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Inhalation and carcinogenic exposure profiles of atmospheric polycyclic aromatic hydrocarbons at a semi-arid region of India

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Abstract
Polycyclic Aromatic Hydrocarbons (PAHs) are toxic air pollutants (TAPs) that have the potential to cause serious and adverse health effects and damage to the environment. These pollutants differ from other primary air pollutants such as sulphur dioxide, nitrogen dioxide, total suspended particulates (TSP) and respiratory suspended particulate matter (RSPM) in the respect that even at substantially lower concentrations; their health effects are carcinogenic in nature. Aerosols from an urban residential site of Agra city (a semi-arid region) were sampled for a span of one year (2010-2011) in order to study the concentration and exposure profiles of PAHs. The samples were extracted with dichloromethane using an automated Soxhlet Extraction. The extracts were analyzed for 17 target Polycyclic Aromatic Hydrocarbons (PAHs). The total PAH (TPAH) concentrations were 27.96 ± 2.3, 22.24 ± 2.1, and 34.38 ± 3.0 ng m^{-3}, respectively, during summer, monsoon and winter seasons. The combined mean concentration of TPAH was 28.19 ngm^{-3} for all seasons. Benzo(b)fluoranthene, Benzo(g,h,i)perylene, Benzo(b)fluoranthene and Indeno(1,2,3-cd)pyrene were the chief PAHs found in the samples collected from this urban region. The average TPAH percentage based on the number of rings were 1% (2-ring), 2% (3-ring), 24% (4-ring), 48% (5- ring), and 25% (6-ring). Dibenz(a,h)anthracene contributed the highest carcinogenic exposure equivalent (4.09 ng m^{-3}) followed by Benzo(a)pyrene (2.80 ng m^{-3}), Benzo(b)fluoranthene (0.453 ng m^{-3}) and Benzo(k)fluoranthene (0.338 ng m^{-3}), accounting for approximately 50%, 35%, 6% and 4% of the total carcinogenicity of PAHs in this urban region, respectively.
Keywords*: Atmospheric PAHs, TEFs, Carcinogenic exposure, Semi-arid region*

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I. INTRODUCTION

Atmospheric aerosols are ubiquitous in the troposphere and play an important role in climate and atmospheric chemistry. They include inorganic and organic chemical species, which could have harmful effects on human health. Aerosols resulting from the emission of harmful gases and particles are serious environmental air pollution problems in many of the world's urban areas [1,2]. Nowadays, there is a lot of concern about air quality in large urban areas, and it has been the subject of many studies. The harmful effects of Polycyclic Aromatic Hydrocarbons (PAHs) on human health have been especially studied, since some of these are carcinogenic and mutagenic, and some are associated with acute and chronic health problems [3,4,5]. PAHs are organic compounds of carbon and hydrogen arranged in combinations of aromatic rings. The contribution from natural sources of PAHs is limited, being restricted to spontaneous forest fires and volcanic emissions [6]. They belong to the group - Persistent Organic Pollutants (POPs) known for their chemical carcinogenicity [7]. In many circumstances the environmental occurrence of PAHs has been associated with adverse effects on public health [8]. In cities, the sources of PAHs are exclusively anthropogenic, i.e. they are formed during incomplete combustion and pyrolysis of organic matter, such as coal, oil, wood and fuels like diesel and petrol. Automobile exhaust has been recognized as the major PAHs contributor in urban areas [9,10]. Their elevated concentrations in urban environments pose a risk of exposure to inhabitants [11]. Atmospheric PAHs are partitioned between the particulate matter and gas phases, depending on the PAH molecular weight. Low molecular weight PAHs have higher concentrations in the vapor phase while high molecular weight PAHs are often associated with particles [2]. The increasing demand for fossil fuels over the past decades due to urbanization, industrial development, increase in population and vehicular traffic has caused a rise in the emission of PAHs to the atmosphere. Studies of the behavior of PAHs in non-tropical regions began in the 1970s, but these studies cannot be fully applied to tropical regions because of climate differences.

In this study, we have chosen to index this mixture in terms of particulate polycyclic aromatic hydrocarbons; they being ubiquitous in the urban atmosphere, a concern has been raised regarding the possible adverse effects on human health in particular with respect to their mutagenic and carcinogenic properties. Therefore, exhaustive and relevant information on their concentrations and their emission sources is necessary to achieve two main goals: *Scientific* for better comprehension of air chemistry and *political* for the design of emission control strategies and their impact. In Agra, the most important source of PAHs is expected to be vehicular emission since motor vehicles contributed as much as 60% of pollution. There are also other important potential sources of PAHs such as smoke coming from diesel generators which are in use because of erratic power supply. The aim of this study is to investigate the concentration and distribution of particulate PAHs at this agricultural site in Agra.

II. METHODS AND MATERIALS

Agra, the city of Taj (27°10'N 78°02'E) is situated on the West bank of river Yamuna in the north central part of India about 200 kilometers (km) south of Delhi in the Indian state of Uttar Pradesh. As the home of the Taj Mahal, Agra is one of the most famous tourist spots in the world. Agra district is around 10,863 sq.km, having a population of 4,418,797 [12]. **Fig. 1** shows the different locations on the map of Agra. **Table 1** shows the meteorological conditions measured during the study period. Three national highways (NH 2, NH 3 and NH 11) cross the city. The atmospheric pollution load is high because pollutants are transported downwind, mainly from an oil refinery situated in Mathura, 50 km northwest from the center of Agra City.

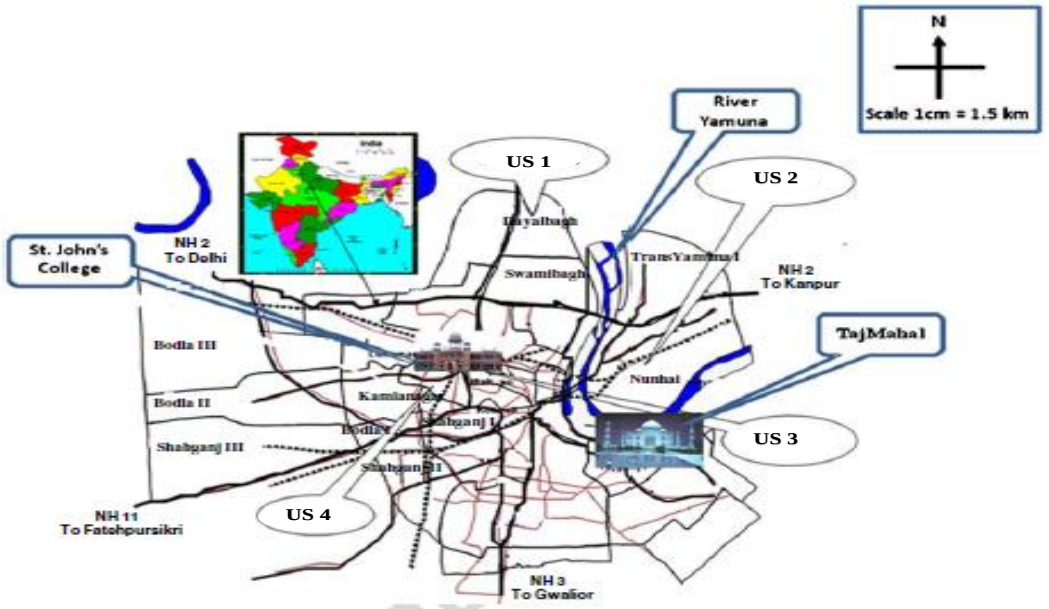

Figure 1: Map of Agra showing different sites

The station was monitored for ambient air quality twice a month in a scheduled manner. Aerosols were collected on 20.3 x 25.4 cm^2 glass fiber filter paper (EPM-2000) using respirable dust samplers (RSPM Envirotech Sampler RDS, 460 DX, New Delhi, India) at the rate of 1.0 cubic meter per minute (m^3/min). The air suction rate was verified every week using calibrated rotameters with an accuracy of ±1%. Samples were stored in a cool, dark place until analysis. Samples and blanks were extracted with 140 milliliters (mL) methylene chloride by Soxtherm®. Blank spike/blank spike duplicate (BS/BSD) samples (spiked with PAH spiking solution) were extracted using clean fibreglass thimbles. The gas chromatograph (GC) oven was temperature programmed to separate the method analytes on a fused silica column, which were then detected with a mass spectrometer (MS) [13].

Table 1: Meteorological conditions measured during the study period

Meteorological parameter	Mean	S.D.	Range
Summer			
Wind speed (m s^{-1})[a]	4.7	0.62	0.2-9.2
Air temperature (°C)	33.2	3.4	15.4-48.8
Relative humidity (%)	48.3	7.1	18.4-62.7
Monsoon			
Wind speed (m s^{-1})[a]	3.7	0.66	0.2-6.9
Air temperature (°C)	31.8	2.4	14.5-38.2
Relative humidity (%)	89.3	7.7	35.6-96.2

Winter			
Wind speed (m s^{-1})[a]	3.8	0.83	0.1-7.5
Air temperature ($^{\circ}$C)	19.1	1.7	3.5-29.5
Relative humidity (%)	78.8	8.4	38.4-88.7

[a] *Dominant Wind direction during summer, monsoon and winter seasons remained as west and north- northwest, east and south-southwest and west-northwest and north-northwest respectively*

III. RESULTS AND DISCUSSION

The statistical data is shown in **Table 2**. Total PAH concentration ranged from 09.57 ng m^{-3} to 63.64 ng m^{-3}. The average concentration with standard deviation of total PAH for the entire sample collected in ambient air was 28.19 ± 6.31ng m^{-3}, and the median was 15.59 ng m^{-3}. **Figure 2** illustrates the average total PAH concentrations during winter, summer and monsoon.

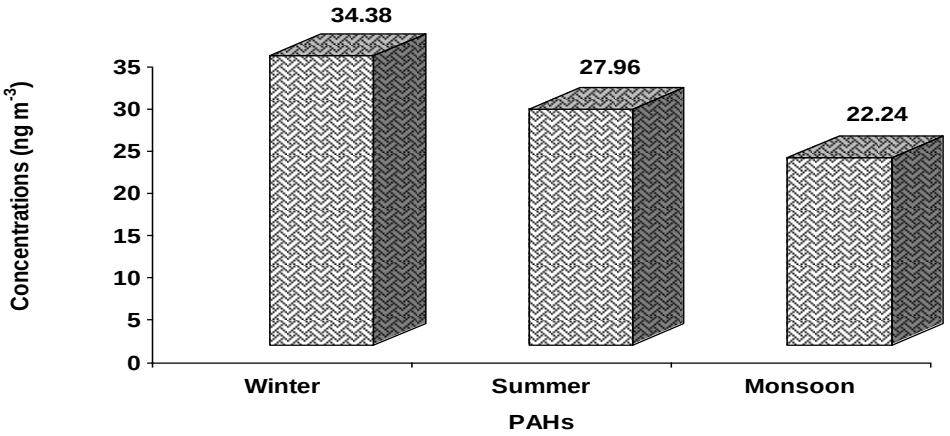

Figure 2: Average total PAH concentrations during winter, summer and monsoon

Table 2: Mean, Median, and Range of PAH (ng m^{-3})

PAHs	Mean ± S.D.	Median	Range
Naphthalene	0.07 ± 0.02	0.05	0.03 – 0.16
Acenaphthylene	0.08 ± 0.02	0.06	0.04 – 0.20
Fluorene	0.03 ± 0.01	0.02	0.01 – 0.09
Phenanthrene	0.04 ± 0.01	0.03	0.02 – 0.10
Anthracene	0.29 ± 0.03	0.20	0.05 – 1.83
Carbazole	0.09 ± 0.02	0.07	0.06 – 0.87
Fluoranthene	0.96 ± 0.04	0.57	0.07 – 1.07
Pyrene	1.38 ± 0.5	0. 96	0.54 – 4.25
Benzo(a)anthracene	1.21 ± 0.4	0. 83	0.42 – 3.29
Chrysene	2.20 ± 0.5	1. 79	0.76 -3.76
Benzo(b)fluoranthene	4.53 ± 0.7	2.96	1.97 – 9.44
Benzo(k)fluoranthene	3.38 ± 0.6	2.07	0.98 – 7.12
Benzo(e)pyrene	2.65 ± 0.6	1.94	0.82 – 5.24
Benzo(a)pyrene	2.80 ± 0.3	1.98	0.89 – 5.47
Indeno(1,2,3-cd)pyrene	4.09 ± 1.5	2.89	1.63 – 9.14
Dibenz(a,h)anthracene	1.14 ± 0.1	0.7	0.37 – 3.88
B(g,h,i)P	3.24 ± 1.0	2.05	0.91 – 7.73
TOTAL	**28.19 ± 6.31**	**15.59**	**09.57- 63.64**

PAH concentrations measured in aerosol at the urban site during different seasons are presented in **Table 3**. Benzo(b)fluoranthene, Benzo(g,h,i)perylene, Benzo(b)fluoranthene and Indeno(1,2,3-cd)pyrene were the predominant compounds at this particular urban site.

Table 3: Seasonal average concentration of PAHs (ng m^{-3})

PAHs	Monsoon	Summer	Winter
Naphthalene	ND	0.12	0.09
Acenaphthylene	0.11	0.13	ND
Fluorene	0.10	ND	ND

Phenanthrene	ND	0.11	ND
Anthracene	0.20	0.24	0.43
Carbazole	0.09	0.18	ND
Fluoranthene	0.71	0.94	1.24
Pyrene	1.22	1.35	1.58
Benzo(a)anthracene	1.13	1.14	1.35
Chrysene	2.12	2.16	2.32
Benzo(b)fluoranthene	3.27	4.35	5.97
Benzo(k)fluoranthene	2.54	3.62	3.98
Benzo(e)pyrene	1.89	2.76	3.3
Benzo(a)pyrene	2.26	2.72	3.43
Indeno(1,2,3-cd)pyrene	2.65	3.8	5.83
Dibenz(a,h)anthracene	0.88	1.13	1.42
Benzo(g,h,i)perylene	3.07	3.21	3.44
TOTAL	**22.24**	**27.96**	**34.38**

ND: Not Detected

Figure 3 shows the trends of individual PAHs throughout the year. The major contributors to TPAH in the aerosol of urban site were 4-rings followed by 5-ring and 6-ring PAHs in all the seasons.

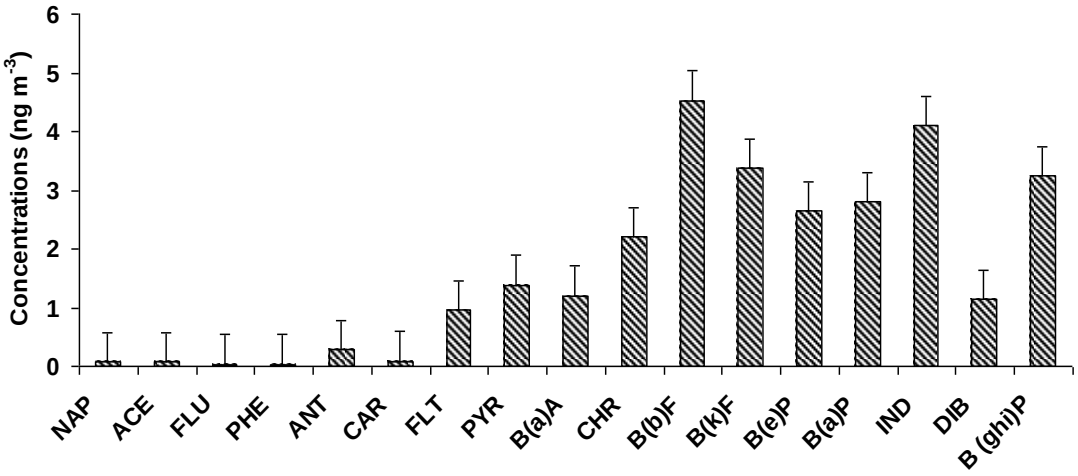

Figure 3: Pattern of individual PAHs throughout the year

3.1 Seasonal variation of PAHs

Summer season comprises of four months i.e. from March to June (temperature ranges from 30.4°C to 48.8°C). Monsoon season comprises of four months *i.e*. from July to October (temperature ranges from 20.5°C to 35.2°C). Winter season comprises of four months i.e. from November to February (temperature ranges from 2.5°C to 28.5°C). The differences in PAH concentration in aerosol is due to the characteristics of individual sites. Although the trend of seasonal variation of all PAH at all the sites is similar in nature i.e. maximum concentration of PAH were found to be in winter (34.38 ng m^{-3}) followed by summer (27.96 ng m^{-3}) and monsoon (22.24 ng m^{-3}) seasons. **Figure 4** illustrates the seasonal pattern of PAHs.

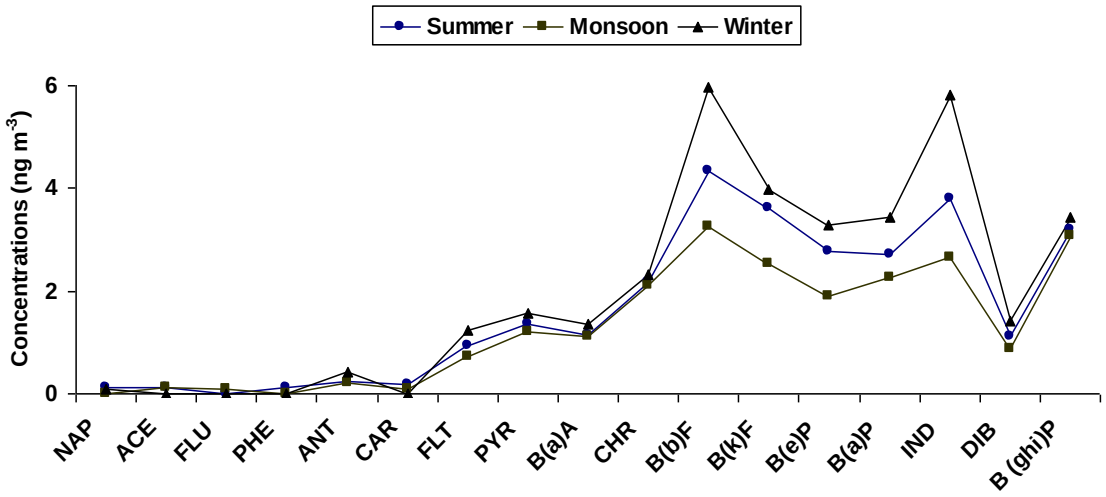

Figure 4: Seasonal trend of PAHs at urban site of Agra

During winter season, people use to make fires using wood and coal (Briquettes) in order to make themselves and their surroundings cozy, enhancing the levels of persistent organic pollutants very high. In summers, the supply of electricity is very much erratic due to high electric loads as the people use air conditioners. Thus, a massive number of diesel generator sets are used to produce adequate current, and as a result the levels of these organic pollutants get augmented. Apart from domestic source contribution in different seasons, the greater degree of photo degradation and chemical decomposition under bright sunshine and higher environmental temperature of summer season must be the influential factors for low PAHs levels, whereas the highest PAHs levels during winter season may be attributed to the factors like additional increase in emissions from domestic sector, meteorological factors like high inversion frequency and low wind speed resulting to slow dispersion rate, low environmental temperature and sunshine resulting to reduced photo degradation of PAHs [14,2]. During the months of monsoon season the region is generally experienced with the recurrent intense rain showers and washout effects of pollutants. Thus, in addition to dry deposition, wet deposition (rain) of PAHs may be accredited to the lowest particulate PAH levels during monsoon season [8,10]. **Figure 5** shows the relative contribution of 2-, 3-, 4-, 5-, and 6-ring PAHs in aerosols, investigated in this study. The average TPAH percentage based on the number of rings were 1% (2-ring), 2% (3-ring), 24% (4-ring), 48% (5- ring), and 25% (6-ring).

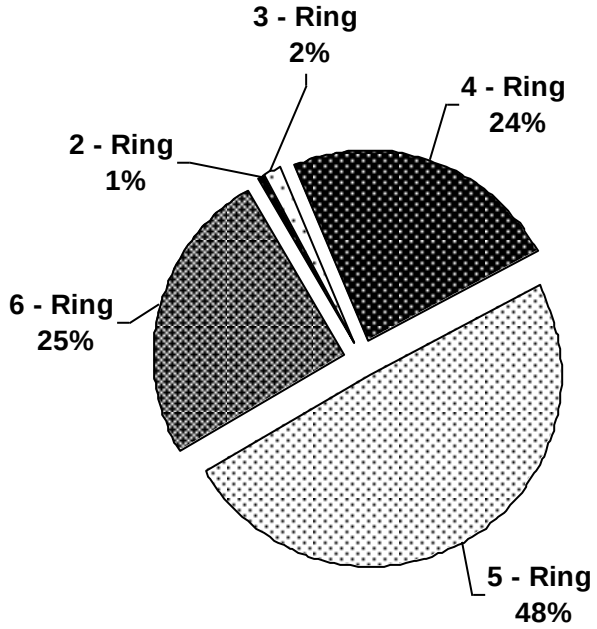

Figure 5: Relative contribution (%) of 2, 3, 4, 5 and 6-ring PAHs

3.2 Source apportionment

Principal Component Analysis (PCA) is a well-established tool for analyzing structure in multivariate data sets [15,7]. A varimax rotated factor analysis was performed to identify the main sources influencing the concentration of studied pollutants at the sampling sites. In this statistical method a set of multiple inter-correlated variables is replaced by small number of independent variables (factors) by orthogonal transformations (rotations). The varimax procedure was adopted for rotation of the factor matrix to transfer the initial matrix into one that was easier to interpret. In the present study, the Statistical Package for Social Scientist (SPSS version 11.0) computer software was used to perform factor analysis. As presented in **Table 4**, shows the factor analysis of the measured PAHs at the residential site. It reveals four factors with Eigen value > 1 accounting for 100% of the variance. The first factor, which accounts for 30.8% of the total variance contains B(a)A, CHR, B(b)F, B(k)F, IND, and may be attributed to vehicular emission as the houses are built very near to roads. The second factor, contributing 29.9% of the variance, contains NAP, PHE, FLT and DIB with incineration as the probable source. The third factor contributes 25.1% of the total variance and may be explained by the use of heavy diesel generators to generate electricity. The fourth factor contributes 14.2% of the total variance and contains CAR and CHR. This may be related to combustion activities (cooking). Differences in concentration of PAH in aerosols can be explained with the different meteorological conditions of these seasons. Due to the volatile nature of these persistent pollutants, temperature is a very important factor in determining their concentrations. The concentration of PAH at present site increases with increasing temperature as photo degradation and volatility accompanied by rising temperature [16], resulting the lower PAH in the summer than in winter season. In contrast, in the winter season at low temperature photo degradation of PAH is decreased, resulting the higher concentration of PAH at this season. While during the months of monsoon season the region is generally experienced with the frequent rain showers and washout effects of pollutants. [8].

Table 4: Results of factor analysis with varimax rotation on PAHs

PAHs	Factor 1	Factor 2	Factor 3	Factor 4
NAP	.311	.577	.489	-576
PHE	-112	.962	-178	.174
CAR	.179	.282	-251	.913
FLU	-524E-02	.967	-113	.222
PYR	.113	.217	-965	9.138E-02
B(a)A	.510	-536	.547	-392
CHR	.766	.377	-9.05E-02	.512
B(b)F	.747	-236	.499	.370

B(k)F	.989	-110	-9.58E-02	-4.58E-03
B(e)P	-1.85E-02	-113	.985	-132
B(a)P	-529	.225	.770	-276
IND	.703	-590	-303	-256
DIB	-160	.977	-6.65E-02	-123
B(ghi)P	-984	.114	.134	1.759E-02
Eigen Value	4.310	4.189	3.520	1.981
% of Variance	30.8%	29.9%	25.1%	14.2%
Cumulative %	30.8%	60.7%	85.5%	100%
Predicted Sources	**Vehicular activities**	**Incineration**	**Use of Diesel generators**	**Cooking activities (using coal & oil)**

3.3 Carcinogenicity of PAHs

Toxic equivalency factors (TEFs) can be used as a practical tool for regulatory purposes for large groups of compounds. The TEF concept is based on the following assumptions: there is a reasonably well-characterized reference compound, these are qualitatively similar toxic effects for all members of the class, TEFs for different toxic end points are similar, the toxic effects of different compounds in a mixture are additive. In this study, the abbreviation TEF is used to denote the cancer potency of specific PAH compounds relative to the potency of B[*a*]P. The TEF values are not true values but are based on the best available data, which in many cases are scanty. The TEFs should be used with great caution, as studies on mixtures of individual PAHs have shown that they may interact metabolically in a number of ways. For pragmatic purpose, the list of TEFs compiled by Tsai PerngJy et al (A), [17] and EPA Region III (B), [13] was adopted in this study. **Table 5** indicates the mean concentration of TPAHs in the urban area of Agra to be 28.19 ng m^{-3}, which corresponds to a B(a)P equivalent exposure of 5.32 ng m^{-3} and 4.95 ng m^{-3} with respect to carcinogenicity, using TEFs according to (A) and (B), respectively. Regardless of the TEF source, similar B(a)P exposure estimates were generated by the two sets of TEF values. Benzo(a)pyrene contributed the highest carcinogenic exposure equivalent (2.80 ng m^{-3}) followed by Dibenz(a,h)anthracene (1.14 ng m^{-3}), Benzo(b)fluoranthene (0.453 ng m^{-3}), Indeno(123cd)pyrene (0.409 ng m^{-3}), Benzo(k)fluoranthene (0.338 ng m^{-3}) and Benzo(a)anthracene (0.121 ng m^{-3}), accounting for approximately 54%, 22%, 8%, 7%, 6% and 2% of the total carcinogenicity of PAHs in this urban region, respectively.

Table 5: BaP toxic equivalency factors (TEFs) and BaP_{eq} exposure profiles (ng m^{-3})

PAHs	MEAN	*TEFs	$BaP_{exposure}$	+TEFs	$BaP_{exposure}$
Naphthalene	0.07	0.001	0.00007	--	
Acenaphthylene	0.08	0.001	0.00008	--	
Fluorene	0.03	0.001	0.00003	--	
Phenanthrene	0.04	0.001	0.00004	--	
Anthracene	0.29	0.01	0.0029	--	
Carbazole	0.09	--	--	0.001	0.00009
Fluoranthene	0.96	0.001	0.00096	--	--
Pyrene	1.38	0.001	0.00138	--	--
Benzo(a)anthracene	1.21	0.1	0.121	0.1	0.121
Chrysene	2.20	0.01	0.022	0.001	0.0022
Benzo(b)fluoranthene	4.53	0.1	0.453	0.1	0.453
Benzo(k)fluoranthene	3.38	0.1	0.338	0.01	0.0338
Benzo(e)pyrene	2.65	--	--	--	--
Benzo(a)pyrene	2.80	1	2.80	1	2.80
Indeno(1,2,3-cd)pyrene	4.09	0.1	0.409	0.1	0.409
Dibenz(a,h)anthracene	1.14	1	1.14	1	1.14
Benzo(g,h,i)perylene	3.24	0.01	0.0324	--	--
TOTAL	**28.19**		**5.32**		**4.95**

***TEFs cited by Tsai et al. (2004)*

+TEFs cited by EPA Region III (2006)

3.4 Worldwide comparison of total PAHs

Table 6 compares the results of the present study with PAH concentrations measured in ambient air at other locations worldwide. The table clearly illustrates that levels of total PAHs in the current study (28.19 ng m^{-3}) were lower than the concentrations found in USA [Columbus (259.57 ng m^{-3}), Houston (160.00 ng m^{-3}) & Los Angeles (64.00 ng m^{-3})], Italy [Naples (54.80 ng m^{-3})], Chile [Santiago (76.80 ng m^{-3})] and Taiwan [Taichung (231.00 ng m^{-3}) & Taipei (209.00 ng m^{-3})] whereas the total PAHs levels were found to be higher from the concentrations determined in Malaysia [Kualalumpur (6.03 ng m^{-3})], USA [Texas (24.60 ng m^{-3})] and Korea [Seoul (26.30 ng m^{-3})] in comparison of the present study.

Table 6: Worldwide comparison of total PAHs with present study

Study area	PAHs (ng m^{-3})	Reference
Columbus, USA	259.57	[18]
Houston, USA	160.00	[19]
Kuala Lumpur, Malaysia	06.03	[20]
Los Angeles, USA	64.00	[19]
Naples, Italy	54.80	[21]
Santiago, Chile	76.80	[22]
Seoul, Korea	26.30	[16]
Taichung, Taiwan	231.00	[23]
Taipei, Taiwan	209.00	[24]
Texas, USA	24.60	[25]
Agra, India	**28.19**	**Present Study**

IV. CONCLUSION

In this study, the entire 17 target PAHs were determined. The expected seasonal pattern was observed, with winter maxima and monsoon minima. This pattern is explained by the increased emissions of these compounds during winter months. The total PAH concentration were maximum in winters (34.38 ng m^{-3}) followed by summer (27.96 ng m^{-3}) and monsoon (22.24 ng m^{-3}), respectively. The mean concentration of total PAHs in the urban residential area of Agra to be 28.19 ng m^{-3}, which corresponds to a B(a)P equivalent exposure of 5.32 ng m^{-3} and 4.95 ng m^{-3} with respect to carcinogenicity. A varimax rotated factor analysis was performed to identify the main sources influencing the PAH concentration at the sampling sites which reveals that automobile exhaust accounts to be the major factor for PAH concentrations followed by incineration, use of heavy diesel generators and other combustion activities like cooking. Benzo(a)pyrene (54%) contributed the highest carcinogenic exposure equivalent followed by Dibenz(a,h)anthracene (22%).

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