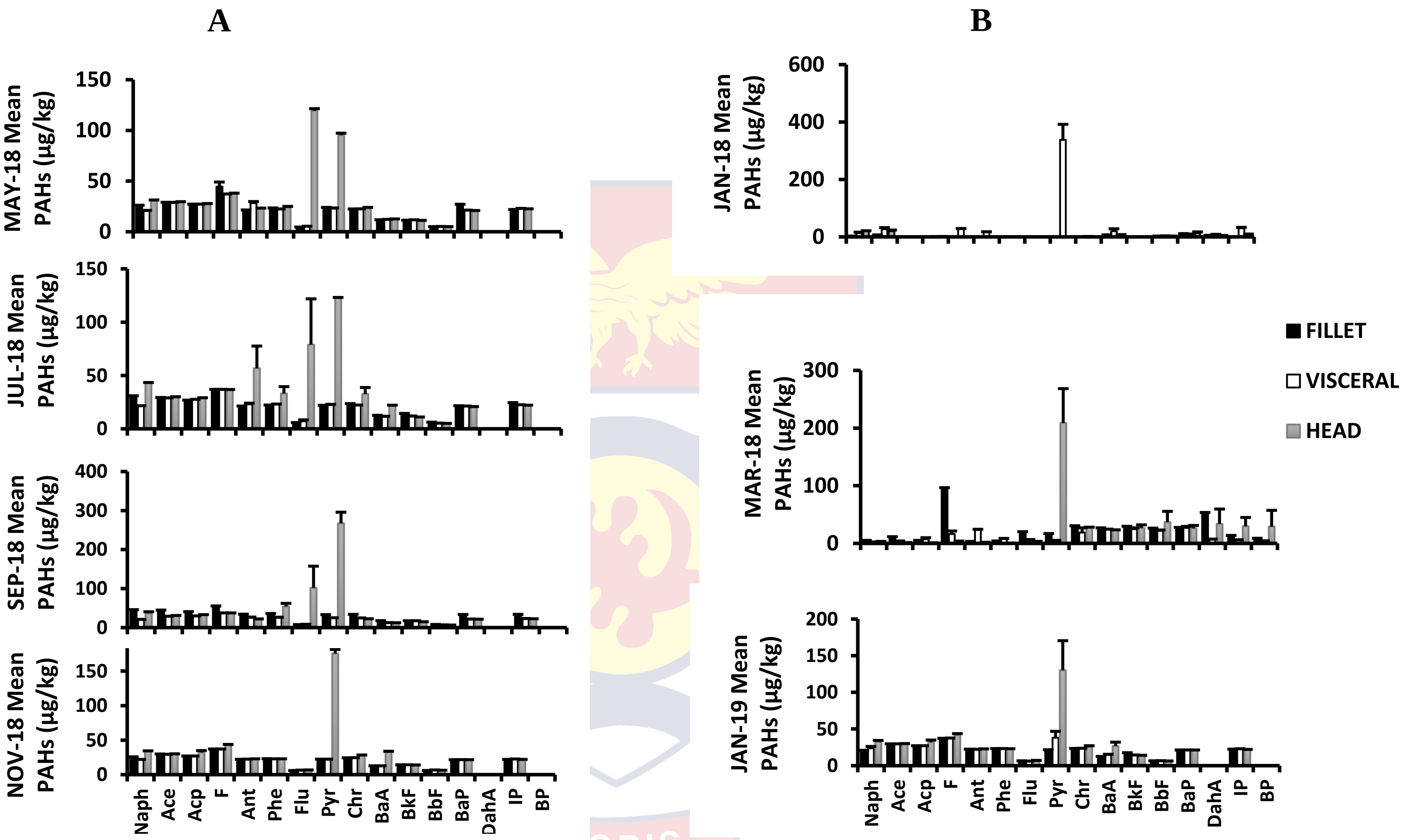

Figure 21: Seasonal variations of mean concentrations of individual PAHs in the fish parts of Brenu Lagoon: **A** composed of wet season months and **B** composed of dry season months.

A Mann-Whitney U test for the season showed all the LMW compounds (Naph, Ace, Acp, F, Ant, and Phe) and some of the HMW compounds (Flu, Pyr, BaP, DahA, IP, BP) to be significantly different in concentrations ($p < 0.05$) (Appendix B6) so far as the fish tissues were concerned. All the LMW compounds significantly higher ($p < 0.05$) in the wet season than in the dry season. The Kruskal-Wallis test also indicated that Naph, Ant, Pyr, BaP, and total PAHs were significantly different in at least one of the fish parts ($p < 0.01$) (Table 8). Naph, Pyr, and total PAHs in BRFH were significantly higher ($p < 0.05$) than that in BRFV. Also, BaP, Pyr, and total PAHs in BRFH were significantly higher ($p < 0.05$) than that in BRFF. For Ant, concentration in BRFV was significantly higher ($p < 0.05$) than in BRFF. Generally, the concentrations detected in BRFF were significantly the lowest ($p < 0.05$) compared with BRFH and BRFV.

Table 8 - *Kruskal–Wallis nonparametric test showing differences among fish parts from Brenu Lagoon*

Compound	Comparisons	Test Statistic	Adjusted Significance ^{a}
Naph	BRFV-BRFF	-9.095	0.324
	BRFV-BRFH	-19.905	0.001
	BRFF-BRFH	-10.810	0.168
Ant	BRFF-BRFH	-4.810	1.000
	BRFF-BRFV	-17.833	0.005
	BRFH-BRFV	-13.024	0.064
Pyr	BRFF-BRFV	-2.619	1.000
	BRFF-BRFH	-24.381	0.000
	BRFV-BRFH	-21.762	0.000
BaP	BRFH-BRFV	-10.786	0.170
	BRFH-BRFF	-16.500	0.011
	BRFV-BRFF	-5.714	0.937
Total PAHs	BRFV-BRFF	-0.667	1.000
	BRFV-BRFH	-17.905	0.005
	BRFF-BRFH	-17.238	0.007

*(a) significance values have been adjusted by the Bonferroni correction for multiple tests.

The composition of total non-carcinogenic PAHs (∑PAHs noncar), total carcinogenic PAHs (∑PAHs car), and total PAHs (∑16PAHs) were depicted in Figure 22. BRFH recorded the highest level of concentration for both ∑PAHs non-car (335.54 μg/kg) and ∑16PAHs (451.99 μg/kg), while BRFV recorded the highest ∑PAHs car (144.19 μg/kg). Furthermore, HMW compounds dominated each fish part, with fillet, visceral and head contributing about 57%, 56% and 69% respectively (Figure 23). Specifically, the 3-ring structures dominated both fillet (≈ 37%) and visceral (≈ 39%), while the 4-ring structures dominated the head (≈ 42%).

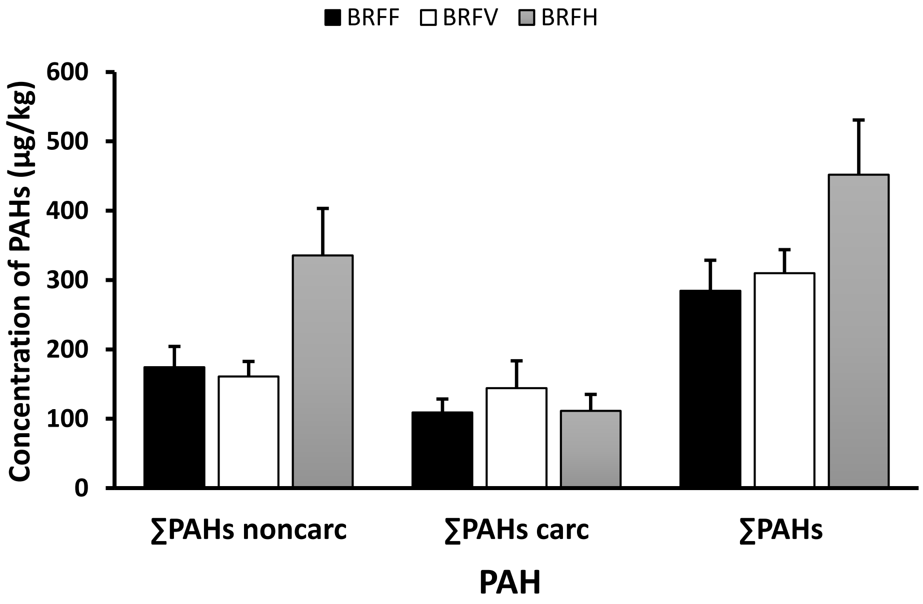

Figure 22: Non-carcinogenic, carcinogenic and Total PAHs in the fillet, visceral, and head of fish from Benya Lagoon.

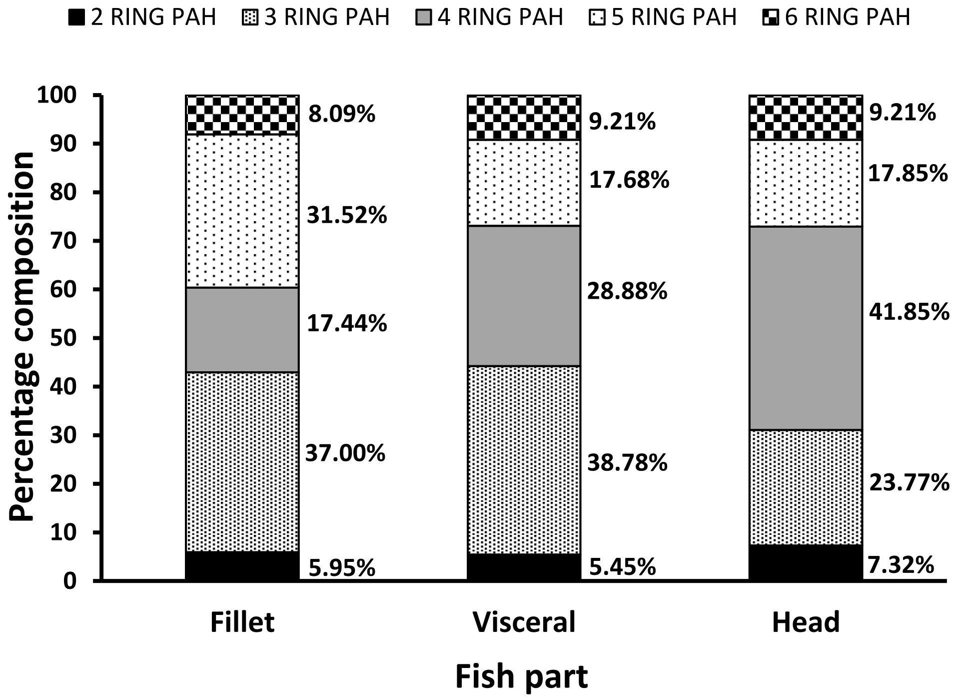

Figure 23: Percentage compositions of 2, 3, 4, 5 and 6-rings PAHs in fish parts.

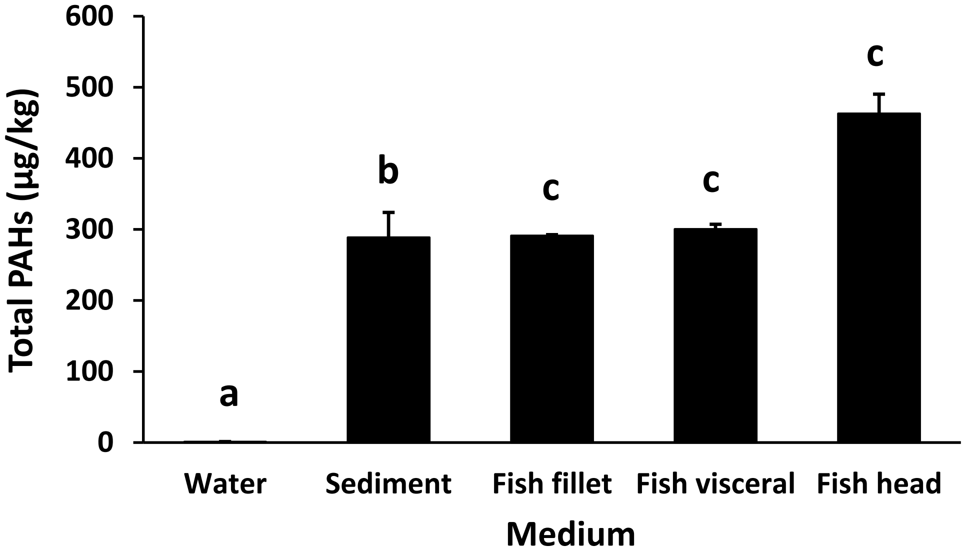

Figure 24: Total PAHs in different media of Brenu Lagoon. Statistical differences among media (p <0.01) are indicated by different letters.

Concerning the different media under study (November 2018 and January 2019 samples), the lowest and highest levels of total PAHs were recorded in the water column (1.18 µg/kg) and head of the fish (463.06 µg/kg) respectively (Figure 24). The ∑16PAHs concentration in the fish head was significantly higher than that in the water and sediment ($p < 0.01$).

4.2.3 Concentrations and distribution of PAHs in the Fosu Lagoon

4.2.3.1 PAHs levels in water and sediment

The results of basic descriptive statistics for the water samples are presented in Table 9. Water samples from Fosu Lagoon recorded ∑16PAHs ranging from 2.29 µg/L to 3.39 µg/L, with a mean concentration of 2.75± 0.40 µg/L. The mean concentration was within the highly polluted range {i.e. above 1.00 µg/L; Li et al. (2015) and Ashayeri et al. (2018)}.

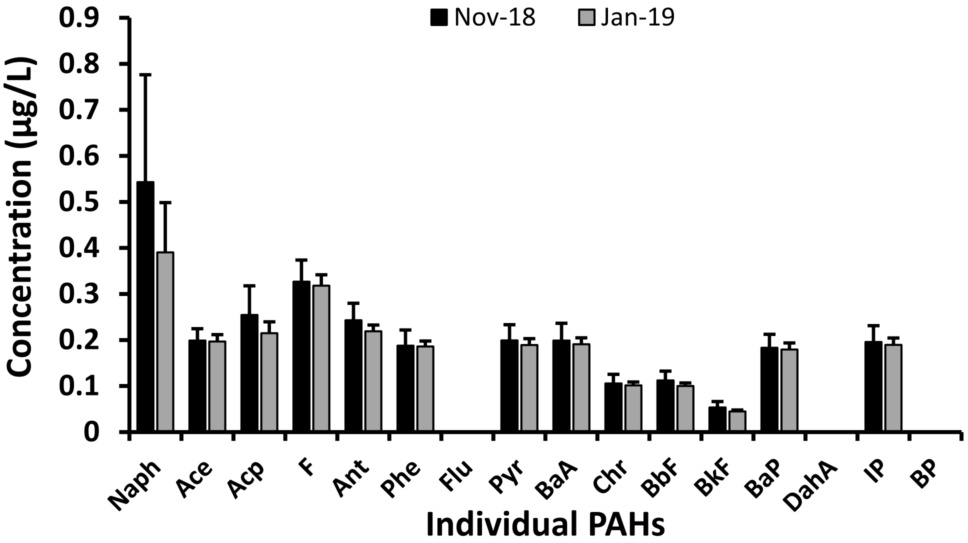

Figure 25: Concentrations of individual PAHs in water samples of Fosu Lagoon.

In Fosu Lagoon, there was no significant difference ($p < 0.05$) for both individual compounds and total PAHs so far as the two sampling months were concerned. Naph and BkF were respectively the highest and lowest compounds detected in the water column. The mean concentrations of Naph during November 2018 and January 2019 were 0.54± 0.23 µg/L and 0.39± 0.19 µg/L respectively. For that of BkF, the mean concentrations in November 2018 and January 2019 were 0.05±0.01 µg/L and 0.05±0.01 µg/L respectively (Figure 25). The LMW compounds (constituting about 61%) dominated in the water column (Figure 5).

Total PAHs of sediment samples from the Fosu Lagoon also ranged from 189.89 µg/kg to 24801.38 µg/kg, with a mean concentration of 728.30±2259.24 µg/kg (Table 9). The compound with the highest recorded concentration was Flu with a mean concentration of 100.48±374.69 µg/kg. Appendix A6 also elaborated on the descriptive statistics of the 3 different stations considered in this lagoon. The total mean concentrations at FST1, FST2, and FST3 were 1248.69± 3880.50 µg/kg, 561.13±227.11 µg/kg, and 375.09±191.18 µg/kg respectively. Flu, BkF, and Pyr were detected as the highest concentrations at FST1, FST2, and FST3 respectively.

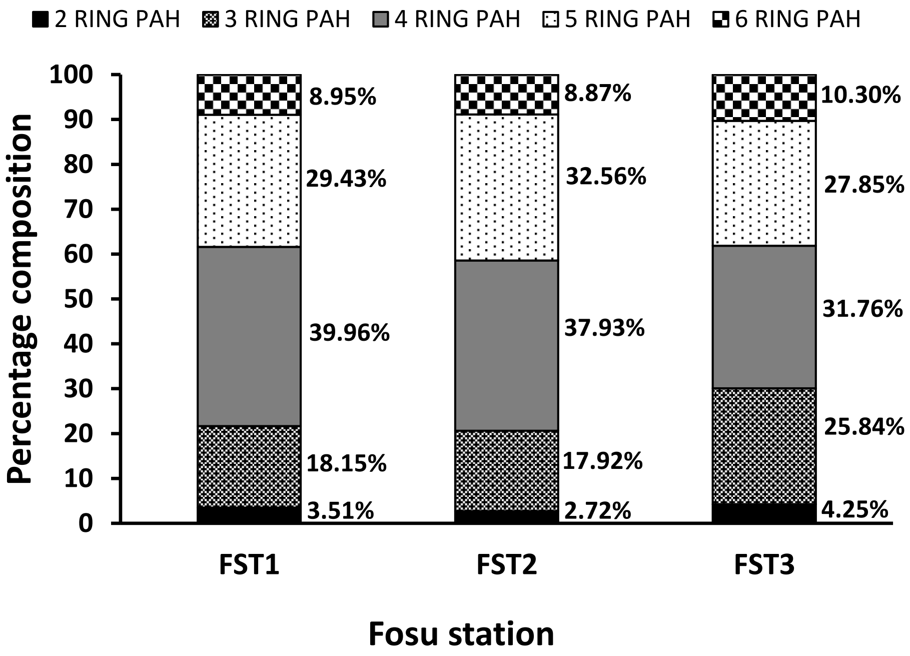

Figure 26: Percentage compositions of 2, 3, 4, 5, and 6 rings to total PAHs in the sediment of Fosu Lagoon.

Table 9 - *Basic descriptive statistics for the PAHs concentration levels in water and sediment (dry weight) of Fosu Lagoon*

	Concentration in water (μg/L)						Concentration in sediment (μg/kg/dry weight)					
Compound	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis
Naph	0.262	0.689	0.467	0.207	0.052	-3.103	BDL	136.575	17.646	19.459	4.149	21.440
Ace	0.181	0.229	0.198	0.023	0.954	-1.859	1.290	85.275	16.374	9.522	3.389	22.801
Acp	0.175	0.327	0.234	0.053	1.156	1.456	0.245	47.345	10.076	7.995	1.420	3.540
F	0.293	0.381	0.322	0.040	1.010	-1.501	0.865	51.690	22.584	11.383	-0.294	-0.351
Ant	0.199	0.280	0.231	0.031	0.696	-0.520	1.510	800.490	37.665	72.882	10.062	105.882
Phe	0.164	0.227	0.187	0.025	0.821	-0.586	BDL	180.835	18.607	16.871	7.771	74.703
Flu	BDL	BDL	BDL	-	-	-	14.565	4081.780	100.483	374.687	10.521	112.648
Pyr	0.174	0.238	0.194	0.027	1.201	-0.295	15.068	3955.425	95.710	362.974	10.546	112.994
Chr	0.177	0.242	0.195	0.028	1.260	-0.146	11.440	1669.005	54.457	152.222	10.465	111.819
BaA	0.092	0.129	0.103	0.015	1.221	-0.146	9.070	3212.055	67.279	294.524	10.675	114.922
BkF	0.089	0.135	0.106	0.016	1.262	1.852	17.460	2431.850	86.358	221.546	10.404	110.971
BbF	0.041	0.068	0.050	0.010	1.873	3.699	3.700	2549.790	52.222	234.595	10.583	113.550
BaP	0.163	0.217	0.181	0.024	1.017	-1.505	7.550	2118.180	58.280	193.404	10.595	113.744
DahA	BDL	BDL	BDL	-	-	-	BDL	237.070	12.418	28.697	4.681	31.866
IP	0.174	0.237	0.193	0.028	1.144	-0.822	8.560	1748.285	48.783	159.990	10.510	112.513
BP	BDL	BDL	BDL	-	-	-	BDL	1640.435	29.363	151.817	10.482	112.052
Total PAHs	2.290	3.390	2.750	0.404	0.508	-1.859	189.890	24801.380	728.304	2259.237	10.609	113.949

*BDL- below detection limit

The sediment from Fosu Lagoon was dominated by HMW compounds, with 4-ring structures being the most prominent at all the 3 stations. Four-ring (4-ring) compounds at FST1 represented about 40% of the total PAHs while that at FST2 and FST3 were 38% and 32% respectively (Figure 26).

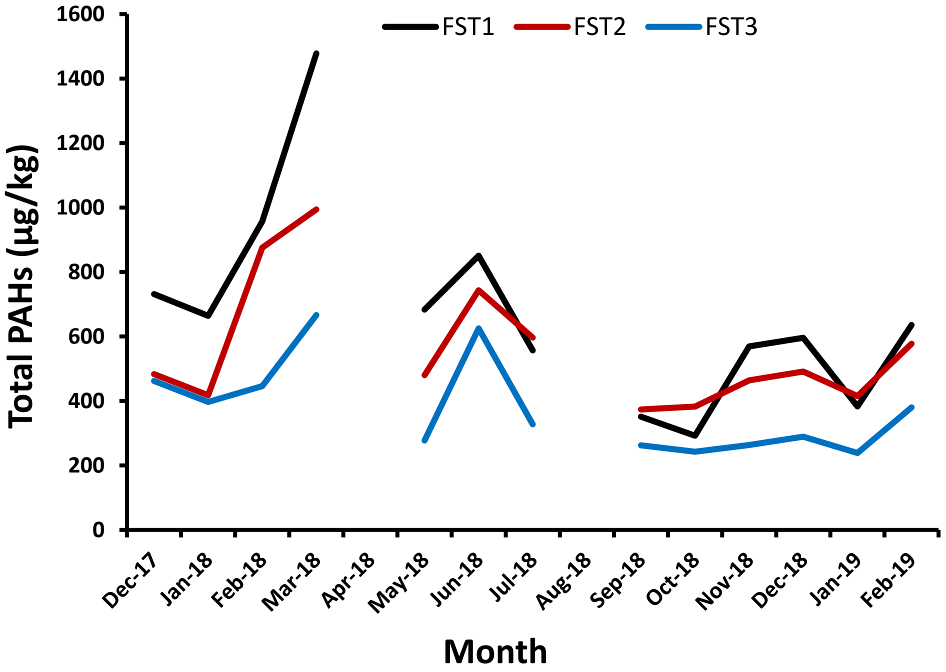

Figure 27: Monthly variations in total PAHs in the sediment of Fosu Lagoon.

The monthly trend for the spatial distribution of total PAHs has been presented in Figure 27. The highest total concentrations of PAHs were recorded in March 2018 for all the stations. The concentrations were 1478.34, 994.06, and 666.27 µg/kg at FST1, FST2, and FST3 respectively. The least total concentrations were also 291.94 µg/kg (FST1), 373.65 µg/kg (FST2), and 238.66 µg/kg (FST3) during October 2018, September 2018, and January 2019 respectively. Moreover, monthly trends for the individual PAHs in sediment have also been presented in Figures 28 and 29.

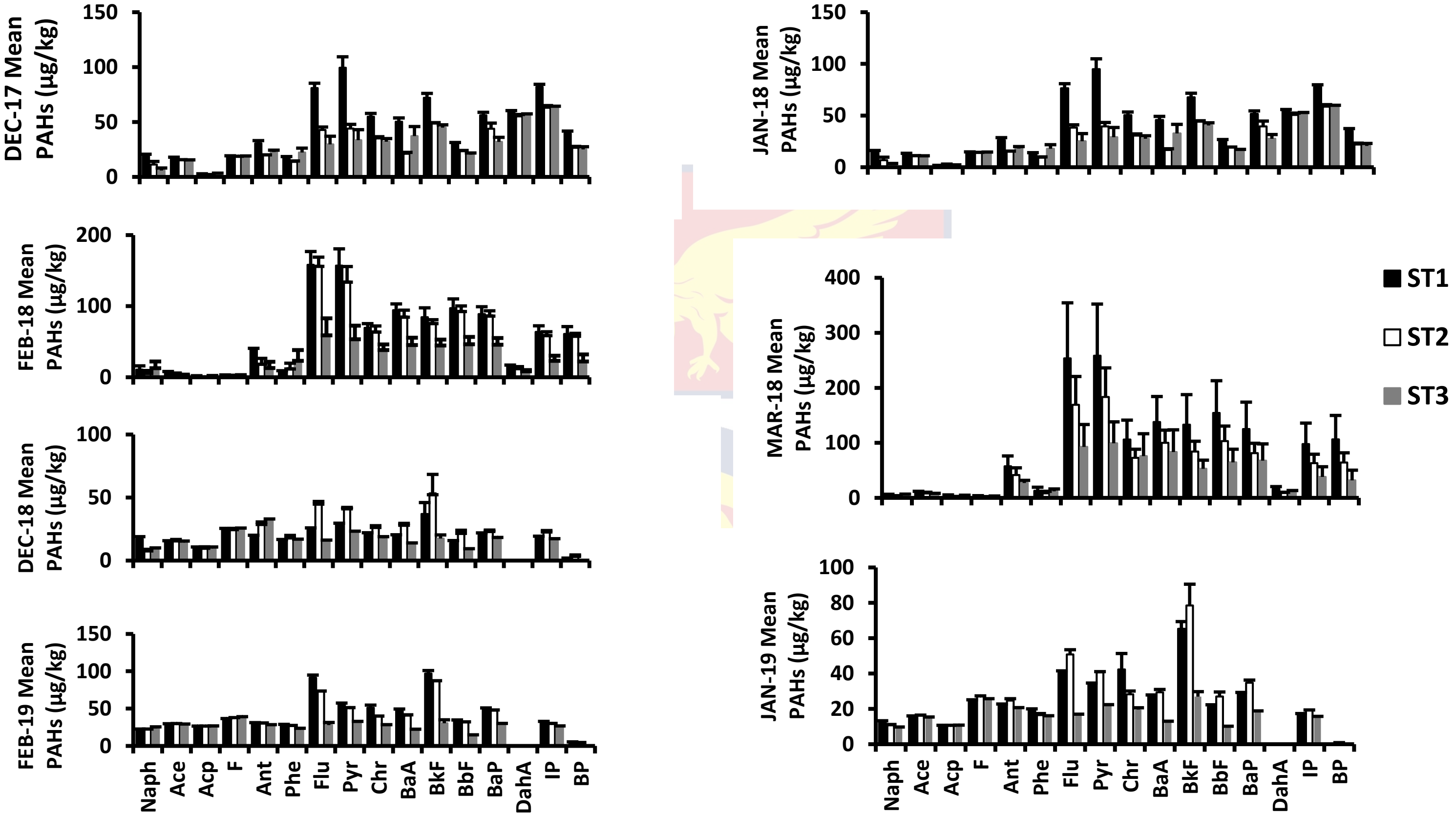

Figure 28: Monthly mean concentrations of individual PAHs in the sediment of Fosu Lagoon for the dry season.

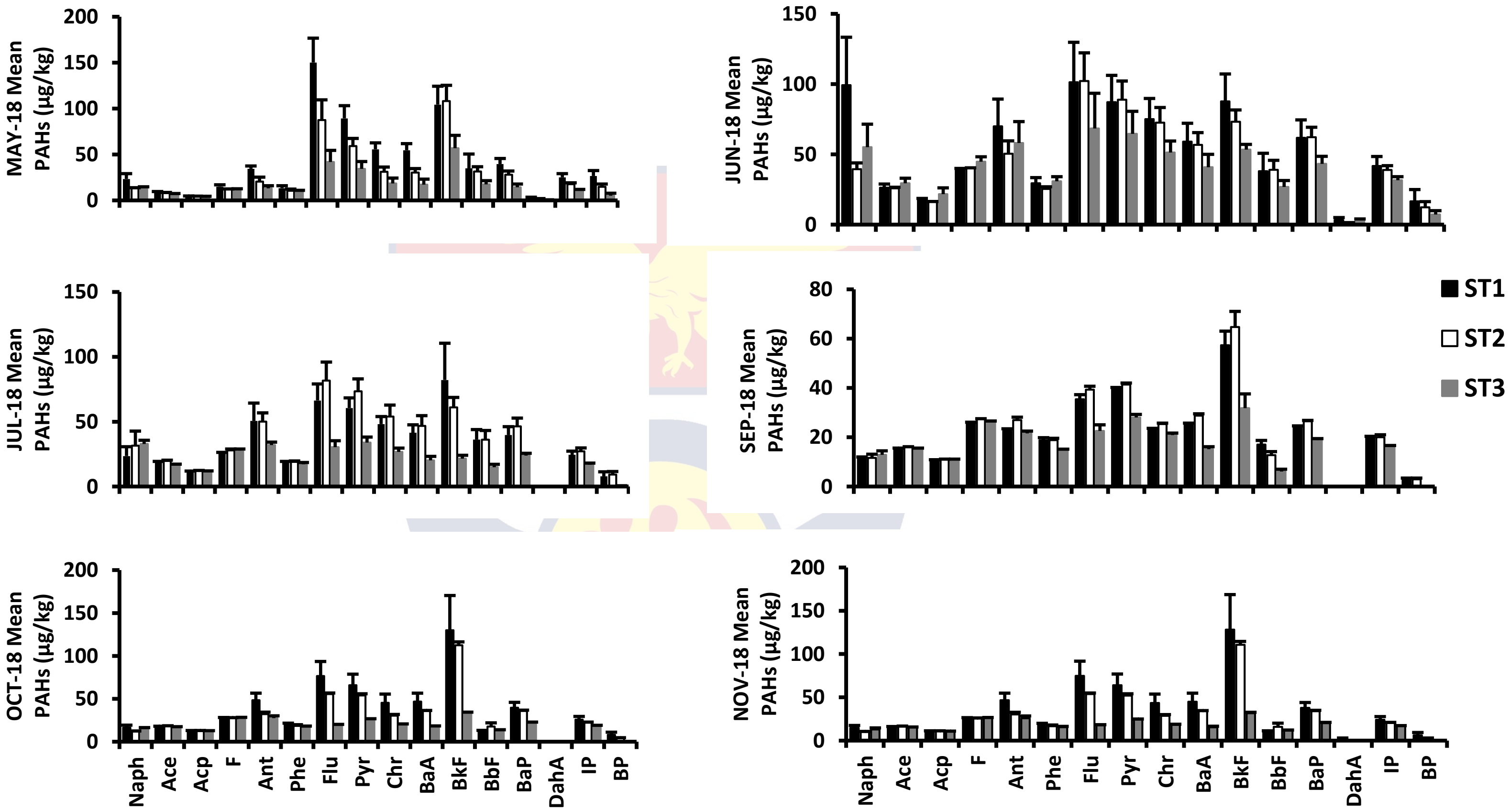

Figure 29: Monthly mean concentrations of individual PAHs in the sediment of Fosu Lagoon for the wet season.

During the dry season (Figure 28; December 2017 to March 2018 and December 2018 to February 2019), either fluoranthene, pyrene or Benzo[k]fluoranthene was detected as the compound with the highest concentration. Such detection was also observed at either FST1 or FST2. By the end of the dry season (December 2018 to February 2019), it was noted that there was a general increase in individual PAHs, except Dibenz[ah]anthracene which was below the detection limit at all stations. The wet season (Figure 29; May 2018 to November 2018) on the other hand, experienced a decrease in the levels of the HMW compounds and an increase in the LMW compounds as compared to the dry season. Samples were not taken in August 2018 due to a constraint, and that is, people were not allowed on the lagoon during the "Fetu Afahye" season at Cape Coast. Fluoranthene was the highest detected compound in May and June 2018, while Benzo[k]fluoranthene was for July to November 2018. The mean concentrations of fluoranthene were 150.12±26.52 µg/kg in May 2018 and 101.26±28.45 µg/kg in June 2018. The mean concentrations of Benzo[k]fluoranthene were 82.15±28.28 µg/kg, 64.68±6.32 µg/kg, 129.96±40.51 µg/kg, and 128.13±40.15 µg/kg in July, September, October, and November 2018 respectively. These two compounds were also observed at either FST1 or FST2. Dibenz[ah]anthracene was found to be below the detection limit in all three stations from July to the end of the wet season.

Through the nonparametric test, recorded sediment concentrations for Naph, Acp, F, Ant, BaA, Chr, BkF, BaP, DahA, IP, BP, and total PAHs were significantly different between the two seasons with resultant $p < 0.05$ (Appendix B7). All the LMW compounds recorded significantly ($p < 0.05$) high concentrations in the wet season compared to the dry season. However, the total

PAHs and HMW compounds were significantly ($p < 0.05$) higher in the dry season than the wet season. For spatial distribution, the Kruskal-Wallis test (Table 10) for sediment samples from Fosu Lagoon indicated that Naph and Ant were the only LMW compounds whose concentrations varied significantly ($p < 0.05$) among the three sampling stations. For both compounds, the concentrations at FST1 were significantly ($p < 0.05$) higher than the other two stations. About 90% of the HMW was also significant ($p < 0.01$) at different stations. The HMW compounds and total PAHs were significantly ($p < 0.01$) lower at FST3 compared to the other two stations (Table 10).

Table 10 - *Kruskal–Wallis nonparametric test showing differences within sampling points*

Compound	Comparison	Test Statistic	Adjusted significance [a]
Naph	FST2-FST3	-3.821	1.000
	FST2-FST1	19.026	0.040
	FST3-FST1	15.205	0.143
Ant	FST3-FST2	11.436	0.410
	FST3-FST1	26.526	0.002
	FST2-FST1	15.090	0.148
Flu	FST3-FST2	39.846	0.000
	FST3-FST1	45.154	0.000
	FST2-FST1	5.308	1.000
Pyr	FST3-FST2	35.436	0.000
	FST3-FST1	43.026	0.000
	FST2-FST1	7.590	0.969
BaA	FST3-FST2	27.603	0.001
	FST3-FST1	40.551	0.000
	FST2-FST1	12.949	0.276

continued

Chr	FST3-FST2	26.436	0.002
	FST3-FST1	38.410	0.000
	FST2-FST1	11.974	0.357
BbF	FST3-FST2	46.821	0.000
	FST3-FST1	49.487	0.000
	FST2-FST1	2.667	1.000
BkF	FST3-FST1	24.410	0.004
	FST3-FST2	28.282	0.001
	FST1-FST2	-3.872	1.000
BaP	FST3-FST2	34.436	0.000
	FST3-FST1	36.641	0.000
	FST2-FST1	2.205	1.000
IP	FST3-FST2	24.513	0.004
	FST3-FST1	26.256	0.002
	FST2-FST1	1.744	1.000
BP	FST3-FST2	25.910	0.002
	FST3-FST1	26.513	0.002
	FST2-FST1	.603	1.000
Total PAHs	FST3-FST2	35.231	0.000
	FST3-FST1	40.923	0.000
	FST2-FST1	5.692	1.000

*a p values have been adjusted by the Bonferroni correction for multiple tests.

4.2.3.2 PAHs levels in fish parts

The descriptive statistics of all the 16 priority PAHs that were detected in the fish parts have been presented in Appendix A7. The Fosu Fish Fillet (FFF), Fosu Fish Visceral (FFV), and Fosu Fish Head (FFH) had a total PAHs ranging from 116.80 to 381.43 µg/kg, 196.73 to 2050.47 µg/kg, and 292.08 to 1606.70 µg/kg respectively. Their mean concentrations were 269.69±78.58 µg/kg (FFF), 558.52±529.63 µg/kg (FFV) and 615.10±360.80 µg/kg (FFH).

The total PAHs in the visceral and head were approximately twice and thrice the concentration in the fillet respectively. In the fillet, visceral, and head, the following compounds recorded the highest concentrations respectively: Ace (28.21±12.62 µg/kg), IP (141.07±325.13 µg/kg), and Flu (139.44±197.85 µg/kg). Benzo[g,h,i]perylene was the compound with the lowest concentrations of 0.08±0.21 µg/kg (in FFF) and 2.43±7.75 µg/kg (in FFH). However, for FFV, DahA was the compound with the lowest concentration of 4.09±7.32 µg/kg. The presence of the carcinogenic compounds compared to the total PAHs was approximately 42%, 48%, and 26% in the fillet, visceral, and head respectively. The dominating carcinogenic compounds were BaP, IP, and IP.

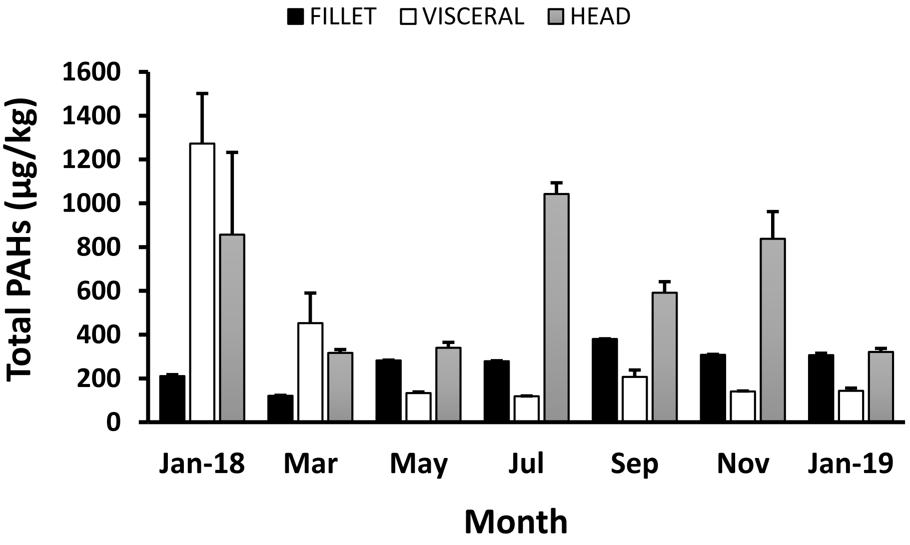

Figure 30: Monthly variations of total PAHs in fish parts from Fosu Lagoon.

From Figure 30, the total PAH in FFH dominated in all the months under study, except January 2018 and May 2018 where the concentrations in FFV dominated. The highest total concentration in FFH was 1,042.15µg/kg and this was recorded in July 2018. However, the highest total concentration in FFV was 856.93 µg/kg and this was recorded in January 2018. Monthly distribution of individual PAHs in the fish tissues was further analysed (Figure 31).

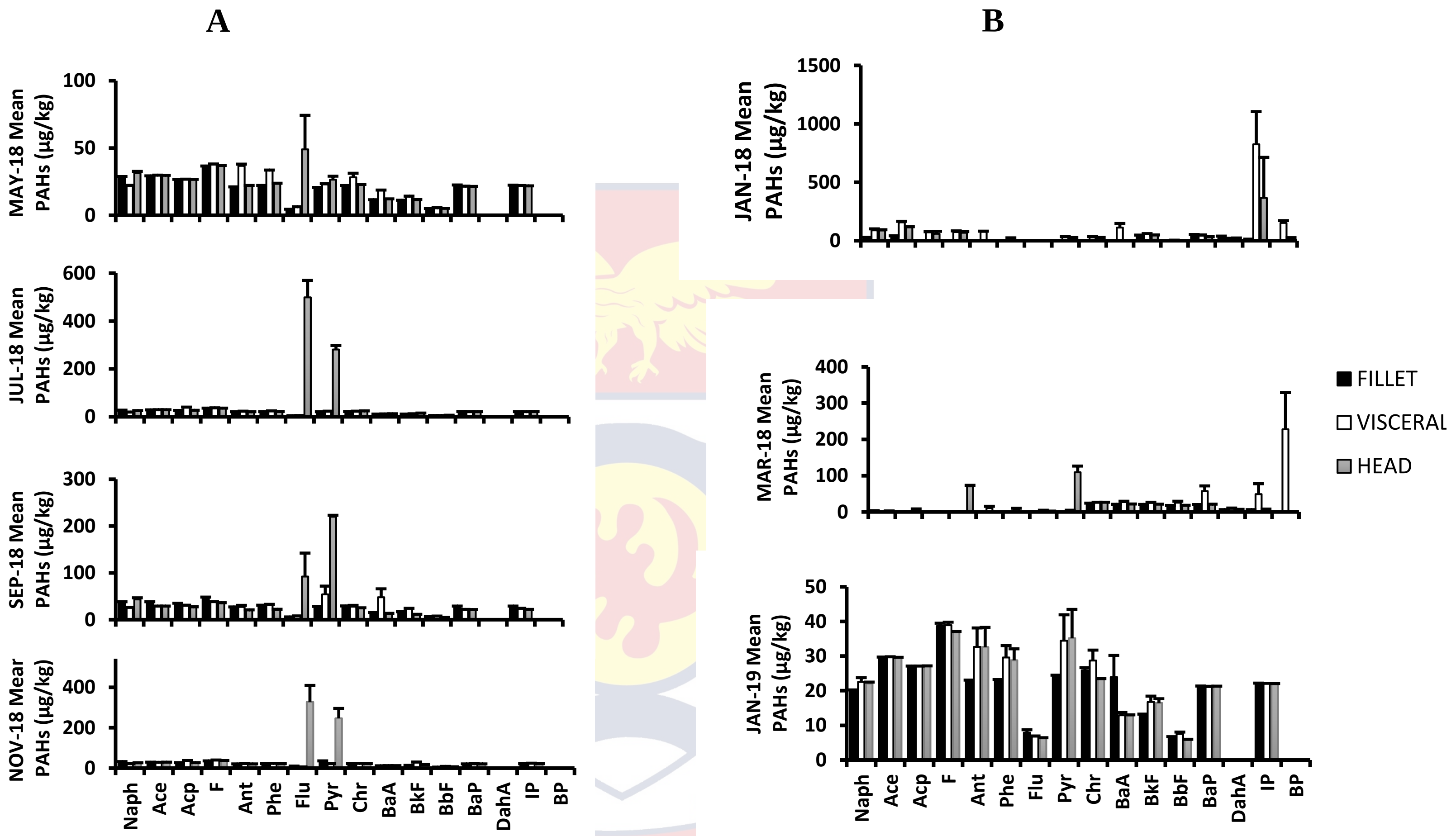

Figure 31: Seasonal variations of mean concentrations of individual PAHs in the fish parts of Fosu Lagoon: **A** composed of wet season months and **B** composed of dry season months.

In the wet season (Figure 31A; May, July, September, and November 2018), DahA and BP were below the detection limit in all the months. Fluoranthene recorded the highest concentration in the head during all the months (except in September 2018 where Pyr dominated). For May 2018, July 2018, and November 2018, the mean concentrations of Flu were 48.99±25.25 µk/kg, 498.92±71.07 µk/kg, and 327.65±81.98 µk/kg respectively. The mean concentration of Pyr in September was 219.98±2.94 µk/kg. During the dry season (Figure 31B; January 2018, March 2018 and January 2019), the highest mean concentrations of individual compounds were 824.74± 280.34 µg/kg (IP), 227.94±101.24 µg/kg (BP), and 38.93± 0.88 µg/kg (F). All these highest recorded compounds in the dry season were measured in the visceral part of the fish.

From the Mann-Whitney U test for the season variations, the concentrations of Naph, Acp, Phe, Flu, Pyr, Chr, BbF, DahA, and BP were significantly different ($p < 0.05$) for the fish parts (Appendix B8). Naph, Acp, Phe, Flu, and Pyr were significantly ($p < 0.05$) lower in the dry season than in the wet season. Chr, BbF, DahA and BP were significantly ($p < 0.05$) higher in the dry season than in the wet season. In using the Kruskal-Wallis test, the following compounds showed significant ($p < 0.05$) differences among the three fish parts: Acp, F, Ant, Phe, Pyr, Chr, BbF, IP, and ∑16PAHs (Table 11). Among these, the concentrations of Acp, F, Ant, Phe, Chr, BbF, IP, and ∑16PAHs in FFF were significantly ($p < 0.05$) lower than their concentrations in FFV. Besides, concentrations of Ant, Phe, Pyr, Chr, and IP in FFH were significantly ($p < 0.05$) higher than their concentrations in FFV.

Table 11 - *Kruskal–Wallis nonparametric test showing differences among fish parts from Fosu Lagoon*

Compound	Comparisons	Test Statistic	Adjusted Significance [a]
Acp	FFF-FFH	-8.857	0.350
	FFF-FFV	-16.857	0.008
	FFH-FFV	-8.000	0.469
F	FFF-FFH	-12.714	0.074
	FFF-FFV	-17.571	0.006
	FFH-FFV	-4.857	1.000
Ant	FFF-FFH	-0.595	1.000
	FFF-FFV	-20.048	0.001
	FFH-FFV	19.452	0.002
Phe	FFF-FFH	-1.429	1.000
	FFF-FFV	-18.214	0.004
	FFH-FFV	-16.786	0.009
Pyr	FFF-FFV	-8.190	0.442
	FFF-FFH	-24.524	0.000
	FFV-FFH	-16.333	0.012
Chr	FFF-FFH	-2.429	1.000
	FFF-FFV	-16.929	0.008
	FFH-FFV	-14.500	0.031
BbF	FFF-FFH	-3.286	1.000
	FFF-FFV	-14.857	0.026
	FFH-FFV	-11.571	0.122
IP	FFH-FFF	0.381	1.000
	FFH-FFV	-15.333	0.020
	FFF-FFV	-14.952	0.025
∑PAHs	FFF-FFV	-19.476	0.002
	FFF-FFH	-26.524	0.000
	FFV-FFH	-7.048	0.638

*a p values have been adjusted by the Bonferroni correction for multiple tests.

The composition of ∑PAHs noncar, ∑PAHs car and ∑16PAHs were also analysed (Figure 32). The highest concentrations for both ∑PAHs noncar and ∑16PAHs were measured in FFH, while that of ∑PAHs car was measured in the FFV. Their concentrations were 453.94 µg/kg (∑PAHs noncar), 270.55 µg/kg (∑PAHs car), and 615.10 µg/kg (∑PAHs). All the fish parts were dominated by the HMW compounds, constituting 56% in the fillet, 62% in the visceral, and 64% in the head (Figure 33).

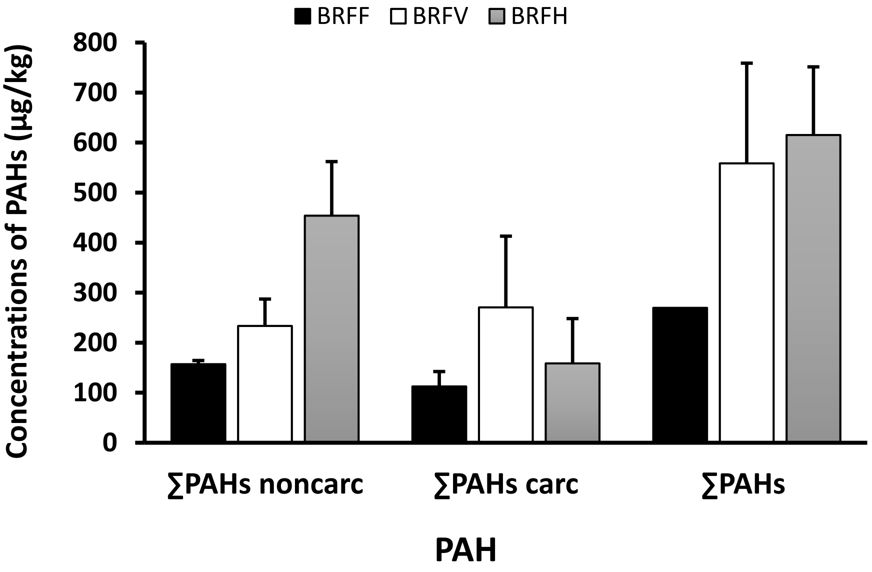

Figure 32: Non-carcinogenic, carcinogenic and total PAHs in the fillet, visceral, and head of fish from Fosu Lagoon.

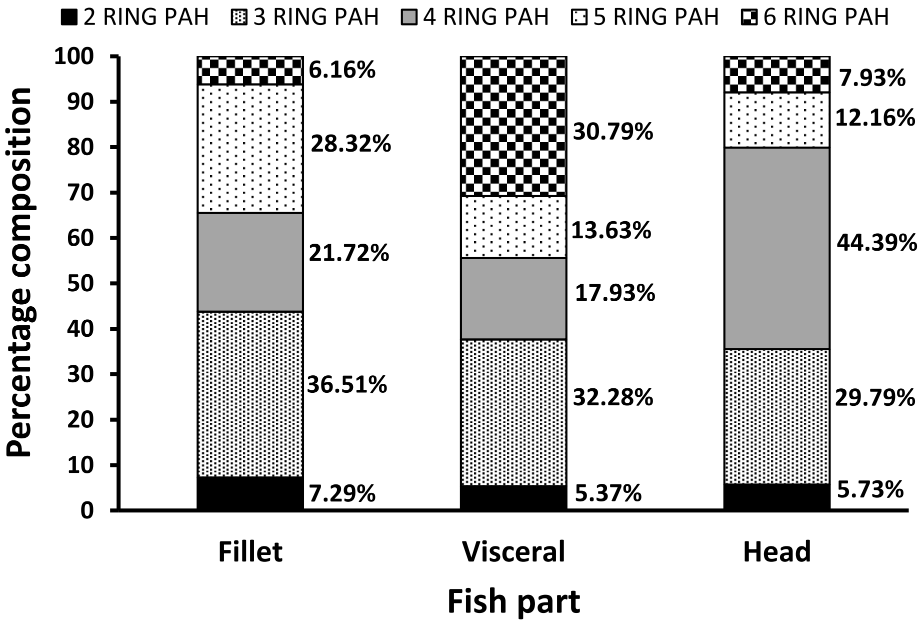

Figure 33: Percentage compositions of 2, 3, 4, 5 and 6-rings PAHs in fish parts.

Additionally, the total PAHs in all the media were considered coherently. It was observed, just like at Benya and Brenu Lagoons that the highest level of total PAHs was measured in the head of the fish (579.01 µg/kg), with water samples (2.66 µg/L) recording the least concentration (Figure 34). However, such concentration in the fish head was not statistically different from the total concentration in the sediment and fish visceral, but significantly higher than that in the fish fillet and water ($p < 0.01$).

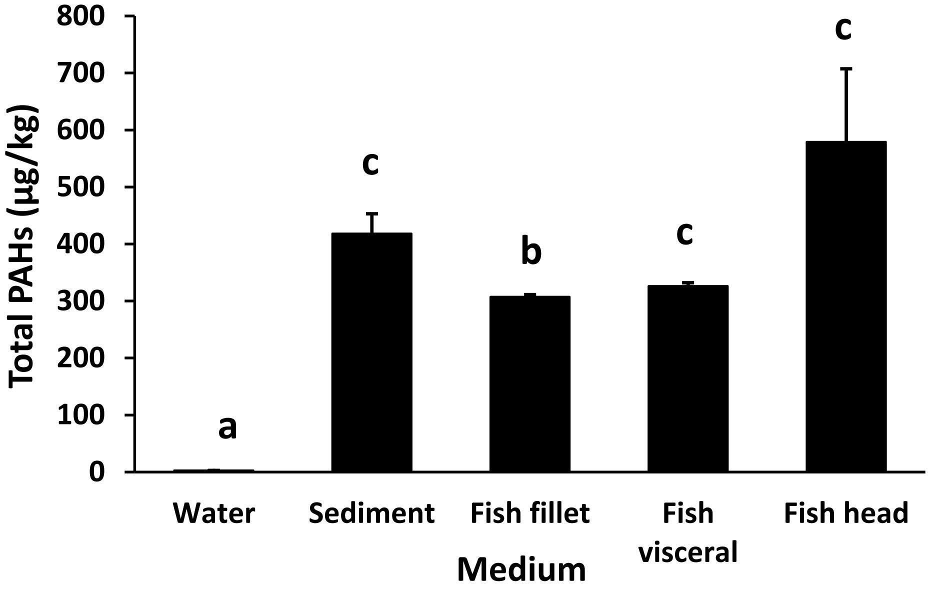

Figure 34: Total PAHs in different media of Fosu Lagoon. Statistical differences among media ($p < 0.01$) are indicated by different letters.

With high affinity to sediments, the measured concentrations of the current study was compared to other coastal sediments in the world (Table 12).

Table 12 - *Comparison of PAH concentrations in the studied sediments to selected worldwide sediments*

Region	**Number of PAHs**	**Concentration (µg/kg)**	**Reference**
Ghana	16	165-728	This study
Côte d'Ivoire	8	1,550-437,520	(Kouakou, Kouassi, Kouassi Kwa-Koffi, Gnonsoro, & Trokourey, 2015)
Togo	21	4-256	(Gnandi et al., 2011)
Cotonou, Benin	14	25-1,450	(Soclo, Garrigues, & Ewald, 2000)
Ologe lagoon, Nigeria	16	350-430	(Obanya et al., 2019)
Morocco	15	22-108	(Giuliani et al., 2015)
Coastal sediments, Thailand	17	6 to 8,399	(Boonyatumanond, Wattayakorn, Togo, & Takada, 2006)
Bizerte coastal sediment, Tunisia	16	83-447	(Zaghden et al., 2007)
Ligurian Sea (Mediterranean)	16	<1-26,247	(Bertolotto et al., 2003)
Western Mediterranean Sea	26	<1-20,500	(Baumard et al., 1998)
Beaufort Sea	Not stated	200-1,200	(Yunker et al., 1996)
Baltic Sea	15	10-1900	(McCready, Birch, & Long, 2006)
Casco Bay, USA	23	16-2,100	(Kennicutt et al., 1994)

4.3 Bioaccumulation

In using total concentrations of PAHs in both fish tissues and sediments, the BAF and BSAF values were obtained (Appendix A8 and A9). The BAF values varied from 0.3 (fish fillet from Fosu) to 3.1 (fish head from Brenu). The BSAFs were 6.7, 1.7 and 2.2 for fish fillet from Benya, Brenu and Fosu Lagoons respectively. It should be noted that BSAFs were measured in fish fillet only. Generally, fish fillet sampled from Brenu Lagoon had the lowest BSAFs for most (≈ 63%) of the individual PAHs. In addition, the low molecular weights compounds generally had higher BSAF values than the high molecular weight compounds (Figure 35).

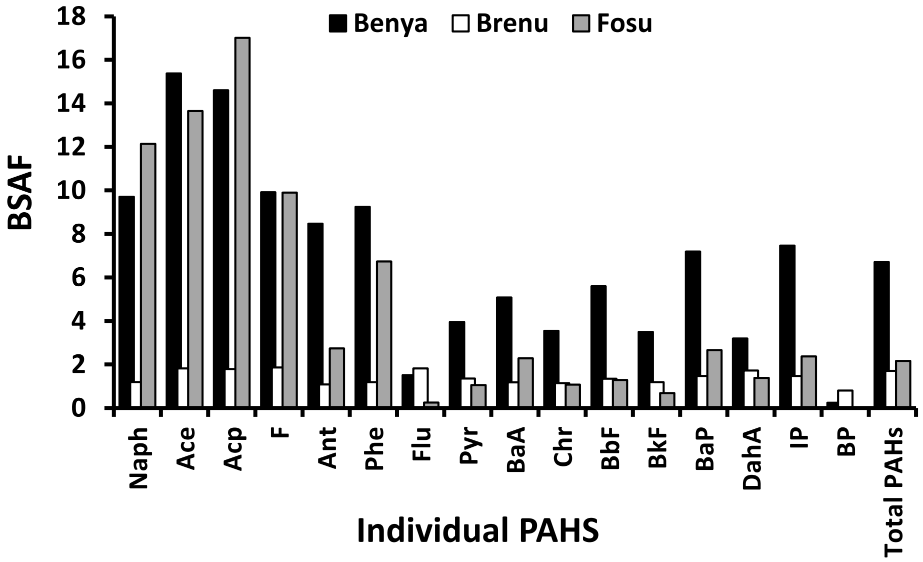

Figure 35: BSAFs for both individual PAHs and total PAHs in fish fillet sampled from Benya, Brenu, and Fosu Lagoons.

4.4 Influence of physicochemical characteristics on total PAHs in sediments

The descriptive statistics (Table 13) and linear regression models (Table 14) were computed for all the 3 lagoons holistically. Both simple and multiple linear regressions were computed to analyse 4 predictors {Total Organic Carbon (%TOC), temperature, pH, and salinity} on total PAHs in lagoon sediments. The overall regression model was significant {F (4, 292) = 18.19, p <0.001, R^2 = 0.20}. However, it was only %TOC and pH that individually, explained the unique significant to the variance in the total PAHs. The results of the simple linear regression indicated that %TOC significantly accounted for 8% of the variance in the total PAHs {F (1, 295) = 24.63, p <0.001, R^2 = 0.08}, with a regression model equation of y = 21.3x + 205.77 (see Figure 36). Also, pH contributed 12% to the variance {F (1, 295) = 40.64, p <0.001, R^2 = 0.12} with a regression model y = 208.4x – 1365.4.

Table 13 - *Descriptive statistics of water and sediment parameters of lagoons*

Parameter	Minimum	Maximum	Mean	Std. Deviation
%TOC	0.15	17.66	4.90	3.40
TEMPERATURE	22.72	33.00	29.84	1.88
pH	7.09	9.09	8.05	0.43
SALINITY	3.40	38.50	21.48	12.85

Table 14 - *Coefficients and contributions of the linear regression model for total PAHs (factor) and the measured physicochemical parameters (predictors)*

Predictor		Coef.	SE coef.	T-value	P-value
Constant		-1325.988	407.849	-3.251	0.001
%TOC		21.309	4.029	5.231	0.000
TEMPERATURE		-4.111	8.238	-.499	0.618
pH		207.210	43.870	4.723	0.000
SALINITY		-0.474	1.483	-.320	0.749
	Adj SS	df	Adj MS	F-value	P-value
Regression	3924202.287	4	981050.572	18.192	0.000
		R^2=20%			

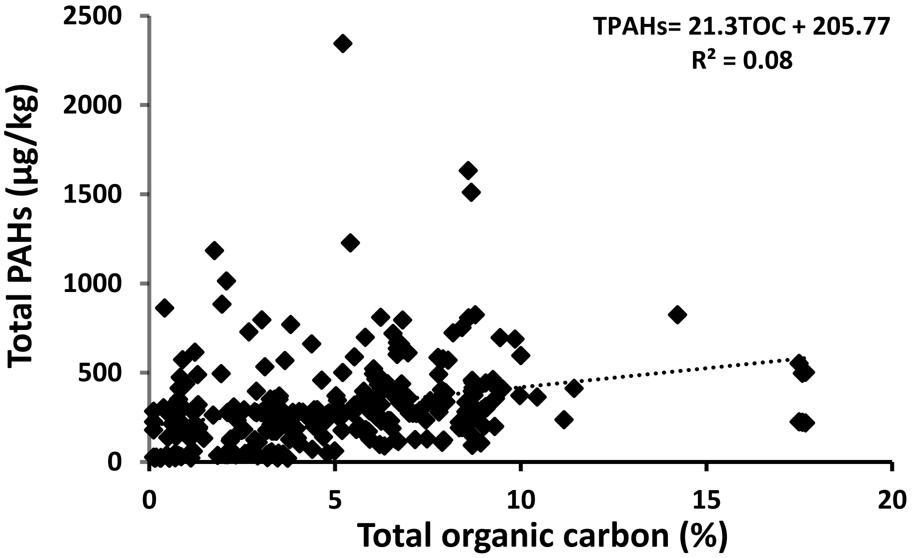

Figure 36: Linear regression model of total organic carbon and total PAHs in sediments.

4.5 Source apportionment of PAHs

In this study, the ratios used for source identification of PAHs have been presented in Figure 37 and Table 15. This analysis focused mainly on sediment and fish samples due to the number of data associated with these samples.

Briefly, in considering the water samples, ∑LMW/∑HMW was above 1 for all the 3 lagoons, ranging from 1.53 (Brenu) to 1.76 (Benya). However, the values for Benya, Brenu, and Fosu were respectively for Ant/Ant+Phe (0.57, 0.57, and 0.55), BaA/BaA+Chr (0.66, 0.66, and 0.65) and Phe/Ant (0.75, 0.76, and 0.81) ratios. The ratio of ∑PAHs(4-rings)/∑PAHs(5-6-rings) was 0.89, 0.92 and 0.93 for Benya, Brenu and Fosu respectively.

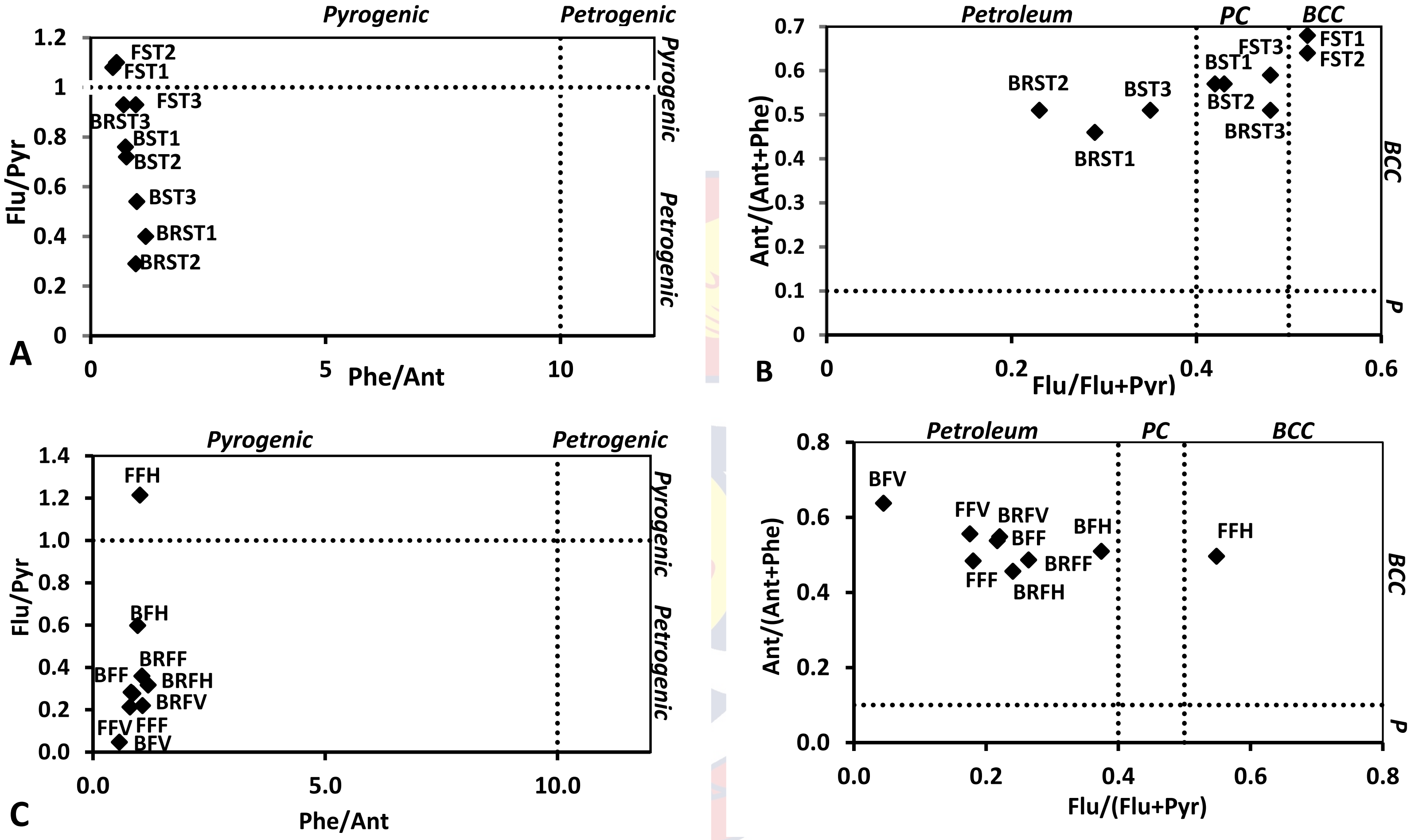

Figure 37: Plots showing PAH isomeric ratios Phe/Ant vs Flu/Pyr and Flu/(Flu+Pyr) vs Ant/(Ant+Phe) for sediments (**A, B**) and fish parts (**C, D**) in Benya, Brenu and Fosu Lagoons. (*P- petroleum, PC- petroleum combustion, BCC- biomass or coal combustion)

Table 15 - *Molecular diagnostic ratios of PAHs in sediment and fish components of three different lagoons*

Stations	∑LMW/ ∑HMW [a]	BaP/BP [b]	∑PAHs(4 rings)/ ∑PAHs(5-6 rings) [c]	Fish parts	∑LMW/ ∑HMW [a]	BaP/BP [b]	∑PAHs(4 rings)/ ∑PAHs(5-6 rings) [c]
BST1	0.46	1.49	1.00	BFF	1.07	12.41	0.68
BST2	0.39	1.53	1.02	BFV	1.19	6.89	1.93
BST3	0.54	1.75	0.75	BFH	0.75	10.16	1.96
BRST1	0.83	2.96	0.68	BRFF	1.08	2.69	0.68
BRST2	0.77	2.90	0.63	BRFV	0.79	1.22	1.25
BRST3	0.86	12.59	1.37	BRFH	0.48	1.06	2.52
FST1	0.22	1.83	1.05	FFF	0.99	37.37	0.75
FST2	0.23	2.61	0.98	FFV	0.56	0.16	0.23
FST3	0.37	1.59	0.84	FFH	0.42	2.70	2.36

[a] The ratios of total LMW chemicals (two- and three-ringed PAHs) to HMW chemicals (four- to six-ringed PAHs). The petrogenic sources are more abundant in LMW PAHs and the pyrogenic sources contain a greater percentage of HMW PAHs.

[b] The ratio BaP/BP could be used to elucidate the traffic (> 0.6) and non-traffic sources (< 0.6).

[c] The ratio of ∑PAHs(4 rings)/∑PAHs(5-6 rings) predicts local source (< 0.9) or longer distance transport (> 0.9).

In the sediment samples, the ratios of Ant/(Ant + Phe), Flu/Pyr, Flu/(Flu + Pyr), and Phe/Ant varied from 0.46 to 0.68, 0.29 to 1.10, 0.23 to 52, and 0.47 to 1.17 respectively (Figure 37; A and B). Specifically, the values determined for Benya sediment were between 0.50 to 0.58, 0.54 to 0.76, 0.35 to 0.43 and 0.74 to 0.98 for Ant/(Ant + Phe), Flu/Pyr, Flu/(Flu + Pyr) and Phe/Ant respectively. In the case of Brenu sediment, the values were respectively 0.46 to 0.51, 0.29 to 0.93, 0.22 to 0.48 and 0.96 to 1.17. For Fosu sediment, the values were 0.59 to 0.68, 0.92 to 1.10, 0.48 to 0.52 and 0.47 to 0.70 respectively. The other diagnostic ratios used in determining the origin of the PAHs in the sediment samples have been presented in Table 15.

In Figure 37; C and D, the Ant/Ant+Phe and Phe/Ant values for all the fish parts were >0.1 and <10 respectively. However, the values of Flu/(Flu + Pyr) were <0.4, except in FFH where the value was 0.55. Also, the values for Flu/Pyr in all the fish parts were <1, except in FFH where the value was >1. Besides, the ratios of BaP/BP and ∑PAHs(4 rings)/∑PAHs(5-6 rings) for all the fish samples ranged from 0.16 to 37.37 and 0.23 to 2.52 respectively (Table 15). It was noted that the values of BaP/BP in all the fish parts were >0.6, except in the FFV (0.16).

4.6 Ecological risk assessment

Ecological risk assessment is of principal importance when developing sediment quality criteria. In this study, the comparison of chemical concentrations of PAHs with the Sediment Quality Guidelines (SQGs) has been presented in Table 16. Contamination levels in all the sampling sites for ∑PAHs were lower than the Effect Range-Low (ERL) (4022 µg/kg). The contamination levels for individual PAHs were also lower than their respective ERL values, except for fluorene (F) and Dibenz[a,h]anthracene (DahA) where contamination levels were between ERL and ERM (Effect Range-Medium) values. The dissimilarities for F were observed at all the sampling sites of both Benya and Fosu sediments. For DahA, the dissimilarities occurred at FST1. However, the Threshold Effect Levels or Probable Effect Levels (TEL/PEL) SQGs told a different story. The values for ∑PAHs at all the sampling sites were below TEL, except for FST1. The ∑PAHs at FST1 (1249 µg/kg) was between TEL and PEL. The values for Acenaphthylene (Ace), Acenaphthene (Acp) and DahA were all above TEL, but below PEL for all the sampling sites. The values of other individual compounds such as F, Anthracene (Ant), Fluoranthene (Flu), Pyrene (Pyr), Benz[a]anthracene (BaA), and Benzo[a]pyrene (BaP) were above TEL in at least, one sampling sites.

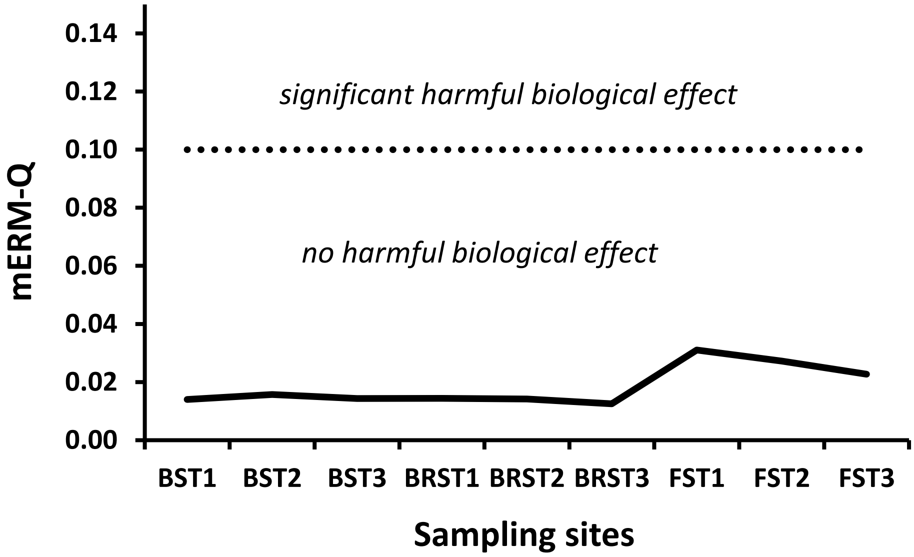

Figure 38: Distribution of mERM-Q in priority levels for sampling sites. The dotted line represents the boundary of the lowest biological effect.

Table 16 - *SQGs values for PAHs (µg/kg dry weight) and relative percentage of sampling sites for Benya, Brenu and Fosu Lagoons*

	SQG ERL-ERM ^{b} (µg/kg)	No. of sampling sites			SQG TEL-PEL^{a} (µg/kg)	No. of sampling sites		
		<ERL	ERL-ERM	>ERM		<TEL	TEL-PEL	>PEL
Naph	160-2100	9	0	0	35-391	9	0	0
Ace	44-640	9	0	0	6-128	0	9	0
Acp	16-500	9	0	0	7-89	0	9	0
F	19-540	3	6	0	21-144	3	6	0
Ant	85-1100	9	0	0	47-245	8	1	0
Phe	240-1500	9	0	0	87-544	9	0	0
Flu	600-5100	9	0	0	113-1494	8	1	0
Pyr	665-2600	9	0	0	153-1398	8	1	0
BaA	261-1600	9	0	0	75-693	8	1	0
Chr	384-2800	9	0	0	108-846	9	0	0
BbF	320-1880	9	0	0	NA	NA	NA	NA
BkF	280-1620	9	0	0	NA	NA	NA	NA
BaP	430-1600	9	0	0	89-763	8	1	0
IP	240-1400	9	0	0	NA	NA	NA	NA
DahA	63-260	8	1	0	6-135	0	9	0
BP	430-1600	9	0	0	NA	NA	NA	NA
∑PAH	4022-44792	9	0	0	655-6676	8	1	0

[a] (Ashayeri et al., 2018; Cardellicchio et al., 2007; Long & MacDonald, 1998)
[b] (Ashayeri et al., 2018; Edokpayi et al., 2016; Long & MacDonald, 1998; NOAA, 1999)
*NA: not available

As shown in Figure 38, the estimated mean ERM quotient (mERM-Q) was lower than 0.1 at all sampling sites and varied from 0.01 (BRST3) to 0.03

(FST1). This resulting index provided a method for evaluating the synergistic effects of contaminants in sediments. The Toxic Equivalent Quotient (TEQ) for total PAHs in sediments ranged from 19.21 µg/kg (BRST3) to 161.93 µg/kg (FST1). The TEQ for total carcinogenic compounds (∑TEQ car) in sediment ranged from 19.02 µg/kg (BRST3) to 160.28 µg/kg (FST1). Another ecological assessment that was considered in this study was Risk Quotient (RQ). In the water samples, the RQ_{NCS} and RQ_{MPCS} for all the individual compounds were <1 (Appendix C1). On the contrary, about 56%, 75% and 56% of individual compounds in Benya, Fosu and Brenu sediments respectively recorded RQ_{NCS} >1. Benya and Brenu Lagoons exhibited very similar distribution in all sites considering the individual PAHs that exhibited RQ_{NCS} <1. At both lagoons, Flu, Chr, BkF, DahA, IP and BP recorded RQ_{NCS} <1. For Fosu sediment, it was only Chr, DahA and BP that had RQ_{NCS} <1. Also, the calculated RQ_{MPCS} of individual PAHs in all the sampling sites at Benya and Brenu were <1, while Pyr concentration at FST1 exhibited RQ_{MPCS} >1. The $RQ_{\sum PAHs\ (NCs)}$ in water were 0.84, 0.63 and 1.43 for Benya, Brenu and Fosu respectively. That of $RQ_{\sum PAHs\ (MPCs)}$ in the water samples were 0.008 (Benya), 0.006 (Brenu) and 0.014 (Fosu). For the lagoon sediments, $RQ_{\sum PAHs\ (NCs)}$ varied from 86 to 118 in Benya, 66 to 72 in Brenu and 132 to 354 in Fosu. For the $RQ_{\sum PAHs\ (MPCs)}$ in sediments, Benya recorded a range of 0.86 to 1.18, Brenu; 0.66 to 0.72 and Fosu; 1.32 to 3.55.

4.7 Human health risk assessment

The potential human health risks through ingestion of water and consumption of fish, as well as dermal contact via water and sediment, were calculated.

4.7.1 Human health risk via ingestion and dermal contact with water

In this study, mean concentrations of BaP in the water samples were 0.11 µg/L, 0.08 µg/L and 0.18 µg/L for Benya, Brenu and Fosu respectively; compared to the established Maximum Contaminant Level (MCL) of 0.2 µg/L for BaP.

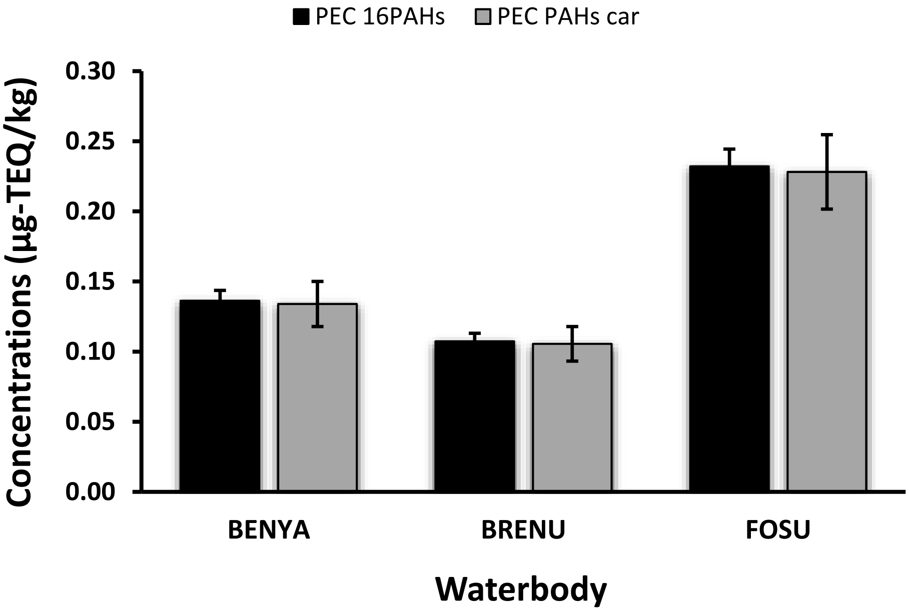

Figure 39: Concentrations of PEC (both ∑16PAHs and ∑PAHs car) in water from Benya, Brenu, and Fosu Lagoons.

The total PEC (µg-TEQ/kg) of the 16 PAHs and the 7 carcinogenic PAHs for water was presented in Figure 39. From the results, the highest values of PEC for both ∑16PAHs and ∑PAHs car in water were from Fosu Lagoon, while Brenu recorded the lowest for both components. The respective values of PEC ∑16PAHs and PEC ∑PAHs car in water samples were 0.14 µg/kg and 0.133 µg/kg (Benya); 0.11 µg/kg and 0.11 µg/kg (Brenu); 0.23 µg/kg and 0.23 µg/kg (Fosu). The PEC values of individual PAHs as presented in Appendix C2 showed that in all the 3 lagoons, BaP was the highest potential toxicity individual compound, contributing more than 50% to PEC ∑PAHs car.

Estimated Incremental Lifetime Cancer Risk (ILCR) for total PAHs via ingestion of water for adults were 1.04×10^{-10} (Benya), 8.16×10^{-11} (Brenu), and 1.76×10^{-10} (Fosu). The ILCR for that of children were 1.45×10^{-10} (Benya), 1.14×10^{-10} (Brenu), and 2.46×10^{-10} (Fosu). For dermal route exposure, ILCR for total PAHs in adults were 1.77×10^{-8} (Benya), 1.40×10^{-8} (Brenu), and 3.02×10^{-8} (Fosu), while that for children were 5.05×10^{-8} (Benya), 3.98×10^{-8} (Brenu), and 8.60×10^{-8} (Fosu). In the water, cancer risk values of PAHs via dermal and ingestion pathways for both adults and children were lower than the threshold set by USEPA (1×10^{-6}) (Appendix C3). Dermal contact risk was generally higher than oral pathway exposure. Risk exposure in children was equally higher than in adults.

4.7.2 Human health risk via dermal contact with sediment

Sediment sampled from the Benya Lagoon had BaP concentrations ranging from 2.16 µg/kg to73.90 µg/kg, with an overall mean concentration of 24.39±14.32 µg/kg. BST2 recorded the highest mean BaP concentration of 27.58±17.36 µg/kg. For Brenu Lagoon BaP concentration ranged from BDL to 23.00 µg/kg, with a mean concentration of 13.62±7.53 µg/kg. BRST2 recorded the highest mean concentration of 13.90±7.36 µg/kg for BaP. However, for Fosu Lagoon, FST1 had the highest mean BaP concentration of 99.92±33.47 µg/kg. The total PEC (µg-TEQ/kg) of the 16 PAHs and the 7 carcinogenic PAHs for sediment was presented in Figure 40. From the result, the highest values of PEC for both ∑16PAHs and ∑PAHs car in the sediments were from Fosu Lagoon, while Brenu recorded the lowest for both components. The values of PEC ∑16PAHs ranged from 19.21 µg/kg (BRST3) to 161.93 µg/kg (FST1). The highest PEC ∑PAHs car in each lagoon was also detected in the following stations: BST2, BRST1 and FST1. Their respective values were 41.54 μg/kg (BST2), 22.81 μg/kg (BRST1), and 160.90 μg/kg (FST1). The PEC values of individual PAHs as presented in Appendix C4 showed that in the sediments of all the lagoons, BaP was the highest potential toxicity individual compound, contributing more than 50% to PEC ∑PAHs car.

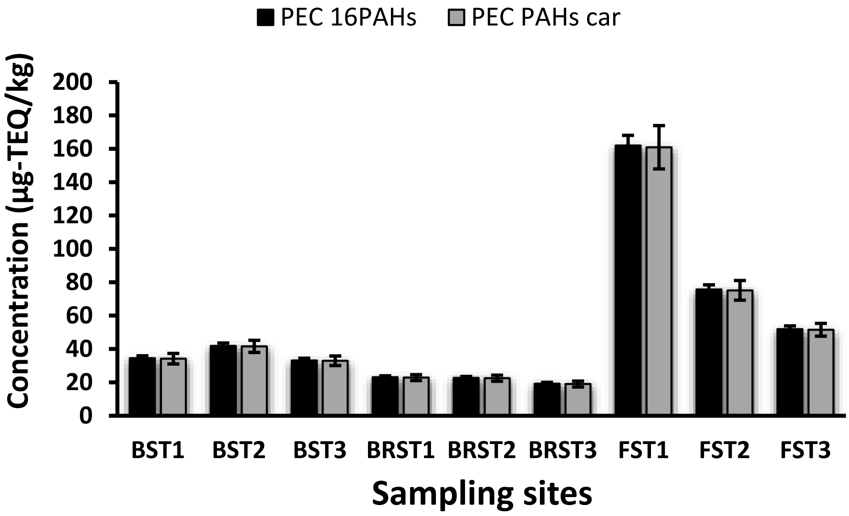

Figure 40: Concentrations of PEC (both ∑16PAHs and ∑PAHs car) in sediments from Benya, Brenu, and Fosu Lagoons.

The ILCRs through dermal exposure of sediments for both individual and total compounds have been presented in the Appendix C5. The ILCR∑16PAHs and ILCR∑PAHs car for adults and children were greater than 1×10^{-6} (the safety level). The ILCR∑16PAHs varied from 2.50×10^{-6} (Brenu) to 2.1×10^{-5} (Fosu) for the adults, and 7.12×10^{-6} to 6.00×10^{-5} for children. For Benya, Brenu and Fosu sediments, adults' ILCR for individual compounds varied from 1.24×10^{-9} (Acp) to 3.59×10^{-6} (BaP), 3.77×10^{-11} (BP) to 1.81×10^{-6} (BaP) and 1.23×10^{-9} (Acp) to 1.30×10^{-5} (BaP) respectively. For both Benya and Brenu sediments, BaP was the only compound at all 3 stations with ILCR $>1\times10^{-6}$. However, for FST1, BaA, BbF, BkF, BaP, DahA and IP were the individual compounds with ILCR >1×10^{-6}. Also, at both FST2 and FST3, the ILCRs for the following compounds were above 1×10^{-6}: BbF, BaP, and DahA.

4.7.3 Human health risk via fish consumption

In considering the fish parts from the various lagoons, the mean concentration of BaP ranged from 19.22±7.66 µg/kg (BRFH) to 32.80±12.79 µg/kg (BFV). It was noted that in all the fish parts the mean concentrations of BaP were above the MCL set by the European Union for fish muscle (2 µg/kg; EFSA, 2008) and that of China for food (5 µg/kg; Shi et al., 2016). In all the fish tissues, the mean concentrations of BaP were approximately ≥ 10 times the MCL set by European Union for fish muscles. The Dietary Daily Intake (DDI) of ∑PAHs car and ∑16PAHs via fish muscle consumption ranged from 8,513 (BRFF) to 8,799 ng/day (BFF) and 21,036 (FFF) to 22,198 ng/day (BRFF) respectively for adults. For the other fish tissues (visceral and head), the DDI of ∑PAHs car and ∑16PAHs ranged from 8,131 (BFH) to 21,103 ng/day (FFV) and 24,164 (BRFV) to 58,142 ng/day (BFV) respectively. Using an ingestion rate of 39 g/capita/day for children, their DDI was half of the adults' DDI.

The total PEC of the 16 PAHs and the 7 carcinogenic PAHs for fish parts in the various lagoons were presented in Figure 41. The highest and lowest PEC ∑PAHs car were found in FFV and BRFH with values of 54.45 µg/kg and 32.78 µg/kg respectively. TEQBaP was the highest recorded PAH. In each lagoon, the highest calculated PEC ∑PAHs car was 52.96 µg/kg (Benya), 37.13 µg/kg (Brenu) and 54.45 µg/kg (Fosu). These were measured in BFV, BRFF and FFV.

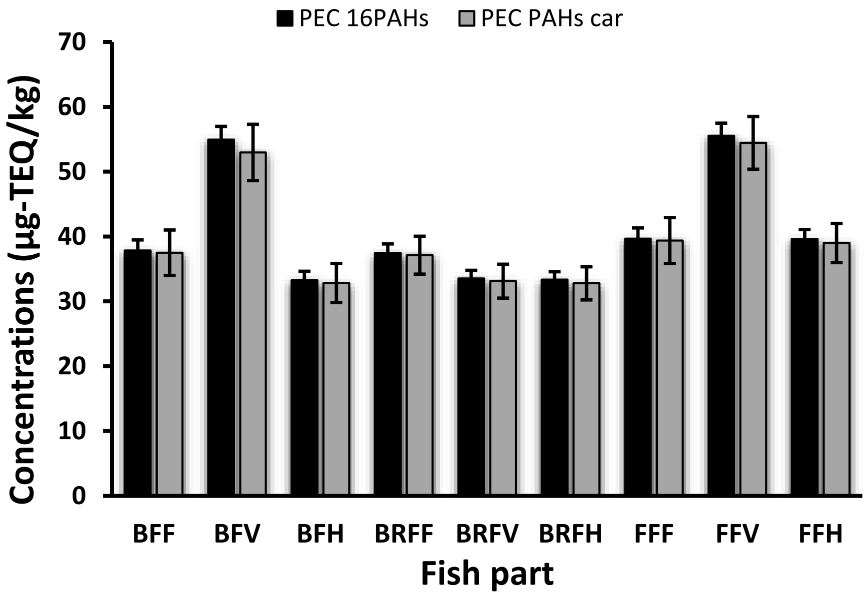

Figure 41: Concentrations of PEC (both ∑16PAHs and ∑PAHs car) in the fish parts from Benya, Brenu and Fosu Lagoons.

The details on the oral or ingestion pathway exposure to fish in these lagoons have been compiled in Appendix C6. ILCRs for ∑16PAHs and ∑PAHs car for both adults and children were greater than the safety threshold, 1×10^{-6}. Considering all the lagoons, ILCR for individual PAHs in adults varied from 1.97×10^{-5} (BFH) to 3.29×10^{-5} (FFV) and that of children, from 2.75×10^{-5} (BFH) to 4.59×10^{-5} (FFV). ILCR for ∑16PAHs varied from 1.97×10^{-5} (BFH) to 3.29×10^{-5} (FFV) in the adults, and 2.75×10^{-5} to 4.59×10^{-5} in children. It was also noted that all the fish tissues had BaA, BaP, DahA and IP as the carcinogenic compounds with ILCR $>1\times10^{-6}$ for both adults and children.

4.8 Sediment remediation

The total concentration of the 16 priority PAHs was at the level of 2.0 ± 0.1 mg/kg. Figure 42 showed freely dissolved concentrations of PAHs (mean ± S.E.) in PAH-contaminated sediment (0% AC) and treated sediment (1%, 3%, and 6% AC), with their corresponding concentrations in the water column. Upon addition of activated charcoal (AC) to PAH-contaminated sediment, there was significant ($p < 0.05$) decrease in the freely dissolved PAHs concentrations in both the water column and sediment (Table 17). In Figure 42, the freely dissolved PAHs concentration at AC dose of 1% was reduced from 1691.8±60.7 µg/L to 1198.8±83.2 µg/L in the water column and from 37.6±4.9 µg/kg to BDL in the sediment. Such significant ($p < 0.05$) reductions of freely dissolved PAH in water column and sediment were equivalent to 29% and 99% respectively (compared to controls without AC). An increase of AC dose from 1% to 3% caused an increase of freely dissolved concentration of PAHs in both media, but was not significantly different from AC doses of 0% and 1%. However, an increase of AC dose from 3% to 6% resulted in a significant ($p < 0.05$) twofold decrease in the sediment and more than a fortyfold in the water column. The mean concentrations at AC dose of 6% were 12.4± 1.0 µg/kg and 31.0±1.8 µg/L for sediment and water column respectively. Also, comparing the decrease of freely dissolved PAHs in the control to AC dose of 6%, concentrations in water column and sediment reduced significantly ($p < 0.05$) to 98% and 67% respectively.

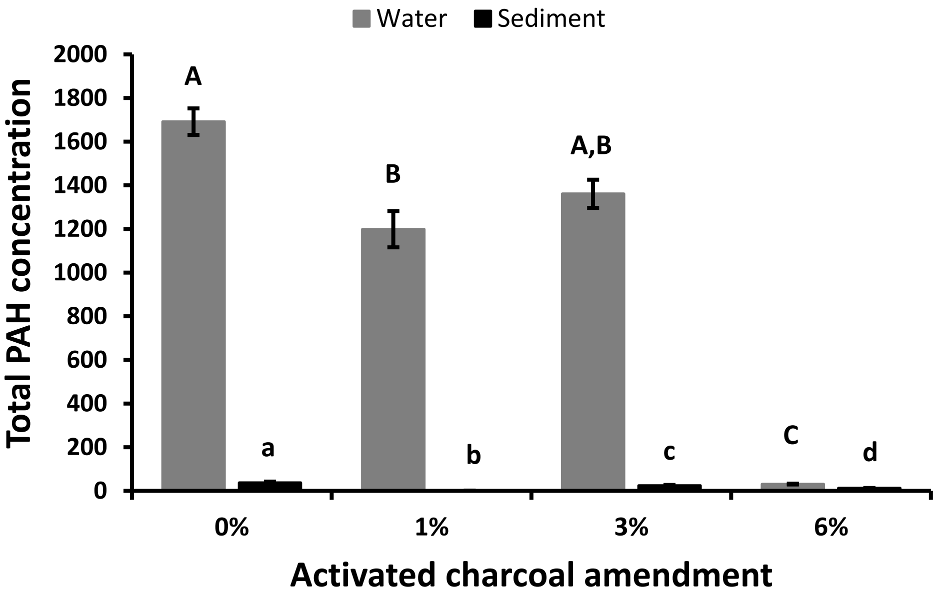

Figure 42: The freely dissolved PAHs concentration (mean ± S.E.) in water column (µg/L) and PAH-contaminated sediment (µg/kg), after activated charcoal (AC) amendment. The upper case alphabets show ANOVA comparison between AC amendments within the water medium. Lower case alphabets show ANOVA comparison between AC amendments within the sediment medium. Means with different alphabets are significantly different (One-way ANOVA: $P < 0.05$).

Table 17 - *Two-way ANOVA for interaction between charcoal amendment and media on total PAHs*

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Charcoal amendment	3	4.4463	1.4821	423.00	0.000
Media	1	20.9030	20.9030	5965.78	0.000
Charcoal amendment × Media	3	10.3639	3.4546	985.96	0.000
Error	16	0.0561	0.0035		
Total	23	35.7693			

CHAPTER FIVE

DISCUSSION

The results presented in Chapter 4 are discussed in this section. The discussion entailed the concentration and distribution of polycyclic aromatic hydrocarbons (PAHs) in the studied lagoons, the possible sources of PAHs to in these lagoons, as well as the eco-toxicological effects on aquatic lives and human health issues associated in consuming fish polluted with PAHs. Furthermore, the possible use of activated charcoal as a remediating material for PAH-contaminated sediment was also discussed.

5.1 Concentrations and distribution of polycyclic aromatic hydrocarbons

There were significant variations in the distribution and concentrations of PAHs in the water column, sediments and fish tissues. Such trend supports the need to critically investigate different media in aquatic ecosystems when monitoring levels of PAHs. One of the reason why this current study chose to survey water, sediments, and fish to evaluate the levels of PAHs in them. In each lagoon, the concentrations and distribution of PAHs in the water column, sediments and fish tissues were discussed.

5.1.1 Concentration of PAHs in water and their distribution

Higher concentrations of PAHs encountered in the Fosu Lagoon among the three study areas may underscore the influence of intense anthropogenic influence on the introduction of PAHs into water bodies. Fosu Lagoon is in close proximity to several commercial and urban activities at the heart of the Cape Coast metropolis. It is known to be the third most polluted lagoon in Ghana, aside the Korle Lagoon in Accra and the Chemu Lagoon in Tema (Akoto et al., 2014; Baffour-Awuah, 2014). Fosu Lagoon has recently been added to the list of water bodies with dead zones, raising concern for the people who depend on it for both sustenance and livelihood. It receives untreated domestic and municipal wastes, burning of wastes, hospital effluents and located under a heavy trafficked bridge. There are other mechanic shops within the vicinity of Fosu Lagoon, which directly discharge used motor oils into the lagoon. All these are potential sources of PAHs to the lagoon. Other studies (Gilbert et al., 2006; Santos et al., 2017; Sarria-Villa, Ocampo-Duque, Páez, & Schuhmacher, 2016) have also reported on the significant contribution of wastewater, burning of biomass/coal and traffic emissions as potential sources of PAHs in the water column of aquatic ecosystems.

The LMW PAHs in this study were predominant in the water samples. This was ascribed to the relatively high solubility nature of the LMW compounds in water. Due to their low octanol-water partition coefficients (Kow), LMW compounds can partition into the water column. The solubility of PAHs in the water column was also ascribed to high temperatures associated with tropical regions. Such a deduction was supported with the fact that these samples were also sampled during the minor wet season (November 2018) and dry season (January 2019), where temperatures are relatively high. With increase in temperature, the solubility of LMW PAHs also increase significantly (Dohányosová, Dohnal, & Fenclová, 2003; Reza, Trejo, & Vera-Ávila, 2002). The dominance of LMW PAHs was also attributed to other characteristics of LMW compounds, such as being less stable, having rapid degradation and evaporation rates compared to the HMW PAHs (Ashayeri et al., 2018; Hu et al., 2010; Omayma et al., 2016). These characteristics coupled with the high levels of LMW compounds in the water column may demonstrate the existence of a local origin or a recent generation through atmospheric deposition or sewage discharge. High levels of LMW PAHs was observed in the Cauca River, Colombia, at stations where there was a point source of burning from a local sugarcane plant (Sarria-Villa et al., 2016).

The present study also revealed that the 3-ring PAHs were abundant in the water samples from all the studied lagoons. The reason could be that, the number of individual PAHs constituting the LMW compounds so far as the 16 priority PAHs are concern is predominantly the 3-ring PAHs. Such abundance was also observed in the works of Jaward et al. (2012) and Sarria-Villa et al. (2016), who also assessed the levels of PAHs in aquatic ecosystems. However, such a comparison should be done with care since the number of individual PAHs reported varied from this study. This present study looked at 16 priority PAHs by USEPA while Jaward et al. (2012) and Sarria-Villa et al. (2016) respectively looked at 11 and 12 individual PAHs out of the 16 priority PAHs. Naphthalene was noted to be the highest detected compound in all the studied lagoons. A compound known to be one of the abundant PAHs in the tropical environments through biological sources (Wilcke, 2007). However, the abundance of Naphthalene in these studied lagoons is of concern. Naphthalene has been reported to potentially act as an endocrine disruptor in fishes. After some days of exposure to Naphthalene, there was a parental transfer of Naphthalene to the larval offspring of fish. Such transfer of contaminant caused deformities in the larval stages with increasing concentrations (Pollino, Georgiades, & Holdway, 2009). Therefore, it is of concern should such contaminant (Naphthalene) become bioavailable to fishes in these lagoons. Further study should be conducted in these current lagoons to investigate this.

The solubility of PAHs due to increase in temperature may have also resulted in the presence of some percentage (at least 37%) of the HMW PAHs in the water column. Another reason to the occurrence of HMW PAHs was ascribed to the presence of suspended particulate matter in the water samples. HMW PAHs have relatively high affinity for suspended particulate matter due to their high partition coefficients. The suspended particulate matter for PAH analysis was not separated from the dissolved pollutants during the extraction of PAHs in the water samples. Unlike another study (Niu et al., 2018) where PAHs were analysed separately for PAHs associated with suspended particulate matter and the dissolved PAHs. They observed higher levels of the HMW compounds in the suspended particulate matter than in the dissolved PAHs. Furthermore, the presence of some HMW compounds that were detected in the water column of this study raises some concern. These are compounds (Benz[a]anthracene, Chrysene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[a]pyrene, and Indeno[1,2,3-cd]pyrene) noted as probable human carcinogens.

Most of the concentrations of the individual PAHs detected (at least, 62%) were above the WHO's limit of 0.05µg/L (Kielhorn & Boehncke, 1998). By using a global classification of PAH contamination in water (Ashayeri et al., 2018; Li et al., 2015), the outcome confirmed that these lagoons are highly polluted.

5.1.2 Concentration of PAHs in sediments and their distribution

Sediments are known to be the main sinks of contaminants in aquatic ecosystems. The protection of sediments is a necessity to achieve a healthy ecological status in the aquatic ecosystems, since contaminated sediments may adversely affect aquatic lives. Another reason sediment was surveyed in this study was its ability to accumulate pollutants over time. It becomes a resource, evident for long-term pollution. PAHs are not usually found as separate entities in environmental media. They are often considered as a mixture and so, the total concentration of their mixture is often used to describe their distribution (Edokpayi et al., 2016). With this understanding, the focus of discussion on the distribution and concentration of PAHs in the sediment samples was mainly on the total PAHs.

Below are discussions on the sediments from Benya, Brenu and Fosu Lagoons.

5.1.2.1 Benya Lagoon

The highest mean ∑PAH concentration was detected at BST2. Benya Lagoon accommodates many fishing vessels due to accessibility to the sea almost on daily basis. It is at this station (BST2) that most canoes and semi-industrial fishing vessels are packed, serving as a fishing quay for many of these fishing vessels. This station is also located under a heavy trafficked bridge. The main activities that goes on at BST2 are extensive coating of fishing vessels with coal tar, discharging of waste oil and domestic wastes directly into the lagoon, as well as fuel emissions (from vehicles and two-stroke engine outboard motors). All these are potential sources of PAHs that may be released into the lagoon. The lowest mean ∑PAH concentration was detected at BST3. PAH contamination at BST3 was attributed to the indiscriminate dumping and burning of solid wastes. A common feature found within the vicinities of most aquatic environments in Ghana. The refuse dump at BST3 is virtually seated on the lagoon, while burning goes on. When it rains, the sewage is directly discharged into the lagoon. The 3-ring PAHs dominated in this station, an indication of a local source of contamination, especially from sewage discharge or petroleum products. Burning of biomass/coal and sewage discharge have been reported by other studies (Arias et al., 2010; Obanya et al., 2019; Sarria-Villa et al., 2016; Souza et al., 2018; Zhang et al., 2016) to be local sources of PAH contamination in aquatic ecosystems. Jaward et al. (2012) and Francisco (2016) also confirmed that contribution to high PAH concentrations in sediments could be attributed to boating activity and spills. Another outcome from this study that confirmed the general input of PAH through boating activity was the presence of chrysene being the highest detected compound at all the 3 stations. Chrysene is known to be a natural constituent of coal tar.

The ∑PAHs contamination in sediment from the global point of view can be categorized into 4 classes: low; 0–100 µg/kg, moderate; 100–1,000 µg/kg, high; 1,000–5,000 µg/kg and very high >5,000 µg/kg (Gnandi et al., 2011; He et al., 2016). The total PAH contamination of sediments in Benya Lagoon (36-824 µg/kg) may be classified as of low to moderate level. This level of contamination was however, lower than Fosu Lagoon, another urban situated lagoon. The lower levels at Benya Lagoon could be because Benya Lagoon is an open lagoon; a characteristic that allows a constant flow into the sea, thereby reducing the chemical load. Boating activities in Benya Lagoon, burning of wastes, traffic emissions, and the improper management of liquid and solid effluents may be among the main sources of PAHs spread in the sediment of Benya Lagoon.

Observed seasonal variations from the studied sediment indicated significantly higher total PAHs and HMW compounds during the dry season (from December 2017 to March 2018) than the wet season (from April 2018 to November 2019). Higher levels of total PAHs in the dry season was in contrast with other similar studies (e.g., Santos et al., 2018; Sarria-Villa et al., 2016). They attributed higher levels of total PAHs during rainy seasons to runoffs. Runoffs are essential transport route for PAHs' input for coastal water bodies. Heavy rainfalls are able to readily transport these pollutants into aquatic systems from point sources, particularly in tropical and subtropical humid coastal areas (Zhang, Liang, Wang, Guan, & Zeng, 2012). The current result of higher levels of total PAHs during the dry season could be due to less influence from land-based sources of PAHs. Another reason was that the sediment was primarily dominated by the HMW compounds which were significantly higher during the dry season. Thus, their abundance may have also contributed to the increase in the total PAHs during the dry season. The abundance of the HMW PAHs was ascribed to their tendency to readily accumulate in sediments. These HMW PAHs have low solubility, high hydrophobicity, stable molecular structures, and less predisposed to degradation (Zakaria & Mahat, 2006). Frequent burning of biomass/coal and petroleum combustion may also have yielded high levels of HMW compounds. Since HMW PAHs are usually derived from combustion processes (Arias et al., 2010; Gilbert et al., 2006; Zhang et al., 2016).

The low levels of the LMW PAHs during the dry season was also ascribed to increased temperatures in this season. Such claim was supported by a significant negative correlation (Pearson R= -0.53, p< 0.001) that existed between temperature and the LMW PAHs. LMW PAHs do not persist for longer periods in sediments compared to the HMW PAHs. Increased temperatures may have led to biodegradation and photo degradation of LMW compounds by planktonic communities, thereby reducing the levels of the LMW PAHs in the sediment. Such dissociation of LMW PAHs from the sediments may have contributed to their solubility in the water column as well. Nguyen et al. (2014) and Zhang et al. (2016) proposed that low PAHs concentration is usually associated with microorganism's seasonal activities, high temperature, low atmospheric pressures and high index of photo degradation.

5.1.2.2 Brenu Lagoon

There are no previous records with respect to total or individual PAH concentrations in the sediments from Brenu Lagoon. However, the recorded values in this study were similar to those typical of coastal areas. According to the global classification of PAH contamination, the sediments from Brenu Lagoon (21–796 μg/kg) may be considered as low to moderately contaminated as registered in other coastal sediments (e.g., Ashayeri et al., 2018; Jaward et al., 2012; Benya sediment of this present study). It was also realised that the concentrations of LMW were significantly lower during the dry season as seen in Benya Lagoon. This was equally ascribed to the increase of temperature during the dry season. In contrast to the other two lagoons of this study, Brenu Lagoon is located in a rural area. It is therefore not under any intense anthropogenic activities compared to Benya and Fosu Lagoons. The level of human activities should have yielded insignificant contamination when compared to the global classification of PAH contamination. However, the level of contamination was within the moderate zone (between 100–1,000 μg/kg). The main source of PAHs within the littoral region of this lagoon could be vehicular emissions. Located near this lagoon is a road that connect the people of Brenu Akyenim and Ampenyi. There are other prevalent activities yielding to the release of PAHs in this lagoon, including, indiscriminate disposal and burning of domestic wastes. Burning of domestic wastes as a contributor to the release of PAHs was supported by the increase levels of HMW PAHs in Brenu sediment. Another reason to the increase levels of PAHs in Brenu Lagoon could be ascribed to PAHs being persistent in the environment and are long range transport pollutant. They are able to travel between the atmosphere and the earth's surface in recurrent, temperature-driven cycles of deposition and volatilisation (Cavalcante et al., 2012; He et al., 2014; Wania & Mackay, 1996).

Among the HMW congeners that were noted to be among the highest recorded compounds in the lagoon sediment were chrysene and Indeno [123-cd] pyrene. These are carcinogenic compounds. Even though Brenu Lagoon seems to be under less PAH- human activities due to its location, monitoring and regulation of PAHs are vital.

5.1.2.3 Fosu Lagoon

PAHs can be found in most urban areas, and the Fosu Lagoon is no exception. Among the three lagoons, the results indicated that Fosu sediment is the most polluted. The contamination level can be described as moderate to very high (190-24,801 μg/kg) according to the classification of PAH contamination. Compared to the global scale, PAHs concentrations have been reported to vary between 1 to 760,000 μg/kg, with modal concentrations of 1,000 to 10,000 μg/kg for rivers, estuaries, harbours, and coastal areas (Zakaria et al., 2002). In

Fosu Lagoon, the sediment sampled from FST1 depicted the highest contamination level. This station is the smallest part of the lagoon and considering the input of pollutant at this section, makes it more vulnerable compared to the other two stations. The level of PAH contamination may be attributed to incinerated household refuse, effluents and sewage discharge, unbent petroleum products from two nearby filling stations, piggery and vehicular emissions from the heavily trafficked bridge. These are all potential sources of PAHs that ultimately ends up in the sediment (Armah, Luginaah, et al., 2012; Gilbert et al., 2006). With Fosu Lagoon being a closed system, fresh input from the sea to dilute such inland-based pollutants is minimal and extremely intermittent.

Generally, the level of PAH contamination as compared to previous studies (Armah, Luginaah, et al., 2012; Gilbert et al., 2006) conducted in this lagoon is low. The reason ascribed to this was some managerial practices that were suggested to curb the rate at which this lagoon was being polluted through anthropogenic activities. In 2008, the Environmental Protection Agency (EPA) Ghana, advised that all activities in the lagoon be put to a halt, due to its highly polluted nature (Adjei et al., 2017). The mechanic shops located at the northern side of the lagoon was advised for example, to reuse their dirty oil in other functions rather than disposing directly into the lagoon; a common practice among the mechanics. Another reason to the low level of PAHs as compared to the previous studies may be due to uptake of PAHs by the outgrown of marshes in the lagoon. PAHs that accumulate in sediments could be used by emergent macrophytes (Ning et al., 2010). Some plants are also known to synthesize PAHs that act as growth hormones (Abdel-Shafy & Mansour, 2016). Therefore, uptake or synthesising of PAHs by outgrown marshes may have led to the depletion of PAHs concentration in the sediment. However, this may not be a conclusive claim. Further study to investigate the residual levels of PAHs in these marshes is needed.

The results from Fosu Lagoon revealed some seasonal variations in the concentrations of PAHs. Low levels of LMW compounds during the dry season was also attributed to increased temperature. Temperatures in the dry season were comparatively higher than those recorded for the wet season. In support of this, there was a significant negative correlation between temperature and LMW compounds (Pearson R= -0.37, $p< 0.001$). In the case of Fosu Lagoon, concentration of total PAHs was significantly higher in the wet season than in the dry season. This was the expected results because for a lagoon which receives almost all domestic and municipal effluents from the Cape Coast metropolis, there is the possibility of increased PAH input via runoffs during the rainy season. Big canals are connected directly to this lagoon. These are canals that receives untreated effluents all over the Cape Coast metropolis, and discharges them directly into the lagoon without any treatment. Runoffs are known to be one of the main sources of high molecular weight compounds in aquatic ecosystems (Essumang, Dodoo, Obiri, & Oduro, 2006; Jaward et al., 2012).

In summary, the total PAHs recorded in the studied sediments demonstrated both spatial and seasonal variations in the distribution of PAHs. The spatial distribution of PAHs in the sediments indicated a higher concentrations of total PAHs in areas with intense anthropogenic activities. The seasonal distribution projected the influence of temperature and runoffs in the dynamics of PAHs in the aquatic ecosystems. The lagoons also exhibited a wide range of PAH concentrations in their sediments, indicating heterogeneous levels of contamination. Such heterogeneity was ascribed to the diverse activities that exist within the littoral areas of these lagoons. The presence of carcinogenic compounds in the study sediments may pose health risks to humans that depend on these lagoons for livelihood.

5.1.3 Concentration and distribution of PAHs in fish tissues

Exposure routes of PAHs in fish include bio-concentration from water across their gills and skin (Gobas et al., 1999; Rojo-Nieto et al., 2014) and ingestion of PAH-contaminated particulate matter along with food materials (Bandowe et al., 2014; Meador et al., 1995). There are difficulties in accurately predicting bioaccumulation levels of PAHs in a whole fish, even with highly sophisticated models (Van der Oost et al., 2003). For this reason, this present study concentrated on residual levels of PAHs in some selected parts of the fish. This selection aided to better understand the mobility of PAHs in fish. So, in this section, residual levels of PAHs in fish fillet, head and visceral samples from Benya, Brenu and Fosu Lagoons were discussed.

5.1.3.1 Fish tissues from Benya Lagoon

Pyrene and fluorene were the abundant compounds detected in the fish tissues. The presence of pyrene is often used as a marker of the total PAHs contamination in fish (Ribeiro, Vollaire, Sanchez-Chardi, & Roche, 2005; Rojo-Nieto et al., 2014). Bandowe et al. (2014) also detected pyrene as one of the dominant compounds in fish tissues sampled along the coast of Ghana’s marine waters. Rojo-Nieto et al. (2014) also detected pyrene as a dominant compound when an integrated study was carried out to assess the fate and effects of PAHs in fish inhabiting a chronically polluted environment. The abundance of these compounds detected in this present study was attributed to two factors. Firstly, by the specific characteristics of Benya Lagoon with respect to PAHs distribution; and, secondly, by the level of biotransformation of some PAHs in this studied fish.

Concerning the study area (Benya Lagoon), the presence of pyrene and fluorene was in accordance with their distribution in the sediments (this study). In Benya sediment, fluorene was the dominant LMW compound and pyrene was among the three PAHs which most frequently showed higher concentrations in the sediment samples. Moreover, the concentrations of pyrene and fluorene were equally among the predominant PAHs that were detected in the water column of Benya Lagoon. Essumang (2010) also reported pyrene to be the dominant PAH detected in the water column of Benya Lagoon, although fluorene was not detected in his study. The presence of these major compounds in the water column of Benya Lagoon may also predict the abundance of these compounds in the atmosphere of the study area. Through dry and wet deposition, they may have been released into the water column. However, this claim can only be an assumption; an atmospheric diagnostic on the levels of PAHs in the study area should be investigated to verify this. Essumang, Ofori, Dodoo, & Adjei (2016) investigated PAHs levels in the Komenda Edina Eguafo Abrem (KEEA), of which the location of Benya Lagoon is found. However, Essumang et al. (2016) focused on indoor levels of PAHs. It should be noted however, that pyrene and fluorene were detected in their samples. All these results suggest that the main presence of these PAHs in all the fish tissues could be partly influenced by the sources of these compounds in the studied area. The two major PAHs (fluorene, pyrene) detected in the fish tissues can be said to be among the dominant PAHs detected in other environmental compartments of the lagoon (atmosphere, water and sediment). More than 80% of the compounds detected in the fish samples were also present in the water and sediment samples of Benya Lagoon.

Biotransformation was the other reason to which the concentrations and distribution of PAHs in fish tissues was ascribed to. For biotransformation, fish do not accumulate large amounts of PAHs due to their low uptake and can easily metabolise PAHs (Hellou & Payne, 1987; Van der Oost et al., 2003; Zelinkova & Wenzl, 2015). However, when fish is found in a chronic exposure environment, it leads to accumulation of PAHs in fish. Such environment generally leads to higher concentrations in the fish tissues (as in the case of this study). The HMW PAHs are metabolised and excreted more readily than the LMW PAHs (Baumard et al., 1999; Hellou & Warren, 1997; Neff, 2002). This probably accounted for the higher concentrations of the LMW PAHs in the fish visceral. The concentration of total PAHs detected in fish visceral was significantly higher than that in the fillet. The general increase of PAHs in the visceral could also be due to higher lipid content in the visceral compared to fillet tissues. Other similar studies (Bandowe et al., 2014; Hellou & Warren, 1997; Neff, 2002; Rojo-Nieto et al., 2014) observed higher concentrations in visceral (presented as gut or liver in other studies) than in the fillet (i.e. muscles in other studies). However, this cannot be a conclusive information as the current study did not analyse the lipid content in the fish visceral. Again, the higher residues of PAHs detected in visceral was an indication of recent and continuous PAH input (Zhao et al., 2014).

Detectable levels of HMW PAHs in all the fish tissues could indicate a less efficient metabolism and a higher potential toxicity. PAH metabolites are known to be more toxic than parent compounds, especially in an environment under constant PAH exposure. The condition factor of the fish samples from Benya Lagoon could be an added support to this claim. Generally, the fillet and visceral were dominated by LMW compounds while that of the head was dominated by HMW compounds. The head may have been enriched with HMW-PAHs through diverse uptake of dissolved contaminants through their gills. As a bentho-pelagic organism, *Sarotherodon melanotheron* could have ingested PAH-contaminated sediment or food (from the sediment compartment), and uptake of PAHs through respiration and ingestion of food (from the water column compartment).

There was also an observed significant seasonal variation. Fluorene was the only LMW compound that exhibited significantly higher concentration in the wet season than the dry season. This higher concentration was ascribed to wet atmospheric deposition of fluorene as well as low temperatures in the wet season, making this compound more stable in the ecosystem.

5.1.3.2 Fish tissues from Brenu Lagoon

Fluorene, acenaphthylene and Benzo[a]pyrene were among the predominant compounds in the fish tissues. The abundance of these compounds was attributed to the distribution of PAHs in the water column and sediment of Brenu Lagoon. Fluorene, acenaphthylene and Benzo[a]pyrene were among the dominant PAHs detected in Brenu sediment, and in the water column, they were not left out either. Generally more than 80% of the compounds detected in the fish samples were also present in the water and sediment samples of Brenu

Lagoon. The level of PAHs in the fish tissues can therefore be said to have been transferred from water, sediment, and the diet. The total PAHs in the fish head was significantly higher than the total PAHs in the visceral and fillet. Zhao et al. (2014) also measured higher levels of PAH residues in fish head (gill) than in the fillet (muscle) of silver carp. Similar results was reported on Benya fish tissues of this current study. The relative higher levels of PAHs in the Brenu fish head may have indicated tissue distribution of PAHs first to the gills and then to the other internal organs of the fish. For fish, the water column is normally the principal source of exposure for organic compounds with low Kow (e.g., the LMW PAHs), while sediment particles (as food) contribute mainly to bioaccumulation for compounds with high Kow (Balk et al., 2011; Van der Oost et al., 2003). The LMW compounds (3-ring PAHs) dominated the fillet and visceral, while the HMW (4-ring PAHs) dominted the head. Again, with higher levels of PAHs detected in the fish head, it is likely that the principal source of PAHs to the fish tissues was from the sediment. However, the higher concentrations of 3-ring compounds (LMW) in both fillet and visceral gave further deductions. The abundance of the LMW PAHs in the fillet can mean a transfer of LMW PAHs from the water column through the skin, since the water column was dominated by the LMW PAHs. Besides, the concentration of fluorene, one of the dominant compouds detected in the water coulmn, was higher in the fillet than its concentration detected in the head and visceral. The adsorption of dissolved contaminants are exposed to the fillet through diffusion of contaminants from water across their skin (Bandowe et al., 2014; Cheung et al., 2007). The higher levels of the LMW PAHs in the visceral also indicated biotransformation of PAHs into water soluble components; a reflection that fish can metabolise PAHs (Hellou & Warren, 1997; Inomata et al., 2012; Tudoran & Putz, 2012; Zelinkova & Wenzl, 2015).

In terms of seasonal variations, the occurrence of all the LMW compounds were significantly higher during the wet season. The high concentration of the LMW compounds in the wet season was attributed to low temperatures associated with such season. Such low temperatures may have contributed to their stability and their resistance to evaporation (Ashayeri et al., 2018; Hu et al., 2010; Omayma et al., 2016).

5.1.3.3 Fish tissues from Fosu Lagoon

As observed from the other two lagoons in this study, the concentrations of PAHs in the fish tissues from Fosu Lagoon also indicated that both the water column and sediments were potential sources of PAHs. The dominance of fluorene, fluoranthene and pyrene in the fish tissues could be related to the distribution of these compounds in the sediment and water column of Fosu Lagoon. Bioconcentration from water through the skin and gills, and ingestion of food or sediments are prospective pathways to accumulate PAHs in fish. The extent of bioaccumulation also hinges on the organism's general behaviour, food habits, trophic level, fat contents and metabolic capabilities (Cheung et al., 2007; Meador et al., 1995; Van der Oost et al., 2003). As a bentho-pelagic organism, *Sarotherodon melanotheron*, may exhibit significantly higher concentrations of PAHs due to its close proximity to substrate sediment with high levels of adsorbed PAHs. Shi et al. (2016) also reported on significantly higher concentrations of PAHs in both blood porgy (demersal fish) and cutlass fish species (bentho-pelagic) than other species in their study. The reason they ascribed to their finding was that these species were more likely to contact and take up PAHs from sediments. Significantly, lower concentration of total PAHs was found in the fish fillet from the Fosu Lagoon as compared to that in the head and visceral samples. Lower lipid contents in the fillet as compared to the head and visceral may partly be the reason for the lowest level of PAHs detected in the fish fillet (Bandowe et al., 2014; Rojo-Nieto et al., 2014; Shi et al., 2016).

The fillet and the visceral were dominated by 3-ring (LMW) PAH compounds, while the head was dominated by 4-ring (HMW) compounds. This result was identical to the findings from Benya and Brenu fish samples, as well as that of Shi et al. (2016). They observed that among the different PAH profiles, 3-ring compounds (LMW) dominated the distribution in fish muscles. It is possible that the source of uptake of the LMW and the HMW compounds by the fish were from the water column and sediment respectively of Fosu Lagoon. Usually, the water column is the main source of exposure for the LMW compounds, while sediment particles (as food) contribute mainly to the bioaccumulation of the HMW compounds (Balk et al., 2011; Van der Oost et al., 2003). The abundance of the the 3-ring compounds in the visceral further confirmed the capability of fish species to metabolise or biotransform PAHs. Based on these findings, it will be essential to investigate the residual levels of PAH metabolites in the fish species from these studied lagoons. With the background knowledge that some of these metabolites are more toxic than the parent PAHs (Jung et al., 2010), makes such investigations even more compelling.

For seasonal trends, the LMW compounds were significantly higher during the wet season, as observed in the other two studied lagoons.

In summary, all the 16 priority PAHs were detected in the fish tissues. The total concentrations of PAHs detected in fish fillet were relatively lower than those detected in the fish head and visceral. Both the water column and sediments were noted to be potential sources of PAHs to the fish. One key observation was that the dominating compounds in both water column and sediments were equally dominating compounds in the different fish tissues, suggesting residual transfer from the fish's immediate environment. Fluorene, acenaphthylene, fluoranthene, pyrene, and Benzo[a]pyrene were among the dominant compounds detected in the studied lagoons. Other factors such as the organism's habitat, metabolism, lipid content, and feeding behaviours, all played a key role in the storage and biotransformation of PAHs in *Sarotherodon melanotheron*.

5.2 Bioaccumulation

Biota-sediment accumulation factors (BSAFs) are known to be appropriate for describing bioaccumulation of contaminants in sediment for aquatic food webs. There are non-equilibrium conditions between both the sediment and biota (fish in this case), and sediment and the overlying water (Ankley et al., 1992; Thomann, Connolly, & Parkerton, 1992). This work however, focussed on non-equilibrium conditions between the sediment and fish (specifically, fish fillet). The fish fillet was the focus because that is mostly the part consumed by both adults and children.

From the results, the bioaccumulation factors (BAFs) of ∑PAHs in all the fish tissues from Fosu Lagoon were generally lower than fish tissues from Benya and Brenu Lagoons. This output was attributed to the fact that the sediment samples of Fosu is mainly dominated by the HMW PAHs, making the sediment composition more hydrophobic. The higher the hydrophobicity, the more readily PAH will bind to the sediments containing higher organic matter (Baumard et al., 1999; Montuori et al., 2016; Ololade et al., 2017; Viguri et al., 2002; Xue et al., 2016). In such a situation, there will be less partition of PAHs from sediment to pore water that may be subsequently accumulated by organisms. These results were further supported by the fact that sediment from Fosu Lagoon recorded the highest percentage of total organic carbon. The capacity of accumulation by the fish was done by taking into account the lipid content. Such evaluation yielded the biota-sediment accumulation factors (BSAFs). The present results showed considerably, greater BSAFs in fish fillet sampled from Benya than those of Brenu and Fosu Lagoons. As an open lagoon, Benya is constantly in contact with the sea, hence, under tidal influence (Biney, 1990). Such influence may lead to a relatively higher mixing of contaminated sediment with the water column as compared to the other two closed lagoons, Brenu and Fosu. Such activity may have led to the readily availability of PAH-contaminated sediment to fish samples in Benya than in Brenu and Fosu Lagoons.

The BSAFs for ∑PAHs of black-chinned tilapia in these studied lagoons were also compared with other studies that used the same method for calculating BSAFs. The BSAFs in this current study were higher than the values reported for sunfish (*Lepomis sp.*) (1×10^{-5}–5×10^{-3}; Thomann & Komlos, 1999), lake trout (*Salvelinus namaycush siscowet*) (1×10^{-4}–7×10^{-3}; Burkhard & Lukasewycz, 2000), and snakehead (0.91; Kwok et al., 2013). For example, snakehead is principally a carnivorous, and feeds on other fish for protein (Paripatananont, 2009). This feeding behaviour allows less exposure to sediment for snakeheads compared to black-chinned tilapia; a bentho-pelagic species. Thus, lower BSAF in snakehead. However, the values of BSAF in the studied fish were lower than that in plankton samples (Froehner et al., 2018). The reason being that PAHs are enzymatically metabolized by fish (Hellou & Warren, 1997; Inomata et al., 2012; Tudoran & Putz, 2012; Zelinkova & Wenzl, 2015). Thus, the lower BSAF in fish might have resulted from biochemical transformation. These findings basically bring out the argument that feeding habit and habitat can be contributing factors to the extent of accumulation of PAHs from sediment to fish.

5.3 Effect of physicochemical characteristics on PAHs

The dynamic of PAHs in an aquatic ecosystem can be determined by the compound’s physicochemical parameters (e.g. vapour pressure, log Kow and solubility) (Ahmed et al., 2017). Yet, some other environmental characteristics should be considered when looking at factors contributing to the behaviour of PAHs in an aquatic ecosystem. In this study, pH, temperature, organic matter content and salinity were considered. These variables play vital role in organic compounds’ adsorption and distribution in an aquatic ecosystem. The relationship between total concentrations of PAHs and these physicochemical parameters were analysed. %TOC ($R^2 = 0.08$) and pH ($R^2 = 0.12$) were the parameters that demonstrated a significant ($p< 0.01$) positive linear relationship with total PAHs in lagoon sediments.

According to several coastal sediments studies, authors have reported a strong positive linear relationship between PAHs abundance in sediments and TOC values (Baumard et al., 1999; Culotta et al., 2006; He et al., 2016; Montuori et al., 2016; Ololade et al., 2017; Viguri, Verde, & Irabien, 2002; Xue et al., 2016). However, in this study, there existed weak positive linear relationship between the total PAHs concentrations and TOC values in the sediments. Lack of strong positive correlation indicated that the PAHs may have been recently generated and, therefore, were yet to fully partition into organic matter in the sediments. It could also mean that the concentration of PAHs in the sediments were related to the sources of contamination more than the organic matter content and pH. The source of contamination being the prominent factor was observed in all the studied lagoons. The concentration of the total PAHs were highest at stations (e.g., BST2, BRST3 and FST1) where pressures of human activities were intense. Notwithstanding, %TOC and pH should not be ignored when monitoring and regulating inputs of PAHs in these studied lagoons. These variables can be controlling factors for the sorption of PAHs onto the sediments. They could affect the distribution of PAHs in an aquatic ecosystem, and consequently, influence the availability and accumulation of PAHs in the biota living in the aquatic ecosystem. Hence, TOC and pH should be considered when looking at the dynamics of PAHs in these sediments.

The fat content of fillet tissues were also analysed to predict the contribution of fat in PAHs absorption in fish. The results showed that there was no correlation between the total PAHs and fat content in fish from Benya and Brenu Lagoons, but there was a significant correlation when fish samples from Fosu Lagoon were analysed. Generally, because PAHs are lipophilic, they prefer to be accumulated in tissues with higher fat contents. Several studies have agreed to this fact; where there existed a correlation between fat content and PAHs (Abdel-Shafy & Mansour, 2016; Bandowe et al., 2014; Essumang et al., 2012; Essumang, Dodoo, & Adjei, 2013; Fairey et al., 1997). However, others have reported otherwise (Otchere, 2005; Zhao et al., 2014). In this current study, both claims were observed. The results of fish samples from Benya and Brenu Lagoons could indicate that fat contents in fillet tissues were not the vital factor in PAHs accumulation in the fish. In essence, the major PAHs uptake in fish from Benya and Brenu Lagoons was not through the direct route of partitioning to the skin (fillet), but through food ingestion. Such claim was further supported by the fact that concentrations in fish head were significantly higher than in fish fillet. For PAHs uptake by fish from Fosu Lagoon, one can say the skin is one of the route exposure. The concentration of total PAHs in the fish fillet from Fosu were significantly higher than those measured from Benya and Brenu Lagoons.

5.4 Source identification by PAHs isomer ratios

Variations in the concentration and type of the individual PAHs at the various sampling sites reflected the main activities yielding to the release of PAHs into these studied lagoons. Accurately identifying the sources of PAHs in a water body can be challenging. There exist countless possible sources and processes that PAHs undergo before being deposited into an environmental media such as sediments, water or fish. For this reason, molecular patterns serving as fingerprints for source identification are generated, making it possible to hypothesise which processes generated which type of PAHs by studying their distribution in that environmental media (Culotta et al., 2006; Santos et al., 2017; Stogiannidis & Laane, 2013; Zakaria et al., 2002). Molecular indices that hinge on the ratios of some PAHs have been extensively used to differentiate PAHs from pyrogenic and petrogenic sources (Ashayeri et al., 2018; Edokpayi et al., 2016; Santos et al., 2017; Yunker et al., 2002). Perylene, usually in lower concentrations is one of the fingerprint to point to biogenic/diagenetic source of PAHs (Bouloubassi et al., 2001; Culotta et al., 2006; Sauer et al., 1993). However, perylene was not detected in any of the lagoons, making pyrogenic and petrogenic to be the two main sources of concern in the studied lagoons.

5.4.1 Sources of PAHs in Benya Lagoon

In Benya sediment, the values of LMW/HMW, Ant/(Ant + Phe) and Phe/Ant suggested that the PAHs found in the sediment were of pyrogenic sources. This was attributed to biomass/coal combustion through indiscriminate disposal and burning of wastes within the vicinity of the lagoon. The values of Flu/(Flu + Pyr) and Flu/Pyr also suggested a source combination of incomplete combustion of petroleum (at BST1 and BST2) and petroleum sources (at BST3). Pyrogenic sources of PAHs through incomplete combustion of petroleum at BST1 and BST2 could be attributed to vehicular emissions. These two stations are located under the heavy trafficked bridge; and the ratio of BaP/BP (>0.6) further supported the potential source of PAHs from traffic sources. Studies by Tay & Biney (2013) and Wilcke (2007) also confirmed that PAH levels dropped exponentially with increase in distance from the roads due to the reduced vehicular emissions. Another contributing factor to the source of PAHs from incomplete combustion of petroleum was the use of the two-stroke engine outboard motors on the lagoon. Intense ship traffic is found at BST1 and BST2 of Benya Lagoon. PAH contamination due to incomplete combustion of petroleum in the Venice lagoon, Italy (Secco et al., 2005), Guan River estuary, China (He et al., 2014) and other coastal water bodies along the coast belt of Ghana (Essumang, 2010) was also ascribed to boat or ship traffic.

At BST3, PAHs emanating from petroleum sources could be related to used oil (from premix fuel) or coal tar directly discharged into this section of the lagoon. Such discharges may have resulted from washing and coal tarring of fishing vessels done at this section of the lagoon. The ratio of ∑PAHs(4 rings)/∑PAHs(5-6 rings) further suggested that contamination of PAHs at this station is of a local source. Aside the petroleum sources, one other activity that may have contributed to local contamination is indiscriminate burning of domestic waste. The refuse dump is actually seated on this part of the lagoon (BST3). When it rains, the sewage discharges directly into the lagoon. Souza et al. (2018) reported that biomass burning was attributed to local source of PAH contamination in Poxim river, Brazil. PAHs of pyrogenic origins are attributed to high temperatures used in incomplete combustion of biomass. Hence, the ratio of Phe/Ant (<10) indicating pyrogenic source could also emanate from the indiscriminate burning of wastes. Other studies (e.g., Ashayeri et al., 2018; Edokpayi et al., 2016; Gilbert et al., 2006) have ascribed the pyrogenic PAHs found in aquatic sediments to be originated from the combustion of biomass.

From the fish samples, sources of PAHs were also attributed to both pyrogenic and petrogenic sources. The values of LMW/HMW in fish samples, further explained these sources. The BFF and BFV portrayed petrogenic sources, while BFH portrayed pyrogenic sources. It also gave an idea of the fishes being polluted by both the water column and the sediment. The LMW dominated the water column of this lagoon, thus, the possibility of uptake of the LMW PAHs by the fish. Also, the sediment was dominated by the HMW compounds, therefore, it could be that the presence of the HMW compounds in the fish head were from ingestion of contaminated sediment. The contribution of PAH contamination by traffic sources in the fish samples were comparatively higher than the traffic contribution to the sediment. The values of BaP/BP in the fish samples were about a magnitude of at least 5, when compared to the sediment samples. Such high values could be attributed to atmospheric deposition or precipitation of vehicular emissions into the water column, where they become readily available to the fish. Although fish can bio-transform PAHs to water-soluble intermediates, the occurrence of parent PAHs in fish tissues had been related to recent episodes of pollution exposure in surrounding environment (Pointet & Milliet, 2000). Also, as to whether the PAHs contamination was of local or longer distance transport, the ratio of ∑PAHs(4 rings)/∑PAHs(5-6 rings) depicted both local and longer distance transport. The local source could suggest uptake from its surrounding environment, while the longer distance transport could be attributed to the semi-volatility nature of PAHs; the ability of PAHs to disperse over long distance (Cavalcante et al., 2012; He et al., 2014; Wania & Mackay, 1996).

5.4.2 Sources of PAHs in Brenu Lagoon

For Brenu sediment, the values of LMW/HMW, Ant/(Ant + Phe) and Phe/Ant suggested that the PAHs found in the sediment were of pyrogenic sources. However, the values of Flu/(Flu + Pyr) and Flu/Pyr suggested a source combination of incomplete combustion of petroleum and petroleum sources. Unlike Benya Lagoon where many two-stroke engine fishing vessels use the lagoon as a fishing quay, this is a close lagoon and the fishing vessels used on this water are small canoes paddled by wooden rudder. Therefore, the activities that may yield petrogenic sources seem not to be clear for Brenu Lagoon. It may be that the petrogenic PAHs are introduced into this lagoon through accidental oil spills, discharge from fishing vessels, municipal and urban runoff (Zakaria et al., 2002). Urban runoff with elevated levels of PAHs have been reported for roadside soils in Kumasi metropolis of Ghana (Essumang et al., 2006). Brenu Lagoon is naturally or artificially opened to the sea when it overflows its bank. During this process, there is an exchange of contaminants such as oil spills, from the marine environment. It should be noted that the sea opposite Brenu Lagoon is as well used for extensive fishing, although it is not as intense as that of Benya Lagoon. At BRST3, the value of Flu/(Flu + Pyr) (between 0.4-0.5) was an expected result. Such value depicted incomplete combustion of petroleum as the source of PAHs to this section of the lagoon. There is a road beside this section which connects the people of Brenu Akyenim and Ampenyi. A busy road used by a lot of vehicles (especially, taxis). The ratio of BaP/BP > 0.6 at all the sampling stations supported the claim that traffic sources contribute to the release of PAHs into this lagoon. At BRST3, the value of BaP/BP was 6 times higher than the other two sampling stations; confirming that this station may receive higher levels of vehicular emissions.

The pyrogenic sources on the other hand, was attributed to indiscriminate disposal and burning of domestic wastes around the Brenu Lagoon. The values of ∑PAHs(4 rings)/∑PAHs(5-6 rings) suggested that PAH contamination in Brenu Lagoon was of both local and long distance sources. The local source could be mainly attributed to the indiscriminate disposal and burning of domestic wastes at the banks of the lagoon, while the long distance source was attributed to the semi-volatile nature of PAHs. As persistent organic compounds, can be transported over long distances (Cavalcante et al., 2012; Francisco, 2016; He et al., 2014; Wania & Mackay, 1996). Therefore, although Brenu Lagoon is not associated with much anthropogenic activities as compared to Benya and Fosu Lagoons, there were still elevated levels of PAHs in its ecosystem that calls for periodic monitoring.

For the fish sampled from Brenu Lagoon, the values of the diagnostic ratios also depicted both petrogenic and pyrogenic. The values of Ant/(Ant + Phe) (>0.1) and Phe/Ant (<10) suggested that the PAHs found in all the fish tissues were of pyrogenic sources, while ∑LMW/∑HMW (>1) of fish fillet suggested a petrogenic source. The ratio of ∑LMW/∑HMW (>1) in the water column also indicated a petrogenic source as well. This can further support the claim that the fishes sampled from this lagoon were exposed to PAHs contamination both in the sediment and the water column. With the fish fillet, there is the possibility of exchange of LMW compounds from the water column through the skin (fillet) of the fish. The values of Flu/(Flu + Pyr) (<0.4) and Flu/Pyr (<1.0) also suggested a petrogenic source.

The BaP/BP was used to primarily, differentiate between combustion sources (either of petroleum products or biomass) in the fish tissues from Brenu Lagoon. The values of BaP/BP >0.6 in the fish tissues suggested potential source of PAHs from vehicular emissions. Thus, through wet and dry deposition, PAHs are released into Brenu Lagoon. The values of ∑PAHs(4 rings)/∑PAHs(5-6 rings) also suggested that the origin of PAHs in the fish tissues was of both local and long distance sources.

5.4.3 Sources of PAHs in Fosu Lagoon

The values of the diagnostic ratios {LMW/HMW, Ant/(Ant + Phe), Phe/Ant, Flu/(Flu + Pyr) and Flu/Pyr} suggested that the PAHs found in the sediment were of both pyrogenic and petrogenic sources as seen in the other two lagoons mentioned above. The main pyrogenic sources in Fosu sediment emanated from indiscriminate disposal of wastes, combustion of domestic and municipal wastes, vehicular emissions and hospital effluents especially, at FST1 and FST2. These sources were also eminent when Gilbert et al. (2006) conducted a research on the characterization and source assessment of PAHs in the sediment of Fosu Lagoon. FST3 was distinguished from the other two stations. The ratios of Flu/(Flu + Pyr) and Flu/Pyr at FST3, suggested that PAHs input at this section of Fosu Lagoon originated mainly from incomplete combustion of petroleum and petrogenic sources. This was supported by the various activities that existed at this station. FST3 is where the mechanic shops are located. Such shops are identified with activities such as, indiscriminate disposal of dirty oil directly into the lagoon, all kinds of vehicular emissions and oil spillage; and all these are potential sources of PAHs.

Among the three stations, local source of PAH contamination was identified with FST3, while the contamination at the other two stations suggested longer distance sources. Local source of contamination could strongly be related to the activities carried out by the mechanic shops along the banks of this lagoon which appears to be the main activity that could release petrogenic PAHs. FST1 and FST2 on the other hand, receive diverse untreated domestic, municipal and hospital effluents through big canals (Appendix D;III and IV) connected directly into the lagoon (Armah, Luginaah, et al., 2012;

Gilbert et al., 2006). The ratio of BaP/BP >0.6, further supported vehicular emissions as potential source of PAHs in Fosu Lagoon. The lagoon is situated in an area that has multiple heavily travelled roadways, activities capable of affecting the levels of PAHs released into this lagoon.

The values of LMW/HMW, Ant/(Ant + Phe) and Phe/Ant in Fosu fish tissues suggested pyrogenic sources of PAHs. That of Flu/(Flu + Pyr) and Flu/Pyr suggested a source combination of petroleum (FFF and FFV) and combustion of biomass/coal (FFH).

FFV was distinguished from the other two fish parts when BaP/BP ratio was considered. The ability of fish to metabolize PAHs may explain why within the visceral, the ratio of BaP/BP indicated non-traffic sources although both the fillet and head suggested otherwise. Consequently, future risk assessment studies should consider the fate and effects of the PAH metabolites to aquatic biota, considering metabolism as a vital excretion pathway (Adeyemo, 2011). Besides, high value of BaP/BP in the fish fillet compared to the values within the sediment could suggest how readily vehicular emissions dissolve in the water column after atmospheric deposition or rain washout. The HMW PAHs of pyrogenic origin reach aquatic environments by direct atmospheric deposition or via contaminated soil (Budzinski, Jones, Bellocq, Piérard, & Garrigues, 1997; Morillo et al., 2007), while LMW pyrogenic PAHs are mainly introduced into aquatic environments by rain washout (Ollivon, Blanchard, & Garban, 1999). The values of ∑PAHs(4 rings)/∑PAHs(5-6 rings) were an indication of both local and long distance sources of PAH contamination. Confirming that human activities that are carried out both far and near are potential sources of PAHs in Fosu Lagoon.

From the above discussion, the main sources of PAHs into the studied lagoons were of both pyrogenic (incomplete combustion of petroleum products and biomass) and petrogenic origins. This identification could aid policy makers in their monitoring and regulation of PAH inputs into these coastal lagoons. Subsequently, such information on the sources could assist policy makers to apportion appropriate measures for remediation activities. It was observed from the evaluation of the diagnostic ratios that the bulk of PAHs in these aquatic ecosystems emanated from sewage outfalls, industrial wastewater, atmospheric fallout, surface runoff from roads and land surfaces, oil spillage, vehicular emissions (e.g., automobiles and trucks), and biomass/coal burning (e.g., solid wastes). They were of both local and distance sources.

5.5 Evaluation of ecological risk

Ecological risk assessment has been shown to be a useful tool to characterize the risk of PAHs to organisms and the ecosystem at large (Li et al., 2015; Wu et al., 2011). Interpretive tools are used to determine if sediment-associated contaminants are at concentrations which could, potentially, impair the designated uses of the aquatic environment. In this respect, the Sediment Quality Guidelines (SQGs), Toxicity Equivalent Quotient (TEQ) and Risk Quotient (RQ) provide scientifically defensible bases for evaluating the potential effects of sediment-associated contaminants on aquatic organisms (Ashayeri et al., 2018; Cao et al., 2010; Cardellicchio et al., 2007; Kalf et al., 1997; Long & MacDonald, 1998).

PAHs present in the water column have the potential to bio-accumulate, especially in fishes or other organisms that freely live in the water column. Thus, posing a potential risk for aquatic ecosystem. The TEQ and RQ were applied in this current study to evaluate the risk of aquatic biota's exposure to PAHs in the water column. The maximum and minimum TEQ values were found at Fosu and Brenu Lagoons respectively. Due to high TEF value for BaP (1), it was the major contributor to TEQ for carcinogenic compounds (TEQ carc) in the ecosystem. The TEQ carc values recorded in the water columns (between 0.11 to 0.23 µg/L) were generally higher than those recorded by Ashayeri et al. (2018) for Shadegan wetland, Iran (0.0022 µg/L and 0.0049 µg/L). Also, the RQ_{NCS} and RQ_{MPCS} for all the individual compounds from all the three lagoons were less than 1, showing low ecological risk level in the study areas. Flu, DahA and BP were not detected in any of the water samples, suggesting that no obvious risk was associated with these compounds so far as the water column is concern. However, for Fosu water samples, the sum of the PAHs for negligible concentrations showed moderate ecological risk for aquatic life ($RQ_{\sum PAHs\ (NCs)} > 1$). The risk associated with the sum of PAHs for maximum concentrations $\{RQ_{\sum PAHs\ (MPCs)}\}$ was less than 1 for all the studied lagoons. $RQ_{\sum PAHs\ (MPCs)} < 1$ also indicated a moderate ecological risk for aquatic life.

5.5.1 Ecological risk assessment for Benya sediment

The concentrations of ∑PAHs in the sediment samples exhibited values lower than ERL, indicating no effect of total PAHs on organisms (Table 16). However, for the individual compounds, fluorene (F) and Dibenz[a,h]anthracene (DahA) concentrations at all the three stations were greater than ERL but below ERM. Such concentrations indicated that F and DahA can pose mild toxic effect on aquatic organisms in Benya Lagoon. The concentration of fluorene in Mvudi River sediment (South Africa) also exceeded ERL, but was below the ERM value (Edokpayi et al., 2016).

Indicating a mild toxic effect on aquatic biota present in the Mvudi River. The results from the TEL/PEL SQGs also revealed that fluorene could cause mild negative biological effects in Benya Lagoon. It should be noted that the values for both ERL and TEL explain similar effects. However, their values may differ for the same sediment being considered (Cardellicchio et al., 2007). In this current study, both SQGs were considered to effectively aid in monitoring and regulating the levels of PAHs in the studied lagoons.

The simultaneous and long-term exposure to a mixture of PAH compounds can cause various adverse biological effects on organisms and aquatic systems, which is not reflected by SQGs. Rather, mERM-Q and Toxic equivalent quotient (TEQ) are used to assess the likely combined toxicity or biological risk for exposure to mixed PAHs in sediments (Ashayeri et al., 2018; Cardellicchio et al., 2007; Long & MacDonald, 1998; Long et al., 1995). As shown in Figure 38, mERM-Q results were lower than 0.1 at all stations. This indicated low probability for negative biological effects in this lagoon. The toxicity assessment of sediments was performed based on both total PAHs and the total concentration of the seven carcinogenic PAHs (TEQ carc) encountered. Consequently, TEQ carc values at all the three stations (32.88 to 41.46 µg/kg) were below the threshold set by Canadian soil quality guidelines for protection of ecosystem and human health according to carcinogenic effects of PAH. A safe TEQ carc value should be below 600 µg/kg (Yu et al., 2015). The Canadian soil quality threshold was considered in this study because it is internationally recognised. Benzo[a]pyrene was the major contributor to TEQ car in the ecosystem, maybe due to its high TEF value of 1 (Ashayeri et al., 2018).

One other ecological assessment that was considered in this study was risk quotient (RQ). At all the three stations, naphthalene, acenaphthylene, acenaphthene, fluorene, anthracene, phenanthrene, pyrene, chrysene, benzo[b]fluoranthene, and Benzo[a]pyrene were the individual compounds that were likely to portray moderate risk to the ecosystem. Such a deduction was made since their $RQ_{NCS} > 1$ and $RQ_{MPCS} < 1$. Cao et al. (2010) and Liu et al. (2013) also used this same method to evaluate the ecological risk of sediments in some rivers in China. Liu et al. (2013) observed that the concentrations of these individual PAHs in Haihe River sediment, China also had the potential to cause moderate risk to aquatic lives. In addition, when total PAHs were considered for this study, the $RQ_{\sum PAHs(NCs)}$ at all the sampling sites exhibited moderate risk. However, $RQ_{\sum PAHs(MPCs)}$ exhibited moderate risk at BST1 and BST2, but not at BST3. The sampling site with the highest value for both $RQ_{\sum PAHs(NCs)}$ and $RQ_{\sum PAHs(MPCs)}$ was also noted to be the most polluted area of Benya Lagoon (BST2). Liu et al. (2013) also reported that in Fuhe and Fuyang River sediments, the most polluted areas were also noted to be the areas with the highest ecological risk. The $RQ_{\sum PAHs(MPCs)}$ in the sediment of Luan River, China was different from the observations in Benya sediment. The $RQ_{\sum PAHs(MPCs)}$ for Luan River sediment was zero (Cao et al., 2010). Therefore, comparing Luan River sediment to Benya sediment, the PAHs pollutants of Benya sediment exhibited higher ecological risk.

5.5.2 Ecological risk assessment for Brenu sediment

For Brenu sediment, both individual and ∑PAHs concentrations in sediment samples at all the three stations exhibited values lower than ERL and TEL, indicating no effect of total PAHs on organisms (Table 16). As observed in Benya, mERM-Q values for Brenu sediment were also lower than 0.1 at all stations. An indication of low probability for negative biological effects in this lagoon when considering mixed toxicity. The sediment of Brenu Lagoon also recorded TEQ carc values below the threshold set by the Canadian soil quality guidelines (600 µg/kg) according to carcinogenic effects of PAH (Yu et al., 2015). Benzo[a]pyrene was also the major contributor to TEQ car in the ecosystem, just as it was seen in Benya sediment. Considering RQ, all the three stations exhibited similar trends. Naphthalene, acenaphthylene, acenaphthene, fluorene, anthracene, phenanthrene, pyrene, benzo[a]anthracene and benzo[b]fluoranthene were the individual compounds that exhibited moderate ecological risk to aquatic life in this lagoon (RQ_{NCs} >1). The ecological risk at all the three stations was a moderate one so far as total PAHs was concern.

5.5.3 Ecological risk assessment for Fosu sediment

The ∑PAHs concentrations in sediment samples at all the three stations exhibited values lower than ERL, indicating that the sum of PAHs are not dangerous to organisms in this lagoon. However, fluorene and dibenz[a,h]anthracene concentrations indicated otherwise. The concentrations of these aforementioned individual PAHs at FST1 showed the possibility of causing harmful effects occasionally, as their values were between ERL-ERM. Also, acenaphthene, acenaphthylene, fluorene, and dibenz[a,h]anthracene had values above TEL but below PEL. These results revealed that these individual compounds could cause occasional negative biological effects in Fosu Lagoon. The concentration of total PAHs (1249±3881 µg/kg) at FST1 also depicted an occasional negative biological effects in the ecosystem.

The value of mERM-Q (Figure 38) was also lower than 0.1 at all stations of Fosu Lagoon. This indicated low probability for negative biological effects in this lagoon. Also, the toxicity assessment of sediments based on TEQ carc for all the sampling stations (ranging from 52 to 162 µg/kg; Appendix C4) were below the threshold set by Canadian soil quality guidelines (Yu et al., 2015). Benzo[a]pyrene was the major contributor to TEQ car in the ecosystem.

Naph, Ace, Acp, F, Ant, Phe, Flu, Pyr, BaA, BbF, and BaP were the individual compounds the exhibited moderate ecological risk (RQ_{NCs} >1) to aquatic life in the Fosu Lagoon. Pyrene at FST1 exhibited significant or high risk (RQ_{MPCs} >1). Pyrene was also recorded to be of high ecological risk (RQ_{MPCs} >1) to Haihe River sediment (Liu et al., 2013). Consequently, the sum of PAHs for both negligible {$RQ_{\sum PAHs(NCs)}$ >1} and maximum permissible {$RQ_{\sum PAHs(MPCs)}$ >1} concentrations showed moderate and high ecological risks respectively for aquatic life. It was also realised that the highest ecological risk observed in Fosu sediment was also at the area (FST1) where the highest pollution occurred.

In summary, water samples from these lagoons exhibited a moderate ecological risk for aquatic life when PAHs were looked at synergistically ($RQ_{\sum PAHs\ (NCs)}$ >1 or $RQ_{\sum PAHs(MPCs)}$ <1). For the sediment samples, all the three lagoons recorded more than 50 % of individual compounds showing a moderate level of ecological risks for aquatic life. Brenu generally recorded the least while Fosu, (especially, FST1) recorded significantly high risk when certain guidelines were considered. Brenu Lagoon’s lower risk to aquatic life could be drawn from the fact that this lagoon is under less influence of human activities as compared to the other two urban lagoons. One other thing that was noted in all the three lagoons was that, BaP was the major contributor to TEQ carc. The elevated levels of BaP in these studied lagoons were ascribed to increase in combustion of biomass/coal within the vicinity of the lagoons. BaP is known to be released into environmental media mainly through the incomplete combustion of biomass/coal (Belis et al., 2011; Hussain et al., 2015; Tu et al., 2018).

It was also noted that different ecological risk assessments gave different outcomes. For policy makers to effectively and comprehensively tackle inputs of PAHs in these lagoons, it will be advisable to consider these possible ecological assessment tools. In this way, monitoring becomes effective and holistic. Some of the assessments suggested that the concentrations of PAHs detected in these ecosystems may occasionally pose toxic effects to some organisms. Some of these toxic effects may include reproduction impairment, photo-toxicity, cancer, and physiological disorders in fish, birds and mammals (Abdel-Shafy & Mansour, 2016; Francisco, 2016; Long et al., 2000; Rengarajan et al., 2015; Santos et al., 2018).

5.6 Evaluation of health implications

Fishes caught in these water bodies are sold to people both far and near, thus, each one stands exposed. To protect human health from the carcinogenic effects of PAH exposure, some thresholds are established. The aim of these established thresholds was to set a non-detectable level (zero concentration for carcinogenic PAHs in environmental media; Ashayeri et al., 2018). However, since it may be impossible to have zero concentration of all PAHs in the environment, certain thresholds are set, above which policy makers or citizens should be concern. The potential risk of PAHs for residents' health was evaluated by using the Incremental Lifetime Cancer Risk (ILCR) model (USEPA, 2004; Zhao et al., 2014) (equations 12, 13 and 14). This was used to calculate the risks of both adults and children exposed to PAHs via water and sediments contact or via fish consumption in these studied lagoons.

From the calculated results of ILCR, the carcinogenic compounds were the major contributors to the calculated TEQ in the ecosystem. For this reason, the discussions on ILCR were based on the total PAHs of carcinogenic compounds (∑PAHscar).

5.6.1 Human health risk via ingestion and dermal contact of water and sediment

For water, this study considered ingestion and dermal intakes as the two major exposure pathways to PAHs for humans. In the case of sediment, dermal exposure pathway was considered (Francisco, 2016; Zhao et al., 2014). The USEPA has established a Maximum Contaminant Level (MCL) for Benzo[a]pyrene (BaP) to be 0.2 µg/L in drinking water (Ashayeri et al., 2018; USEPA, 1982). It is well known that BaP is the most carcinogenic PAH. Benya and Brenu values were lower than the MCL, while that of Fosu was of the same value as the MCL for drinking water. This further confirmed the risk associated with contact with the water column of Fosu Lagoon. Essumang (2010) also reported on the levels of PAHs in the water column of Benya Lagoon. In Essumang's work, BaP was not detected in the water samples. Benzo[b]fluoranthene was the only carcinogenic compound found in the water samples. Therefore, even though the concentration of BaP based on this current study is below the MCL, its detection is of concern.

From the calculated results of ILCR for water samples, dermal and ingestion pathways for both children and adults were lower than the safety threshold, 1×10^{-6} (USEPA, 2004; Man et al., 2013). An indication of very low risk for cancer through ingestion and dermal contact with the water column. This suggested that even if untreated water is consumed (i.e., ingestion pathway), or there is a dermal exposure to PAHs (through recreational bathing activities or dermal absorption by fishermen), there would be a very low risk associated with humans getting cancer (Ashayeri et al., 2018). These findings were lower than those reported for other coastal waters in Ghana (such as lower Volta estuary, Sakumono lagoon, Keta lagoon, and Narkwa lagoon; Essumang, 2010).

The results for the sediment samples on the other hand, demonstrated that cancer risk of ∑PAHs car were greater than the safety level (1×10^{-6}). Meaning, there is a possibility of harmful effects (cancer) of PAH compounds in sediments via dermal absorption for both children (varied from 7.05×10^{-6} to 5.96×10^{-5}) and adults (varied from 2.47×10^{-6} to 2.09×10^{-5}). For children, the results meant that between 7 and 60 out of 1,000,000 are likely to suffer cancer-related sickness. For adults, the possibility to get cancer-related sickness was between 2 and 20 out of 1,000,000 in their lifetime of 70 years. It should be noted that the potential risk for inhabitants will be determined by the synergistic effect with other potential pollutant exposure routes such as inhalation. Making the inhabitants more vulnerable to PAHs exposure. People live in poverty state near these studied water bodies, so their economic situation might enhance their exposure levels to PAHs through sediment and water (Ashayeri et al., 2018).

5.6.2 Human health risk via fish consumption

Usually, risk assessments and regulatory limits on PAHs in fish are based on their wet weight concentrations (Bandowe et al., 2014), and in this current study, all the concentrations of the fish tissues were measured in their wet weights. The mean concentrations of BaP in all the fish tissues from the various lagoons ranged from 19.22±7.66 µg/kg (BRFH) to 32.8±12.79 µg/kg (BFV). The concentrations of BaP in all the fish tissues were above MCL set by European Union for fish muscle (EFSA, 2008) which is 2 µg/kg and that of China for food (5 µg/kg) (Shi et al., 2016). Both thresholds were considered in this study because they are internationally recognised. In all the studied lagoons, mean concentrations of BaP in fish tissues were approximately 10 times higher than the MCL set by European Union for fish muscles. The concentrations of BaP in this study were also higher than those reported by Bandowe et al. (2014) in fish tissues sampled from the marine waters of Ghana

Carcinogenic risks were estimated as the incremental probability of a person developing cancer over a lifetime due to exposure to the potential carcinogen (Essumang, 2010; Essumang et al., 2012). This risk was referred to as Incremental Lifetime Cancer Risk (ILCR) in this study or just carcinogenic risk. Some authors reported it as individual excess lifetime cancer risk (IELCR) (Essumang, 2010) or excess cancer risk (ECR) (Bandowe et al., 2014; Shi et al., 2016; Tongo, Ogbeide, & Ezemonye, 2017; Yoon, Park, Lee, Yang, & Lee, 2007b). Carcinogenic risk resulting from lifetime exposure to the 16 USEPA PAHs through consumption of these fishes were then calculated and compared to the acceptable threshold value of 1.0E-6 set by the USEPA (Ding & Ni, 2012; USEPA, 1982; Yoon et al., 2007a).

ILCR∑16PAHs and ILCR∑PAHs car for both adults and children were greater than the safety threshold of 1×10^{-6} for all the studied lagoons. For the adults' exposure, fillet tissues ranged from 2.2×10^{-5} (BRFF) to 2.33×10^{-5} (FFF) for ILCR∑PAHs car, while that of the children ranged from 3.07×10^{-5} (BRFF) to 3.26×10^{-5} (FFF). This indicated that ≈20 to 25 out of 1,000,000 adults are likely to suffer from cancer-related sickness from eating fillet of *Sarotherodon melanotheron* throughout their life time of 70 years. For children, this ranged from ≈30 to 35 out of 1,000,000. Although these values were higher than the acceptable risk level (1×10^{-6}), they were not above the priority risk level (1×10^{-4}). With the values of visceral and head tissues also being above the acceptable risk, it should be noted that consumption of fish without complete removal of these parts will yield a higher health risk per unit mass of fish consumed.

In summary, there is the likelihood for people that depend on these studied lagoons to be exposed to cancer-related sickness, especially through dermal contact with sediment and consumption of black-chinned tilapia.

5.7 Sediment remediation

This section discussed the effect of activated charcoal addition on the pore-water concentrations of total PAHs. Pore-water concentrations of PAHs, also known as freely dissolved concentrations of PAHs, are the bioavailable concentrations in a water body (Abdel-Shafy & Mansour, 2016; Dong et al., 2012). These are the concentrations taken up by organisms living in the sediment or the water column.

Reduction of freely dissolved concentration of total PAHs was observed in the different compartments (water and sediment) when different AC doses (0%, 1%, 3%, and 6%) were applied. Significant reductions ($p < 0.05$) were observed in the 1% and 6% AC doses. The reductions in the sediment were also significantly ($p<0.05$) lower than those observed in the water column, even in the control where no AC was added. The reduction in the control was ascribed to the presence of organic matter or other naturally occurring carbonaceous materials that may be present in the sediment. This sediment was sampled from Fosu Lagoon, and was enriched with organic matter. Organic matter and carbonaceous materials such as black carbon have been reported to readily bind PAHs, making them immobile (Ghosh et al., 2003; Gustafsson et al., 1997; Jonker & Smedes, 2000; Koelmans et al., 2006; Luthy et al., 1997; Weber et al., 1999). However, there were some levels of PAHs detected in the water column of the control, even though the distilled water used for the experiment had no presence of PAHs. Higher levels of the LMW compounds in the water column could explain the relatively low solubility of PAHs in water even though they are hydrophobic contaminants (Dohányosová et al., 2003; Niu et al., 2018; USEPA, 2000). Again, the presence of PAHs in the aqueous medium may be due to the mobility of contaminants from the sediment to the aqueous medium when distilled water was gently being poured into the beaker of PAH-contaminated sediment.

Upon addition of AC, another carbonaceous material to the PAH-contaminated sediment, further increase in the reduction of freely dissolved PAHs was observed. The AC doses (1% and 6%) significantly yielded a 19% to 98% reduction of freely dissolved PAHs in the water column, and from 35%

to 99% reduction in the sediment. The effectiveness of the activated charcoal in this study was ascribed to its large surface area and a pore structure effective for binding the PAHs. Brändli et al. (2008) and Zimmerman et al. (2004) also reported that when activated carbon was added to contaminated soil and sediment, the concentrations of freely dissolved hydrophobic contaminants available to the water column and the sediment reduced. Brändli et al. (2008) reported on a reduction rate of up to 99% and Zimmerman et al. (2004) also mentioned a reduction rate of 84%. Oleszczuk et al. (2012) also reported on a reduction range of 56% to 95% in PAH-contaminated sewage sludge.

In ecotoxicology, researchers' goal is to apply the least treatment dose that could significantly cause a positive response. Therefore, it is worth noting that the least AC dose (1%) could significantly reduce the freely dissolved PAH in both the water (by 29%) and the sediment (99%). Carbonaceous materials (CM; such as AC) seem to improve the habitat quality for benthic organisms by way of decreasing bioavailable hydrophobic contaminants and their toxicity in sediment (Rakowska et al., 2012). However, its effectiveness should not promote usage of higher concentrations of CM, as in the case of this current study. AC dose of 6% equally resulted in significant reductions of the freely dissolved PAH in both water (by 98%) and sediment (by 67%). However, the positive effects of decreasing toxicity at low CM concentrations most probably outweigh the mild adverse effects detected at higher CM concentrations (Rakowska et al., 2012). Indeed, there is a carbon sequestration effect that is lost in the energy-intensive activation process in the manufacturing of AC. However, a previous study on a complete life-cycle assessment of sediment remediation by different amendment materials projected that biomass-based activated carbon was the optimal material from a global perspective (Sparrevik et al., 2011).

In summary, AC dose (1%) effectively reduce the freely dissolved PAH concentrations to about 29% and 99% in the water column and sediment respectively. For the AC dose of 6%, the freely dissolved PAH concentrations reduced to 98% and 67% in the water column and sediment respectively. The reduction in the sediment was significantly higher than in the water column when compared to their control experiments. Apart from the AC, organic matter in sediments was also a major binding site for PAHs.

CHAPTER SIX

SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

6.1 Summary

The main focus of this study was to assess the dynamics of PAHs in three coastal lagoons in Ghana and determine the efficacy of activated charcoal in sediment remediation under laboratory conditions. The outcome of this provided vital information on the concentrations, distribution, and human and eco-toxicological effects of PAHs in these coastal lagoons. In addition, the extent of transfer of PAHs from the sediment to fish was established. Previous studies had reported on the levels of PAHs in the sediment of Fosu and in the water column of Benya Lagoons. This study provided further details regarding PAHs levels in three main environmental media; water, sediment and fish (*Sarotherodon melanotheron*), from three coastal lagoons for comparative assessment. In order to aid effective environmental policy formulation in the regulation of PAHs, potential sources of PAHs to these ecosystems were identified. The findings showed that increased levels of PAHs in these coastal waters were greatly influenced by the intense human activities that are carried out within the vicinity of the water bodies. From an evaluation process using diagnostic ratios, the bulk of the PAHs in these aquatic ecosystems emanated mainly from petrogenic and pyrogenic sources. A laboratory experiment was also conducted to enhance the mechanistic understanding on the use of activated charcoal as a significant approach in reducing bioavailability of PAHs in coastal sediment.

The outcome of the ecological and human health risks associated with these coastal lagoons calls for monitoring and effective coastal management programs that must be implemented to guarantee the safe usage of the current areas especially, for fishing and bathing purposes.

6.2 Conclusions

The following conclusions were deduced from this current study:

1. The fate of PAHs in the studied lagoons was affected by both seasonal and spatial variations, as well as the transport behaviour of PAHs in the different compartments (water, sediment and fish) of the lagoons.
2. The LMW PAHs were predominant in the water column, with naphthalene being the most detected compound. More than 62% of the individual PAHs detected in the water column were above the WHO's limit of 0.05µg/L.
3. Concentrations of PAHs in sediments were classified as moderate in relation to global levels of PAH contamination.
4. The residual levels of PAHs in fish tissues from Benya, Brenu, and Fosu Lagoons were attributed to two main factors; (1) specific characteristics of the study areas and (2) the level of biotransformation of some PAHs in the studied fish.
5. The levels of PAHs among the studied lagoons suggested that higher concentrations of total PAHs were detected in environments with intense anthropogenic influence, and Fosu Lagoon was noted for that.
6. The lagoons also exhibited a wide range of PAH concentrations in their sediments, indicating heterogeneous levels of contamination. Multiple sources of PAHs contamination in the lagoons were of both pyrogenic and petrogenic sources. These included: burning of biomass, shipping activity, vehicular emissions, sewage, oil spillage, indiscriminate disposal of waste, and mechanic shops.

7. From the ecological risk assessment view point, more than 50 % of individual PAHs in the sediments exhibited a moderate level of ecological risk for aquatic life.
8. The calculated Incremental Lifetime Cancer Risk (ILCR) of these studied lagoons also showed that, the dermal and ingestion pathways of water for both children and adults were lower than the safety threshold (10E-6) for all PAHs. However, the findings on ILCR for sediments were above the safety limit (10E-06); an indication of cancer-related effects via dermal absorption of sediments for both children and adults. For the fish tissues, the mean concentrations of Benzo[a]pyrene were above the maximum contamination level set by European Union for fish (2 μg/kg) and that of China (5 μg/kg). The ILCR of consumption of fish for both adults and children were also above the safety threshold.
9. Activated charcoal was identified to be a promising carbonaceous material for remediating PAH-contaminated sediment of coastal lagoons.

Based on these findings, if regulatory measures are not set or enforced, the health of the whole lagoon ecosystems and the human populations that depend on these ecosystems for sustenance and livelihood could be severely impacted. Inhabitants who virtually have no idea on PAHs and their harmful effects should be educated on the best practices to effect change.

6.3 Recommendations

With the levels of PAHs in these lagoons, there is the urgent need for the removal of the refuse dump virtually situated on the station 3 of Benya Lagoon (BST3). The emissions from the refuse dump was a major contributor to PAHs levels in this lagoon. Inhabitants, and especially fisher folks should be educated on PAHs and their adverse effects, as well as best practices that could minimise the inputs of PAHs through their activities.

The calculated Incremental Lifetime Cancer Risk (ILCR) for dermal and ingestion pathways of the water were lower than the threshold (10E-06) set by USEPA. However, the dermal exposure to the studied sediments were above the threshold. Therefore, people who are exposed to these lagoons especially, the fisher folks, should opt for ways in fishing that minimises their direct contact with the sediments of these lagoons. The calculated ILCR for fish tissues were also above the lower boundary of the threshold (10E-06), but less than the upper boundary of the threshold, 10E-04. Therefore, consumption of *Sarotherodon melanotheron* from these lagoons should also be minimised. As much as possible, other parts (such as the visceral and the head) should be avoided.

The findings from the remediation experiment also portrayed that the use of activated charcoal can be a promising technique to remediate PAH-contaminated sediments. However, effective measures to curb the continuous release of anthropogenic PAHs into these lagoons should be put in place. Such measures will give room for the activated charcoal to bind effectively to the contaminants.

Some vital questions remain unanswered, and these could further explain observations in the present study. Therefore, further studies are recommended to be undertaken to:

1. Investigate the fate and effects of the PAH metabolites to aquatic biota in coastal waters of Ghana.
2. Investigate levels of PAHs in plants species around these lagoons.
3. Evaluate PAH levels in shell fishes and other benthic organisms in these lagoons.
4. Investigate fish tissue's level-response relationship and actual eco-toxicological risk.
5. Investigate possible secondary effects of activated charcoal amendment on benthic organisms in coastal waters in Ghana.
6. Investigate the role of mixing in the efficacy of activated charcoal in sediment remediation.

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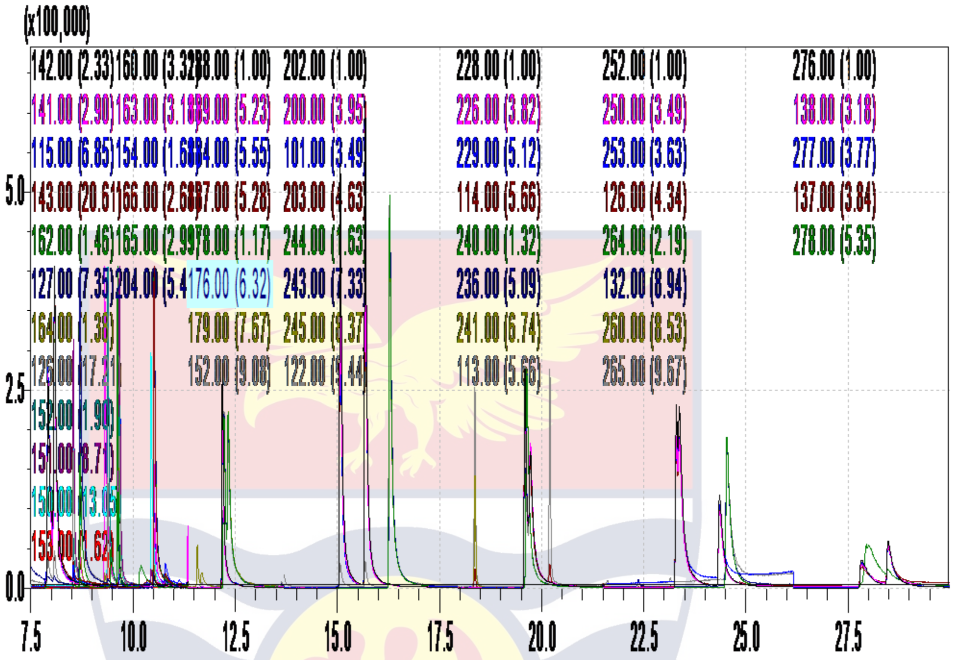
APPENDICES

APPENDIX A- SUPPORTING DATA

Appendix A1 - *Selected ion monitoring chromatogram showing analysed PAHs*



Appendix A2-*Basic descriptive statistics for the PAHs concentrations in the sediment (dry weight) of three stations of Benya lagoon*

	Concentration in BSTI (µg/kg)						Concentration in BST2 (µg/kg)					
Compound	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis
Naph	3.999	47.910	19.661	11.260	1.030	0.669	1.555	48.630	18.436	10.828	1.341	1.612
Ace	0.066	28.935	12.122	9.126	0.049	-1.099	0.258	28.943	12.269	9.357	0.047	-1.194
Acp	0.380	26.415	9.505	7.232	0.648	0.223	0.735	26.520	9.680	7.374	0.672	0.069
F	0.372	36.915	18.954	13.196	-0.235	-1.595	1.130	37.035	20.173	12.154	-0.189	-1.459
Ant	0.605	36.100	16.164	7.590	-0.182	0.416	BDL	38.440	15.412	8.531	0.239	0.237
Phe	0.109	21.953	11.665	7.576	-0.345	-1.385	1.010	26.445	12.146	7.454	-0.139	-1.317
Flu	0.550	145.390	23.258	27.940	2.870	9.637	5.520	55.495	22.951	15.280	0.919	-0.632
Pyr	2.270	122.900	29.242	21.053	2.935	10.608	9.625	66.120	31.065	13.895	0.737	-0.123
Chr	3.690	62.490	25.008	13.113	0.979	0.945	6.065	69.840	32.923	18.427	0.681	-0.714
BaA	1.110	214.935	30.406	42.604	3.038	9.983	4.120	591.650	62.884	131.608	3.322	10.189
BkF	2.985	68.895	20.525	15.395	1.628	2.068	5.782	63.504	27.075	16.574	1.122	0.349
BbF	0.105	82.580	17.186	18.314	1.946	3.786	3.953	46.084	17.111	14.543	0.821	-0.914
BaP	2.155	73.900	23.703	13.721	1.733	4.285	4.475	68.493	27.577	17.364	1.275	0.660
DahA	BDL	13.060	2.169	4.023	1.734	1.542	BDL	18.242	3.705	5.952	1.377	0.490
IP	BDL	52.890	16.537	10.679	1.232	3.108	BDL	39.458	18.353	10.308	0.298	0.078
BP	BDL	50.005	6.511	11.606	2.231	5.249	BDL	39.697	8.126	13.511	1.479	0.640
Total PAHs	35.820	824.460	282.618	149.529	1.908	4.803	89.43	807.53	339.888	169.436	1.108	0.962

Continued Concentration in BST3 (µg/kg)

Compound	Min.	Max.	Mean	Std. Deviation	Skewness	Kurtosis
Naph	2.605	65.970	17.347	12.482	2.07	5.332
Ace	0.061	28.853	11.892	9.038	0.094	-1.075
Acp	0.050	26.460	9.846	6.818	0.726	0.822
F	0.157	36.480	18.665	13.058	-0.252	-1.517
Ant	BDL	23.555	12.336	8.099	-0.196	-1.56
Phe	BDL	22.283	11.497	7.174	-0.346	-1.128
Flu	0.945	61.075	10.287	12.622	2.902	8.606
Pyr	1.451	70.730	18.057	12.830	2.323	8.109
Chr	3.215	53.333	24.944	13.943	0.665	-0.515
BaA	0.800	50.120	17.804	12.070	0.988	0.223
BkF	3.190	51.415	17.965	12.585	1.064	0.35
BbF	0.080	80.820	15.502	16.677	1.93	4.924
BaP	2.160	50.365	21.896	10.903	0.608	0.382
DahA	BDL	15.873	3.389	5.130	1.138	-0.205
IP	BDL	32.345	15.758	7.956	-0.531	-0.174
BP	BDL	37.220	4.208	8.429	2.458	6.354
Total PAHs	43.56	501.21	231.395	90.38619	0.507	1.845

*BDL- below detection limit

Appendix A3- *Basic descriptive statistics for the PAHs concentrations in fish parts (wet weight) of Benya lagoon*

	Concentration in fillet (µg/kg)						Concentration in visceral (µg/kg)					
Compound	Min.	Max.	Mean	Std. Deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. Deviation	Skewness	Kurtosis
Naph	3.770	80.920	28.774	18.188	1.348	2.752	0.660	88.595	27.027	23.368	1.739	2.578
Ace	0.205	41.505	27.883	12.418	-1.498	1.734	11.180	177.085	47.212	51.843	2.149	3.053
Acp	BDL	36.280	19.888	12.759	-0.897	-1.044	0.800	274.720	57.400	83.292	2.155	3.157
F	0.365	49.410	30.102	15.445	-1.113	-0.133	6.395	39.375	32.592	10.797	-2.068	2.756
Ant	BDL	33.395	19.950	9.063	-1.410	1.757	21.938	980.240	153.449	257.272	2.453	5.524
Phe	BDL	29.950	17.129	9.754	-0.955	-0.744	22.808	317.975	87.146	98.932	1.411	0.415
Flu	BDL	10.830	4.245	2.850	0.075	0.155	BDL	23.370	7.497	7.028	0.934	0.086
Pyr	BDL	31.550	16.206	10.430	-0.799	-0.987	BDL	1489.935	160.419	392.492	2.767	7.135
BaA	4.425	35.550	21.862	7.236	-1.165	1.874	23.183	115.730	34.479	21.870	3.206	10.403
Chr	BDL	24.010	12.336	6.594	-0.249	0.616	11.843	41.460	21.406	9.526	1.277	0.729
BbF	11.123	51.325	19.793	13.048	1.817	2.060	12.233	108.175	32.985	28.743	1.934	2.611
BkF	BDL	18.760	7.793	5.539	1.017	0.239	5.205	76.050	15.500	16.642	2.833	8.971
BaP	21.375	44.565	26.132	7.499	1.861	2.145	20.798	71.315	32.800	12.786	1.449	2.687
DahA	BDL	22.250	4.244	6.916	1.586	1.490	BDL	66.170	9.157	18.731	2.321	4.517
IP	7.300	41.740	20.649	7.728	0.457	2.277	9.835	46.895	24.973	8.630	0.192	1.403
BP	BDL	4.180	0.508	1.078	2.484	6.360	BDL	7.275	1.367	2.319	1.404	0.649
Total PAHs	173.035	433.288	277.494	57.289	0.290	2.632	300.799	3726.158	745.407	957.222	2.456	5.101

Continued Concentration in head (µg/kg)

Compound	Minimum	Maximum	Mean	Std. Deviation	Skewness	Kurtosis
Naph	1.705	102.040	30.393	23.592	2.118	5.026
Ace	1.570	199.145	42.259	46.022	2.508	6.609
Acp	2.870	46.465	25.511	9.916	-0.997	2.079
F	2.700	39.820	31.537	12.256	-1.729	1.467
Ant	BDL	29.783	17.336	9.528	-1.107	-0.239
Phe	BDL	27.255	17.658	9.766	-1.054	-0.642
Flu	BDL	140.768	37.843	47.894	1.192	0.237
Pyr	12.560	186.645	78.145	56.220	0.502	-1.099
BaA	5.740	27.230	21.814	5.070	-2.514	6.274
Chr	BDL	24.275	12.093	6.376	-0.293	0.819
BbF	11.213	47.680	18.745	12.362	1.934	2.355
BkF	BDL	18.105	6.595	5.171	1.376	1.651
BaP	21.130	28.435	22.652	2.364	1.984	2.598
DahA	BDL	16.435	3.440	5.762	1.310	0.106
IP	5.660	22.710	18.908	5.836	-1.754	1.707
BP	BDL	2.230	0.114	0.485	4.583	21.000
Total PAHs	178.411	553.608	385.044	94.736	-0.435	-0.375

*BDL- below detection limit

Appendix A4- *Basic descriptive statistics for the PAHs concentrations in the sediment (dry weight) of three stations of Brenu lagoon*

	Concentration in BRST1 (μg/kg)						Concentration in BRST2 (μg/kg)					
Compound	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis
Naph	BDL	41.010	10.069	9.157	1.470	2.709	BDL	32.025	9.688	8.544	1.086	1.109
Ace	BDL	28.890	12.073	8.815	0.131	-0.965	0.060	28.875	12.455	9.019	0.122	-1.160
Acp	BDL	26.745	9.054	8.080	0.585	-0.461	BDL	26.550	8.885	8.239	0.585	-0.600
F	0.001	38.805	17.310	13.701	-0.018	-1.462	0.001	37.185	17.641	13.927	-0.074	-1.578
Ant	BDL	45.300	11.771	10.731	0.856	1.034	BDL	25.545	10.753	8.965	-0.048	-1.591
Phe	BDL	70.350	14.149	16.125	2.112	5.167	BDL	22.245	10.285	8.311	-0.056	-1.631
Flu	BDL	38.850	4.806	8.101	3.709	13.850	BDL	7.485	3.169	2.198	0.097	-1.011
Pyr	BDL	36.900	11.011	9.223	0.518	-0.146	BDL	22.710	10.479	8.278	-0.005	-1.637
Chr	BDL	28.200	15.232	8.978	-0.500	-1.006	BDL	25.710	15.257	8.425	-0.662	-0.898
BaA	0.029	23.245	9.207	7.308	0.430	-0.701	0.029	23.370	9.024	7.023	0.580	-0.267
BkF	BDL	37.500	11.584	7.978	1.561	2.568	3.130	33.300	11.964	7.502	1.373	1.560
BbF	BDL	19.530	6.231	6.489	1.144	-0.072	BDL	19.435	6.148	6.241	1.154	-0.003
BaP	BDL	21.960	13.682	7.532	-0.644	-1.033	2.160	22.950	13.893	7.364	-0.681	-1.040
DahA	BDL	56.250	4.303	12.493	3.925	14.975	BDL	56.400	3.694	12.517	4.086	15.912
IP	BDL	55.650	14.317	11.898	1.969	5.529	BDL	54.150	14.231	12.106	1.730	4.435
BP	BDL	16.800	0.029	3.777	3.600	12.651	BDL	15.750	1.040	3.507	4.016	15.461
Total PAHs	22.06	474.500	166.062	106.733	0.715	0.942	21.520	283.800	158.606	88.675	-0.182	-1.167

Continued Concentration in BRST3 (µg/kg)

Compound	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis
Naph	BDL	59.670	12.677	14.786	2.151	4.770
Ace	BDL	29.220	12.401	9.348	0.190	-1.329
Acp	BDL	26.528	9.017	8.411	0.498	-0.818
F	0.001	36.675	17.107	14.888	0.035	-1.740
Ant	BDL	21.900	9.915	8.941	0.001	-1.854
Phe	BDL	21.878	9.915	8.725	0.031	-1.764
Flu	BDL	375.203	17.048	66.079	4.902	24.791
Pyr	BDL	168.923	16.496	29.598	4.244	19.972
Chr	BDL	25.650	14.668	9.109	-0.567	-1.197
BaA	0.029	23.210	9.009	7.130	0.470	-0.455
BkF	2.93	22.085	10.342	5.548	0.969	0.280
BbF	BDL	19.130	4.948	5.717	1.625	1.825
BaP	2.126	21.920	13.271	7.869	-0.513	-1.438
DahA	BDL	6.920	1.474	2.412	1.389	0.381
IP	2.475	22.335	11.861	7.640	0.082	-1.641
BP	BDL	1.900	0.214	0.478	2.268	4.225
Total PAHs	20.810	796.110	170.363	149.037	2.259	7.750

*BDL- below detection limit

Appendix A5- *Basic descriptive statistics for the PAHs concentrations in fish parts (wet weight) of Brenu lagoon*

	Concentration in fillet (µg/kg)						Concentration in visceral (µg/kg)					
Compound	Min.	Max.	Mean	Std. Deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. Deviation	Skewness	Kurtosis
Naph	1.046	47.000	21.672	14.068	-0.014	-0.744	BDL	27.840	17.430	7.961	-1.356	0.639
Ace	6.170	46.417	25.422	11.923	-0.392	-0.681	3.300	34.000	25.236	9.548	-1.821	1.807
Acp	BDL	42.036	21.523	13.333	-0.643	-0.873	BDL	29.940	20.743	11.440	-1.144	-0.562
F	0.001	97.135	43.447	27.064	0.510	0.657	0.001	37.785	28.926	14.387	-1.397	0.252
Ant	BDL	35.643	17.438	11.215	-0.539	-0.865	21.000	31.703	24.456	3.068	1.165	0.553
Phe	BDL	37.476	18.395	11.622	-0.543	-0.830	7.275	26.798	20.120	5.974	-1.365	0.590
Flu	BDL	20.860	6.978	5.661	1.460	1.868	BDL	9.450	5.081	2.879	-0.812	-0.263
Pyr	BDL	34.619	19.432	9.284	-1.092	1.120	BDL	53.340	19.006	13.410	0.346	0.932
BaA	BDL	34.964	22.117	9.941	-1.547	1.790	1.955	411.566	67.551	116.981	2.452	4.845
Chr	BDL	27.510	13.180	7.330	-0.018	0.621	BDL	24.625	12.676	6.594	-0.077	0.831
BbF	6.908	30.250	15.303	6.521	1.099	0.925	11.317	36.246	16.176	6.430	2.066	4.173
BkF	BDL	27.070	8.072	7.690	1.771	2.336	BDL	24.755	7.439	6.405	1.674	2.548
BaP	2.198	34.405	21.529	8.818	-1.263	1.324	3.352	29.445	19.636	7.185	-1.627	2.222
DahA	BDL	54.135	8.916	18.633	2.057	2.714	BDL	10.265	2.392	3.902	1.166	-0.482
IP	3.020	34.726	20.021	8.449	-0.534	0.020	5.890	23.220	18.322	7.200	-1.051	-0.937
BP	BDL	9.495	1.149	2.903	2.323	4.002	BDL	35.458	4.604	10.806	2.248	3.730
Total PAHs	32.366	457.017	284.595	116.502	-1.304	1.476	176.995	577.371	309.793	90.206	1.765	4.052

Continued Concentration in head (µg/kg)

Compound	Minimum	Maximum	Mean	Std. Deviation	Skewness	Kurtosis
Naph	1.595	43.500	28.774	12.912	-1.090	0.460
Ace	0.545	30.923	24.440	10.657	-1.707	1.344
Acp	BDL	37.050	22.003	14.376	-0.884	-1.105
F	0.330	43.890	28.596	17.642	-0.956	-1.052
Ant	BDL	92.865	21.202	21.156	2.110	6.286
Phe	BDL	67.530	22.423	18.652	0.627	0.382
Flu	BDL	198.293	45.223	63.071	1.171	0.029
Pyr	BDL	322.985	142.877	91.828	0.270	-0.097
BaA	BDL	43.230	22.834	10.630	-1.162	1.784
Chr	BDL	34.620	18.444	10.823	-0.240	-0.748
BbF	7.229	37.730	14.034	6.466	2.668	9.071
BkF	BDL	74.240	9.356	15.530	4.002	17.116
BaP	2.517	36.265	19.217	7.662	-1.036	2.531
DahA	BDL	85.270	6.685	18.815	4.011	17.076
IP	2.792	58.090	20.723	11.159	1.406	6.193
BP	BDL	85.650	5.162	18.670	4.412	19.831
Total PAHs	65.380	845.846	451.992	208.145	-0.332	0.094

*BDL- below detection limit

Appendix A6- *Basic descriptive statistics for the PAHs concentrations in the sediment (dry weight) of three stations of Fosu lagoon*

	Concentration in FST1 (µg/kg)						Concentration in FST2 (µg/kg)					
Compound	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis
Naph	0.120	136.575	22.32045	27.200	3.724	13.947	BDL	48.300	14.251	11.650	1.602	2.150
Ace	2.630	85.275	18.12221	12.807	3.935	20.316	4.385	29.955	15.915	6.864	0.324	-0.135
Acp	0.245	47.345	10.734	9.220	1.894	5.574	0.330	26.670	9.486	7.100	0.783	0.595
F	0.865	41.168	22.770	10.620	-0.569	-0.123	1.475	40.328	22.329	11.534	-0.408	-0.613
Ant	19.715	800.490	56.824	123.619	6.030	37.116	6.005	68.025	30.004	13.516	1.293	2.099
Phe	0.270	180.835	22.266	26.920	5.634	33.883	3.220	28.448	16.550	6.507	-0.132	-0.436
Flu	21.698	4081.780	188.769	642.111	6.175	38.392	34.118	270.505	76.662	48.846	2.145	5.567
Pyr	29.348	3955.425	178.605	622.855	6.178	38.417	32.018	286.985	69.572	48.483	2.901	10.254
Chr	21.330	1669.005	91.389	260.052	6.186	38.494	24.600	103.810	41.550	19.670	1.607	2.220
BaA	16.703	3212.055	130.369	507.126	6.219	38.776	14.118	145.380	42.771	26.443	2.073	5.162
BkF	21.09	2431.85	144.540	377.984	6.138	38.078	27.240	142.110	77.060	26.469	0.418	-0.390
BbF	3.70	2549.79	99.358	403.916	6.187	38.498	8.423	157.330	36.432	30.476	2.321	6.018
BaP	20.205	2118.18	99.923	332.470	6.199	38.603	21.360	116.325	45.452	21.492	1.573	2.497
DahA	BDL	237.07	17.186	41.587	4.205	21.024	BDL	56.850	9.995	18.994	1.842	1.760
IP	16.598	1748.285	83.378	274.882	6.156	38.227	14.61	95.040	35.696	19.263	1.136	0.677
BP	BDL	1640.435	62.135	260.732	6.146	38.132	BDL	99.815	17.402	21.858	2.044	4.534
Total PAHs	262.670	24801.380	1248.690	3880.504	6.196	38.575	351.180	1510.560	561.128	227.109	2.331	7.186

Continued Concentration in FST3 (μg/kg)

Compound	Min.	Max.	Mean	Std. deviation	Skewness	Kurtosis
Naph	1.030	87.765	16.365	15.635	2.792	10.744
Ace	1.290	36.443	15.086	7.784	0.610	0.747
Acp	0.290	30.473	10.008	7.675	1.020	0.901
F	1.060	51.690	22.653	12.232	-0.029	-0.181
Ant	1.510	80.865	26.166	13.342	2.238	8.052
Phe	BDL	44.250	17.006	9.011	0.585	1.776
Flu	14.565	173.365	36.019	31.952	2.854	9.235
Pyr	15.068	176.790	38.952	28.756	3.330	13.785
Chr	11.440	157.520	30.431	23.604	4.385	22.862
BaA	9.070	163.930	28.695	26.317	3.808	18.462
BkF	17.460	84.570	37.473	15.597	1.341	2.566
BbF	4.470	112.260	20.876	19.492	3.177	12.519
BaP	7.550	128.730	29.466	19.642	3.659	17.223
DahA	BDL	57.750	10.073	19.715	1.868	1.791
IP	8.560	74.805	27.274	18.095	1.440	0.719
BP	BDL	68.840	8.551	14.353	2.440	7.439
Total PAHs	189.890	1227.690	375.094	191.181	2.777	10.044

*BDL- below detection limit

Appendix A7- *Basic descriptive statistics for the PAHs concentrations in fish parts (wet weight) of Fosu lagoon*

	Concentration in fillet (µg/kg)						Concentration in visceral (µg/kg)					
Compound	Min.	Max.	Mean	Std. Deviation	Skewness	Kurtosis	Min.	Max.	Mean	Std. Deviation	Skewness	Kurtosis
Naph	BDL	38.680	23.921	11.943	-0.896	-0.032	BDL	104.645	28.791	27.415	1.972	3.584
Ace	BDL	41.535	28.214	12.616	-1.502	1.639	0.260	173.650	43.677	48.983	2.030	2.966
Acp	BDL	35.430	20.399	13.542	-0.838	-1.081	BDL	77.180	33.570	21.089	0.631	0.802
F	0.001	48.450	28.174	18.437	-0.834	-1.082	0.070	91.375	38.093	20.801	0.284	2.152
Ant	BDL	27.350	16.129	10.658	-0.862	-1.096	3.910	84.560	32.967	20.672	1.412	1.645
Phe	BDL	31.030	17.179	11.501	-0.761	-1.070	BDL	35.565	23.798	10.287	-1.297	1.153
Flu	BDL	11.108	4.606	3.325	0.004	-0.730	BDL	8.715	5.043	2.614	-1.178	0.320
Pyr	BDL	36.930	18.460	12.873	-0.508	-1.109	3.215	84.660	27.627	17.912	1.664	4.464
BaA	BDL	29.710	20.867	9.099	-1.813	2.265	20.975	42.185	26.619	5.209	1.600	2.716
Chr	BDL	34.785	13.600	8.143	0.444	1.475	11.550	180.955	34.787	40.740	2.681	8.071
BbF	11.093	48.645	19.115	12.310	1.907	2.329	11.663	65.245	25.151	14.330	1.621	2.285
BkF	BDL	18.350	6.719	5.153	1.311	1.571	2.305	33.590	9.010	7.446	2.374	5.687
BaP	20.570	56.340	26.439	10.089	2.079	3.503	20.700	80.675	29.827	16.321	2.166	4.206
DahA	BDL	40.210	6.173	13.019	2.118	3.058	BDL	21.910	4.086	7.318	1.617	1.239
IP	6.155	29.380	19.621	7.009	-0.759	-0.066	5.790	1118.500	141.067	325.132	2.839	6.961
BP	BDL	0.735	0.075	0.210	2.984	7.650	BDL	351.250	54.406	106.693	1.963	2.905
Total PAHs	116.995	381.431	269.691	78.578	-0.702	0.001	196.728	2050.468	558.519	529.631	2.284	4.175

Continued Concentration in head (µg/kg)

Compound	Minimum	Maximum	Mean	Std. Deviation	Skewness	Kurtosis
Naph	0.770	98.400	34.036	25.687	1.307	1.483
Ace	7.050	124.260	38.674	33.066	1.964	2.769
Acp	BDL	98.845	27.952	19.823	2.136	8.439
F	36.330	85.145	46.048	16.059	1.417	0.496
Ant	BDL	42.480	16.796	11.976	-0.195	-0.300
Phe	BDL	34.515	17.012	11.371	-0.753	-0.980
Flu	BDL	621.923	139.439	197.854	1.327	0.504
Pyr	8.010	330.870	133.983	111.779	0.383	-1.449
BaA	18.265	30.350	24.226	2.343	0.233	2.890
Chr	BDL	22.270	12.078	5.992	-0.673	1.124
BbF	11.175	50.020	19.829	12.388	1.827	2.167
BkF	BDL	18.390	6.603	5.273	1.441	1.779
BaP	21.000	37.950	22.925	4.157	2.925	8.723
DahA	BDL	26.410	3.977	7.641	2.052	3.480
IP	6.125	1061.565	69.092	227.466	4.578	20.974
BP	BDL	34.340	2.432	7.747	3.932	16.082
Total PAHs	292.084	1606.695	615.102	360.798	1.286	1.239

*BDL- below detection limit

Appendix A8- *Bioaccumulation factors (BAFs) for the different fish tissues sampled from Benya, Brenu, and Fosu lagoons*

	Benya			**Brenu**			**Fosu**		
PAH	**fillet**	**visceral**	**head**	**fillet**	**visceral**	**head**	**fillet**	**visceral**	**head**
Naph	1.6	1.5	1.7	1.4	1.1	1.8	1.7	2.1	2.5
Ace	2.6	4.4	3.9	2.1	2.1	2.0	1.9	3.0	2.7
Acp	2.5	7.1	3.2	2.0	2.0	2.1	2.4	4.0	3.3
F	1.7	1.8	1.8	2.1	1.4	1.4	1.4	1.9	2.3
Ant	1.4	11.0	1.2	1.2	1.7	1.5	0.4	0.8	0.4
Phe	1.6	8.0	1.6	1.4	1.5	1.7	1.0	1.3	1.0
Flu	0.3	0.5	2.3	2.1	1.5	13.5	0.0	0.0	1.1
Pyr	0.7	6.6	3.2	1.6	1.5	11.4	0.1	0.2	1.1
BaA	0.9	1.4	0.9	1.3	4.1	1.4	0.3	0.4	0.4
Chr	0.6	1.0	0.6	1.3	1.3	1.8	0.2	0.4	0.1
BbF	0.9	1.6	0.9	1.5	1.6	1.4	0.2	0.2	0.2
BkF	0.6	1.2	0.5	1.4	1.3	1.6	0.1	0.1	0.1
BaP	1.2	1.5	1.1	1.7	1.5	1.5	0.4	0.4	0.3
DahA	0.5	1.2	0.4	2.0	0.5	1.5	0.2	0.1	0.1
IP	1.3	1.5	1.2	1.7	1.5	1.7	0.3	2.4	1.2
BP	0.0	0.1	0.0	0.9	3.7	4.1	0.0	1.0	0.0
Total PAHs	1.1	3.0	1.6	2.0	2.1	3.1	0.3	0.6	0.7

Appendix A9- *Biota-sediment accumulation factors (BSAFs) for fish fillet sampled from Benya, Brenu, and Fosu lagoons*

Compound	**Benya**	**Brenu**	**Fosu**
Naph	9.7	1.2	12.1
Ace	15.4	1.8	13.6
Acp	14.6	1.8	17.0
F	9.9	1.9	9.9
Ant	8.5	1.1	2.7
Phe	9.2	1.2	6.7
Flu	1.5	1.8	0.3
Pyr	4.0	1.4	1.0
BaA	5.1	1.2	2.3
Chr	3.5	1.1	1.1
BbF	5.6	1.3	1.3
BkF	3.5	1.2	0.7
BaP	7.2	1.5	2.7
DahA	3.2	1.7	1.4
IP	7.5	1.5	2.4
BP	0.2	0.8	0.0
Total PAHs	6.7	1.7	2.2

APENDIX B – PARAMETRIC AND NONPARAMETRTIC TABLES

Appendix B1 - *Results of Mann-Whitney test for PAHs' interaction between dry and wet seasons for Benya sediment samples*

Null Hypothesis	**P-value**
The distribution of Naphthalene is the same across categories of SEASON.	0.001
The distribution of Acenaphthylene is the same across categories of SEASON.	0.000
The distribution of Acenaphthene is the same across categories of SEASON.	0.000
The distribution of Fluorene is the same across categories of SEASON.	0.000
The distribution of Anthracene is the same across categories of SEASON.	0.000
The distribution of Phenanthrene is the same across categories of SEASON.	0.000
The distribution of Fluoranthene is the same across categories of SEASON.	0.049
The distribution of Benzo[a]anthracene is the same across categories of SEASON.	0.000
The distribution of Chrysene is the same across categories of SEASON.	0.000
The distribution of Benzo[b]fluoranthene is the same across categories of SEASON.	0.000
The distribution of Benzo[k]fluoranthene is the same across categories of SEASON.	0.000
The distribution of Benzo[a]pyrene is the same across categories of SEASON.	0.000
The distribution of Dibenzo[ah]anthracene is the same across categories of SEASON.	0.000
The distribution of Indeno[123cd]pyrene is the same across categories of SEASON.	0.000
The distribution of Benzo[ghi]perylene is the same across categories of SEASON.	0.000
The distribution of Total PAHs is the same across categories of SEASON.	0.002

Appendix B2 - *Results of Mann-Whitney test for PAHs' interaction between dry and wet seasons for Benya fish samples*

Null Hypothesis	P-value
The distribution of Fluorene is the same across categories of SEASON.	0.001
The distribution of Fluoranthene is the same across categories of SEASON.	0.000
The distribution of Chrysene is the same across categories of SEASON.	0.005
The distribution of Benzo[b]fluoranthene is the same across categories of SEASON.	0.000
The distribution of Benzo[k]fluoranthene is the same across categories of SEASON.	0.006
The distribution of Dibenzo[ah]anthracene is the same across categories of SEASON.	0.000
The distribution of Indeno[123-ed]pyrene is the same across categories of SEASON.	0.004
The distribution of Benzo[ghi]perylene is the same across categories of SEASON.	0.000

Appendix B3 - *Results of Kruska-Wallis test for PAHs' interaction among fillet, visceral and head of Benya fish samples*

Null Hypothesis	P-value
The distribution of Acenaphthene is the same across categories of FISH TISSUES.	0.001
The distribution of Anthracene is the same across categories of FISH TISSUES.	0.000
The distribution of Phenanthrene is the same across categories of FISH TISSUES.	0.000
The distribution of Fluoranthene is the same across categories of FISH TISSUES.	0.032
The distribution of Pyrene is the same across categories of FISH TISSUES.	0.000
The distribution of Benzo[a]anthracene is the same across categories of FISH TISSUES.	0.000
The distribution of Chrysene is the same across categories of FISH TISSUES.	0.000
The distribution of Benzo[b]fluoranthene is the same across categories of FISH TISSUES.	0.005
The distribution of Benzo[k]fluoranthene is the same across categories of FISH TISSUES.	0.001
The distribution of Benzo[a]pyrene is the same across categories of FISH TISSUES.	0.006
The distribution of Indeno[123-ed]pyrene is the same across categories of FISH TISSUES.	0.001
The distribution of Total PAHs is the same across categories of FISH TISSUES.	0.000

Appendix B4 - *Results of independent samples test for PAHs' interaction between dry and wet seasons for Brenu water samples*

PAH	**F**	**t**	**df**	**P-value (2-tailed)**	**Mean Difference**
Fluorene	1.328	3.650	4	0.022	0.0019
Pyrene	0.818	9.675	4	0.001	0.0023
Indeno[123-cd]pyrene	1.509	6.538	4	0.003	0.0008

Appendix B5 - *Results of Mann-Whitney test for PAHs' interaction between dry and wet seasons for Brenu sediment samples*

Null Hypothesis	**P-value**
The distribution of Naphthalene is the same across categories of SEASON.	0.001
The distribution of Fluorene is the same across categories of SEASON.	0.000
The distribution of Anthracene is the same across categories of SEASON.	0.001
The distribution of Phenanthrene is the same across categories of SEASON.	0.028
The distribution of Fluoranthene is the same across categories of SEASON.	0.008
The distribution of Pyrene is the same across categories of SEASON.	0.001
The distribution of Benzo[a]anthracene is the same across categories of SEASON.	0.024
The distribution of Benzo[b]fluoranthene is the same across categories of SEASON.	0.000
The distribution of Benzo[a]pyrene is the same across categories of SEASON.	0.006
The distribution of Dibenzo[ah]anthracene is the same across categories of SEASON.	0.000
The distribution of Benzo[ghi]perylene is the same across categories of SEASON.	0.000

Appendix B6 - *Results of Mann-Whitney test for PAHs' interaction between dry and wet seasons for Brenu fish samples*

Null Hypothesis	P-value
The distribution of Naphthalene is the same across categories of SEASON.	0.000
The distribution of Acenaphthylene is the same across categories of SEASON.	0.001
The distribution of Acenaphthene is the same across categories of SEASON.	0.000
The distribution of Fluorene is the same across categories of SEASON.	0.018
The distribution of Anthracene is the same across categories of SEASON.	0.008
The distribution of Phenanthrene is the same across categories of SEASON.	0.000
The distribution of Fluoranthene is the same across categories of SEASON.	0.004
The distribution of Pyrene is the same across categories of SEASON.	0.001
The distribution of Benzo[a]pyrene is the same across categories of SEASON.	0.028
The distribution of Dibenz[ah]anthracene is the same across categories of SEASON.	0.000
The distribution of Indeno[123-ed]pyrene is the same across categories of SEASON.	0.000
The distribution of Benzo[ghi]perylene is the same across categories of SEASON.	0.000

Appendix B7 - *Results of Mann-Whitney test for PAHs' interaction between dry and wet seasons for Fosu sediment samples*

Null Hypothesis	P-value
The distribution of Naphthalene is the same across categories of SEASON.	0.000
The distribution of Acenaphthene is the same across categories of SEASON.	0.000
The distribution of Fluorene is the same across categories of SEASON.	0.004
The distribution of Anthracene is the same across categories of SEASON.	0.018
The distribution of Benz[a]anthracene is the same across categories of SEASON.	0.010
The distribution of Chrysene is the same across categories of SEASON.	0.016
The distribution of Benzo[k]fluoranthene is the same across categories of SEASON.	0.001
The distribution of Benzo[a]pyrene is the same across categories of SEASON.	0.000
The distribution of Dibenzo[ah]anthracene is the same across categories of SEASON.	0.000
The distribution of Indeno[123-cd]pyrene is the same across categories of SEASON.	0.000
The distribution of Benzo[ghi]perylene is the same across categories of SEASON.	0.000
The distribution of Total PAHs is the same across categories of SEASON.	0.009

Appendix B8 - *Results of Mann-Whitney test for PAHs' interaction between dry and wet seasons for Fosu fish samples*

Null Hypothesis	P-value
The distribution of Naphthalene is the same across categories of SEASON.	0.012
The distribution of Acenaphthene is the same across categories of SEASON.	0.047
The distribution of Phenanthrene is the same across categories of SEASON.	0.003
The distribution of Fluoranthene is the same across categories of SEASON.	0.000
The distribution of Pyrene is the same across categories of SEASON.	0.023
The distribution of Chrysene is the same across categories of SEASON.	0.044
The distribution of Benzo[b]fluoranthene is the same across categories of SEASON.	0.000
The distribution of Dibenzo[ah]anthracene is the same across categories of SEASON.	0.000
The distribution of Benzo[ghi]perylene is the same across categories of SEASON.	0.000

APPENDIX C- TOXICOLOGICAL ASSESSMENT

Appendix C1- *Negligible concentrations and maximum permissible concentrations of PAHs in water samples from Benya, Brenu and Fosu lagoons*

PAH	**Negligible Concentrations (NCs)**			**Maximum Permissible Concentrations (MPCs)**		
	Benya	**Brenu**	**Fosu**	**Benya**	**Brenu**	**Fosu**
Naph	0.2279	0.1286	0.3336	0.0023	0.0013	0.0033
Ace	0.0883	0.0750	0.1650	0.0009	0.0008	0.0017
Acp	0.1208	0.0917	0.1950	0.0012	0.0009	0.0020
F	0.1458	0.1233	0.2683	0.0015	0.0012	0.0027
Ant	0.1125	0.0892	0.1925	0.0011	0.0009	0.0019
Phe	0.0200	0.0161	0.0367	0.0002	0.0002	0.0004
Flu	BDL	BDL	BDL	BDL	BDL	BDL
Pyr	0.0875	0.0742	0.1617	0.0009	0.0007	0.0016
Chr	0.0010	0.0008	0.0018	0.0000	0.0000	0.0000
BaA	0.0150	0.0131	0.0286	0.0002	0.0001	0.0003
BbF	0.0158	0.0133	0.0294	0.0002	0.0001	0.0003
BkF	0.0010	0.0010	0.0021	0.0000	0.0000	0.0000
BaP	0.0040	0.0031	0.0067	0.0000	0.0000	0.0001
DahA	BDL	BDL	BDL	BDL	BDL	BDL
IP	0.0017	0.0015	0.0033	0.0000	0.0000	0.0000
BP	BDL	BDL	BDL	BDL	BDL	BDL
Total PAHs	0.8415	0.6308	1.4247	0.0084	0.0063	0.0142

Appendix C2- *Potency Equivalent Concentration (PEC) values for water samples from Benya, Brenu and Fosu lagoons*

PAHs	BENYA	BRENU	FOSU
Naph	0.000319	0.00018	0.000467
Ace	0.000106	0.00009	0.000198
Acp	0.000145	0.00011	0.000234
F	0.000175	0.000148	0.000322
Ant	0.00135	0.00107	0.00231
Phe	0.000102	0.000082	0.000187
Flu	BDL	BDL	BDL
Pyr	0.000105	0.000089	0.000194
Chr	0.00103	0.00089	0.00195
BaA	0.0054	0.0047	0.0103
BbF	0.0057	0.0048	0.0106
BkF	0.0025	0.0024	0.005
BaP	0.109	0.084	0.181
DahA	BDL	BDL	BDL
IP	0.0103	0.0088	0.0193
BP	BDL	BDL	BDL
Total PAHs	0.1362	0.1074	0.2321

Appendix C3- *ILCR of adults and children exposed to PAHs in lagoon water through ingestion or dermal routes*

	Adults exposure via ingestion			Children exposure via ingestion		
Compound	Benya	Brenu	Fosu	Benya	Brenu	Fosu
Naph	2.42E-13	1.37E-13	3.55E-13	3.38E-13	1.91E-13	4.95E-13
Ace	8.05E-14	6.84E-14	1.50E-13	1.12E-13	9.55E-14	2.10E-13
Acp	1.10E-13	8.36E-14	1.78E-13	1.54E-13	1.17E-13	2.48E-13
F	1.33E-13	1.12E-13	2.45E-13	1.86E-13	1.57E-13	3.42E-13
Ant	1.03E-12	8.13E-13	1.76E-12	1.43E-12	1.14E-12	2.45E-12
Phe	7.75E-14	6.23E-14	1.42E-13	1.08E-13	8.70E-14	1.98E-13
Flu	BDL	BDL	BDL	BDL	BDL	BDL
Pyr	7.98E-14	6.76E-14	1.47E-13	1.11E-13	9.44E-14	2.06E-13
BaA	7.83E-13	6.76E-13	1.48E-12	1.09E-12	9.44E-13	2.07E-12
Chr	4.10E-12	3.57E-12	7.83E-12	5.73E-12	4.99E-12	1.09E-11
BbF	4.33E-12	3.65E-12	8.05E-12	6.05E-12	5.09E-12	1.12E-11
BkF	1.90E-12	1.82E-12	3.80E-12	2.65E-12	2.55E-12	5.30E-12
BaP	8.28E-11	6.38E-11	1.38E-10	1.16E-10	8.91E-11	1.92E-10
DahA	BDL	BDL	BDL	BDL	BDL	BDL
IP	7.83E-12	6.69E-12	1.47E-11	1.09E-11	9.34E-12	2.05E-11
BP	BDL	BDL	BDL	BDL	BDL	BDL
∑16PAHs	1.04E-10	8.16E-11	1.76E-10	1.45E-10	1.14E-10	2.46E-10
∑PAHscar	1.02E-10	8.02E-11	1.73E-10	1.42E-10	1.12E-10	2.42E-10

Continued	Adults exposure via dermal			Children exposure via dermal		
Compound	Benya	Brenu	Fosu	Benya	Brenu	Fosu
Naph	4.15E-11	2.34E-11	6.08E-11	1.18E-10	6.67E-11	1.73E-10
Ace	1.38E-11	1.17E-11	2.58E-11	3.93E-11	3.34E-11	7.34E-11
Acp	1.89E-11	1.43E-11	3.04E-11	5.37E-11	4.08E-11	8.67E-11
F	2.28E-11	1.93E-11	4.19E-11	6.49E-11	5.48E-11	1.19E-10
Ant	1.76E-10	1.39E-10	3.01E-10	5.00E-10	3.97E-10	8.56E-10
Phe	1.33E-11	1.07E-11	2.43E-11	3.78E-11	3.04E-11	6.93E-11
Flu	BDL	BDL	BDL	BDL	BDL	BDL
Pyr	1.37E-11	1.16E-11	2.52E-11	3.89E-11	3.30E-11	7.19E-11
BaA	1.34E-10	1.16E-10	2.54E-10	3.82E-10	3.30E-10	7.23E-10
Chr	7.03E-10	6.11E-10	1.34E-09	2.00E-09	1.74E-09	3.82E-09
BbF	7.42E-10	6.24E-10	1.38E-09	2.11E-09	1.78E-09	3.93E-09
BkF	3.25E-10	3.12E-10	6.51E-10	9.26E-10	8.89E-10	1.85E-09
BaP	1.42E-08	1.09E-08	2.35E-08	4.04E-08	3.11E-08	6.71E-08
DahA	BDL	BDL	BDL	BDL	BDL	BDL
IP	1.34E-09	1.14E-09	2.51E-09	3.82E-09	3.26E-09	7.15E-09
BP	BDL	BDL	BDL	BDL	BDL	BDL
∑16PAHs	1.77E-08	1.40E-08	3.02E-08	5.05E-08	3.98E-08	8.60E-08
∑PAHscar	1.74E-08	1.37E-08	2.97E-08	4.96E-08	3.91E-08	8.45E-08

Appendix C4- *PEC values for sediment samples from Benya, Brenu and Fosu lagoons*

Compound	BST1	BST2	BST3	BRST1	BRST2	BRST3	FST1	FST2	FST3
Naph	0.020	0.018	0.017	0.010	0.010	0.013	0.022	0.014	0.016
Ace	0.012	0.012	0.012	0.012	0.012	0.012	0.018	0.016	0.015
Acp	0.010	0.010	0.010	0.009	0.009	0.009	0.011	0.009	0.010
F	0.019	0.020	0.019	0.017	0.018	0.017	0.023	0.022	0.023
Ant	0.162	0.154	0.123	0.118	0.108	0.099	0.568	0.300	0.262
Phe	0.012	0.012	0.011	0.014	0.010	0.010	0.022	0.017	0.017
Flu	0.023	0.023	0.010	0.005	0.003	0.017	0.189	0.077	0.036
Pyr	0.029	0.031	0.018	0.011	0.010	0.016	0.179	0.070	0.039
BaA	2.501	3.292	2.494	1.523	1.526	1.467	9.139	4.155	3.043
Chr	0.304	0.629	0.178	0.092	0.090	0.090	1.304	0.428	0.287
BbF	2.053	2.708	1.797	1.158	1.196	1.034	14.454	7.706	3.747
BkF	1.719	1.711	1.550	0.623	0.615	0.495	9.936	3.643	2.088
BaP	23.703	27.577	21.896	13.682	13.893	13.271	99.923	45.452	29.466
DahA	2.169	3.705	3.389	4.303	3.694	1.474	17.186	9.995	10.073
IP	1.654	1.835	1.576	1.432	1.423	1.186	8.338	3.570	2.727
BP	0.065	0.081	0.042	0.000	0.010	0.002	0.621	0.174	0.086
∑16PAHs	34.453	41.819	33.143	23.010	22.628	19.213	161.932	75.647	51.935
∑PAHscar	34.167	41.538	32.922	22.814	22.448	19.019	160.901	75.123	51.517

Appendix C5- *ILCR of adults and children exposed to PAHs in lagoon sediment through dermal routes*

	Adults exposure via dermal contact								
Compound	BST1	BST2	BST3	BRST1	BRST2	BRST3	FST1	FST2	FST3
Naph	2.56E-09	2.40E-09	2.26E-09	1.31E-09	1.26E-09	1.65E-09	2.90E-09	1.85E-09	2.13E-09
Ace	1.58E-09	1.60E-09	1.55E-09	1.57E-09	1.62E-09	1.61E-09	2.36E-09	2.07E-09	1.96E-09
Acp	1.24E-09	1.26E-09	1.28E-09	1.18E-09	1.16E-09	1.17E-09	1.40E-09	1.23E-09	1.30E-09
F	2.47E-09	2.62E-09	2.43E-09	2.25E-09	2.30E-09	2.23E-09	2.96E-09	2.91E-09	2.95E-09
Ant	2.10E-08	2.01E-08	1.60E-08	1.53E-08	1.40E-08	1.29E-08	7.39E-08	3.90E-08	3.40E-08
Phe	1.52E-09	1.58E-09	1.50E-09	1.84E-09	1.34E-09	1.29E-09	2.90E-09	2.15E-09	2.21E-09
Flu	3.03E-09	2.99E-09	1.34E-09	6.25E-10	4.12E-10	2.22E-09	2.46E-08	9.97E-09	4.69E-09
Pyr	3.80E-09	4.04E-09	2.35E-09	1.43E-09	1.36E-09	2.15E-09	2.32E-08	9.05E-09	5.07E-09
BaA	3.25E-07	4.28E-07	3.25E-07	1.98E-07	1.98E-07	1.91E-07	1.19E-06	5.41E-07	3.96E-07
Chr	3.96E-08	8.18E-08	2.32E-08	1.20E-08	1.17E-08	1.17E-08	1.70E-07	5.56E-08	3.73E-08
BbF	2.67E-07	3.52E-07	2.34E-07	1.51E-07	1.56E-07	1.35E-07	1.88E-06	1.00E-06	4.88E-07
BkF	2.24E-07	2.23E-07	2.02E-07	8.11E-08	8.00E-08	6.44E-08	1.29E-06	4.74E-07	2.72E-07
BaP	3.08E-06	3.59E-06	2.85E-06	1.78E-06	1.81E-06	1.73E-06	1.30E-05	5.91E-06	3.83E-06
DahA	2.82E-07	4.82E-07	4.41E-07	5.60E-07	4.81E-07	1.92E-07	2.24E-06	1.30E-06	1.31E-06
IP	2.15E-07	2.39E-07	2.05E-07	1.86E-07	1.85E-07	1.54E-07	1.08E-06	4.64E-07	3.55E-07
BP	8.47E-09	1.06E-08	5.47E-09	3.77E-11	1.35E-09	2.78E-10	8.08E-08	2.26E-08	1.11E-08
∑16PAHs	4.48E-06	5.44E-06	4.31E-06	2.99E-06	2.94E-06	2.50E-06	2.11E-05	9.84E-06	6.76E-06
∑PAHscar	4.45E-06	5.40E-06	4.28E-06	2.97E-06	2.92E-06	2.47E-06	2.09E-05	9.77E-06	6.70E-06

Continued Children exposure via dermal contact

Compound	BST1	BST2	BST3	BRST1	BRST2	BRST3	FST1	FST2	FST3
Naph	7.29E-09	6.83E-09	6.43E-09	3.73E-09	3.59E-09	4.70E-09	8.27E-09	5.28E-09	6.06E-09
Ace	4.49E-09	4.55E-09	4.41E-09	4.47E-09	4.62E-09	4.60E-09	6.72E-09	5.90E-09	5.59E-09
Acp	3.52E-09	3.59E-09	3.65E-09	3.36E-09	3.29E-09	3.34E-09	3.98E-09	3.52E-09	3.71E-09
F	7.02E-09	7.48E-09	6.92E-09	6.41E-09	6.54E-09	6.34E-09	8.44E-09	8.27E-09	8.39E-09
Ant	5.99E-08	5.71E-08	4.57E-08	4.36E-08	3.98E-08	3.67E-08	2.11E-07	1.11E-07	9.70E-08
Phe	4.32E-09	4.50E-09	4.26E-09	5.24E-09	3.81E-09	3.67E-09	8.25E-09	6.13E-09	6.30E-09
Flu	8.62E-09	8.51E-09	3.81E-09	1.78E-09	1.17E-09	6.32E-09	7.00E-08	2.84E-08	1.33E-08
Pyr	1.08E-08	1.15E-08	6.69E-09	4.08E-09	3.88E-09	6.11E-09	6.62E-08	2.58E-08	1.44E-08
BaA	9.27E-07	1.22E-06	9.24E-07	5.64E-07	5.65E-07	5.44E-07	3.39E-06	1.54E-06	1.13E-06
Chr	1.13E-07	2.33E-07	6.60E-08	3.41E-08	3.34E-08	3.34E-08	4.83E-07	1.59E-07	1.06E-07
BbF	7.61E-07	1.00E-06	6.66E-07	4.29E-07	4.43E-07	3.83E-07	5.36E-06	2.86E-06	1.39E-06
BkF	6.37E-07	6.34E-07	5.74E-07	2.31E-07	2.28E-07	1.83E-07	3.68E-06	1.35E-06	7.74E-07
BaP	8.78E-06	1.02E-05	8.11E-06	5.07E-06	5.15E-06	4.92E-06	3.70E-05	1.68E-05	1.09E-05
DahA	8.04E-07	1.37E-06	1.26E-06	1.59E-06	1.37E-06	5.46E-07	6.37E-06	3.70E-06	3.73E-06
IP	6.13E-07	6.80E-07	5.84E-07	5.31E-07	5.27E-07	4.40E-07	3.09E-06	1.32E-06	1.01E-06
BP	2.41E-08	3.01E-08	1.56E-08	1.07E-10	3.85E-09	7.93E-10	2.30E-07	6.45E-08	3.17E-08
∑16PAHs	1.28E-05	1.55E-05	1.23E-05	8.53E-06	8.39E-06	7.12E-06	6.00E-05	2.80E-05	1.92E-05
∑PAHscar	1.27E-05	1.54E-05	1.22E-05	8.45E-06	8.32E-06	7.05E-06	5.96E-05	2.78E-05	1.91E-05

Appendix C6- *ILCR of adults and children exposed to consumption of fish contaminated with PAHs*

	Adults exposure via fish consumption								
Compound	BFF	BFV	BFH	BRFF	BRFV	BRFH	FFF	FFV	FFH
Naph	1.71E-08	1.60E-08	1.80E-08	1.28E-08	1.03E-08	1.71E-08	1.42E-08	1.71E-08	2.02E-08
Ace	1.65E-08	2.80E-08	2.50E-08	1.51E-08	1.50E-08	1.45E-08	1.67E-08	2.59E-08	2.29E-08
Acp	1.18E-08	3.40E-08	1.51E-08	1.28E-08	1.23E-08	1.30E-08	1.21E-08	1.99E-08	1.66E-08
F	1.78E-08	1.93E-08	1.87E-08	2.57E-08	1.71E-08	1.69E-08	1.67E-08	2.26E-08	2.73E-08
Ant	1.18E-07	9.09E-07	1.03E-07	1.03E-07	1.45E-07	1.26E-07	9.56E-08	1.95E-07	9.95E-08
Phe	1.02E-08	5.16E-08	1.05E-08	1.09E-08	1.19E-08	1.33E-08	1.02E-08	1.41E-08	1.01E-08
Flu	2.52E-09	4.44E-09	2.24E-08	4.14E-09	3.01E-09	2.68E-08	2.73E-09	2.99E-09	8.26E-08
Pyr	9.60E-09	9.51E-08	4.63E-08	1.15E-08	1.13E-08	8.47E-08	1.09E-08	1.64E-08	7.94E-08
BaA	1.30E-06	2.04E-06	1.29E-06	1.31E-06	4.00E-06	1.35E-06	1.24E-06	1.58E-06	1.44E-06
Chr	7.31E-08	1.27E-07	7.17E-08	7.81E-08	7.51E-08	1.09E-07	8.06E-08	2.06E-07	7.16E-08
BbF	1.17E-06	1.95E-06	1.11E-06	9.07E-07	9.59E-07	8.32E-07	1.13E-06	1.49E-06	1.18E-06
BkF	4.62E-07	9.19E-07	3.91E-07	4.78E-07	4.41E-07	5.54E-07	3.98E-07	5.34E-07	3.91E-07
BaP	1.55E-05	1.94E-05	1.34E-05	1.28E-05	1.16E-05	1.14E-05	1.57E-05	1.77E-05	1.36E-05
DahA	2.52E-06	5.43E-06	2.04E-06	5.28E-06	1.42E-06	3.96E-06	3.66E-06	2.42E-06	2.36E-06
IP	1.22E-06	1.48E-06	1.12E-06	1.19E-06	1.09E-06	1.23E-06	1.16E-06	8.36E-06	4.09E-06
BP	3.01E-09	8.10E-09	6.76E-10	6.81E-09	2.73E-08	3.06E-08	4.44E-10	3.22E-07	1.44E-08
∑16PAHs	2.24E-05	3.26E-05	1.97E-05	2.22E-05	1.99E-05	1.98E-05	2.35E-05	3.29E-05	2.35E-05
∑PAHscar	2.22E-05	3.14E-05	1.94E-05	2.20E-05	1.96E-05	1.94E-05	2.33E-05	3.23E-05	2.31E-05

Continued Children exposure to fish consumption

Compound	BFF	BFV	BFH	BRFF	BRFV	BRFH	FFF	FFV	FFH
Naph	2.38E-08	2.24E-08	2.52E-08	1.79E-08	1.44E-08	2.38E-08	1.98E-08	2.38E-08	2.82E-08
Ace	2.31E-08	3.91E-08	3.50E-08	2.10E-08	2.09E-08	2.02E-08	2.33E-08	3.61E-08	3.20E-08
Acp	1.65E-08	4.75E-08	2.11E-08	1.78E-08	1.72E-08	1.82E-08	1.69E-08	2.78E-08	2.31E-08
F	2.49E-08	2.70E-08	2.61E-08	3.60E-08	2.39E-08	2.37E-08	2.33E-08	3.15E-08	3.81E-08
Ant	1.65E-07	1.27E-06	1.43E-07	1.44E-07	2.02E-07	1.75E-07	1.33E-07	2.73E-07	1.39E-07
Phe	1.42E-08	7.21E-08	1.46E-08	1.52E-08	1.66E-08	1.86E-08	1.42E-08	1.97E-08	1.41E-08
Flu	3.51E-09	6.20E-09	3.13E-08	5.77E-09	4.20E-09	3.74E-08	3.81E-09	4.17E-09	1.15E-07
Pyr	1.34E-08	1.33E-07	6.47E-08	1.61E-08	1.57E-08	1.18E-07	1.53E-08	2.29E-08	1.11E-07
BaA	1.81E-06	2.85E-06	1.81E-06	1.83E-06	5.59E-06	1.89E-06	1.73E-06	2.20E-06	2.00E-06
Chr	1.02E-07	1.77E-07	1.00E-07	1.09E-07	1.05E-07	1.53E-07	1.13E-07	2.88E-07	9.99E-08
BbF	1.64E-06	2.73E-06	1.55E-06	1.27E-06	1.34E-06	1.16E-06	1.58E-06	2.08E-06	1.64E-06
BkF	6.45E-07	1.28E-06	5.46E-07	6.68E-07	6.16E-07	7.74E-07	5.56E-07	7.46E-07	5.46E-07
BaP	2.16E-05	2.71E-05	1.87E-05	1.78E-05	1.62E-05	1.59E-05	2.19E-05	2.47E-05	1.90E-05
DahA	3.51E-06	7.58E-06	2.85E-06	7.38E-06	1.98E-06	5.53E-06	5.11E-06	3.38E-06	3.29E-06
IP	1.71E-06	2.07E-06	1.56E-06	1.66E-06	1.52E-06	1.71E-06	1.62E-06	1.17E-05	5.72E-06
BP	4.20E-09	1.13E-08	9.43E-10	9.51E-09	3.81E-08	4.27E-08	6.21E-10	4.50E-07	2.01E-08
∑16PAHs	3.13E-05	4.55E-05	2.75E-05	3.10E-05	2.77E-05	2.76E-05	3.28E-05	4.59E-05	3.28E-05
∑PAHscar	3.10E-05	4.38E-05	2.72E-05	3.07E-05	2.74E-05	2.71E-05	3.26E-05	4.51E-05	3.23E-05

Appendix D- *Sources of PAHs within the peripheral of the lagoons under study (I: domestic waste from households in the background, II: fishing activities, III: solid wastes dumped into the lagoons and IV: a drainage that discharges untreated municipal waste directly into the lagoon)*