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DETERMINATION OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHS) IN GOMTI RIVER, LUCKNOW, INDIA

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ABSTRACT

The Gomti river, one of the major tributaries of the river Ganga serves as one of the major source of drinking water for the Lucknow City, the State capital of Uttar Pradesh, India. The river receives the untreated wastewater and effluents from Lucknow city directly in its course through more than 25 drains. Polycyclic aromatic hydrocarbons have been identified as exhibiting toxic/hazardous properties.Our study shows that Gomti river is highly contaminated with PAHs and poses high risk to the aquatic life. The study area covers seven different locations. Grab samples of water and bed sediments were collected from each of the locations in the month of November and December. Total PAHs in water at all the seven locations studied ranged between0.467-5.342 µg/l. In the river water the most abundant hydrocarbon was acenaphthene followed by acenaphthene. The most abundant hydrocarbon among all the analysed hydrocarbon in the river sediments was acenaphthene.

Keywords: The River Gomti, Lucknow City, PAHs

INTRODUCTION

Polycyclic aromatic hydrocarbons are neutral, non-polar organic molecules that comprise two or more benzene rings arranged in various configurations. Members of this class compounds have been identified as exhibiting toxic/hazardous properties (Masih *et al*, 2010) moreover they are the major culprits in urban areas causing human lung cancer (Lighty *et al.*, 2000). On the basis of their molecular masses and properties, three classes of PAHs can be distinguished. 2-3 ring PAHs are defined as low molecular weight PAHs (LMW-PAHs), 4 ring PAHs are known as middle molecular weight PAHs (MMW-PAHs), whereas 5-6 ring PAHs are defined as high molecular weight PAHs (HMW-PAHs). The LMW-PAHs and MMW-PAHs have a significant acute toxicity, while some of HMW-PAHs show high carcinogenic and mutagenic potentials. Most of these are formed during incomplete combustion of organic materials such as wood and fossil fuels, petroleum products, coal and the composition of PAHs mixture varies with the source(s) and also due to selective weathering effects in the environments (Neff 1979). Forest fires, which may or may not be the consequence of human activity, are a significant and usually unpredictable source of PAHs. In general, environmental contamination is by complex mixtures of PAHs, not by single compounds. The Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous pollutants frequently found in different environment such as freshwater and marine sediment (Fernandez *et al*, 2000).

PAHs have been detected in the atmosphere, water, soil, sediments and food (Masih & Taneja, 2006). During their atmospheric residence, PAHs are redistributed between the gas and particulate phase (Bourotte *et al*, 2005) and therefore be transported through atmosphere over long distances and enter water bodies by wet and dry deposition and/or gas-water interchange (Fang *et al.*, 2004). On entering the aquatic systems, most of the PAHs are adhered to the suspended particles due to their hydrophobic properties, and therefore sink to the bottom and accumulate in the sediments (Masih, 2011) and from a long-term source of potential pollution. Atmospheric deposition has been regarded as a main pathway for the loading of PAHs to many water bodies (Masih *et al*, 2008).

Due to their wide distribution, the environmental pollution by PAHs has aroused global concern. Combination of their physico-chemical properties such as low aqueous solubility, moderate vapour pressure, high octanol-water partition coefficient (Kow) and persistence in environment make them capable of long range transport. Once in the atmosphere, they can travel long distances and deposit in remote areas such as mountains, lakes and even the Arctic and Antarctic (Fernandez *et al*, 1999). Atmospheric transport is a major pathway for the loading of these contaminants to the systems in remote regions with no history of their use. Wet and dry depositions are the common removal modes for PAHs. The critical effect of many PAHs in mammals is their carcinogenic potential. Benzo(a)pyrene, benzo(a)anthracene, dibenzo (a,h)anthracene, benzo(b)fluoranthene and indeno(1,2,3-cd) pyrene have been classified as probable possible carcinogens to humans (IARC, 1987). Due to tendency of PAHs to accumulate in the food chain, their release during dredging operations, episodes of high scouring, or leaching from confined disposal facilities poses a threat to aquatic ecosystem and consequently a potential threat to human health (Tabak *et al*, 2003).Although several studies on behaviour of PAHs in various aquatic systems (Fernandez *et al*, 1999; Watanabe *et al*, 2003; Dodder *et al*, 2003) have been conducted but data on distribution and level of PAHs in the Indian water resources are rare.

This study was undertaken with a view to generate data on distribution of PAHs in river system in India and presents information on the concentration and distribution of PAHs in the water and bed sediments of the river Gomti, Uttar Pradesh.

MATERIALS AND METHOD

The Gomti river, one of the major tributaries of the river Ganga originates from a natural reservoir in the swampy and densely forested area (Miankot, elevation of about 200m; North latitude $28^0$34' and east longitude $80^0$07') in Pilibhit district of Uttar Pradesh, India. The soil sediments here are silty sands. The river flowing through the districts of Pilibhit, Shajahnpur, Sitapur, Lucknow , Barabanki , Sultanpur , Janpur and Ghazipur in Uttar Pradesh traverses a total distance of about 730 km before finally merging with the Ganga river in Ghazipur district about 30 km north of Varansi**.** Throughout its stretch , there are a few small tributaries (Kathna, Sarayan,Reth, Kalyani and Sai) originating within short distances and carrying the wastewater and industrial effluents from different towns and industrial units in the basin. Lucknow, Sultanpur and Janpur are the three major urban settlements on the banks of the river and there are several industrial units in the catchments of the river in this region. Further, the river serves as one of the major source of drinking water for the Lucknow City, the State capital of Uttar Pradesh with a population of about 3.5 million. The river receives the untreated wastewater and effluents from Lucknow city directly in its course through more than 25 drains. The study area covers seven different locations namely Gaughat (I), Mohan Meaking (II), Martyr's Memorial (III), Hanuman Setu (IV), Nisatganj Bridge (V), Pipraghat (VI) and Malhaur (VII) **(fig. 1)** on the river spread over about 30 km stretch of the river in Lucknow city (State capital).

Grab samples of water and bed sediments were collected from each of the locations in the month of November and December. The sediments samples were collected from three points (1/4,1/2 and 3/4) across the river width at each of the seven locations using Ekman sediments sampler. The samples collected in the polyethylene bags were transported to the laboratory, pebbles, shells and vegetable matter were removed and the samples were air-dried. The air-dried samples then ground with pestle-mortar and sieved to 200 BSS mesh size. The organic carbon content of the sediment was determined using back titration method (Walkely and Black 1934, Pandey *et al.*2013a). The river water samples collected from midstream of the river (30 cm below the surface) in brown glass bottles were transported to the laboratory under ice conditions. The Total organic Carbon of the water was determined using TOC Analyzer (Schimadzu, Model No. -5000A).

Extraction

For PAHs analysis 10.0 ± 0.05 g sediment sample from each location were soxhlet extracted with 100 ml of dichloromethane for 16 hr. the extracts were demoisturised using activated anhydrous granular sodium sulphate and concentrated in a rotary evaporator and go for Silica-gel column cleanup**.**

For PAHs analysis, 1 Lit. of the water sample from each location was triply extracted by liquid- liquid extraction in a 2 Lit. Separatory funnel using 50, 30 and 30 ml of dichloromethane solvent. The combined solvent extracts were demoisturised using anhydrous granular sodium sulphate and concentrated in a rotary evaporator and go for Silica gel column cleanup.

Cleanup

Exchanged the Dichloromethane extract with Cyclohexane. After setting the column Eluted column by 15 ml. n-Pentane. Now, passed the sample through the column. Transfered by rinsing flask three times by 2 ml (Cyclohexane) then eluted the column by 25 ml n-Pentane. Threw away the above two elutes. Now, eluted the column by 4:6 mixture of Methylene chloride plus n-Pentane (25ml.). Collected the elute and reduced upto 2 ml. The working solvent was exchanged to acetonitrile making up final volume to 2 ml.

The samples were stored in dark at 4^0C till the analysis was performed. All the sediments and water samples were analysed for 9 PAHs viz. acenaphthylene , acenaphthene , phenanthrene, anthracene, fluoranthene ,pyrene, chrysene , benzo(a)pyrene and benzo(g,h,i) perylene using the high purity grade water-acetonitrile solvent system on HPLC (Waters. USA) equipped with UV-VIS detector (Model 2486). The PAHs standards (99.9 % purity) were supplied by Sigma-Aldrich,USA. All the analysis were carried out in duplicate and the recoveries of individual PAHs were determined through spiked sample method which were found between 90-93 percent. Recovery correction factors were applied to the final results. The results given in Table 1 and 2 is the mean value of three points (1/4, ½ and ¾) at each sampling sites.

RESULTS AND DISCUSSION

The level of different individual and total PAHs contents in water and sediments of Gomti river at all the selected locations in winter season are presented in table 1 and 2 respectively. Total PAHs in water at all the seven locations studied ranged between 0.467-5.342 µg/l. In the river water the most abundant hydrocarbon was acenaphthylene followed by acenaphthene. Highest concentrations of ΣPAHs in the river water at all the locations were found higher than the Bureau of Indian Standards (BIS) guideline value of 0.2 µg/l (BIS 1982). The

ΣPAHs contents in the river sediments at different locations ranged between 290.3-25457 ng/g. The most abundant hydrocarbon among the entire analysed hydrocarbon in the river sediments was acenaphthene in winter. The highest concentration of ΣPAHs in the river sediment was found at Site- V (25457 ng/g, in winter). Between Gaughat and Malhaur location, there are some 25 drains carrying about 400 million litres per day (mld) of untreated sewage and industrial wastewater from different parts of the city, discharging directly into the river. Moreover, a little upstream of the Pipraghat, there is barrage to restrict the river flow to maintain water level in the river for abstraction at Gaughat for urban water supply. The ΣPAHs content in the sediment at Pipraghat was found to be 12390 ng/g in winter at upstream of the Pipraghat (downstream of Lucknow). There is a crematoria on the river bank, where funeral activities are carried out throughout the year and may be a major source of the hydrocarbon to the river through surface run-off and atmospheric transport. A comparisons of the ΣPAHs burden of Gomti river with other aquatic resources (Table-3) suggests that Gomti river is heavily polluted.

There was found a correlation in winter (R2=0.2343) between the concentration of ΣPAHs in the water and sediment of the river Gomti (Fig. 2 –a). Further, at each of the locations, ΣPAHs contents of the sediments were found several times higher as compared to the ΣPAHs as contents in the water of the Gomti river. The reason for the lower concentrations of PAHs in river water as compared to the sediment may be due to the low aqueous solubility, volatilization, and affinity of these compounds to the organic matter. The spatial and temporal distribution of ΣPAHs generally followed the distribution pattern of organic carbon (%) content as reflected by observed correlation in winter (R2=0.1991) among the ΣPAHs and the organic carbon (%) contents of the sediments of the Gomti river (Fig. 2-b). It was observed that these possible carcinogenic hydrocarbons contributed about 8.67 % and 7.33 % of the total PAHs contents of the river water and sediments, respectively. It was observed that the 3-ringed hydrocarbons were the most abundant ones followed by the 4- and 5-ringed hydrocarbons, both in the sediments and in the water of the Gomti River. But in winter (water) 4-ringed hydrocarbons were the most abundant ones followed by the 3- and 5-ringed hydrocarbon.

In the Todos Santos Bay, more than 75 % of the total PAHs concentrations were represented by three- and four- ringed hydrocarbons (Zamora *et al*. 2002). The abundance of three- and four- ringed hydrocarbons may be attributed to the partitioning of these hydrocarbons from the dissolved to organic carbon-rich settling particles, whereas higher molecular weight PAHs exist primarily in the particulate phase in both the atmosphere and water (Bidleman 1988).

Some molecular ratio of specific hydrocarbons has been developed to distinguish between the PAHs originating from various sources (pyrolytic, petroleum hydrocarbons and diagenetic). Combustion of organic matter at high temperature generates PAHs characterized by a low Phenanthrene/Anthracene ratio (<10), whereas the slow maturation of organic matter during catagenesis leads to much higher Phenanthrene/Anthracene ratio (>15) (Soclo 1986). Catagenesis is the process by which organic material in sedimentary rocks is thermally altered by increasing temperature resulting in the generation of oil and gas. The isomer ratio of fluoranthene and pyrene concentrations greater than 1 is characteristic of pyrolytic origin, whereas, values lower than 1 are related to the petroleum hydrocarbons (Sicre *et al*. 1987).

In the Gomti river sediments, ratio of fluoranthene to pyrene concentrations in winter season was given in Table-5. The ratio which was found <1 are related to petroleum hydrocarbon but > 1 shows its pryolytic origin at all the locations which also suggests that majority of the hydrocarbons have originated from combustion and open burning of common activities in the region and these have been identified as among the major contributors to the PAHs release in the region (UNEP 2003).

CONCLUSION

Our study shows that the Gomti river is highly contaminated with PAHs and poses high risk to the aquatic life. The findings support that the PAHs present in the river system at most of the locations have their origin from combustion processes in the catchments.

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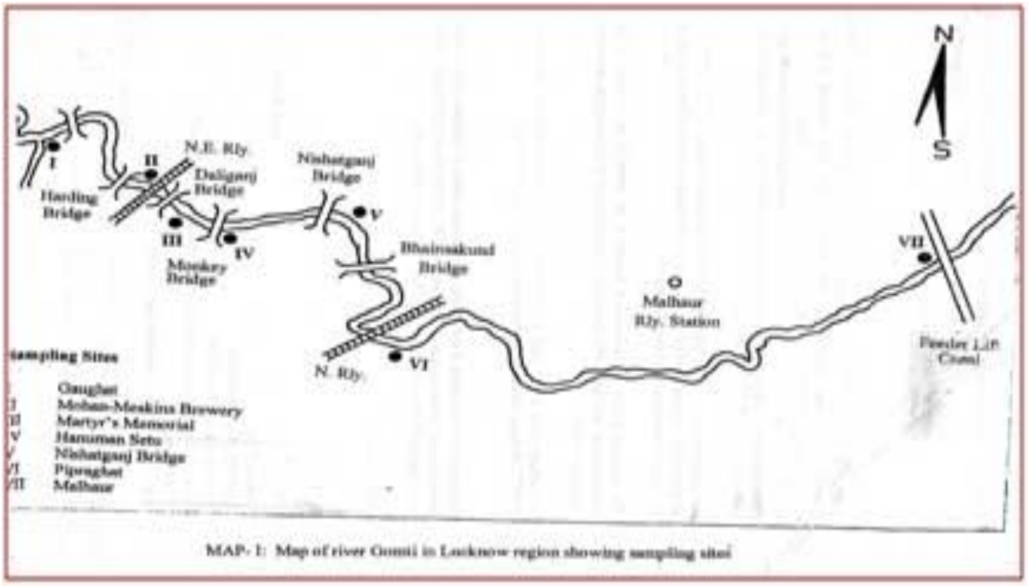
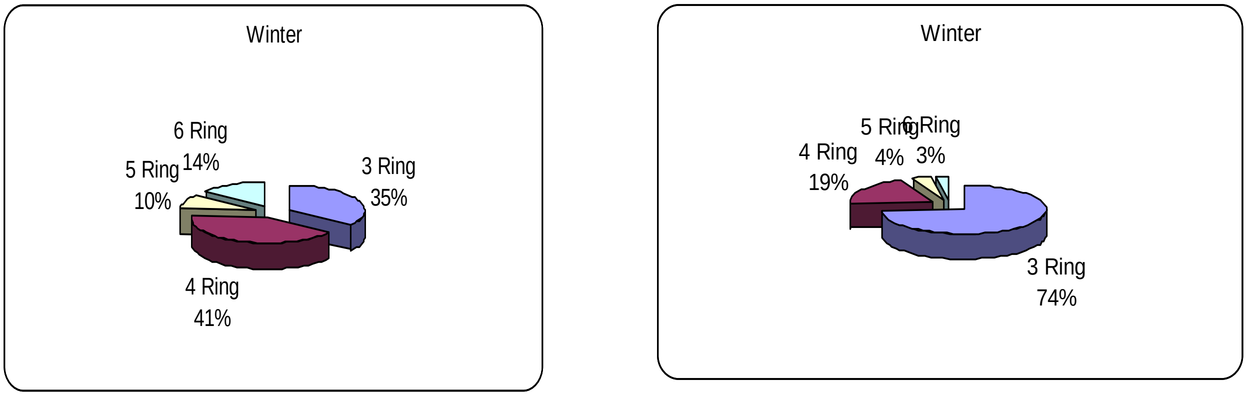


Fig.1:Map of river Gomti in Lucknow region showing sampling sites



(a) **Water** (b) **Sediments**

Fig 2: Contribution of the different groups of PAHs to the total PAHs burden in river water and sediments in winter.

Table-1: Concentration of different PAH in water (ng/ml) of river Gomti

Sampling Site	Acenaphthylene	Acenaphthene	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Chrysene	Benzo (a) Pyrene	Benzo (g,h,i) Perylene
I	1.437 (0.00-4.310)	0.688 (0.000-1.710)	0.067 (0.010-0.163)	0.106 (0.003-0.203)	0.218 (0.00-0.547)	0.541 (0.00-1.164)	1.971 (0.00-4.125)	0.000 (0.00-0.00)	0.356 (0.00-0.196)
II	0.098 (0.062-0.149)	0.098 (0.029-0.213)	0.079 (0.069-0.095)	0.004 (0.00-0.007)	0.538 (0.190-1.130)	0.050 (0.013-0.106)	0.026 (0.023-0.031)	0.091 (0.023-0.202)	0.045 (0.00-0.133)
III	0.211 (0.098-0.639)	0.042 (0.022-0.072)	0.166 (0.146-0.200)	0.002 (0.00-0.004)	0.576 (0.187-1.140)	0.021 (0.007-0.033)	0.029 (0.015-0.047)	0.056 (0.023-0.082)	0.107 (0.003-0.278)
IV	0.000 (0.000-0.000)	0.568 (0.375-0.897)	0.001 (0.000-0.002)	0.041 (0.039-0.045)	0.070 (0.014-0.103)	0.044 (0.000-0.133)	0.073 (0.048-0.112)	0.019 (0.00-0.056)	0.000 (0.00-0.00)
V	0.067 (0.000-0.127)	0.140 (0.000-0.273)	0.005 (0.00-0.010)	0.030 (0.00-0.054)	0.019 (0.017-0.021)	0.154 (0.097-0.204)	0.048 (0.029-0.061)	0.001 (0.00-0.004)	0.003 (0.00-0.005)
VI	0.142 (0.00-0.307)	0.039 (0.000-0.117)	0.013 (0.00-0.032)	0.019 (0.002-0.053)	0.082 (0.019-0.190)	0.134 (0.005-0.215)	0.029 (0.000-0.058)	0.018 (0.00-0.046)	0.000 (0.000-0.000)
VII	0.407 (0.025-0.785)	0.544 (0.257-1.098)	0.010 (0.005-0.017)	0.000 (0.00-0.001)	0.058 (0.034-0.097)	0.063 (0.029-0.127)	0.025 (0.012-0.049)	0.184 (0.018-0.343)	0.032 (0.00-0.071)

Table-2: Concentration of different PAH in sediment (ng/g)of river Gomti

Sampling Site	Acenaphthylene	Acenaphthene	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Chrysene	Benzo (a) Pyrene	Benzo (g,h,i) Perylene
I	112.097 (2.27-212.68)	116.763 (59.13-181.93)	0.330 (0.0-0.99)	3.739 (2.75-4.428)	16.630 (14.16-19.34)	39.060 (23.14-52.58)	0.987 (0.52-1.35)	0.233 (0.0-0.70)	0.356 (0.00-0.196)
II	275.217 (175.61-390.24)	724.633 (447.59-1246.52)	0.000 (0.0-0.0)	14.883 (8.94-21.30)	44.993 (29.99-55.81)	166.217 (94.36-217.10)	5.250 (4.19-6.41)	0.707 (0.0-1.93)	0.045 (0.00-0.133)
III	235.283 (151.87-362.64)	597.270 (194.22-1398.29)	1161.837 (0.0-3399.50)	284.247 (0.0-836.16)	1217.873 (27.84-1929.68)	159.310 (87.77-262.73)	86.670 (7.70-149.79)	95.977 (0.0-283.37)	0.107 (0.003-0.278)
IV	356.413 (0.0-927.90)	262.887 (0.0-583.83)	1750.463 (0.0-5191.02)	9.430 (0.0-18.57)	922.037 (0.0-2726.12)	1387.030 (158.93-3801.13)	52.087 (0.0-144.63)	109.347 (0.56-310.36)	0.000 (0.00-0.00)
V	167.983 (0.0-345.99)	23623.177 (44.63-59176.12)	28.557 (0.0-85.67)	5.997 (0.0-17.99)	1336.327 (850.19-1627.17)	22.213 (15.45-28.64)	179.517 (123.77-275.38)	90.993 (60.49-120.01)	0.003 (0.00-0.005)
VI	112.887 (0.0-338.66)	10185.053 (0.0-30376.44)	2.627 (0.0-7.88)	46.533 (15.76-123.84)	1530.617 (9.79-3297.84)	161.017 (0.0-357.11)	67.183 (0.0-196.37)	92.360 (0.0-277.08)	0.000 (0.000-0.000)
VII	131.093 (0.0-393.28)	443.383 (0.0-875.36)	1163.363 (0.0-3453.30)	34.207 (0.0-102.62)	701.187 (9.14-1110.02)	25.493 (0.0-38.59)	25.080 (0.0-61.74)	100.743 (0.0-200.99)	0.032 (0.00-0.071)

Table 3: Concentration (ng/g dry wt) of possible mutagenic/carcinogenic hydrocarbons in the sediments of Gomti river and other aquatic systems.

Aquatic System	**n***	**PAHs (ng/g dry wt.)**	**Reference**
Gomti River. India	9	10.51-35953.16	Present Study
East River. USA	18	1069910	Tabak et al. 2003
NY/NJ Harbor. USA	4	1100	Tabak et al. 2003
Todos Santos Bay. Mexico	16	7.6-813.1	Zamora et al. 2002
Morava river. Czeck Republic	16	636- 13205	Vondracek et.al 2001
Remote Mt. Lakes	-	13000-18000	Fernandez et al.1999
Passic River.USA	19	0.22-8000	Huntley et al.1995

n*= Number of hydrocarbon taken in the analysis.

Table 4 : Phenanthrene/Anthracene & Fluoranthene/Pyrene Ratio in the sediments of the Gomti River in Winter Season.

Location	Phenanthrene/Anthracene	Fluoranthene/Pyrene
I	0.09	0.43
II	0.0	0.27
III	4.09	7.64
IV	185.63	0.66
V	4.76	60.17
VI	0.06	9.51
VII	34.01	27.51