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Analyses of Polycyclic Aromatic Hydrocarbons (PAHs) in the Ganga River Water in Uttar Pradesh, India

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) were analyzed in the Ganga River water samples collected from three cities. Jajmau (Kanpur), Dala Khera (Fatehpur), and Kara Kachar (Kaushambi) of Uttar Pradesh, India. At Jajmau (Kanpur), out of sixteen PAHs, eight were found in the Ganga river water in concentration (µg.L^{-1}) order: acenaphthylene (3.8356) > pyrene (0.5878) > fluorene (0.5752) > anthracene (0.2806) > benzo(b)fluoranthene (0.1960) > phenanthrene (0.0526) > benzo(a)pyrene (0.0234) > naphthalene (0.006). In contrast, in Dala Khera (Fatehpur), two PAHs: anthracene (0.2806) and fluorene (0.07894), were observed. In Kara Kachar (Kaushambi), only single phenanthrene (0.04507) was detected. It was noticed that the three-ring types of PAHs occur commonly in all three sites. It is concerning because the river water sampled had PAH concentrations, namely Acy, Flu, Phe, and Pyr, that were relatively higher than those recommended (0.05 µg.L^{-1}) by WHO (1998) in surface water. In contrast, the amounts of Nap, Ant, BbF, and Bap were recorded within the safe levels in Kanpur, while in the other two cities (Fatehpur and Kaushambi), Phe and Ant were detected lower than their permissible limit. Flu was measured as higher than its recommended value by WHO (1998). Similarly, the concentration of Acy, Ant, Pyr, BbF and BaP in river water samples at Jajmau, Kanpur were higher than their safe limits suggested by RIVM report 607711007/2012 for inland surface water while Nap, Flu, and Phe were lower than their recommended values. However, at, Dala Khera and Kara Kachar (Fatehpur and Kaushambi respectively), the concentrations of Ant, Flu and Phe were lower than their prescribed limits given by RIVM 607711007/2012.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are the priority pollutants that have imposed severe ecological and human health risks due to their non-biodegradable and lipophilic nature (Kafilzadeh et al. 2011, Wolska et al. 2012, Duttagupta et al. 2020a). Chemically, they bear two to six fused aromatic rings of carbon and hydrogen atoms. They enter into the environment via anthropogenic activities or natural sources such as industrial emissions, automobiles fuel burning, incomplete combustion of fossils fuels like petroleum and coal, domestic, agricultural, and urban waste-water discharges or volcanic eruption, forest fires, and natural oil seepage respectively (Agarwal et at. 2006, Farooq et al. 2011, Li & Ran 2012, Awe et al. 2020, Ofori et al. 2020). Though more than hundreds of PAHs have been identified in water, soil, and air, sixteen of them, namely Naphthalene (Nap), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene(Phe), Anthracene (Ant), Fluoranthene (Flu), Pyrene (Py), Benzo(a)anthracene (BaA), Chrycene (Chy), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF), Benzo(a)pyrene (BaP), Dibenzo(a.h)anthracene (DbA), Benzo(ghi)perylene (Bper), and Indeno(1,2,3-cd) pyrene (IP) were reported to exhibit carcinogenic effects (USEPA 2013). Despite being toxic, they are extensively utilized in the market to manufacture various industrial and commercial goods. They are preferred because PAHs containing raw materials are readily available, cost-effective, and have peculiar characteristics that endow waterproofing, abrasion resistance, binding ability, moisture and UV radiation protection, flexibility, elasticity, adhesiveness, tensile strength to the products that increase their shelf lives (USEPA 2013, Purr et al. 2016). Consequently, vast amounts of effluents containing PAHs from these industrial types get released into the aquatic ecosystem directly or indirectly

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(Sogbanmu et al. 2019, Duttagupta et al. 2020b). However, most of the studies were confined to the analysis of PAHs concentrations in the sediments of the aquatic ecosystem such as Hooghly river estuary (Zanardi-Lamardo et al. 2019), Cochin estuary (Ramzi et al. 2017), Adyar river, Cooum river, Ennore estuary, and Pulicat lake (Goswami et al. 2016), Yamuna river (Agarwal et al. 2006, Kumar et al. 2014) and Hooghly and Brahmaputra river (Khuman et al. 2018). Only few workers have measured it in surface freshwater resources (Kafilzadeh et al. 2011, Hussain et al. 2014, Singare 2016, Srivastava et al. 2017). Some researchers also demonstrated PAHs' detrimental effects on aquatic fauna. For instance, physiological malfunctions such as muscles atrophy in fish (Olayinka et al. 2019), CAT and LPO in *Mya arenaria* (Frouin et al. 2007), GST and CAT *Mytilus trossulus* and *Neries* (Pempkowiak et al. 2006). Since humans directly consume fish as one of the primary proteinaceous sources, it could be dangerous to their lives. Hence, PAH analysis in the aquatic ecosystem should be monitored regularly (Incardona et al. 2015, Wu et al. 2011, Patel et al. 2020). For the current purpose, the concentration of sixteen toxic PAHs was analyzed in the Ganga river water samples collected from three major cities of Uttar Pradesh, India: Kanpur, Fatehpur, and Kaushambi. River water sampling was conducted from the Ganga in Uttar Pradesh (UP), India, because the Ganga traverses a maximum (of 1000 km) in UP, and several industrial estates drain their waste directly into the river leading to its contamination. Since Ganga is globally worshipped as "Ganga Ma" and is extensively used by millions of people for ritual activities, drinking, bathing, and domestic purposes. Therefore, it becomes imperative to analyze PAHs in the Ganga river water (Haritash et al. 2016, Nagpure et al. 2017).

MATERIALS AND METHODS

Chemicals

Sixteen mix PAHs external standard (2000 $\mu g.mL^{-1}$) was purchased from Chem Service Inc. Germany. Dichloromethane (CAS No. 75-09-2), Acetonitrile (CAS No. 75-05-8), and n-hexane (CAS No. 110-54-3) were procured from Merk Life Sciences. Silica gel (CAS No. 112926-00-8) was bought from Sigma-Aldrich, high purity grade (7734), pore size 60Å, 70-230 mesh.

Instrumentation

An Agilent 1220 Infinity HPLC was equipped with a UV detector (254nm). Operating conditions were: flow rate- 0.9 $mL.min^{-1}$, sample volume - 20 µL, run time- 45 min, column: ZORBAX Eclipse C18 (250mm long × 5µm ID), mobile phase was water and acetonitrile as ratio of 60:40, programme was set same as APHA AWWA (2008).

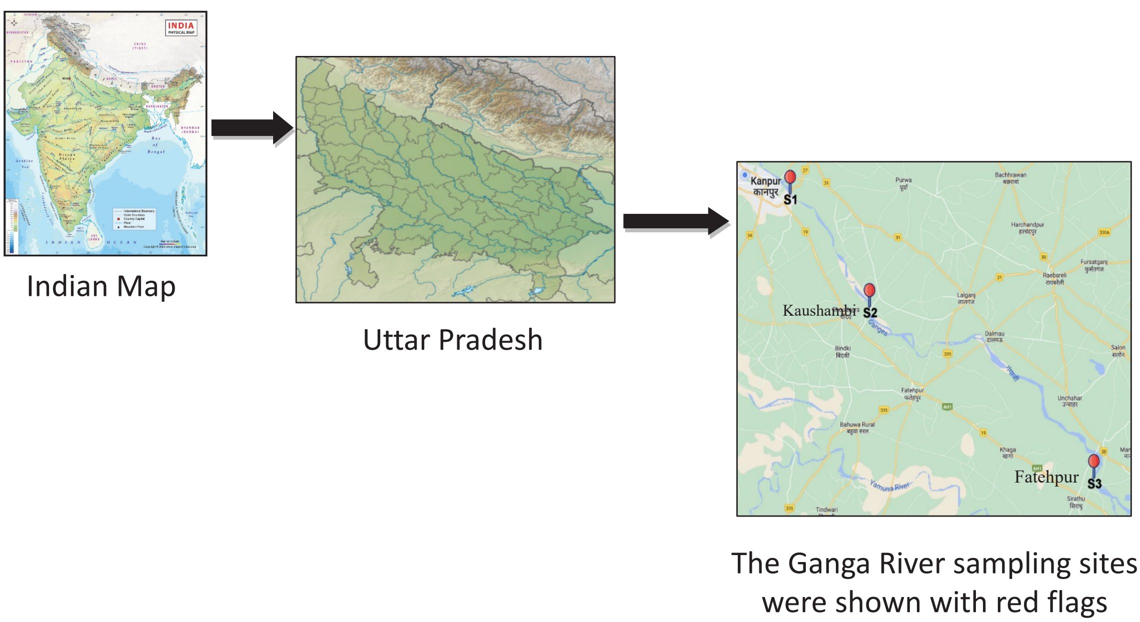

Fig. 1: Map of the Ganga River showing the sampling sites.

Study Area

The present study was carried out about the stretch of 183 km from Kanpur to Kaushambhi, lying in the basin of Ganga (Fig. 1). The sampling sites were Jajmau (Kanpur, Latitude 26.435391° and Longitude 80.408684°), Dala Khera (Fatehpur, Latitude (26.103616° and Longitude 80.66689°), and Kara Kachar (Kaushambi, Latitude 25.706076° and Longitude 81.366626°). Approximately 200 large and small industries are located in Kanpur along the Ganga river bank, having significant sludge sources generated by industries. Kanpur poured 18 drains (domestic and industrial waste) directly into River Ganga. A common effluent treatment plant (CETP) is located at Jajmau, but it is insufficient for treating industrial, tannery, and domestic effluent. Fatehpur, which is the meeting point of the Pandu River to the Ganga, drifts most of the domestic effluents of rural and urban areas. River Pandu joins seven drains named Panki TTP drain, ICI drain, Ganda Nalla, COD Nalla, Halwakhanda Nalla, Ratanpur Nalla, and Panki thermal power by passing through Kanpur. Almost 210,000 tons of fly ash coal-based power plant was discarded in Pandu River in Kanpur. Pandu receives domestic wastes from villages/towns (Sultanpur, Kaindepur, Matinpur, Pure Dayal, Saurajpur, Kotla, Khalispur, Baghauli, Lahangi Aht) located at Fatehpur (UPPCB 2015), and these effluents ultimately dump into river Ganga. District Kaushambi is rich in many historical places in which most of the area is covered by agricultural land. A few famous temples, such as Karadham, Sheetla Maa, and the Jain, are the primary source of tourist fascination. Kara Kachar is the spot of the present study, obtaining agricultural and ritual waste.

Water Sampling

The present study is conducted in the rainy season from 25-08-2021 to 30-10-2021. We collected river samples to the midstream of the Ganga River below 30 cm of the surface in 1-liter amber glass bottles for PAHs analysis by following APHA-AWWA (2008) and immediately transferred to the ice box and brought them to the laboratory for further extraction and cleanup.

Extraction and Cleanup of the Sample

First, by adding Acetonitrile to the stock solution, we prepared calibration standards solution at four concentration levels, 1%, 3%, 5%, and 7%. We extracted the sample within seven days of collection and thoroughly analyzed it within forty days of extraction according to EPA method 610. The samples were extracted with the liquid-phase extraction method of APHA AWWA (2008) The extraction procedure involved a 1 L water sample in a 2 L beaker added 80ml of dichloromethane. After adding DCM, shake the sample for 15 min.

Furthermore, pour it into 2 L of the separatory funnel and hold it for 5 min to separate aqueous and non- aqueous phase. After the separation, the nonaqueous extract was collected in another conical flask and repeated this process thrice. The pooled extract was filtered with Whatman 42 filter paper. After extraction, a sample cleanup was performed by making the slurry of 10 g activated silica gel in n-hexane and placed in a 50 mL and 10 mm internal diameter (ID) chromatographic glass column and placing 1-2 cm anhydrous sodium sulfate to the top of the column for demoisturized the sample. Before loading the sample, wash the column by DCM three to four times. PAHs were eluted by applying the mixture (50 mL) of n-hexane and dichloromethane in the ratio (30:20, v/v). The extracted sample was concentrated in a rotatory evaporator till the sample remained at 0.5 µL and made up the sample 1 mL by adding 0.5 µL Acetonitrile.

RESULTS

Table 1 represents the concentration of various PAHs found in the Ganga river water samples at all three different sampling cities, namely Jajmau (Kanpur), Dalakhera (Fatehpur), and Kara Kachar (Kaushambi) of Uttar Pradesh, India. Results represent the triplicate analysis of the samples as the mean value. It was noted that Ace, Fln, BaA, Chy, Bkf, DbA, BPer, and IP PAHs were not present in the Ganga river water samples at any of the three cities. However, the rest eight PAHs, namely Nap (two-rings), Acy, Flu, Phe, Ant (three-rings), Pyr (four-rings), and BbF, BaP (five-rings), were found in Kanpur. Flu and Ant were present in the Ganga river water samples at Fatehpur, and only Phe was detected in Kaushambi (Table 1). The concentrations ($\mu g.L^{-1}$) of eight PAHs in Kanpur followed the order: Nap (0.006) < BaP (0.0234) < Phe (0.0526) < BbF (0.1960) < Ant (0.2806) < Flu (0.5752) < Pyr (0.5878) < Acy (3.8356) (Table 1). The results conclude that PAH concentration exceeds at the urban site than in semi-urban and rural areas. Kanpur is known for anthropogenic pollution due to local vehicle combustion, industrial coal combustion, and biomass burning. Therefore, surface run-off, vehicle emissions, or industries may contaminate rivers primary with PAHs. The amounts ($\mu g.L^{-1}$) of Flu and Ant were 0.07894 and 0.0017, respectively, at Fatehpur, while the quantity of Phe was 0.04507 $\mu g.L^{-1}$ in Kaushambi (Table 1). The finding of high variation in PAH distribution at all three cities can be due to the resuspension of road dust by wind and vehicles. Further, Ganga river water samples at Kanpur were noticed to be contaminated with both low molecular weight (LMW) and high molecular weight (HMW) PAHs having rings ranging from one to five. It was observed that 3-ringed PAHs were commonly present at all three sites (Table 1). According to Ravindra et al. (2008), three PAH rings correspond to

Table 1: Concentration ($\mu g.L^{-1}$) of PAHs in the Ganga River water samples in the three different studied sites.

S. No.	Polycyclic aromatic hydrocarbons	Jajmau (Kanpur)	Dala Khera (Fatehpur)	Kara Kachar (Kaushambi)
1.	Naphthalene	0.006±0.00072	ND	ND
2.	Acenaphthylene	3.8356±0.3009	ND	ND
3.	Acenaphthene	ND	ND	ND
4.	Fluorene	0.5752±0.0462	0.07894±0.009	ND
5.	Phenenthrene	0.0526±0.01163	ND	0.04507±0.015
6.	Anthracene	0.2806±0.0288	0.0017±0.003	ND
7.	Fluoranthene	ND	ND	ND
8.	Pyrene	0.5878±0.097	ND	ND
9.	Benzo(a)anthracene	ND	ND	ND
10.	Chrysene	ND	ND	ND
11.	Benzo(b)fluoranthane	0.1960±0.0237	ND	ND
12.	Benzo(k)fluoranthane	ND	ND	ND
13.	Benzo(a)pyrene	0.0234±0.0017	ND	ND
14.	Dibenzo(a,h)anthracene	ND	ND	ND
15.	Benzo(ghi)perylene	ND	ND	ND
16.	Indeno(1,2,3-cd)pyrene	ND	ND	ND

ND = Not detected

pyrogenic sources such as diesel, creosote, and coal tar pitch used for transportation. All three studied cities are suggested to contribute to PAH contamination by pyrogenic sources. The concentration of Acy, Flu, Phe, and Pyr in river water samples was comparatively higher than their levels recommended (0.05 $\mu g.L^{-1}$) by WHO (1998) in surface water. Although on the contrary, the amounts of Nap, Ant, BbF, and Bap were recorded within the safe levels of WHO (1998). In the other two cities (Fatehpur and Kaushambi), Phe and Ant were detected lower than their permissible limit, while Flu was measured at higher levels compared to their recommended values at Dala Khera (Fatehpur) than WHO (1998) limits. Similarly, the concentration of Acy, Ant, Pyr, BbF, and BaP in a river water sample at Kanpur (Fig. 2) was higher than their safe limit suggested by the RIVM report (607711007/2012) in inland surface water while Nap,

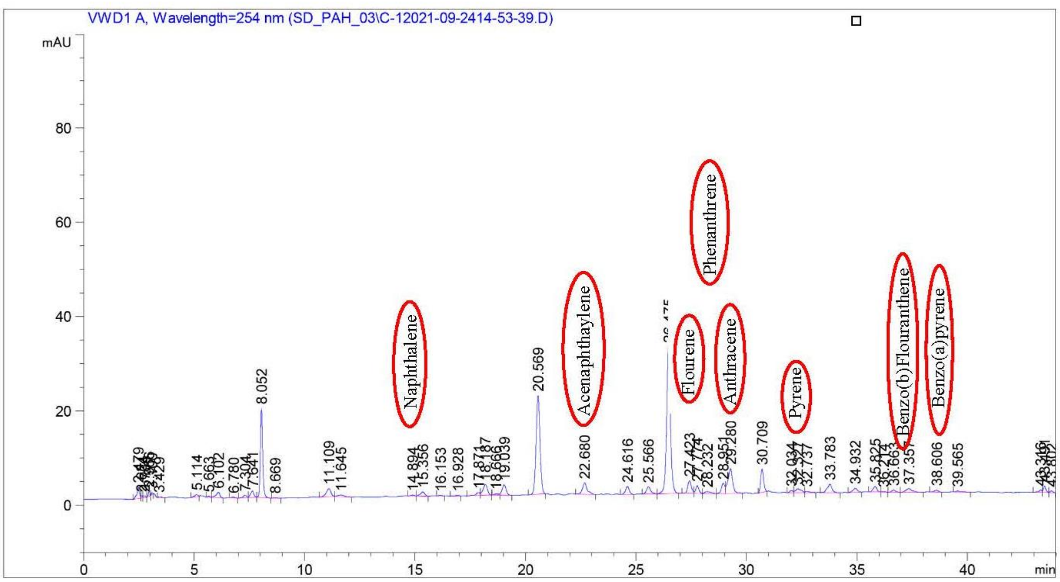

Fig. 2: Graph showing identified PAHs at Jajmau (Kanpur), UP, India.

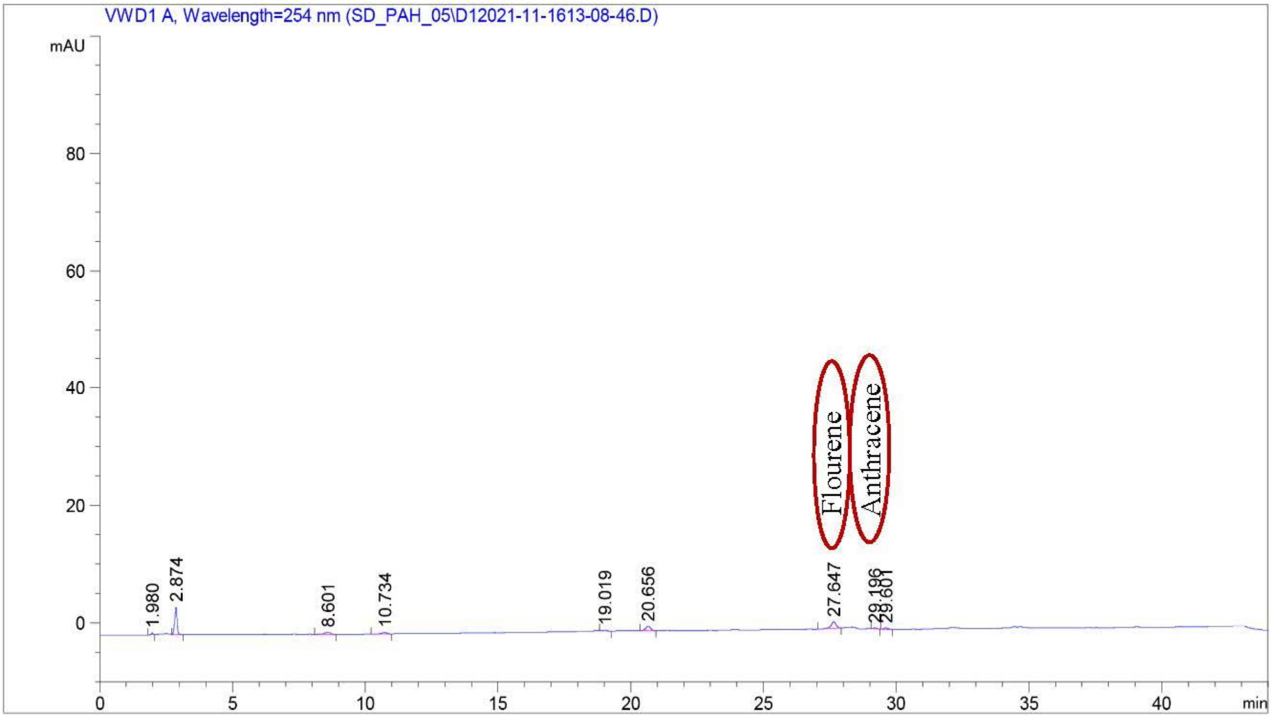

Fig. 3: Graph showing identified PAHs at Dala Khera (Fatehpur), UP, India.

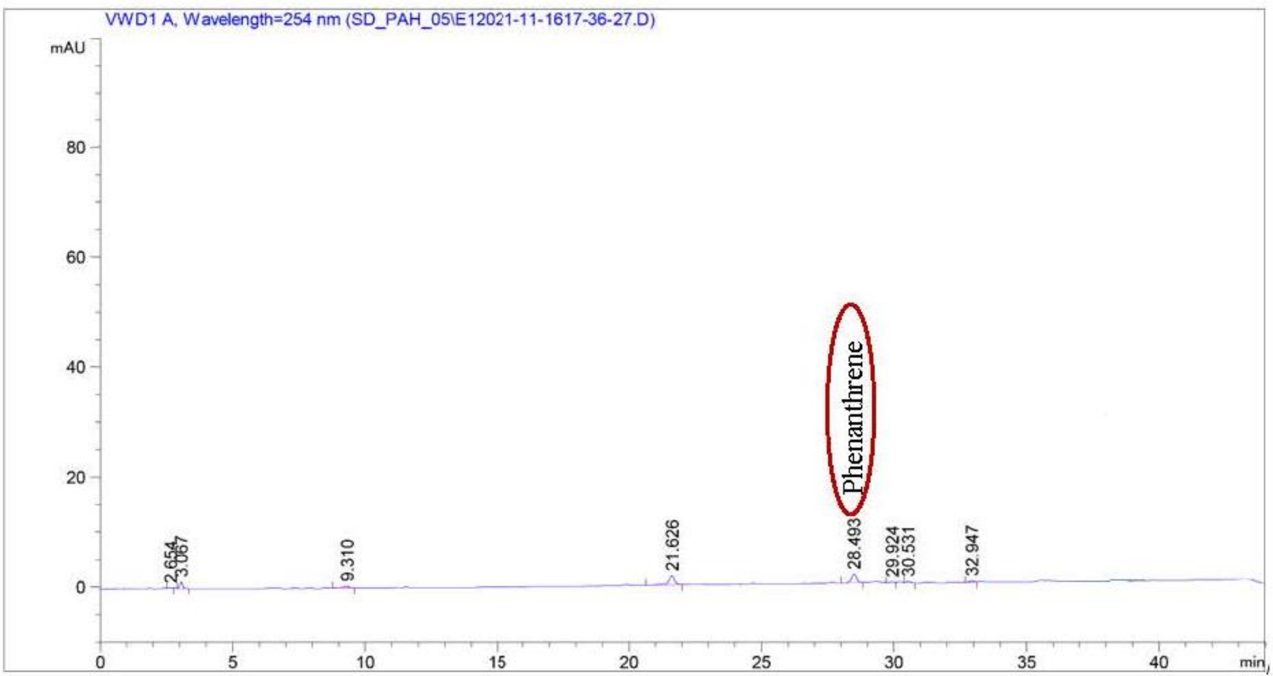

Fig. 4: Graph showing identified PAHs at Kara dham (Kaushambi), UP, India.

Table 2: Sixteen studied PAHs and their retention time.

S. No.	Polycyclic aromatic hydrocarbons (PAHs)	Retention time (min)
1.	Nap	14.698
2.	Ace	22.13
3.	Acy	24.020
4.	Flu	27.156
5.	Phe	28.385
6.	Ant	29.292
7.	Fln	31.182
8.	Pyr	32.240
9.	BaA	34.552
10.	Chy	34.744
11.	BbF	37.428
12.	BkF	37.777
13.	BaP	38.586
14.	DbA	39.601
15.	Bper	41.078
16.	IP	41.430

PAHs, while individual PAHs are less studied (Table 3). The observed PAH concentration in the present study was compared with the other river of the world, including India. The PAH concentration was comparable with the Vall River, Klip River, Chenab River, and Nile River. In India, some authors have previously revealed different types of PAHs in the Ganges River water ranging from 0.001 to 10.6 $\mu g.L^{-1}$ (Srivastava et al. 2017, Sharma & Singh 2018; Duttagupta 2020a). The Nap and Phe concentrations in Ganga River at Jajmau (Kanpur) and Kara Kachar (Kaushambi) were lower than the concentration 4.9 – 10.6 $\mu g.L^{-1}$ and 3.32 – 6.61 $\mu g.L^{-1}$ noticed by Duttagupta et al. (2020b) in the Ganga River, Western Bengal Basin, India.

Further, the concentration of BaP (0.0234 $\mu g.L^{-1}$) was comparatively higher than BaP in the Nile River of Egypt (Refai et al. 2022). BaP was investigated in Jajmau, Kanpur, which has been described as the most potent carcinogenic and used as a standard indicator for all PAHs (Masih et al. 2019). BaP measures the potential toxicity of the other priority PAH congeners by multiplying the PAH concentration by its corresponding toxic equivalent factor

Table 3: Occurrence of PAHs in rivers globally.

S.No.	PAHs	River	Concentration	Reference
	PAHs concentration in the Indian River			
1.	∑PAHs	Mithi river (Mumbai, India)	157.96±18.99 $\mu g.L^{-1}$	Singare (2016)
2.	∑PAHs	Ganga river (India)	0.05-65.9 $ng.L^{-1}$	Sharma & Singh (2018)
3.	∑PAHs	Sea water, Mumbai Harbour (India)	83.3-377.5 $ng.L^{-1}$	Pandit et al. (2006)
5.	Naphthalene Phenanthrene	Ganga river basin (India)	4.9-10.6 $\mu g.L^{-1}$ 3.32-6.61 $\mu g.L^{-1}$	Duttagupta et al. (2020a, 2020b)
6.	Acenaphthylene, Fluorene, Phenanthrene, Anthracene, Pyrene, Benzo(a)anthracene, Chrycene, Benzo(b)fluoranthene, Benzo(a)pyrene, Dibenzo(a,h)anthracene, Indeno(1,2,3-ed)pyrene, Benzo (g,h,I,) perylene	Ganga river (India)	0.001-1.78 $\mu g.L^{-1}$	Srivastava et al. (2017)
7.	∑PAHs	Ganga river (India)	0.06-84.21$\mu g.L^{-1}$	Malik et al. (2011)
8.	∑PAHs	Municipal drain water Delhi, (India)	2.5-300 ppb	Kumar et al. (2020)
	Globally, PAHs concentration in river water			
9.	∑PAHs	Densu river basin (Ghana)	37.1 $\mu g.mL^{-1}$	Amoako et al. (2011)
	∑PAHs	Luan river (China)	99.4 $ng.L^{-1}$	Cao et al. (2018)

Flu, and Phe were measured lower than its recommended value. However, the concentration of Ant, Flu, and Phe was lower than their recommended safe limit by RIVM (607711007/2012). Table 2 shows the retention time of the 16 PAH studied.

DISCUSSION

Literature studies revealed that generally, PAH levels contamination occurred in rivers of the world (Table 3). Most of the research has been done as the sum of the sixteen

S.No.	PAHs	River	Concentration	Reference
10.	Acenaphthalene	Vall river (South Africa)	0.00815-0.0828 mg.L^{-1}	Moja et al. (2013)
	Phenanthrene		0.0214-0.0263 mg.L^{-1}	
	Anthracene		0.0073-0.0092 mg.L^{-1}	
	Fluoranthene	Klip river (South Africa)	0.098-0.1205 mg.L^{-1}	
	Naphthalene		0.0339-0.0382 mg.L^{-1}	
	Acenaphthalene		0.001-0.0073 mg.L^{-1}	
	Phenanthrene		0.0487-0.0521 mg.L^{-1}	
	Anthracene		0.03582-0.4072 mg.L^{-1}	
	Fluoranthene		0.0552-0.0593 mg.L^{-1}	
11.	Nap	Chinab river (Pakistan)	192.6 ng.L^{-1}	Farooq et al. (2011)
	Acy		53.78 ng.L^{-1}	
	Ace		47.23 ng.L^{-1}	
	Flu		92.22 ng.L^{-1}	
	Phe		158.3 ng.L^{-1}	
	Anth		32.23 ng.L^{-1}	
	Fluo		35.65 ng.L^{-1}	
	Pyr		63.45 ng.L^{-1}	
	BaA		1.21 ng.L^{-1}	
	Chr		3.08 ng.L^{-1}	
	BbF		1.75 ng.L^{-1}	
	BkF		2.34 ng.L^{-1}	
	BaF		1.32 ng.L^{-1}	
	Ind		0.25 ng.L^{-1}	
	dBahAn		0.02 ng.L^{-1}	
	BghiP		1.38 ng.L^{-1}	
	∑PAHs		686.92 ng.L^{-1}	
12.	∑PAHs	Raba river (Hungary)	41-437 ng.L^{-1}	Nagy et al. (2013)
13.	Banzo(a)pyrene	Nile river (Egypt)	0.07 µg.L^{-1}	Refai et al. (2022)
	∑PAHs		0.87 µg.L^{-1}	
	Fluoranthene		0.14±0.14 µg.L^{-1}	
14.	∑PAHs	Kor river (Iran)	51.42-291.4 ng.L^{-1}	Kafilzadeh et. al (2011)
15.	∑PAHs	Danube river (Europe)	25-357 ng.L^{-1}	Nagy et al. (2012)
16.	∑PAHs	Warri river (Nigeria)	34 ng.L^{-1}	Asagbra et al. (2015)
17.	∑PAHs	Luanhe river (China)	309.75 ng.L^{-1}	Li et al. (2010)

(TEF). The concentration (µg.L^{-1}) of Nap (0.006), Acy (3.8356), Phe (0.0526), and Ant (0.2806) in our reports were lower than Vall river (South Africa) (Table 3). The concentration (µg.L^{-1}) of Acy, Flu, Ant, Pyr, and BbF in our result was higher than in the Chenab river noted by Farooq et al. 2011 (Table 3). However, the level of PAHs studied in the Ganga River is considerably lower than in other rivers investigated globally (Table 3). The lowest amount of PAHs was detected at Kaushambi, located in wide-open areas with low levels of human impact. There is almost no traffic, combustion activities, or direct discharges of industrial effluents. Globally, LMW PAHs are most studies usually formed from low-temperature processes. HMW PAHs are nonvolatile and non-biodegradable, originating from high-temperature processes. The overall assessment showed that PAHs in the Ganga River water samples in all three studied sites at different cities might be due to their emission from diverse sources but more predominantly have pyrogenic origin than petrogenic.

CONCLUSION

The present study revealed that the Ganga River was contaminated with several kinds of PAHs at Jajmau, Dala Khera and Kara Kachar studied sites at Kanpur, Fatehpur, and Kaushambi. Further, the three-ringed PAHs were comparatively more than five and six-ringed PAHs. According to Jinadasa et al. (2020), three-ringed PAHs are highly damaging for fish in the early life stage; therefore, it may lead to an imbalance in the ecosystem. Hence, it is imperative to investigate PAHs distribution and sources in the Ganga River water from urban or rural areas. The findings of this study provide a new dimension of PAHs distribution in the river Ganga.

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