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Polycyclic aromatic hydrocarbons in sediments and fish species from the White Nile, East Africa: Bioaccumulation potential, source apportionment, ecological and health risk assessment☆

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ABSTRACT

The impact of oil exploration and production activities on the environment of sub-saharan African countries is not well studied. This study aimed at determining concentrations, sources, and bioaccumulation of 13 polycyclic aromatic hydrocarbons (PAHs) in sediments and fish from the White Nile near Melut oil fields, South Sudan. The study also assessed the ecological and human health risk associated with PAHs in this aquatic system. Total ($\sum_{13}$) PAH concentrations ranged from 566 to 674 ng g^{-1}dry weight (dw) in sediments, while those in fish were 191−1143 ng g^{-1} wet weight (ww). $\sum_{13}$PAH concentrations were significantly higher in *C. gariepinus* than in other fish species. Low molecular weight PAHs (LPAHs) dominated the profile of PAHs in sediments (constituted 95% of $\sum_{13}$PAHs) and fish (97% of $\sum_{13}$PAHs). Compared to Sediment Quality Guidelines of the United States Oceanic and Atmospheric Administration, the levels of LPAHs in this study were all above the threshold effect limits, but below the probable effect level, while those of high molecular weight PAHs (HPAHs) were all below the lowest effect levels. The carcinogenic potency equivalent concentrations of PAHs in *L. niloticus* and *C. gariepinus* were above the US EPA screening level; suggesting consumption of these species could adversely affect human health. Biota-sediment accumulation factor values (range: 0.006−3.816 g OC g^{-1} lipid) for PAHs showed high bioaccumulation of LPAHs in fish muscle, and that bioaccumulation decreased with increase in hydrophobicity of the compounds. This is possibly because LPAHs have higher aqueous solubilities which increases their bioavailability through water-gill transfers compared to HPAHs. Profiles of PAHs in the White Nile environment indicate predominant contribution from petrogenic sources, which could be attributed to presence of crude oil reservoirs and oil production operations. More research into the levels of other environmental pollutants in the oil-rich area is recommended.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) in the environment are of global concern due to their ubiquity, environmental persistence, long-range transport, toxicity, and potential adverse effects on human health and the environment (Net et al., 2015; Zeng et al.,

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2018). Some of the adverse health effects to humans and other living organisms include carcinogenicity, teratogenicity, mutagenicity and hepatotoxicity (IARC, 2010; Liu et al., 2017). There is evidence that exposure of humans to PAHs through ingestion and/or inhalation can result in cardiovascular diseases and abnormal sperm morphology (Pei et al., 2018; Ramesh et al., 2011). Due to these deleterious effects of PAHs in the ecosystem, the United States Environmental Protection Agency (US EPA) has included 16 PAHs on its list of priority pollutants (Keith and Telliard, 1979; Keith, 2015).

Anthropogenic activities such as incomplete combustion of fossil fuels, vehicle emissions, crude oil spills, and burning of wood and coal are the major sources of PAHs (Neff et al., 2005; Ravindra et al., 2008). However, natural vegetation fires (forest and savanna), volcanic eruptions, oil seepages, biosynthesis and diagenesis also contribute to PAHs burden in the environment (Liu et al., 2017). Once these pollutants are emitted, they can find their way into water bodies via runoff and/or atmospheric deposition (Oliva et al., 2017). In the aquatic environment, PAHs partition between the various phases (water, sediments and biota) according to their inherent physicochemical properties (Meador et al., 1995; Neff et al., 2005; Srogi, 2007). Because of their low aqueous solubility, a large proportion of PAHs in the aquatic environment are sorbed onto organic matter in sediments (Frapiccini et al., 2018). Moreover due to their lipophilicity, PAHs can also bioaccumulate across the food chain and build up in top predators (such as humans) that consume contaminated biota (Ofori et al., 2020). Consequently, PAHs pose ecotoxicological hazards to the aquatic system and to humans and livestock that depend on it (Lindén and Pålsson, 2013; Logan, 2007).

The extraction and export of natural resources such as oil is a major source of national revenue for several sub-Saharan African countries. Such oil production activities can potentially release pollutants such as PAHs, toxic elements, salts and radioactive substances into the environment (Kuorwel et al., 2018). Despite the rapid development of the oil industry and drilling activities in Africa, limited literature is available on the potential impact of these activities in the African environment. The detrimental impacts of such oil exploration, production, transport and refining on the environment have been reported in some oil producing countries such as Nigeria and Malaysia (Osuagwu and Olaifa, 2018; Zakaria et al., 2001). For instance, in Nigeria oil spills contribute significantly to infant mortality (Bruederle and Hodler, 2019).

Oil production in South Sudan's Upper Nile and Unity states is the major contributor to the nation's economy and contributes over 95% of the nation's total revenue (Grant and Thompson, 2013; Le Billon and Savage, 2016). The Nile River System in South Sudan is susceptible to severe environmental pollution due to emissions from oil production activities (Fallet, 2010; Sudan, 2018; Tiitmamer, 2016). Not much is known about how emissions from oil production activities interact with climatic and biogeochemical factors to influence PAHs (levels, composition profiles, spatial distribution, balance of sources, health and ecological risks) in aquatic systems located in tropical East Africa. The White Nile River being part of the same Nile River system, and close to the Melut oil fields, presents a suitable case to understand the dynamics and impact of this type of pollution. The objectives of this study were to [1] establish levels and sources of PAHs in sediments and fish species from a section of White Nile [2] estimate bioaccumulation potential of the pollutants in the fish relative to the sediments, [3] identify the processes and mechanisms that drive the accumulation of PAHs in sediments and fish, and [4] estimate the potential human health risks posed through consumption of fish contaminated with PAHs. The data will guide the formulation of policies for the management of the impact of oil production on the aquatic environments, as well as aid comparison studies on the dynamics of PAHs in the different ecosystems worldwide.

2. Materials and methods

2.1. Study area and sampling

The study area was a section of White Nile River which is located in vicinity of Melut town (10.440° N, 32.202° E), a small administrative town in South Sudan's Upper Nile state (Fig. 1). This section is also near a wastewater treatment plant at site C, and oil installations within the Melut oil fields (Adar, Moleeta, Gumry, and Paloch) where oil drilling is ongoing. It is possible that airborne emissions and effluents containing PAHs could be reaching White Nile via atmospheric deposition, accidental spills, runoff and/or natural seepages.

Between May and August 2019, a total of 30 surface sediments

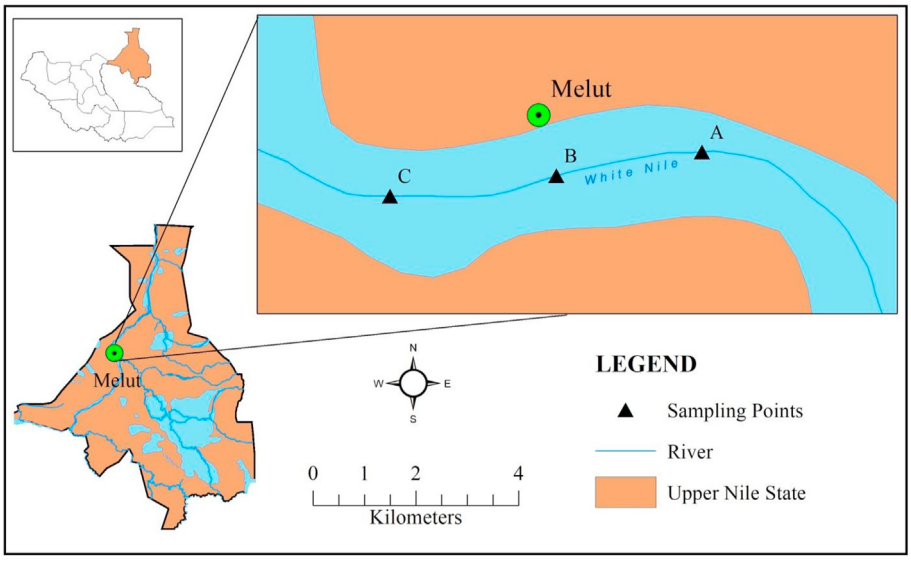

Fig. 1. Map showing study area and sampling sites.

(top 0–10 cm depth) were collected from three sampling stations (A, B and C; 10 samples from each site). The stations were 2 km from each other. Within each sampling station, samples were collected at distances of approximately 200 m from each other. Samples were then transferred into precleaned glass bottles. Three different species of fish (*Lates niloticus, Oreochromis niloticus* and *Clarias gariepinus*) were collected using gill nets from the same localities as the sediments. These species were chosen because they are commercially important and are commonly consumed locally (Sudan, 2018). A total of 66 fish samples were collected; *L. niloticus* (N = 24), *O. niloticus* (N = 18) and *C. gariepinus* (N = 24), and their weights and lengths measured. 20 g of edible muscle was sliced from each fish, wrapped in aluminum foil and labeled. The fish and sediment samples were transferred to UN boxes packed with dry ice and transported to the pesticide residue analysis laboratory at the Department of Chemistry, Makerere University, Uganda. In the laboratory, the samples were kept in a freezer at −20 °C to avoid microbial degradation before extraction.

2.2. Reagents and standards

Pesticide grade dichloromethane (DCM) and *n*-hexane were procured from Sigma-Aldrich. Other chemicals used included activated copper powder, anhydrous sodium sulfate, silica gel, sulfuric acid, potassium dichromate, ferrous ammonium sulfate and 1, 10 phenanthroline monohydrate – ferrous sulfate (ferroin), all of analytical grade. The 13 PAHs analysed were Naphthalene (Nap), Acenaphthene (Ace), Acenapthylene (Acy), Fluorene (Flu), Anthracene (Ant), Fluoranthene (Flt), Pyrene (Pyr), Benz[a]anthracene (BaA), Chrysene (Chr), Benzo[b] fluoranthene (BbF), Benzo[k] fluoranthene (BkF), Benzo[a]pyrene (BaP) and Dibenzo[a,h] anthracene (DahA). Analytical standards of 13 PAHs were purchased from Chiron (Trondheim, Norway).

2.3. Analytical procedures on the samples

2.3.1. Extraction and analysis of samples

Before extraction, all samples were allowed to thaw. Sediment samples (20 g) were freeze dried, homogenized and sieved using a 1 mm mesh. Extraction of sediment samples was done as described by Sun et al. (2016) with slight modifications. Briefly, 3 g of the freeze dried and homogenized sample was ultrasonicated in 30 ml of *n*-hexane/dichloromethane (1:1, v/v) for 30 min at room temperature. After adding copper granules, the sample was then centrifuged, and the supernatant was decanted off. The extraction procedure was repeated once more. The combined extracts were concentrated to 1 ml and then cleaned-up on a glass column (1 cm internal diameter, 30 cm length) packed with 8 g silica gel (60–230 mesh) and 3 g anhydrous sodium sulfate added on top. The column was eluted with 50 ml *n*-hexane/DCM (7:3, v/v). The eluate was evaporated to near dryness using a gentle stream of nitrogen, and then reconstituted to 1 ml with *n*-hexane and kept for instrumental analysis.

Fish samples were extracted using a method reported by Yu et al. (2016) with minor modifications. Briefly, 5 g fish muscle was ground with anhydrous sodium sulfate (20 g). The mixture was ultrasonicated in 20 ml of *n*-hexane/DCM (2:1, v/v) for 30 min. The extraction was repeated once more. The combined extracts were concentrated on a rotary evaporator and then reconstituted with 4 ml of *n*-hexane. 10 ml of sulfuric acid (60%, v/v) was added to the extract and the mixture was then centrifuged. The supernatant was cleaned on a glass column (1 cm internal diameter, 30 cm length) packed with 8 g silica gel (60–230 mesh) and 3 g anhydrous sodium sulfate. The column was eluted with 40 ml of a mixture of *n*-hexane/DCM (4:1, v/v). The collected eluate was evaporated to near dryness using a gentle stream of nitrogen gas and then reconstituted into *n*-hexane (1 ml) before gas chromatographic analysis.

Instrumental analysis was done using a Shimadzu gas chromatograph (GC) equipped with a mass spectrometer (MS) operated in multiple reactions monitoring mode. Separation of the target analytes was achieved with a ZB-5MSi capillary column (30 m × 0.25 mm internal diameter × 0.25 μm film thickness). Details of the temperature program have been presented in the supplementary material. Analyte identification was based on comparison of retention times of peaks, ion pairs and collision energies from the standards with those obtained from the sample extracts (Table S1; Fig. S3, S4). After confirmation, quantification was done using an external calibration method (R^2 = 0.999 or better).

2.3.2. Determination of total organic carbon in sediments and lipid content in fish

Total organic carbon (OC) in the sediment samples was determined as described by Okalebo et al. (2002). Briefly, 1 g of the sample was transferred into a digester tube, and 5 ml of potassium dichromate solution and 7.5 ml of conc. H_2SO_4 acid were added. The tube was then heated at 145–155 °C for 30 min, and allowed to cool. The digest was transferred to a conical flask. 0.3 ml of ferroin indicator was added and the mixture stirred. The mixture was then titrated with ferrous ammonium sulfate solution. The endpoint was reached with a colour change from greenish to brown. The blank-corrected titre value (T) was recorded and OC was calculated according to equation (1).

$$\textit{Organic carbon}\ (\%) = \frac{T \times 0.3 \times 0.2}{\textit{Sample weight}} \quad (1)$$

The lipid content in fish was determined as reported by Sun et al. (2016). Briefly, 5 g of each fish sample was extracted using ethyl acetate (20 ml) in an ultrasonic bath for 30 min. The extraction was repeated once. The solvent was evaporated to dryness from the combined extract using a rotary evaporator and lipid content determined gravimetrically.

2.4. Quality control and quality assurance

To ensure quality of the data, all samples were analysed in triplicate. Recovery studies were conducted by spiking procedural blanks, as well as fish and sediment samples (with known amounts of target chemicals) with PAH standards and applying the same analytical procedure as the samples. The average percentage recoveries of the spiked PAH target compounds ranged from 65 to 100% and 72–98% for sediment samples at spiking levels of 0.3 and 0.8 ng g^{-1}, respectively. For fish samples, the mean percentage recoveries varied from 63 to 97% and 74–105% at the two spiking levels, respectively (Table S2). As majority of the recoveries were within the acceptable range (60–120%) (Kerebba et al., 2017), the data was not corrected for recovery. For every set of five samples, blanks were analysed to check for cross contamination. Naphthalene was the only analyte detected in the blanks (detection frequency; 20%), with a maximum concentration of 2.6 ng g^{-1}. The rest of the analytical blanks did not show any peaks corresponding to any target chemicals. Fig. S1 in the supplementary information shows a mass chromatogram of a typical procedural blank. In terms of naphthalene, all data was blank-corrected by subtracting an average of all procedural blank values from the sample values. Limits of detection and quantification were calculated as 3- and 10-times the signal to noise ratio, respectively. Limits of detection varied from 0.002 to 0.148 ng g^{-1} and limits of quantification ranged from 0.006 to 0.448 ng g^{-1} (Table S3).

2.5. Statistical analysis

Descriptive statistics reporting the mean concentrations and standard deviations was used. Arithmetic means were calculated from only positive quantifiable samples. One sample *t*-test was used to determine differences between means of individual PAHs, paired samples *t*-test was used to display difference between means of two groups (LPAHs and HPAHs) and one-way analysis of variance (ANOVA) was used to display differences in levels among sites and fish species. To assess relationships between PAH levels and OC or lipid content in the samples, Pearson correlation coefficients between log-transformed PAH levels and the sample parameters. In all cases, normality of the data was confirmed using the Shapiro-Wilk test and statistical analysis were deemed significant at $p < 0.05$. For PCA, varimax factor rotation was used and only principal components with eigen values greater than 1 were retained. All statistical analyses were done using SPSS 23 (IBM, Chicago IL, USA).

2.6. Source identification

2.6.1. Diagnostic ratios

Diagnostic ratios of PAHs are commonly used as qualitative source identification to differentiate between pyrogenic and petrogenic sources and also specific fuels from which PAHs in an environmental compartment are derived (Neff et al., 2005; Tobiszewski and Namieśnik, 2012; Yunker et al., 2002). The use of diagnostic ratios (DRs) is based on the assumption that PAH isomers have similar physicochemical properties and their transformation and degradation in the environment take place at the same rate, preserving the relation that exists during emission, sample collection and until analysis (Santos et al., 2017; Yunker et al., 2002). Therefore, the ratio of the two isomers remains constant from the time of emission to the sample analysis (Tobiszewski, 2014). Diagnostic ratios which were used include Flt/(Flt + Pyr), BaA/(BaA + Chr), LPAHs/HPAHs and BbF/ BkF. Normally, Flt/(Flt +Pyr) value < 0.4 indicates petrogenic source and >0.4 shows pyrogenic origin. BaA/(BaA +Chr) > 0.35 suggests pyrogenic origin, while <0.35 indicates petrogenic source (Wattayakorn and Boonperm, 2013; Yunker et al., 2002). LPAHs/HPAHs value < 1 shows pyrogenic source, whereas > 1 indicates petrogenic origin. The ratio BbF/BkF < 0.6 indicates petrogenic source while >0.6 indicates the source is of pyrogenic origin (Tobiszewski and Namieśnik, 2012). Diagnostic ratios were only calculated if the PAHs of concern were quantifiable in the samples.

2.6.2. Principal component analysis

Principal component analysis (PCA) is used to reduce large data by transforming a large number of correlated variables to a smaller number of uncorrelated components to allow easy visualization of the similarities and differences in the data sets (Retnam et al., 2013). In environmental studies, PCA is used as an exploratory tool to identify major sources of PAHs (Cao et al., 2011; Guo et al., 2004). Loading of each source marker on each principal component gives an idea of the source of pollution (Ravindra et al., 2008). For instance high loading of Ant, Phe, Pyr and BaA indicates coal combustion, while Ant, Phe, Flt and Pyr indicates wood combustion (Khaustov and Redina, 2017). In this study, data for sediments were combined and samples with PAHs not detected were assigned zeros as their concentrations (Commendatore et al., 2012), and those with PAHs levels below the limits of detection were assigned concentration half the limits of detection. Only principal components for which the eigen value was greater than one were retained.

2.7. Estimation of bioaccumulation of PAHs

Biota-Sediment Accumulation Factor (BSAF) model is used to estimate the degree to which hydrophobic organic compounds accumulate in biota relative to sediments (Meador et al., 1995; Wong et al., 2001). The model is based on equilibrium partitioning between the sediment organic carbon and organism's lipid (Ssebugere, 2015). Normally, BSAF is expressed as the ratio of the analyte's concentration in the organism's tissue normalised to lipid content in the organism to the concentration of the same analyte in sediments normalised to total organic carbon content (Burkhard, 2009). In the current study, BSAF for each PAH in fish was calculated using equation (2) as reported by Aamir et al. (2017).

$$BSAF = \frac{C_o/f_l}{C_s/f_{soc}} \quad (2)$$

where:

C_o is the PAH concentration (ng g^{-1} ww) in the fish muscle
f_l is the lipid fraction of the muscle (g lipid g^{-1} ww)
C_s is the PAH concentration (ng g^{-1} dw) in sediment and
f_{soc} is the sediment organic carbon (g organic carbon g^{-1} dw)

2.8. Estimation of potential health risks associated with PAHs in the White Nile

The human cancer risk from the exposure to PAHs in fish was estimated using the BaP equivalent levels (BaPeq) of PAHs. For each sample, BaPeq was calculated as the product of individual concentrations of the PAH congeners and their toxicity equivalency factors (TEFs, Table S4) (Nisbet and Lagoy, 1992). All the individual BaPeq were then added up to give a carcinogenic potency equivalent concentration (PEC) of all the PAHs (equation (3)) according to Nisbet and Lagoy (1992).

$$PEC = \sum(TEF \times concentration) \quad (3)$$

The PECs for each fish species were then compared with a screening value (SV) (0.67 ng g^{-1} ww) for carcinogenic PAHs. The SV is a threshold level of total PAHs in fish muscle that is of potential public health concern (Cheung et al., 2007; Tongo et al., 2018). The SV was calculated according to Russell et al. (1997) (equation (4)).

$$SV = \left[\frac{RL}{SF} \times BW\right] \Big/ CR \quad (4)$$

where:

RL is maximum acceptable risk level (dimensionless)
SF is US EPA oral slope factor for PAHs (μg g^{-1} $day)^{-1}$
BW is the average body weight for a male adult (kg)
CR is consumption rate of fish (g day^{-1})

In the current study, RL was estimated that if a person weighing 70 kg consumed 142.2 g of fish per day with the same concentration of contaminant for 70 years, the increased risk would be at most one additional cancer death per 100,000 persons (Means, 1989; Wang et al., 2010). SF is used to estimate the upper-bound probability of an individual developing cancer as a result of lifetime (70 years) exposure to a particular level of a potential carcinogen (Cheung et al., 2007). Therefore, RL, SF, BW and CR were 10^{-5}, 7.30 (μg g^{-1} $day)^{-1}$, 70 kg and 142.2 (g day^{-1}), respectively (Cheung

Table 1
Mean levels (ng g^{-1} dw) of the 13 PAHs in sediment samples and SQGs (ng g^{-1} dw).

PAH	Sampling sites			SQGs			
	A (n = 10)	B (n = 10)	C (n = 10)	TEL	PEL	ERL	ERM
Nap	184 ± 22.7 (7)	345 ± 239 (6)	300 ± 132 (5)	30	390	160	2100
Acy	16.8 ± 15.2 (6)	21.6 ± 20.5 (6)	15.9 ± 13.9 (5)	10	130	44	640
Ace	79.3 ± 72.3 (6)	112 ± 111 (6)	74.1 ± 20.1 (4)	10	90	16	500
Flu	156 ± 148 (7)	150 ± 147.7 (6)	125 ± 101 (4)	20	140	19	540
Ant	129 ± 121 (6)	17.2 ± 14.5 (1)	22.7 ± 21.8(1)	50	240	85	1100
∑LPAHs	**565**	**644**	**537**				
Pyr	18.6 ± 16.9 (6)	15.2 ± 9.73 (6)	13.6 ± 12.1 (4)	150	1400	670	2600
Flt	15.6 ± 14.9 (6)	12.5 ± 10.8 (4)	12.9 ± 10.2 (4)	110	1490	600	2600
BaA	0.85 ± 0.71 (3)	Nd	0.05 ± 0.16 (1)	70	690	261	1600
Chr	2.19 ± 2.02 (7)	0.60 ± 0.59 (6)	0.26 ± 0.17(4)	110	850	384	2800
BbF	0.80 ± 0.14 (6)	0.78 ± 0.49 (4)	1.20 ± 0.56(4)	70	710	320	1880
BkF	0.84 ± 0.44 (4)	0.25 ± 0.24 (2)	0.19 ± 0.11 (2)	60	610	280	1620
BaP	2.84 ± 2.74 (6)	0.62 ± 0.47 (3)	0.64 ± 0.24 (4)	90	760	430	1600
DahA	0.05 ± 0.15 (1)	0.01 ± 0.04 (1)	nd	6.22	135	63	260
∑HPAHs	**41.9**	**29.9**	**28.90**				
∑PAHs	**607**	**674**	**566**				
%TOC	0.95 ± 0.09	0.71 ± 0.58	3.5 ± 0.62				

nd = non-detectable, n = number of sediment samples, levels reported as mean ± standard deviation (positive samples), SQG values adapted from Long and MacDonald (1998) and Darilmaz et al. (2019), TEF- Threshold effect level, PEL- Probable effect level, ERL- Effect range low, ERM- Effect range median.

et al., 2007).

3. Results and discussion

3.1. PAHs in sediments

Levels of the 13 PAHs in the sediment samples from White Nile are presented in Table 1. Total $(\sum)_{13}$PAHs were 608, 674 and 566 ng g^{-1} dry weight (dw) at stations A, B and C, respectively.

These levels did not differ significantly amongst sampling locations ($p > 0.05$). At the studied sites, low molecular weight PAHs (LPAHs; 2- and 3-ring PAHs) were detected at concentrations ranging from 537 to 644 ng g^{-1} dw, accounting for 95% of ∑PAHs. Naphthalene was the most dominant congener amongst the LPAHs (contributed 33–56% to the ∑LPAHs), followed by Flu, Ace, and Ant. The levels of LPAHs were significantly higher ($p < 0.05$, *t*-test) than those of higher molecular weight (4- and 5- ring) PAHs (HPAHs: range 29–42 ng g^{-1} dw) at all the sites. The similarity in the distribution patterns of PAHs among different sites is indicative of a common source of PAHs pollution (Li et al., 2010). The pattern of PAHs observed in the current study is similar to those reported in studies of PAHs in sediments and soils from the oil producing region (Niger Delta Region) of Nigeria (Asagbra et al., 2015; Inam et al., 2018; Sojinu et al., 2010a). Predominance of LPAHs over HPAHs in the environmental samples can be attributed to unburnt petroleum sources such as petroleum-related activities (Fourati et al., 2018). In the present study, crude oil source and/or petroleum exploration and production activities taking place in the vicinity of the White Nile could be releasing PAHs and related compounds into the environment through accidental spills and airborne emissions. These compounds may also find their way into the White Nile through surface runoff, natural seepage and/or atmospheric deposition.

Some of the PAH molecular diagnostic ratios for sampling sites A, B and C are presented in Table S5 (supplementary information). The ratios $\frac{BaA}{(BaA+Chr)}$, $\frac{\sum LPAHs}{\sum HPAHs}$ and $\frac{BbF}{BkF}$ were 0.28, 17.66 and 0.39, respectively in sediments at sampling site A, suggesting unburnt petroleum as the major source of PAHs in the White Nile. However, the value 0.46 for $\frac{Flt}{(Flt+Pyr)}$ ratio suggested a pyrogenic source of PAHs (Souza et al., 2018). In station B, the ratio $\frac{Flt}{(Flt+Pyr)}$ was 0.45 suggesting a pyrogenic source. Similarly, in station C, the ratios showed majorly petrogenic source with minor contribution from pyrogenic source. Generally, diagnostic ratios also suggest that the sources of PAHs in the sediments of the White Nile are mainly petrogenic. This could be attributed to the petroleum activities near the study area.

Principal component analysis (PCA) of the PAHs in sediment samples generated three principal components (PC1 to PC3) which explained 49.59%, 16.68% and 14.16% of the total variance in the data set, respectively as shown in Table S6 (Supplementary information). PC1 was heavily loaded with Acy, Ace, Nap and Flu, and moderately loaded with Pyr, BbF and Flt. Nap, Acy, Ace and Flu (PAHs with 2–3 rings) are mainly from petroleum origin or from biomass burning (Neff et al., 2005). Pyr, Flt and BbF are mainly from high temperature combustion (Ravindra et al., 2008; Tobiszewski and Namieśnik, 2012). Thus, PC1 could be likely derived from mainly petrogenic source and biomass combustion. PC2 was loaded with Chr, Bap, BkF and Ant. Chr and BkF were associated with gasoline/diesel vehicle emissions (Tobiszewski and Namieśnik, 2012; Yali et al., 2019). PC3 was loaded with BaA, DahA, Ant and BkF. BaA is source marker for gasoline combustion, while Ant is associated with biomass combustion. Therefore, PCA confirmed that the main source of PAHs in sediment samples from the White Nile, South Sudan were petrogenic with minor contributions from biomass combustion and vehicular emissions. The petrogenic source could be due to the petroleum activities in the area. Biomass combustion source may be attributed to burning of wood for fuel and grass burning around Melut town, while vehicular emissions could be as a result of combustion of gasoline or diesel-powered vehicles in the area. Furthermore, it could also be brought by long range atmospheric deposition.

In comparison to other related studies, the $\sum_{13}$PAHs levels (566–674 ng g^{-1} dw) in sediments of the current study were in the same range of data as that in some studies on the African continent such as in Egypt (161–874 ng g^{-1}dw) (El-Kady et al., 2018), Nigeria (365–1432 ng g^{-1} dw) (Sogbanmu et al., 2016) and Senegal (2–636 ng g^{-1} dw) (Net et al., 2015). The measured concentrations in sediments in the current study were lower than those reported from South Africa (1,168,000–10,469,000 ng g^{-1} dw) (Adeniji et al., 2019), but higher than those recorded in Uganda (17–80 ng g^{-1} dw) (Kerebba et al., 2017), Togo (4–257 ng g^{-1} dw) (Gnandi et al., 2011) and Morocco (22–108 ng g^{-1} dw) (Giuliani et al., 2015). Compared

Table 2
Mean levels (ng g^{-1} ww) of the 13 PAHs in fish samples.

PAHs	A			B			C		
	L. niloticus	*O. niloticus*	*C. gariepinus*	L. niloticus	*O. niloticus*	*C. gariepinus*	L. niloticus	*O. niloticus*	*C. gariepinus*
	n = 8	n = 6	n = 8	n = 8	n = 6	n = 8	n = 8	n = 6	n = 8
Nap	281 ± 184 (6)	415 ± 263 (5)	957 ± 293 (7)	516 ± 341 (5)	413.2 ± 402 (4)	812 ± 643 (6)	662 ± 413 (6)	136 ± 91 (2)	720 ± 494 (5)
Acy	9.57 ± 1.60 (5)	9.35 ± 1.76 (5)	10.9 ± 5.21(7)	9.94 ± 4.19 (5)	9.26 ± 4.77 (4)	8.37 ± 5.76 (7)	10.7 ± 4.29(5)	3.0 ± 2.77(2)	6.82 ± 5.93(5)
Ace	39.7 ± 12.6 (5)	34.0 ± 13.9 (5)	79.9 ± 39.7 (7)	41.7 ± 32.55(5)	37.1 ± 23.7 (4)	60.±39.8 (6)	49.1 ± 28.8(5)	33.9 ± 9.52(2)	45.1 ± 42.7 (5)
Flu	56.4 ± 12.3 (5)	47.5 ± 21.8(5)	68.1 ± 32.5 (7)	53.9 ± 39.33(5)	59.8 ± 50 (4)	53.3 ± 37.1(6)	64.3 ± 36.1(5)	13.2 ± 8.17(4)	38.1 ± 36.8 (4)
Ant	79.3 ± 8.60 (3)	21.1 ± 20.8 (2)	5.98 ± 7.67(1)	4.47 ± 5.65(1)	nd	16 ± 9.67(2)	17.3 ± 19.8(1)	nd	10.9 ± 12.9(1)
∑LPAHs	**466**	**527**	**1124**	**626**	**520**	**950**	**804**	**186**	**820**
Pyr	2.46 ± 0.45 (5)	2.09 ± 0.92 (4)	6.51 ± 2.31(7)	4.25 ± 2.68(5)	3.78 ± 3.45(4)	10.2 ± 8.74(6)	2.98 ± 0.92(5)	0.78 ± 0.26(2)	4.52 ± 2.30(5)
Flt	2.46 ± 1.57 (2)	8.73 ± 1.88(4)	9.07 ± 6.14(6)	13.7 ± 6.64(3)	9.54 ± 4.84(4)	11.4 ± 6.07(5)	8.44 ± 1.2(4)	3.13 ± 1.87(2)	7.88 ± 6.09(5)
BaA	nd	nd	0.12 ± 0.57(1)	nd	0.10 ± 0.14(1)	1.39 ± 0.37(3)	0.08 ± 0.09(1)	nd	0.10 ± 0.28(1)
Chr	1.19 ± 0.06(5)	1.05 ± 0.55(5)	0.84 ± 0.45(7)	1.05 ± 0.50(5)	0.72 ± 0.65(4)	1.34 ± 1.19(6)	1.30 ± 0.10(5)	0.44 ± 0.39(2)	0.96 ± 0.04(5)
BbF	nd	0.78 ± 0.22(2)	0.57 ± 0.31(3)	2.42 ± 0.13(3)	0.48 ± 0.18(2)	0.33 ± 0.26(3)	0.13 ± 0.27(1)	nd	0.19 ± 0.23(1)
BkF	0.10 ± 0.30(1)	0.20 ± 0.40(1)	0.11 ± 0.30(1)	0.89 ± 0.28(2)	0.16 ± 0.24(1)	0.78 ± 0.43(4)	0.98 ± 0.06(3)	0.19 ± 0.36(1)	0.05 ± 0.07(1)
BaP	0.02 ± 0.04(1)	0.09 ± 0.10(1)	1.06 ± 2.24(1)	nd	0.06 ± 0.02(2)	0.68 ± 0.31(4)	0.19 ± 0.08(2)	0.16 ± 0.28(1)	0.63 ± 0.58(2)
DahA	nd	nd	nd	nd	0.04 ± 0.03(2)	0.13 ± 0.04(2)	0.03 ± 0.21(1)	nd	nd
∑HPHs	**6.23**	**12.94**	**18.28**	**22.3**	**14.9**	**26.24**	**14.1**	**4.70**	**14.3**
∑PAHs	**472**	**540**	**1143**	**648**	**534**	**976**	**817**	**191**	**835**
%Lipid	5.79 ± 1.85	2.18 ± 1.97	2.32 ± 0.89	3.83 ± 2.16	1.78 ± 1.27	2.40 ± 1.04	2.67 ± 1.75	1.47 ± 0.84	2.41 ± 0.45

nd-non detectable (below the limit of detection), n = number of fish samples, levels reported as mean ± standard deviation (Positive samples), %lipid reported as mean ± standard deviation.

to studies outside Africa, the levels of PAHs in the present study were in the same range with studies reported from South China (340–710 ng g^{-1} dw) (Sun et al., 2016), Korea (207–2670 ng g^{-1} dw) (Yim et al., 2005), but lower than those concentrations recorded from Malaysia (869–1637 ng g^{-1} dw) (Nasher et al., 2013). The variations of the PAH levels in the different studies may be attributed to differences in pollution sources and number of studied PAH analytes.

Comparison of the PAH levels in sediments in this study against the sediment quality guidelines (SQGs) developed by the United States Oceanic and Atmospheric Administration (US NOAA) (Hahladakis et al., 2013; Long and MacDonald, 1998), showed that the concentrations of most of the individual LPAHs were above the threshold effect levels (TELs) (10–50 ng g^{-1} dw) and effect range low (ERL) limits (16–160 ng g^{-1} dw), but below the probable effect levels (PELs; 90-30 ng g^{-1} dw) and effect range medium (ERM; 500–2100 ng g^{-1} dw) limits (Table 1). Hence, the concentrations of LPAH levels in the sediments of the White Nile sediments might pose ecological risks. The concentrations of all the individual HPAHs levels were below the TEL (6.2–150 ng g^{-1} dw) and ERL (63–670 ng g^{-1} dw) limits.

3.2. PAHs in fish samples

Levels of PAHs in the three fish species (*L. niloticus, O. niloticus* and *C. gariepinus*) are presented in Table 2.

The $\sum_{13}$PAHs ranged from 472 to 819 ng g^{-1} ww, 191–540 ng g^{-1} ww and 835–1143 ng g^{-1} ww in *L. niloticus, O. niloticus* and *C. gariepinus*, respectively. In the current study, the levels of the LPAHs (Nap, Ace, Acy, Ant and Flu; range; 186–1125 ng g^{-1} ww) in the fish species were higher compared to the levels of the HPAHs (Pyr, Flt, BaA, Chr, BbF, BkF, BaP and DahA; range; 5–26 ng g^{-1} ww). LPAHs accounted for 98% of ∑PAHs in fish. Nap, Flu, Ace and Ant contributed most to the total PAH content in all the species. The dominance of LPAHs in the fish muscles has been observed in other studies (Bandowe et al., 2014; Jafarabadi et al., 2019; Soltani et al., 2019) and may suggest pollution of the fish by a petroleum sources such as direct discharge of unburnt fuel and tankers transporting oil (Oliva et al., 2017; Soltani et al., 2019). The dominance of 2–3 ring PAHs especially naphthalene was also reported in higher plants sampled from Niger Delta region (oil production activity site) of Nigeria (Sojinu et al., 2010b). It could also be attributed to the relatively higher water-gill transfer efficiencies of LPAHs (Jafarabadi et al., 2019). Since LPAHs have high aqueous solubility and lower octanol-water partition coefficients (Log K_{ow}< 5.0), their concentrations in water are higher and they are more easily absorbed by the fish compared to HPAHs. Moreover, HPAHs are more quickly metabolized by fish than the LPAHs (Bandowe et al., 2014; Abdallah, 2017). This lowers the levels of HPAHs detected.

In this study, the concentrations of PAHs in *C. gariepinus* were significantly higher ($p < 0.05$, ANOVA) compared to those in other fish species (Table 2). This could be attributed to differences in feeding habits between *C. gariepinus* and other species (Lyytikäinen et al., 2007). The feeding habits of *C. gariepinus* vary from being a benthic omnivore to a piscivore (Dadebo et al., 2014; Meri et al., 2018). This leads to a higher level of exposure of *C. gariepinus* to PAHs and could have resulted to the high concentrations in the species given that PAHs bioaccumulate across the food chain.

When compared to other studies in Africa, the $\sum_{13}$PAHs concentrations (191–1143 ng g^{-1} ww) in fish tissues in the present study are comparable to ranges of PAH levels in fish muscle reported from Egypt (285–1008 ng g^{-1} ww) (Abdallah, 2017) and Nigeria (43–937 ng g^{-1} ww) (Effiong et al., 2016), but higher than those reported from Madagascar (2–63 ng g^{-1} ww) (Rumney et al., 2011) and Ghana (2–167 ng g^{-1} ww) (Nyarko and Klubi, 2011). When compared to studies outside Africa, the concentrations of PAHs in this study are higher than those reported in China (53–248 ng g^{-1} ww) (Su et al., 2015) and in St. Helena (2–20 ng g^{-1} ww) (Rumney et al., 2014), but lower than levels reported from Brazil (26,520–2055,000 ng g^{-1} ww) (Froehner et al., 2018). The variations in the levels of PAHs in fish tissues in the different locations could be because of variability in the fish species studied and sources of PAH pollution.

3.3. Variation of PAH concentrations with TOC in sediment samples and lipid in fish samples

We explored the relationship between the PAH levels in sediments and fish with TOC and lipid content, respectively (supplementary information, Tables S7 and S8). At site A, the LPAH concentrations had a significant positive correlation with TOC in

sediments ($r > 0.79$, $p < 0.05$, $n = 10$), whereas no significant correlation was observed between all the HPAHs and TOC ($p > 0.05$). The PAH levels in sediments did not correlate significantly with TOC at sites B and C, except Chr at site C. Similarly, no significant correlation was observed between PAH concentrations in the fish species with the lipid content, except for BbF in *O. niloticus*, and Bap which had negative correlation.

The poor correlation between PAHs and TOC may indicate that the distribution of PAHs in the White Nile is not invariant due to the dynamic nature of sorption and desorption processes in river systems (Zhu et al., 2008). In addition, the poor correlation could suggest that the PAHs in these sediments come from more than one source and that their levels are more influenced by direct inputs rather than by sedimentary characteristics (Ameur et al., 2010; Nouira et al., 2013). Since the PAH profiles at all sites suggested petrogenic sources, this poor correlation suggested that the oil producing activities in the region could be responsible for spillages of a number of petrogenic substances such as crude oil or refined products like gasoline, asphalt, or coal, all of which contribute to the PAH load in the environment. Similarly, the non-significant correlation between PAHs concentrations in fish with lipid content indicated that lipid content was not a key factor for the PAHs accumulation in the fish tissues, and that the levels of PAHs in fish muscle may not have reached chemical equilibrium. These findings are consistent with those of Devier et al. (2005) who observed no correlation and Granby and Spliid (1995) who observed negative correlation between PAHs concentrations in organism's tissues and lipid content. However, some studies such as Jafarabadi et al. (2019) observed strong correlations between PAH concentrations in fish with lipid content, showing lipid was a key factor for the PAHs accumulation.

3.4. Bioaccumulation of PAHs in fish from the White Nile

The accumulation of PAHs in fish relative to sediments was estimated by calculating biota-sediment accumulation factors (BSAFs). BSAFs for the PAHs in the three fish species (*L. niloticus*, *O. niloticus* and *C. gariepinus*) at sampling sites A, B and C are shown in Table 3.

BSAF values for the PAHs for *L. Niloticus* ranged from 0.006 g OC g^{-1}lipid for BbF to 3.816 g OC g^{-1} lipid for Nap, while for *O. niloticus*, the BSAFs values varied from 0.029 g OC g^{-1} lipid for BkF to 3.799 g OC g^{-1} lipid for Nap. For *C. gariepinus*, BSAFs values ranged from 0.154 g OC g^{-1} lipid for BbF to 3.209 g OC g^{-1} lipid for Nap. BSAFs for BaA and DahA have not been presented because they were detected in either the fish species or sediment in less than three samples. Generally, BSAF values decrease with increase in ring size of the PAHs in all the fish species. In addition, negative linear relationships were observed between the BSAFs values for the PAHs in the fish species and their respective Log K_{ow} values (Fig. 2). The Log K_{ow} values adopted in this study were taken from Stogiannidis and Laane (2015).

Our findings were consistent with those of Kwok et al. (2013) and Liang et al. (2007), who reported lower BSAF values for HPAHs than LPAHs in fish samples from Mai Po Marshes in Hong Kong. The decreasing trend of BSAFs with increasing number of aromatic rings in the PAHs can be attributed to the strong affinity of HPAHs to sediments which contain organic and black carbon (which are strong sorbents for PAHs). This lowers their bioavailability and bioaccumulation in the fish tissues. LPAHs on the other hand have higher water solubility which enhances direct water-gill transfer and hence bioaccumulation rates (Jafarabadi et al., 2019). Moreover, HPAHs have relatively higher rates of conversion into their metabolites which reduces their capacity to bioaccumulate (Abdallah, 2017; Zhang et al., 2015).

Among the sampling sites, the values of the BSAFs were significantly higher at site C than the other sites. Site C is close to Melut water treatment plant whose effluents may have contributed to PAHs presence and the TOC content in the sediments. Studies have reported decreased bioavailability of hydrophobic organic compounds (HOCs) such as PAHs in sediments with high TOC content (Ferraro et al., 1990). However, the opposite observation showed by the present study might be attributed to the nature of organic carbon responsible for PAHs binding at site C. HOCs in sediments may be more or less available depending on the type of organic carbon which determines how weak or strong the compounds are sorbed to the sediment organic matter (Ghosh, 2007).

Among the fish species, *C. gariepinus* had the highest BSAF values for the PAHs. This could have resulted because of the feeding habits of the fish since it is a benthic omnivore and piscivore (Meri et al., 2018). *L. niloticus* had higher values of BSAFs than *O. niloticus*. The probable reason could be the difference in their trophic levels. *L. niloticus* is a piscivore unlike *O. niloticus* which eats zooplankton and some invertebrates (Ssebugere, 2015).

3.5. Human health risk posed by the consumption of PAH-contaminated fish

The average levels of BaP (0.03–0.79 ng g^{-1} ww) in all the fish tissues analysed were below the maximum limit of 2 ng g^{-1} ww set by European Union (EU) for fish muscle, which is recommended to be safe for human consumption (Larsen, 2008). The BaPeq concentrations (due to the concentrations of 13 PAHs) in the three fish species are presented in Table S4 (supplementary information). The PEC values ranged from 0.65 to 2.21 ng g^{-1} ww. The PECs for *L. niloticus* and *C. gariepinus* were higher than the SV level, thus

Table 3
BSAFs (g OC g^{-1} lipid) values for PAHs in the fish species.

PAH	*L. niloticus*			*O. niloticus*			*C. gariepinus*		
	A	B	C	A	B	C	A	B	C
Nap	0.18 ± 0.13	0.50 ± 0.27	3.82 ± 0.27	0.61 ± 0.07	1.51 ± 1.15	3.80 ± 1.33	1.00 ± 0.63	0.99 ± 0.47	3.21 ±1.05
Acy	0.08 ± 0.02	0.13 ± 0.02	1.02 ± 0.58	0.53 ± 0.34	0.38 ± 0.20	1.63 ± 0.30	0.69 ± 0.20	0.90 ± 0.18	1.67 ±1.13
Ace	0.12 ± 0.08	0.41 ± 0.06	1.56 ± 1.31	0.46 ± 0.21	1.09 ± 0.15	2.02 ± 0.02	0.76 ± 0.13	0.80 ± 0.36	1.80 ± 1.15
Ant	0.08 ± 0.02	NA	NA	NA	NA	NA	0.69 ± 0.14	NA	NA
Flu	0.13 ± 0.09	0.44 ± 0.22	2.07 ± 1.89	0.31 ± 0.16	1.46 ± 1.14	3.01 ± 0.39	1.00 ± 0.84	0.34 ± 0.13	2.35 ±0.95
Pyr	0.05 ± 0.04	0.11 ± 0.02	0.75 ± 0.38	0.25 ± 0.10	0.26 ± 0.19	1.34 ± 0.88	0.57 ± 0.23	0.24 ± 0.10	1.56 ± 1.03
Flt	0.08 ± 0.04	0.11 ± 0.08	0.87 ± 0.54	0.24 ± 0.13	0.27 ± 0.18	1.51 ± 0.53	0.46 ± 0.24	0.22 ± 0.18	1.60 ± 0.18
Chr	0.04 ± 0.02	0.08 ± 0.07	0.61 ± 0.35	0.20 ± 0.16	0.16 ± 0.08	0.89 ± 0.12	0.30 ± 0.17	0.22 ± 0.09	1.25 ± 0.40
BbF	0.01 ± 0.00	0.06 ± 0.02	0.60 ± 0.38	0.11 ± 0.01	0.13 ± 0.07	1.08 ± 0.05	0.30 ± 0.05	0.15± 0.03	1.07 ± 0.30
BaP	0.02 ± 0.01	0.03 ± 0.02	0.37 ± 0.19	0.17 ± 0.10	0.06 ± 0.01	1.31 ± 0.88	0.27 ± 0.11	0.17 ± 0.10	0.93 ± 0.24
BkF	0.03 ± 0.00	0.03 ± 0.01	0.46 ± 0.37	0.03 ± 0.01	0.13 ± 0.06	0.57 ± 0.05	0.21 ± 0.10	0.18 ± 0.12	0.83 ± 0.38

BSAFs reported as mean ± standard deviation, N = 3 paired samples, NA = not available (compound not detected either in fish or sediment).

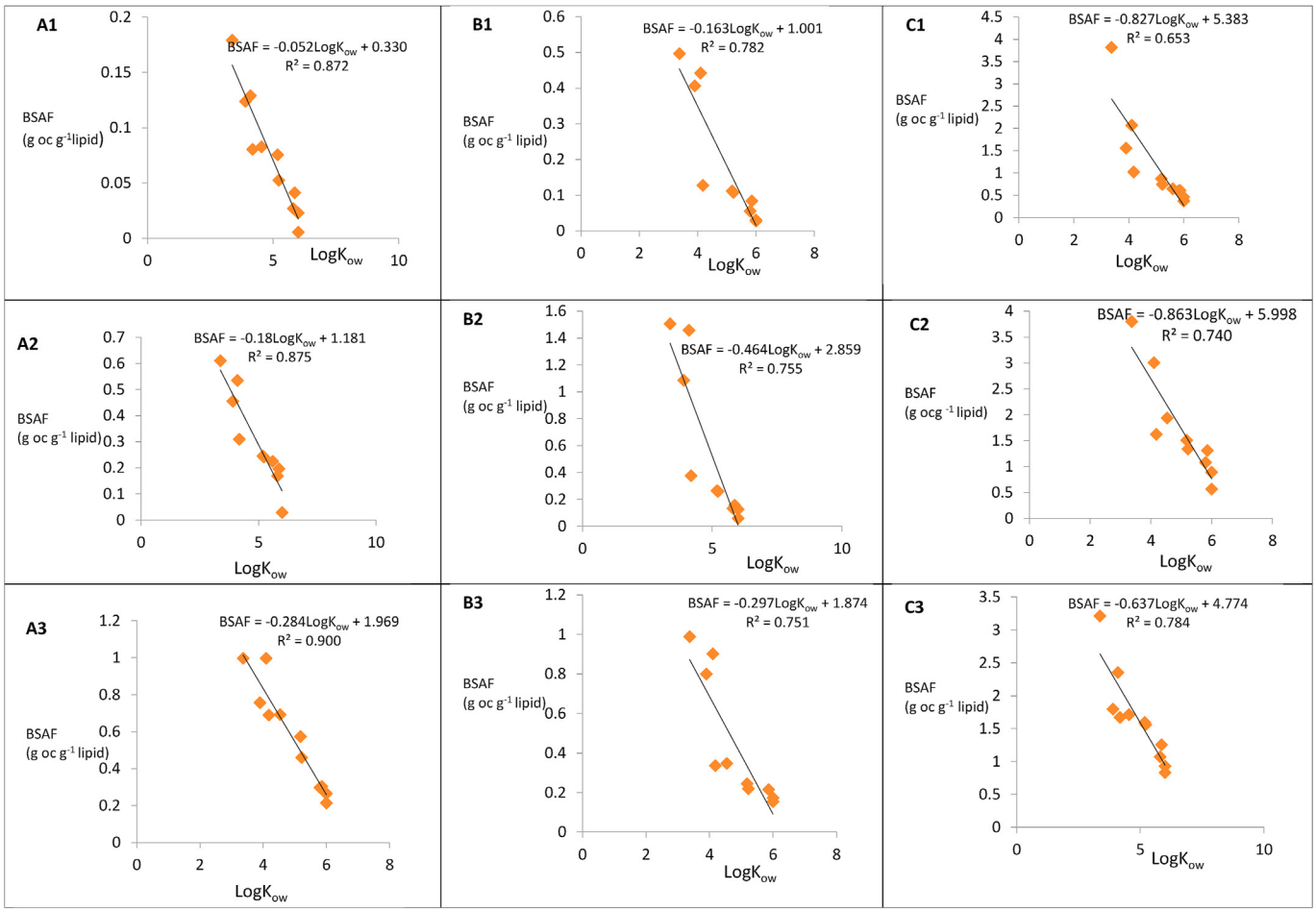

Fig. 2. Relationships between BSAFs for PAHs and their individual $LogK_{ow}$ values for *L. niloticus* (A1, B1, C1), *O. niloticus* (A2, B2, C2), and *C. gariepinus* (A3, B3, C3) at sites A, B and C, respectively.

human consumption of these species could present a health risk with respect to PAHs. The PEC values recorded in this study were higher than studies in Ghana (0.003–0.012 ng g^{-1}) (Nyarko and Klubi, 2011) and Nigeria (0.024–0.12) (Nkpaa et al., 2013), suggesting that the health risk resulting from the consumption of contaminated fish is higher in South Sudan than in Ghana or Nigeria. However, our PEC values were lower than those reported in Poyang Lake, China (1.89–24.9 ng g^{-1}) (Zhao et al., 2014). This variation might be attributed to differences in sources and concentrations of PAHs (to which the fish are exposed) in the different study locations.

4. Conclusions

The study investigated, for the first time, the dynamics of PAHs in sediments and fish from the part of the White Nile near Melut Oil fields. $\sum_{13}$PAHs in sediments and fish ranged from 566 to 674 ng g^{-1} dw and 191–1143 ng g^{-1} ww, respectively. In both sediments and fish, $\sum$LPAHs/$\sum$HPAHs ratios were >1). SQG values indicated that the levels of LPAHs in sediments might pose ecological risks. PECs for *L. niloticus* (0.76 ng g^{-1}ww) and *C. gariepinus* (2.21 ng g^{-1}ww) were also higher than the threshold level (0.67 ng g^{-1} ww) for $\sum_{13}$PAHs in fish muscles. This suggested potential health risks to the fish consumers. However, BaP average levels in all the fish species studied were lower than the maximum limit of 2 ng g^{-1} ww set by European Union (EU) for fish tissue, which is recommended to be safe for human consumption. Molecular diagnostic ratios and PCA revealed the sources of the PAHs were mainly from petrogenic origin with minor contributions from biomass combustion and vehicular emissions. The petrogenic source could be related to the crude oil reservoirs and/or oil production activities around Melut. Biota-sediment accumulation factors (BSAFs) for the PAHs in the fish tissues demonstrated greater bioaccumulation for LPAHs than the HPAHs. Overall, our study sheds light on the effects of the oil drilling activities on the quality of sediments and fish in White Nile ecosystem. We recommend that follow up studies should collect samples from a larger number of sampling locations within the Nile River system and its catchment (sediments, fish, invertebrates, riparian plants, soils and air samples) and determine an extended list of PAHs and their derivatives in these samples. This will further help to improve our understanding of the magnitude and spatial distribution of PAHs, as well as their fate, bioaccumulation, potential negative effect in the White Nile environment.

Author contribution statement

Juma John Moses Abayi: Conceptualization, Methodology, Formal analysis, Investigation; Methodology, Writing – original draft. **Christopher Tombe Gore**: Conceptualization, Writing – review & editing and Funding acquisition. **Christine Nagawa**: Writing – review & editing, Supervision, Funding acquisition.

Benjamin A. Musa Bandowe: Writing – review & editing. **Henry Matovu**: Conceptualization, Investigation; Methodology, Writing – review & editing. **Edward Mubiru**: Writing – review & editing. **Emily Chelangat Ngeno**: Writing – review & editing. **Silver Odongo**: Writing – review & editing. **Mika Sillanpää**; Supervision, Writing – review & editing. **Patrick Ssebugere**: Conceptualization, Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

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