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Determination of Polycyclic Aromatic Hydrocarbons in Atmospheric Particulate Matters ($PM_{2.5}$) in Urban and Rural Areas of Bangladesh

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A B S T R A C T

Polycyclic aromatic hydrocarbons (PAHs) also known as polynuclear aromatic hydrocarbons are stable organic organic pollutants that have carcinogenic, mutagenic and teratogenic properties. Due to these properties as well as their persistence in the environment, they have been placed on the priority pollutants list by the United States Environmental Protection Agency (US-EPA) and European Environment Agency. These compounds are semi-volatile organic compound (SVOCs) that could be distributed in the various environmental samples and maybe deposited in the air particulate matters which could be transported over transboundary regions. This study deals with the determination of the polycyclic aromatic hydrocarbons (PAHs) in the atmospheric particulate matters ($PM_{2.5}$) in the heavily polluted mega city and rural areas in Bangladesh. Aerosol particulate matters ($PM_{2.5}$) were collected with high volume air sampler, Environtech (Model APM 550) on Quartz filters and later PAHs compounds were extracted using established method. The average concentration of $PM_{2.5}$ in the urban and rural areas were 234.5±56.6 μgm^{-3} and 82.5±19.6μgm^{-3} respectively. The average concentration of PAHs pollutants, Naphthalene, Phenanthrene, Anthracene, Flourene were found 0.46±0.23 ngm^{-3}, 0.70±0.37 ngm^{-3}, 0.73 ± 0.40 ngm^{-3}, 0.55 ± 0.55 ngm^{-3} in the urban area. Fluoranthene, Pyrene, Benzo(a)Pyrene, Benzo(e)Pyrene, Perylene, Chrysene were below detection limit in the urban area. The average concentration of Naphthalene was 0.033 ngm^{-3} in the rural area in Bangladesh. Phenanthrene, Anthracene, Flourene, Chrysene, Fluoranthene, Benzo(a)pyrene, pyrene were below detection limit in the rural area. Different multivariate techniques like Pearson's correlation and diagnostic ratios were applied to identify the possible sources of PAHs emission.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are compounds consist of at least two fused aromatic rings. This class of compounds primarily originate from incomplete combustion of carbon-bearing materials such as wood, fat, tobacco, fossil fuels and therefore have both point (e.g., oil spill) and nonpoint sources (e.g., vehicle

emissions) (1). These compounds are semi-volatile organic compound (SVOCs) that are ubiquitous in different environmental entities. They are formed by both anthropogenic (e.g., fossil fuel burning) and natural activities like volcanic eruption or by biological releases (2-5). Vehicles, industrialization, power generation, waste/agriculture incineration are the major sources of anthropogenic PAHs emission in urban cities (6,7). In atmosphere, PAHs could be distributed as gaseous form or may be deposited on the particulate matter (PM) and depending on meteorological condition, they could be airborne and could be transported over transboundary regions (8-10). PAHs evolving from residential and industrial sources could travel to distant regions such as Arctic and Antarctic regions through long-range atmospheric transport (LRAT) (11-13). Sixteen PAHs are considered priority pollutant by USEPA of which some are confirmed carcinogenic and mutagenic and owe considerable concern due to their long term detrimental human health effects (1, 14-15). Increase of respiratory disease associated with the exposure to ambient air pollution that may contain particulate matter ($PM_{2.5}$ and PM_{10}), sulphur dioxide (SO_2), nitrogen dioxide (NO_2) according (16,17). Exposure to particulate matters is a serious concern because of these particles may contain PAHs (18,19), sulphate (SO4-), nitrate (NO3-), black carbon, mineral dust, sodium chloride, ammonia, water (20,17), nitro-PAHs (21) and metals like lead (Pb), chromium (Cr), aluminium (Al) and copper (Cu) (7). A report published (2016) by World Health Organization (WHO) shows that air pollution is the single most cause to premature death and annually seven million people died from it (22). Another report shows that 4.2 million people died from outdoor air pollution and 91% of these deaths belong to the middle- and low- income countries (WHO 2020). The particulate air pollution is measured through estimation of $PM_{2.5}$ and PM_{10} that could contain PAHs which are known for mutagenic, and carcinogenic effect (23,24), PAHs metabolites are able to form DNA-adduct that causes mutation (25-28). A study shows that fuel and lubricant have considerable effect on particle-bound PAHs emission and PAHs emission increases with the increase of engine load (29). Particle-associated PAHs content significantly differ from gasoline exhaust to diesel exhaust and mostly 2-4 ring PAHs were found in the gaseous phase than particle phase (30). Another study shows that PAHs emission rate from non-catalyst automobiles is about 26-fold higher than that of catalyst-assisted automobiles and emission rate is ~4 times higher for diesel driven truck per kilometer than that of catalyst-assisted vehicle (31). The PAHs content of street dust and street is affected by vehicular PAHs emission and asphalt contribute a little in the PAHs content (32). The light PAHs are mainly high temperature emission that found in gaseous phase whereas the heavy PAHs are the combustion products like Pyrene and Fluorene (33). Study on size distribution of PAHs containing particles shows that PAHs content is found mostly on 3-micron diameter particles, and it depends on the sampling site from urban to sub urban to rural areas (34). Particle size distribution also depends on aging process of aerosol that showed the tendency to shift towards larger particle size upon atmospheric transport but particles larger than 10 microns contribute minor to the PAHs content (35,36). Due to the adverse effect of PAHS on human health, it is important to estimate their concentration, source profiles to have a regional as well as global air pollution control strategy (37,38,39).

In the present study, we measured the concentration of atmospheric particulate matter, $PM_{2.5}$ and the concentrations of four accumulated USEPA priority listed particulate PAHs at the heavily traffic surrounded unban area, the capital city Dhaka, and the rural area, Horipur, Nurnagar Station, Shyamnagar, Satkhira, Bangladesh during the period of winter. Different multivariate techniques like Pearson's correlation and diagnostic ratios were applied to identify the possible sources of PAHs emission. In addition, the variation of PAHs level was evaluated in relation to backward air mass trajectories to study the regional/global impact on the pollution level. This understanding of the pollution behavior across the transboundary region provides important support in managing the effective regional/global air pollution control strategies.

Materials and Methods

Study Area

The present study emphasized understanding the dynamics of particulate PAHs pollution in terms of transboundary as well as regional basis. Keeping this goal in our mind, we selected Haripur (Latitude 22.31834° north and Longitude 89.04637° east, Elevation 23 feet), a rural area located at Shyamnagar upazila, Satkhira district of Bangladesh (Fig-1) nearby Kolkata, India and an urban area, Mukarram Hussain Science Building, University of Dhaka (Latitude: 23.72839° North, Longitude: 90.39819° East, Elevation: 34.0 Meters), located at the capital city Dhaka, Bangladesh (Fig-2). The common sources of air pollution in Dhaka

city may be attributed by the industrialization and modernization of society, motorized vehicles, explosion of the human population, massive traffic, burning of fuel, burning of coal in the brick kilns and power plant, burning of different types of plastic, dust particles from the different construction sites. The climate in Bangladesh is characterized by high temperature and high humidity throughout the year with a distinct seasonal variation of precipitation. The approximate annual rainfall in the city varies generally in between 159 mm (85 inches) and 3,048 mm (120 inches). On the average, approximately 80% of the yearly rainfall occurs during the monsoon season, May to September. Wind direction in Dhaka city is mainly from west and south-west direction at pre-monsoon/monsoon, and from north and north-west at winter (83).

Figure.1 Map of university of Dhaka area indicating sampling location in red sing. Source: Google

Sampling Procedure

Samples for particulate matter $PM_{2.5}$ were collected from both urban (n=9) and rural (n=4) areas through quartz filters (Gelman, Membrane Filters, Type TISSU Quartz 2500QAT-UP, 47 mm diameter) using high volume air sampler, Environtech (Model APM 550) for 24:00 hours. Before sampling, filters were prepared at about 800^0C for 200 min to reduce the organic species background level from filters. The instrument was installed at about 34 m height on the roof of Mukarram Hussain Science Building, Department of Chemistry, University of Dhaka, Bangladesh.

Particulate matters ($PM_{2.5}$) was determined from the difference between loaded and unloaded filters with a digital balance after conditioning them in a desiccator for 3 hours. The loaded quartz filters wrapped in aluminum foil to prevent PAHs loss were kept in the refrigerator at 4^0C until chemical analysis.

Fast Analytical Approach

A fast and efficient extraction procedure was followed to extract particulate PAHs. The samples were subjected to Soxhlet extraction by 40mL of dichloromethane solvent and then sonication was done for 24 hours. After sonication the extract was filtered through Whatman 41 filter paper and collected in a clean volumetric flask. The filtrate solution was pre-concentrated through Kuderna-Danish evaporator and reduced to below 2 mL solution which was then subjected to clean-up.

Clean-up Procedure

A clean-up column was prepared with internal diameter 1 cm. 20g pre-activated silica gel was placed on top of the cotton sitting down the column and then anhydrous sodium sulphate was topped on it by 1.5 cm height in the column. 5 mL dichloromethane was passed through the column and the pre-concentrated solution was introduced

on the top of column which was then washed with 60 mL of dichloromethane solvent. The collected eluent was later evaporated using K-D evaporator.

Chromatographic Analysis

The GC-MS analysis of the crude extract was performed using a Varian GC-MS (Model Varian CP 3800, USA) equipped with a VF-5 fused silica capillary column (30 m × 0.25 i. d., film thickness 0.25 µm, Varian, USA). An electron ionization system with the ionization energy of 70 eV was used for the detector of GC-MS. Helium gas was used as carrier gas with constant flow rate of 1 mL/min. Injector and mass transfer line temperature were set at 250°C and 280°C, respectively. Diluted samples (1/100 v/v, in methanol) of 1 µL were manually injected at 20% split injector mode with a solvent delay time of 4 min. The initial column oven temperature was started at 50°C held for 1 min, raised to 210°C at 8°C/min and finally raised to 280°C at 10°C/min. Identification of the extract's PAHs were done by comparing the GC retention time and area of a particular PAH with that of standard GC retention time and area as well as co-injecting the standard PAHs (40). Total Ion Chromatogram (TIC) mode was preferred for the analysis of samples. The concentration of the PAHs molecules was calculated (41) by the following equation:

$$S = \frac{(\mathrm{As} - \mathrm{Ab})\ x\ Cstd}{\mathrm{Astd}}$$

Where *S* is the sample strength, As and Astd represent the peak areas of the analyte and the standard component respectively, *Cstd* represents the concentration of the standard solution and Ab is the peak area of blank sample.

Quality Assurance

The method validation was done by Inter- and intra-day replicates to check the accuracy and precision. The system accuracy was evaluated by recovery studies in which the known amount of standard solution was added in the extract. The sample blank and sample blind were injected to verify the system suitability. Standards calibration curves were established by plotting the peak areas against four different concentrations of the reference compounds. The external standard method was used for quantification of the markers in the PAHs samples extracts. Four different PAHs compounds (Naphthalene, Anthracene, Phenanthrene and Fluorene) were used to construct the standard calibration curve. Recovery studies were done at three different standard concentrations.

Results and Discussion

Concentration of $PM_{2.5}$

The particulate matter ($PM_{2.5}$) samples were collected from both urban and rural area. The sampling duration in urban area was during the end of January to the early-February. Total nine samples were collected from the urban region. Aerosol particulate matters ($PM_{2.5}$) were collected on the quartz filters with Environtech (Model APM 550) on the roof of the Mukarram Hossain Khundhkur Biggan Bhaban at the Department of Chemistry, University of Dhaka, Bangladesh. The highest concentration of $PM_{2.5}$ was 315.40 µgm-3 and lowest concentration was 157.60 µgm-3. The average concentration was 234.5±56.5 μgm^{-3}. On the other hand, four aerosol particulate matters ($PM_{2.5}$) samples were collected from the rural area on the quartz filters with Environtech (Model APM 550) at Horipur, Nurnagar Station, Shyamnagar, Satkhira. The highest concentration of $PM_{2.5}$ was 111.60 µgm-3 and lowest concentration was 69.20 µgm-3. The average concentration was 82.50 ± 19.59 µgm-3 (19.59 µgm-3 was standard deviation). From the result, we have seen that the atmospheric particulate matters ($PM_{2.5}$) was three times lower in the rural area than the urban area. Due to the lack of massive traffics, lack of industries, lack of population and lack of air pollution creating sources the atmospheric particulate matters ($PM_{2.5}$) concentration was lower in the rural area. Due to the massive growth of population, industrialization, vehicles, construction of different infrastructure in the urban area the atmospheric particulate matters ($PM_{2.5}$) much higher in the urban area.

PAHs Concentrations Associated with $PM_{2.5}$

Four PAHs molecules, Naphthalene, Phenanthrene, Anthracene and Fluorene were investigated associated with particulate matters ($PM_{2.5}$) which were found in different concentration in the collected samples. Among the measured four PAHS, the average concentrations of Anthracene, 0.73±0.40 and Naphthalene, 0.46±0.23 were found highest and lowest PAHs respectively in the urban area (Table 3). As individual PAHs, Fluorene was found the highest concentration, 1.9368 ngm^{-3} and Naphthalene 0.1144 ngm^{-3} was found the lowest concentration in a

single day PAHs contribution in the urban area (Table 3). On the basis of single day total PAHs contribution, the highest amount was 3.6687 ngm^{-3} and the lowest amount was 1.1500 ngm^{-3} (Table 4).

Table.1 Measurements of daily variation and average concentration of atmospheric particulate matters ($PM_{2.5}$) in the urban area, Dhaka, Bangladesh

Date	$PM_{2.5}$ (μgm^{-3})	Average $PM_{2.5}$ (μgm^{-3})
28/01/2013	157.60	
29/01/2013	253.90	
30/01/2013	195.90	
31/01/2013	242.00	
01/02/2013	315.40	234.53 ± 56.6
02/02/2013	313.80	
03/02/2013	252.90	
05/02/2013	172.60	
06/02/2013	207.10	

Table.2 Measurements of daily variation and average concentration of atmospheric particulate matter ($PM_{2.5}$) in the rural area Satkhira, Bangladesh

Date	$PM_{2.5}$ (μgm^{-3})	Average $PM_{2.5}$ (μgm^{-3})
10/03/2012	73.40	
12/03/2012	111.60	82.50 ± 19.6
14/03/2012	69.20	
16/03/2012	75.80	

Table.3 Average concentration of different PAHs in the atmospheric particulate matters ($PM_{2.5}$) in the urban area Dhaka

Naphthalene (ngm^{-3})	Average Naphthalene (ngm^{-3})	Phenanthrene (ngm^{-3})	Average Phenanthrene (ngm^{-3})	Anthracene (ngm^{-3})	Average Anthracene (ngm^{-3})	Fluorene (ngm^{-3})	Average Flourene (ngm^{-3})
0.3912		0.7190		0.7400		0.2471	
0.4742		1.0103		0.8628		0.5675	
0.6228		0.4016		0.4126		0.1695	
0.7981		1.4284		1.0663		0.6941	
0.1171	0.46±0.23	0.4195	0.70±0.37	0.4300	0.73±0.40	0.1755	0.55±0.55
0.4484		0.6326		0.6509		1.9368	
0.1144		0.2666		0.2736		0.4954	
0.5084		0.9248		1.5796		0.3331	
0.6621		0.5225		0.5374		0.3573	

Figure.2 Variation of atmospheric particulate matters ($PM_{2.5}$) in the urban area Dhaka, Bangladesh

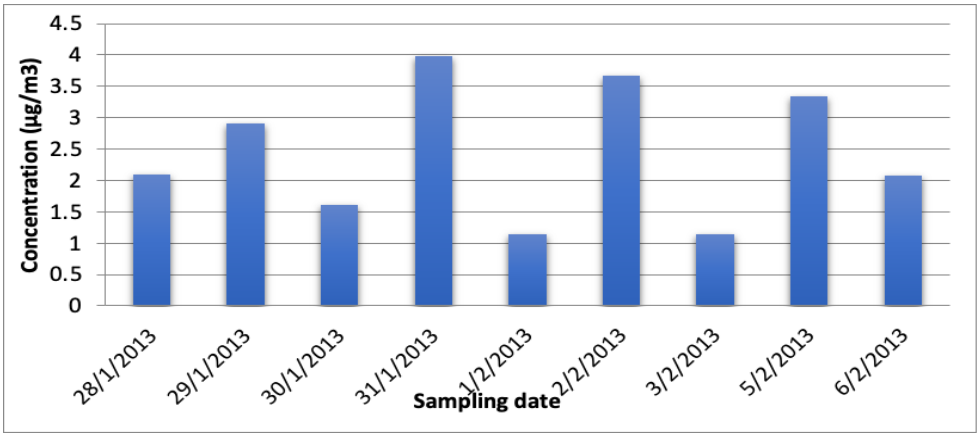

Figure.3 Variation of atmospheric particulate matters ($PM_{2.5}$) in the rural area, Satkhira, Bangladesh

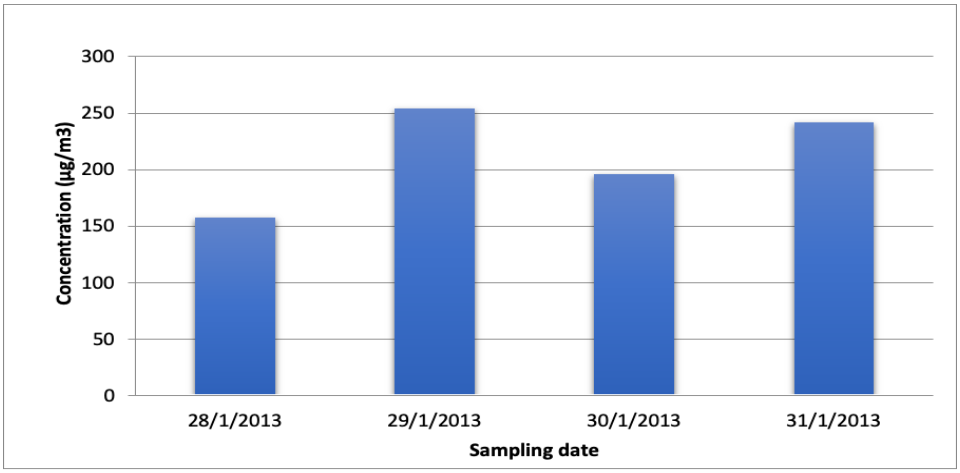

Figure.4 Variation of the average concentration of different PAHs in the atmospheric particulate matters ($PM_{2.5}$) in the urban area Dhaka

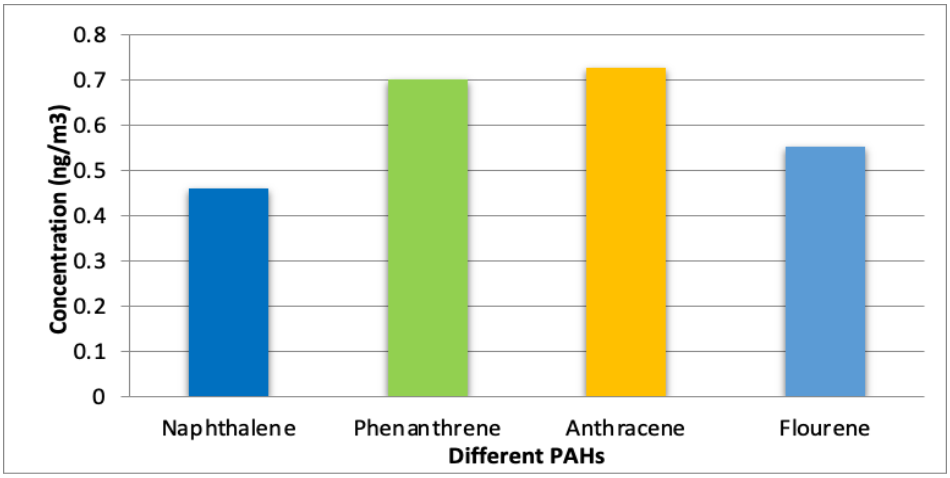

Figure.5 Daily variation of total 4 PAHs (Naphthalene, Anthracene, Phenanthrene and Fluorene) concentration in the atmospheric particulate matter ($PM_{2.5}$) of urban area Dhaka

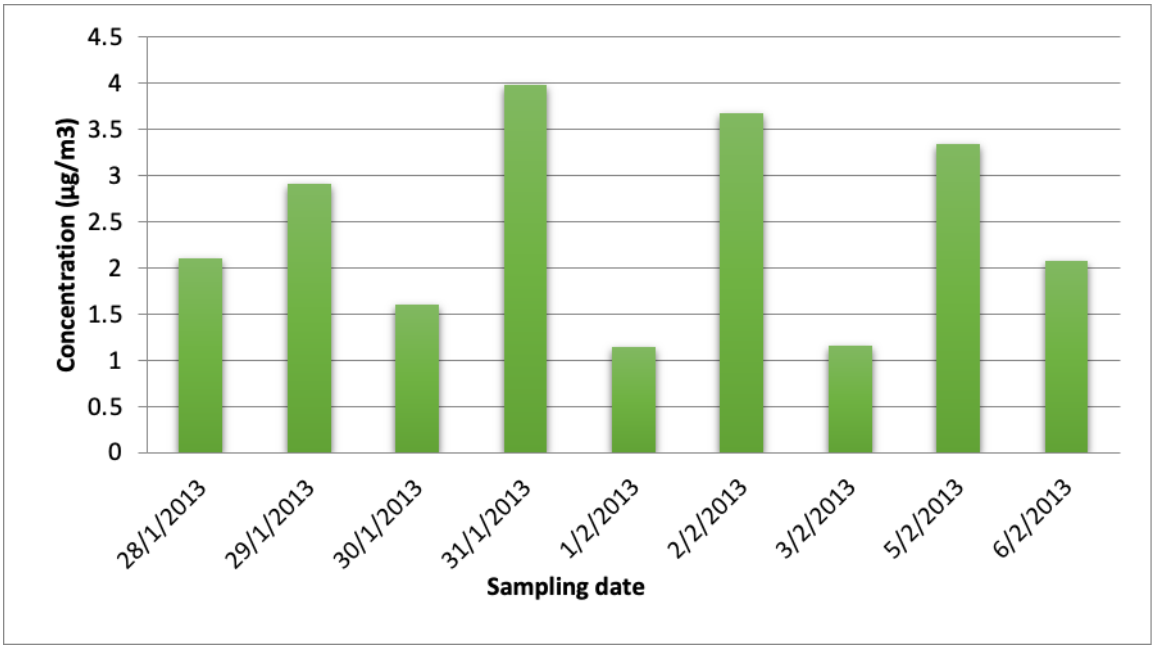

Table.4 Daily variation of total PAHs concentration in the atmospheric particulate matter ($PM_{2.5}$) of urban area Dhaka, Bangladesh

Sampling date	Naphthalene (ngm^{-3})	Anthracene (ngm^{-3})	Phenanthrene(ngm^{-3})	Flourene (ngm^{-3})	Total PAHs (ngm^{-3})
28/01/2013	0.3912	0.74	0.719	0.2471	2.0973
29/01/2013	0.4742	0.8628	1.0103	0.5675	2.9148
30/01/2013	0.6228	0.4126	0.4016	0.1695	1.6065
31/01/2013	0.7981	1.0663	1.4284	0.6941	3.9869
01/02/2013	0.1171	0.43	0.4195	0.1755	1.1421
02/02/2013	0.4484	0.6509	0.6326	1.9368	3.6687
03/02/2013	0.1144	0.2736	0.2666	0.4954	1.1500
05/02/2013	0.5084	1.5796	0.9248	0.3331	3.3459
06/02/2013	0.6621	0.5374	0.5225	0.3573	2.0793

Pearson's Correlation

	Naphthalene	Phenanthrene	Anthracene	Flourene
Flourene	0.09	0.15	0.03	1
Anthracene	0.44	**0.76**	1	
Phenanthrene	**0.63**	1		
Naphthalene	1			

The highest average concentration of Anthracene followed by Phenanthrene, Naphthalene and Flourene. As for the PAHs-to-PAHs correlation Phenanthrene was highly correlated with Anthracene (r^2 = 75.95), Phenanthrene was also correlated with Naphthalene (r^2 = 62.60), Naphthalene was lowly correlated with Anthracene (r^2 = 44.01). So Naphthalene, Phenanthrene and Anthracene were from the same sources. But Fluorene was very low correlation with Naphthalene, Phenanthrene and Anthracene. So the sources of Fluorene were different from the sources of Naphthalene, Phenanthrene and Anthracene.

In conclusion, this study aimed to address air pollution aspects due to the increased rate of mortality and morbidity and also multifarious effects of particulate pollution on our environment. The average concentration of $PM_{2.5}$ in the urban area was 234.5±56.6 $\mu gm^{-3}$and in the rural area was 82.50±19.59 µgm-3. The concentration of four PAHs (Naphthalene, Phenanthrene, Anthracene, Flourene) were measured through Gas Chromatography coupled with Mass Spectrometer (GC-MS). The average concentration of Naphthalene, Phenanthrene, Anthracene, Flourene were 0.46±0.23 ngm^{-3}, 0.70±0.37 ngm^{-3}, 0.73±0.40 ngm^{-3}, 0.55±0.55 ngm^{-3} in the urban area in Bangladesh, respectively. The average concentration of total four PAHs was 2.44±1.07 ngm^{-3}. The average concentration of Naphthalene was 0.03258 ng/m^3 in the rural area (Horipur, Nurnagar Station, Shyamnagar, Satkhira) of Bangladesh. Phenanthrene, Anthracene, Flourene were not found in the rural area (Horipur, Nurnagar Station, Shyamnagar, Satkhira) of Bangladesh. The correlation between Naphthalene and Phenantherene was (r^2 = 62.60), Naphthalene and Anthracene was (r^2 = 62.60), Phenanthrene and Anthracene was (r^2 = 75.95), Phenanthrene and flourene was (r^2= 0.15), Flourene and Naphyhalene was (r^2= 0.09), Flourene and Anthracene was (r^2= 0.03). Those compounds having correlation with r^2=60 or more were from the same sources. The average concentration of $PM_{2.5}$ in the urban area was three times higher than the rural area. From this study, our finding is that the industrialization and modernization of society, introduction of massive motorized vehicles, burning of fossil fuel in the brick kilns and power plant, burning of different types of plastic, dust particles from the different construction sites were the sources of the air pollution as well as atmospheric particulate matters ($PM_{2.5}$) in the urban areas of Bangladesh.

Data availability

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

Author contributions

Most Rayhana Rahman: Sample Collection, Methodology; K.M. Fayek Itteshaf Rownok: Sample Collection, Editing; Md Atiar Rahman: Data Analysis; Md. Jamiul Kabir: Editing, Review; Mahbuba Begum: Sample Collection; Md Mahbubul Haque: Data Curation; M S Rahman: Editing, Review; Yeasmin Nahar Jolly: Final Review.

Declarations

Ethical Approval Not applicable.

Consent to Participate Not applicable.

Consent to Publish Not applicable.

Conflict of Interest The authors declare no competing interests.

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