

## Original Paper

# Source and Distribution of Polycyclic Aromatic Hydrocarbons (PAHs) in Water from Mboppi River in Douala–Cameroon

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Received: October 17, 2021    Accepted: November 5, 2021    Online Published: November 16, 2021

doi:10.22158/se.v6n4p1

URL: <http://dx.doi.org/10.22158/se.v6n4p1>

### **Abstract**

*Having a source attributed to anthropogenic activities such as incomplete combustion or pyrolysis of organic materials, polycyclic aromatic hydrocarbons are toxic organic pollutants that the ubiquity is no more to be proved. The purpose of this work is to identify and quantify the hydrocarbons pollution of the Mboppi River. Eight (8) samples of water were collected in the river during the dry and the rainy season (4samples for each season). Hydrocarbon fraction was extracted by magnetic agitation of the mixture water/hexane followed by clean-up, fractionation and subsequently, analysis using gas chromatography coupled to a Flame Ionization Detector (GC-FID). Total concentrations of polycyclic aromatic hydrocarbons in the samples were ranged between 196.3-1040.19 µg/L in the river. Polycyclic aromatic hydrocarbons with more than four rings showed the highest concentrations in the river independently from the seasonal variation while the polycyclic aromatic hydrocarbons with 2 or 3 rings were usually present in low concentrations or sometimes undetectable. From the data, it was also possible to conclude that there is predominance of petroleum sources, and essentially closed to the more industrialized areas. Mboppi River can then be considered as being among the most polycyclic aromatic hydrocarbons polluted environment in comparison with some rivers and estuaries.*

### **Keywords**

*Cameroon, gas chromatography, mboppi River, polycyclic aromatic hydrocarbons, water body*

## 1. Introduction

Defined as earth system predominantly influenced by humans, Anthropocene include dispersion of pollutants in all environmental components (Mbusnum et al., 2020). The same as coastal management becomes a real issue, the management of surface water plan in general and City Rivers in particular are of most interest. Urban waste has been highly discussed and various techniques of management has been developed. Due to the population that grows rapidly and city expansion, new environmental issues are rising up among which hydrocarbon pollution (Wijesiri et al., 2019). Human activities such as industrial activities, transportation, etc., have incidents like oil spill, waste mismanagement that leads to air, soil and water pollution. Many studies have shown that water bodies besides industries were receiving pollutants from these last (Maskaouia et al., 2002; Shi et al., 2005; Zhifeng et al., 2008; Brum et al., 2009). Polycyclic Aromatic Hydrocarbons (PAHs) are produced by natural processes and anthropogenic activities and introduced into the environment through various routes. Anthropogenic inputs can originate from incomplete combustion, oil spills, domestic and industrial wastewater discharges, as well as atmospheric fallout of vehicle exhaust and industrial stack emission (Montuori et al., 2020). It has also been demonstrated that there are some water characteristics that have an influence on the behavior of PAHs in water among which Conductivity (Jing et al., 2017), Turbidity (Abuhelou et al., 2017; Guangchao et al., 2021), Organic matter content (Raber & Kogel-Knabner, 1997; Yang et al., 2010; Nascimento et al., 2017; Li et al., 2018), and pH (Zeledón-Toruño et al., 2007; Lamichhane et al., 2016) can be listed out.

Studies conducted by Meva'a (2007) illustrated the appearance of oily dark scum at certain points of the Mboppi River. Let's stress that this river is flowing beside some companies or structures Railway station, Petrol stations, etc. Water plant pollution by hydrocarbons is of interest since it is known that any life have a high need water. The alarming fact is that among polluting hydrocarbons, there are those which are not biodegradable, persistent, or carcinogenic (Aljerf & Al-Masri, 2018). PAHs, because of their bioaccumulation capacity and environmental persistence, special attention has been paid on these pollutants. The solubility of PAHs in water is low and decreases with the increase of molecular weight (Haritash & Kaushik, 2009). Lighter PAHs, like naphthalene, are more water-soluble and volatile than the heavier compounds, and will therefore be less persistent in the environment, being subject to dilution, evaporation and bacterial degradation (Page et al., 1999). It has been shown that water bodies going across an industrial zone or a developing city is polluted by PAHs. Thus, Montuori et al. (2020) recorded the PAHs concentration range of 318.1-1429.1 ng.L<sup>-1</sup> in water from Volturno River of Southern Italy. The level of pollution by PAHs in some water bodies around the world are listed in the Table 1 below. It has been shown the seasons have an influence on the concentration variation of PAHs in water bodies (Chizhova et al., 2020). In contrary to what appears in Table 1 that there is no African country mentioned, Ofori et al. (2020) made a systematic review on the occurrence and levels of PAHs in African environments. After a selection of published articles, 121 of them were retained and distributed through 19 countries out of 54, with Nigeria having up to 56. The total PAHs

in aquatic environment from this systematic review ranged from 0 to 10,469,000  $\mu\text{g}/\text{kg}$  (Ofori et al., 2020). It comes out from it that Cameroon is not counted among countries where published studies on PAHs have been done before. Firstly, it has been pointed out that the total concentration of PAHs in coastal sediments from Ngoua River-Cameroon ranged from 140.42  $\mu\text{g}/\text{g}$  to 229.47  $\mu\text{g}/\text{g}$  dry matters during the rainy season and from 48.89  $\mu\text{g}/\text{g}$  to 333.49  $\mu\text{g}/\text{g}$  dry matters during the dry season (Jessie et al., 2014). Secondly, persistent organic pollutants, among which PAHs, in sediments of the Wouri Estuary Mangrove, Cameroon: Levels, patterns and ecotoxicological significance have also been studied (Mbusnum et al., 2020). Effectively, they showed that the total PAHs concentration in the Wouri Estuary Mangrove is in the range of 83-544  $\text{ng}/\text{g}$ . Comparing to Nigeria, it can be accepted that many studies have not yet been done and published on Cameroon water bodies.

**Table 1. Level of River Pollution by PAHs in Some Developed Countries**

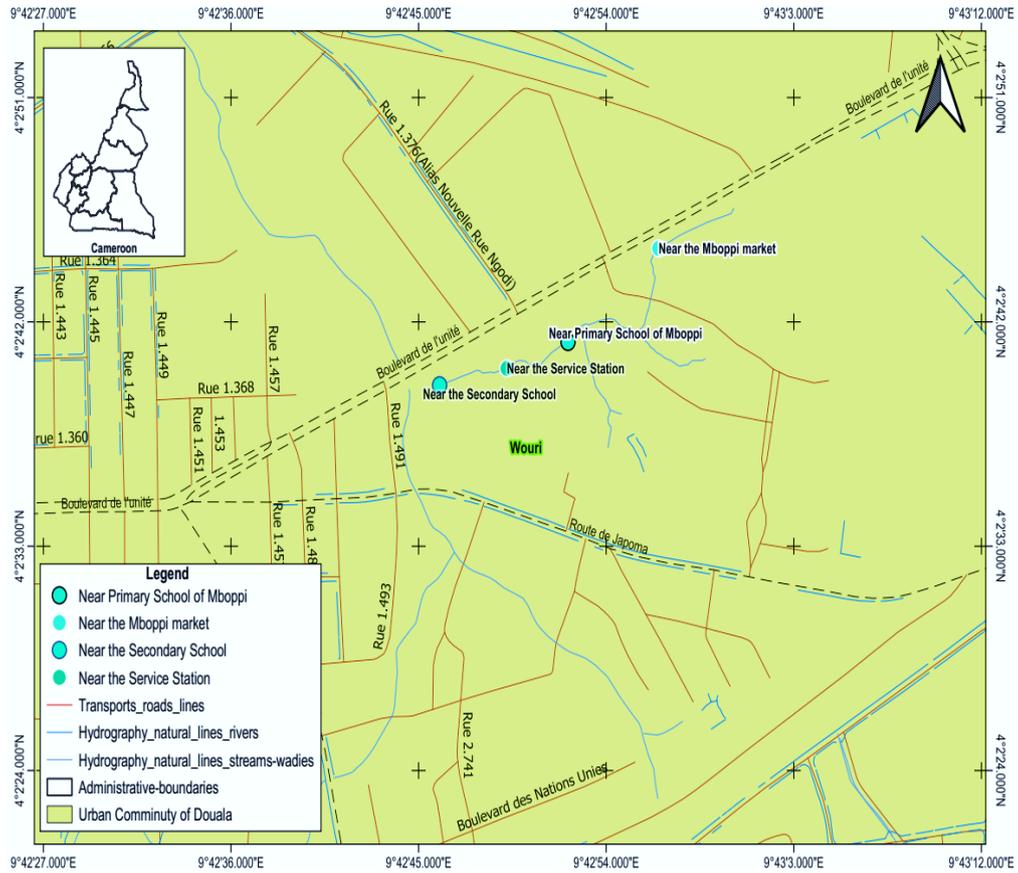
Country	Water body	PAHs concentration	Reference
China	Jiulong River	0.017 $\mu\text{g}\cdot\text{L}^{-1}$	Maskaoui et al., 2002
China	Yellow River	2.18 $\mu\text{g}\cdot\text{L}^{-1}$	Gongchen et al., 2006
China	Dialo River (watershed)	6.47 $\mu\text{g}\cdot\text{L}^{-1}$	Guo et al., 2007
China	Dialo River (Estuary)	0.48 $\mu\text{g}\cdot\text{L}^{-1}$	Bin Men et al., 2009
China	Pearl River (Estuary)	2.6 – 39.1 $\text{ng}\cdot\text{L}^{-1}$	Xiao-Jun et al., 2008
China	Pearl River (Delta)	4.3 – 68.7 $\mu\text{g}\cdot\text{L}^{-1}$	Zhao et al., 2015
China	Tianjin	45.81 – 1272 $\text{ng}\cdot\text{L}^{-1}$	Shi et al., 2005
China	Songhua River	13.9 – 161 $\text{ng}\cdot\text{L}^{-1}$	Wan-Li et al., 2013
China	Tonghui River	192.5 – 2651 $\text{ng}\cdot\text{L}^{-1}$	Zhang et al., 2004
Colombia	Cauca River	4476.5 $\text{ng}\cdot\text{L}^{-1}$	Sarria-Villa et al., 2016
France	Huveaune River	572 – 4235 $\mu\text{g}\cdot\text{kg}^{-1}$	Kanzari et al., 2014
France	Seine River	450 – 5650 $\mu\text{g}\cdot\text{kg}^{-1}$	Motelay-Massei et al., 2004
Italy	Volturno River	318.1 – 1429.1 $\text{ng}\cdot\text{L}^{-1}$	Montuori et al., 2020
Japan	Partizanskaya River	0.028 t/year	Chizhova et al., 2020
Japan	Tumen River	2.5 t/year	Chizhova et al., 2020
Nigeria	Ovia River	2.33 – 25.83 $\mu\text{g}\cdot\text{L}^{-1}$	Tongo et al., 2017
South Africa	Bufalo River	14.91 – 206 $\mu\text{g}\cdot\text{L}^{-1}$	Adeniji et al., 2019
South Africa	Diep River	72.38 $\mu\text{g}\cdot\text{L}^{-1}$	Adetunji et al., 2020
Taiwan	Gao-ping River	0.43 $\mu\text{g}\cdot\text{L}^{-1}$	Doong and Lin, 2004
Taiwan	Salt River	0.485 – 10.2 $\mu\text{g}\cdot\text{L}^{-1}$	Chen et al., 2020
USA	New York estuary	0.031 $\mu\text{g}\cdot\text{L}^{-1}$	Gigliotti et al., 2002
USA	Mississippi River	62.9 – 144.7 $\text{ng}\cdot\text{L}^{-1}$	Zhang et al., 2007

The main general objective of this study is to monitor the Mboppi River in the Cameroon town with the highest population and the most industrialized named Douala. Its actual population is reaching 3 million inhabitants and by 2030 may be up to 4 million inhabitants (Mbusnum et al., 2020). The population increase, the urbanization, industrialization and the commercial business growth exerts high pressure on the environment among which water bodies. The Mboppi River received most attention during the recent years because of the increase of commercial business affecting the quality of the water of this river. However, only little data of Polycyclic Aromatic Hydrocarbon (PAHs) in this surface water are available. The study has the aim to determine the levels of PAHs and their sources in Mboppi River during rainy and dry seasons.

## **2. Materials and Methods**

### *2.1 Materials*

The study was carried out in Douala at the area located between the Global Position System (GPS) coordinates: 4° 02' 51" N –4° 02' 33" N and 9° 42' 45" E-9° 43' 03" E (Figure 1). The Mboppi River is located in the commercial zone of Douala which is the economic town of Cameroon. Sampling of water was realized at different sites along the Mboppi River. The river also pass through a military camp in which there are primary school, health centre, and agricultural farms. Two campaigns were conducted, one during the rainy season (July and August) and one during the dry season (January and February). The total of eight (8) samples was collected. The coordinates of the sampling points which were recorded and sequentially numbered along the river, were obtained by GPS (Global Position System) are represented on Figure1. Figures 2 and 3 illustrates how the first sampling point looks like during each sampling campaign, in rainy and dry season.



**Figure 1. Study Area with the Location of the Sampling Points on the Mboppi River**



**Figure 2. Sampling site n°1 in Rainy Season**



**Figure 3. Sampling Site n°1 in Dry Season**

Prior the sampling, all glassware were carefully decontaminated by washing followed by several rinsing with ultra-pure water and baked overnight at 200°C. Samples of water were transported to the laboratory under freezing ( $T=20^{\circ}\text{C}$ ) and kept at this temperature till analysis.

In situ physic-chemical analyses were realized on water samples and the values of these characteristics are summarized in the Table 2 below. The values at the point B1 during the rainy season could not be determined due to the unavailability of the analysis equipment at that moment.

**Table 2. Physico-Chemical Characteristics of Water Samples from Mboppi River**

Physico-chemical characteristics	Seasons	Mboppi market (B1)	Primary school (B2)	Service station (B3)	Secondary school (B4)
pH	Rainy	-	$6.59 \pm 0.07$	$6.32 \pm 0.04$	$6.04 \pm 0.43$
	Dry	$6.26 \pm 0.06$	$6.20 \pm 0.08$	$6.13 \pm 0.07$	$6.53 \pm 0.09$
Conductivity (mS/Cm <sup>3</sup> )	Rainy	-	$0.73 \pm 0.00$	$0.75 \pm 0.00$	$0.82 \pm 0.00$
	Dry	$2.24 \pm 0.00$	$0.95 \pm 0.00$	$1.13 \pm 0.01$	$2.27 \pm 0.01$
Turbidity (NTU)	Rainy	-	$402.67 \pm 0.09$	$601.67 \pm 0.36$	$98.67 \pm 0.17$
	Dry	$435.34 \pm 0.04$	$4.34 \pm 0.67$	$4.67 \pm 0.09$	$22.67 \pm 1.04$
Organic matter content (mg.L <sup>-1</sup> )	Rainy	-	$29.67 \pm 1.19$	$21.67 \pm 0.09$	$26.67 \pm 0.05$
	Dry	$36.67 \pm 0.04$	$26.67 \pm 2.04$	$20.00 \pm 1.02$	$28.34 \pm 0.09$

## 2.2 Methods

### 2.2.1 PAHs Extraction and Clean-Up

The PAHs extraction from water was conducted as described by Brun et al. (2009). For each homogenized sample, an aliquot of 300 mL was mixed to 25 mL of hexane in an Erlenmeyer flasks under magnetic stirring during 40 min. The mixture was then introduced in a separatory funnel in order to separate the three phases (hexane, water, and solid particles). Extracts of the same sample were combined and concentrated in a rotary evaporator giving final volumes of 2 mL, then transferred to vials and kept in a freezer until further analysis. Samples were always extracted and analyzed in independent triplicates.

The clean-up procedure was conducted following the modified method described by Jessie et al. (2014). The hexane extract was subject to a 1:2 alumina/silica gel glass column for cleanup and fractionation. The column was eluted with 15 mL of hexane and the eluted was discarded. The *n*-alkane fraction was eluted with 10 ml of hexane, the second fraction containing the mixture of *n*-alkane and PAH was eluted with 10 ml of hexane/ethyl acetate mixture (83:17, v/v) and the third fraction containing only the PAHs was eluted with 20 mL of hexane/ethyl acetate mixture (65:35, v/v). The last fraction containing only PAHs was used for further analyses of identification and quantification.

### 2.2.2 Gas-Chromatographic Analysis

The analysis of PAHs was carried out using SHIMADZU GC-14B Gas Chromatograph (GC) equipped with a Flame Ionization Detector (FID) and a Hewlett Packard integrator. The GC-FID was using a ZB-5 capillary column of 30m length, 0.32mm film thickness, and 0.25µm of internal diameter (US PATENT, Varian, Walnut Creek, CA, USA). Nitrogen (99 %) was used as the GC carrier gas at a constant flow of 1.0 ml/min. 1-µL samples were injected into the chromatograph. The column temperature was programmed from 40°C for 5min to 320°C at a rate of 6°C/min. The 16 US-EPA priority PAHs were analyzed: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fl), phenanthrene (Phe), anthracene (An), fluoranthene (Fla), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo(k)fluoranthene (BkF), benzo(b)fluoranthene (BbF), benzo(a)pyrene (BaP), indeno (1, 2, 3-cd) pyrene (IP), dibenzo(a, h)anthracene (DahA), and benzo(g, h, i) perylene (BghiP).

### 2.2.3 Principal Component Analysis

The analytical results of samples and sites replicates were statistically studied by using Principal Component Analysis (PCA) which enables the extraction of the systematic variations in one data set (Qualls & Haines, 1992). This method can be used to depict information from a large data set as well as to help in data interpretation. In this work, PCA was employed to understand the interaction between the concentration of PAHs with the samples location and the seasonal variation.

## 3. Results and Discussion

### 3.1 Levels of PAHs

The total concentration of the sixteen US-EPA PAHs gives a very wide variation range from “Not detected” (Nd) to 1040.19 µg/L in the Mboppi River during the dry and rainy seasons. The Figure 4 presents the concentration of total PAHs ( $\Sigma 16$  PAHs), while Table 3 shows the total concentration of high molecular weight and low molecular weight of PAHs identified in the Mboppi River.

In the Mboppi River, the total concentration of PAHs ranged from Nd to 316.46 µg/L in the rainy season and from 234.29 to 1040.19 µg/L in the dry season. The PAH having the highest concentration was Anthracene (200.32 µg/L) found in sample B1 which is close to the first market of Central Africa (Mboppi market). High concentrations of total PAHs were found in the samples B2 (316.46 µg/L) in the rainy and in B1 (1040.19 µg/L) in the dry season.

**Table 3. Concentration of PAHs (µg/L) in Water from the Mboppi River**

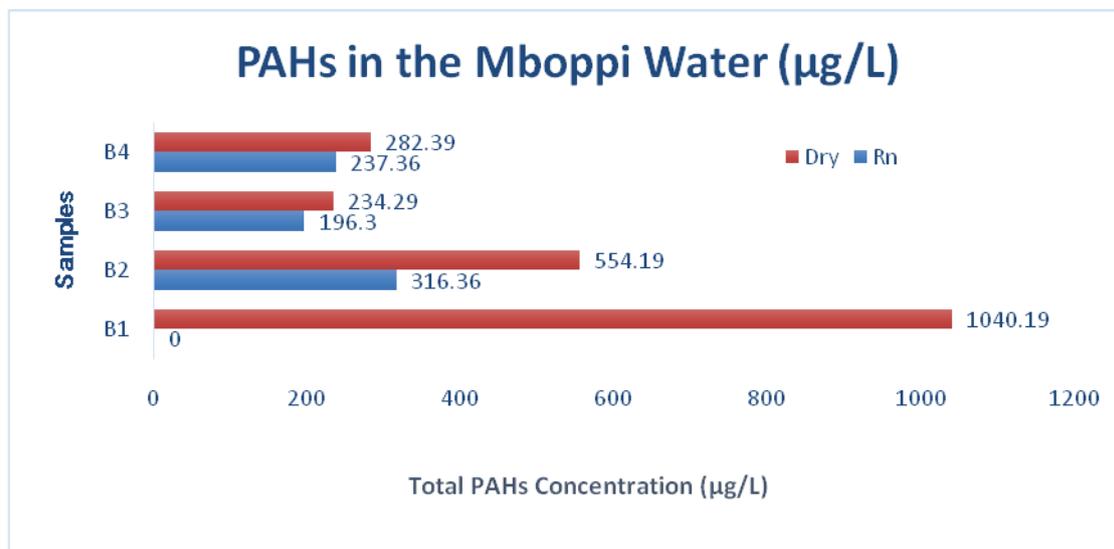
PAHs Compounds	Mboppi Water							
	B1		B2		B3		B4	
	Rn	Dry	Rn	Dry	Rn	Dry	Rn	Dry
Naphtalene	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd

Acenaphthylene	Nd	50.99	2.34	24.56	Nd	32.11	6.79	32.58
Acenaphthene	Nd	32.87	14.57	22.36	10.67	20.54	9.38	33.02
Fluorene	Nd	91.67	16.17	66.54	14.17	32.3	11.7	41.96
Phenanthrene	Nd	82.06	29.28	33.32	15.52	10.48	19.46	12.76
Anthracene	Nd	200.32	3.85	98.42	4.38	24.67	6.2	39.29
$\Sigma$ of (LMW)	<b>Nd</b>	<b>582.06</b>	<b>66.21</b>	<b>235.02</b>	<b>44.74</b>	<b>100.1</b>	<b>53.53</b>	<b>150.52</b>
Fluoranthrene	Nd	154.15	34.37	90.76	34.52	24.62	15.4	10.91
Pyrene	Nd	167.81	45.12	80.02	26.75	30.07	45.19	11.73
Benzo[a]anthracene	Nd	32.44	6.7	17.4	3.78	Nd	8.36	10.78
Chrysene	Nd	10.18	13.37	7.5	10.46	7.67	15.66	10.79
Benzo[b]fluoranthene	Nd	15.45	6.2	6.07	8.07	8.26	13.61	9.95
Benzo[k]fluoranthene	Nd	36.91	34.65	19.04	15.67	10.54	12.03	10.09
Benzo[a]pyrene	Nd	59.96	48.09	26.53	14.08	17.8	20.21	23.76
Indeno[1,2,3-cd]pyrene	Nd	41.51	31.17	17.56	17.69	10.68	18.46	24.34
Dibenzo[ah]anthracene	Nd	43.33	20.13	15.2	12.12	9.16	19.17	17.46
Benzo[ghi]perylene	Nd	50.54	10.45	39.23	8.42	15.39	15.74	12.97
$\Sigma$ of (HMW)	<b>Nd</b>	<b>458.13</b>	<b>250.25</b>	<b>319.31</b>	<b>151.56</b>	<b>134.19</b>	<b>183.83</b>	<b>142.77</b>
LMH/HMW	<b>Nd</b>	<b>1.27</b>	<b>0.26</b>	<b>0.73</b>	<b>0.29</b>	<b>0.74</b>	<b>0.29</b>	<b>1.05</b>
UCM	Nd	4674.22	2941.66	3213.44	1567.45	2034.21	2101.28	2254.76
$\Sigma$ 16 PAHs	<b>Nd</b>	<b>1040.19</b>	<b>316.46</b>	<b>554.51</b>	<b>196.3</b>	<b>234.29</b>	<b>237.36</b>	<b>282.39</b>
Fl/(Fl+Pr)	<b>0</b>	<b>0.48</b>	<b>0.43</b>	<b>0.53</b>	<b>0.56</b>	<b>0.45</b>	<b>0.25</b>	<b>0.48</b>

**Rn**: Rainy season; **Dry**: Dry season; **N**: Ngoua Water; **Nd** : no detected.

**LMW / HMW**, low molecular weight / high molecular weight; **UCM**: Unresolved Complex mixture

The Mboppi River was polluted by the two categories of PAHs known as Low Molecular Weight (LMW) and High Molecular Weight (HMW). The PAHs of LMW represent 22.55 to 55.95 % of the total PAHs concentration, while the PAHs of HMW are ranged from 44.04 to 57.27 % of the total PAHs concentration.



**Figure 4. Total Concentration of PAHs in the Water from Mboppi River**

**Rn:** rainy season; **Dry:** dry season.

Comparing the results found in the present study to what other researcher published, it can be said that the MboppiRiver (ND to 1040.1µg/L) is a highly polluted water body, but not as much as Huveaune River and Seine River in France in which total PAHs concentration reaches 4235 µg.kg<sup>-1</sup> and 5650 µg.kg<sup>-1</sup> respectively (Motelay-Massei et al., 2004; Kanzari et al., 2014). Ofori et al. (2020) made a systematic studies on PAHs level of pollution of African water bodies in which it appears that there are some of them that are greatly polluted than Mboppi River. The Table 4 below, born from Ofori et al. (2020) presents African water bodies that have higher total PAHs concentration than Mboppi River with their respective country, total PAHs concentration and initial authors.

**Table 4. Some African water Bodies with high Total PAHs Concentration (Ofori et al., 2020)**

Country	Water body	PAHs concentration	Initial author
Tunisia	Golf of Gabès	175 to 10,769 µg/kg	Zaghden et al., 2017
South Africa	Mvudi River	16,585 µg/L	Edokpayi et al., 2016
South Africa	Nzhelele River	15,134 µg/L	

All rivers presenting high concentrations of total PAHs have the similarity of going across cities with high population and/or high industrial activities along their bed.

In the Mboppi River, the absence of the biodiversity (fishes, shrimps, etc.) was observed during the sampling. In fact, the disappearance of these biodiversity is surly due to the high concentration of PAHs and other pollutants in the Mboppi River. By the other hand, a part of being a threat to the aquatic biodiversity, the Mboppi River also represents a danger for the population who is using this

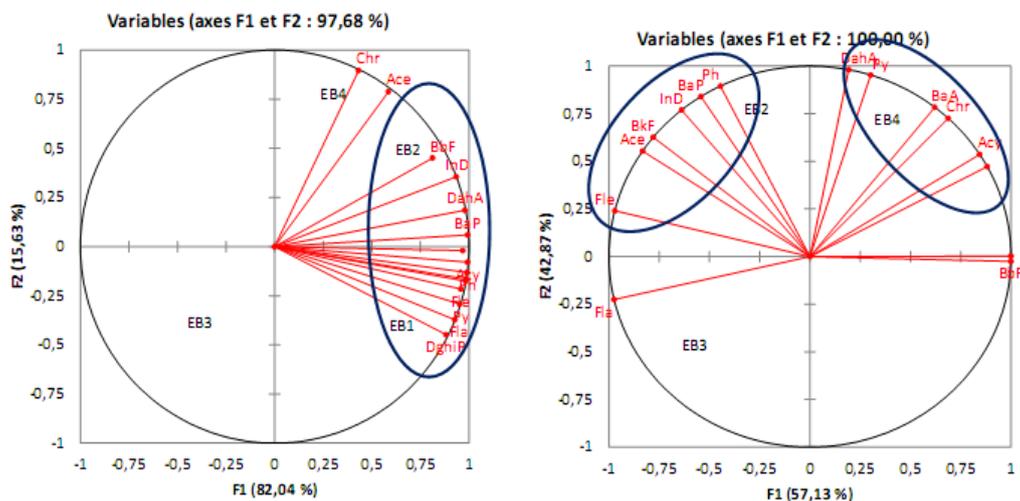
river for other activities such as and extraction since some PAHs can be transformed into other compounds called PAHs metabolites (Awanthaand Shaw, 2010) when absorbed by human.

### 3.2 PAHs Pattern and Source

The profile of PAHs of LMW and HMW of the studied area are function of the type of activities around the river. In the Mboppi River, the result shows that this area is polluted by the mixture of Low and high molecular weight (Table 1). Several molecular ratios of selected PAH compounds, such as the abundance ratio of 2–3-ring hydrocarbons to 4–6-ring hydrocarbons (LMW/HMW) including Fluoranthene/ (Fluoranthene + Pyrene) that have been developed to aid in the interpretation of PAH compositions and in their possible sources. If Fluoranthene/ (Fluoranthene + Pyrene) ratio  $<0.40$ , it indicates that the source is petroleum, if between  $0.40-0.50$ , it indicates that the source is petroleum combustion, and if  $>0.50$ , it indicates that the source is combustion of coal, grasses and wood (Doong & Lin, 2004).

In the Mboppi River, the ratio of Fluoranthene/(Fluoranthene + Pyrene) is 0 in B1, 0.43 in B2, 0.56 in B3 and 0.25 in B4 during the rainy season respectively, and 0.48 in B1, 0.53 in B2, 0.45 in B3 and 0.48 in B4 during the dry season respectively. In rainy season, an increasing value is observed from B1 to B3 followed by a decrease up to 50% of B3 in B4. This can be justified by the run-off waters from the floor of places where hydrocarbons are manipulated such as cars and bikes repairs garages along the river bed and the addition of two smaller rivers that join Mboppi River. The situation differs in dry season because during this season, the run-off is completely absent, the flow of smaller rivers has reduced and the water evaporation phenomenon is also taking place. The comparison of these values for rainy season against dry season lets us note an increase of the ratio is observed for sampling points B1, B2, and B4 while there is a slight decrease of the ratio for point B3. These results show that petroleum and petroleum combustion are the principal sources of PAHs pollution in the Mboppi River. These pollutions from petroleum and petroleum combustion are directly linked with the various activities around the river which include the petrol station, the railway station, etc.

The Principal Component Analysis (PCA) (Figure 5) shows that the distribution of PAHs in the dry and rainy seasons. The distribution of individual PAHs were concentrated into two samples independently of the seasonal variation. In the rainy and dry seasons, the most polluted sample was B2. B2 represents the sample collected near the Mboppi market and it's the most PAHs polluted with high molecular weight PAHs including Fluoranthene, Pyrene, Benzo[a]pyrene, and Benzo[ghi]perylene.



**Figure 5. Principal Component Analysis Loading Plot Water for Mboppi River**

Dry season Rainy season.

#### 4. Conclusion

The aim of this work was to identify and quantify the hydrocarbons pollution of the Mboppi River located in a commercial zone in order to point out the degree of pollution and toxicity of the river. The total concentrations of polycyclic aromatic hydrocarbons in the samples were ranged between 196.3-1040.19  $\mu\text{g/L}$  in the river. Polycyclic aromatic hydrocarbons with more than four rings showed the highest concentrations in the river independently from the seasonal variation while the polycyclic aromatic hydrocarbons with 2 or 3 rings were usually present in low concentrations or sometimes undetectable. The PCA analysis revealed that the PAHs levels in the Mboppi River were coming from petroleum and petroleum combustion. This study also revealed that the Mboppi River can be considered as among the most PAHs polluted environment in comparison with other rivers and estuaries.

#### Compliance with ethical standards

**Conflict of interest:** The authors declare that they have no conflict of interest.

**Ethical approval:** This article does not contain any studies with human participants or animals performed by any of the authors.

**Data availability statement:** Data will be made available on reasonable request.

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