

Human health risk exposure with respect to particulate-bound polycyclic aromatic hydrocarbons at mine fire-affected coal mining complex

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Abstract Particulate-bound poly-aromatic hydrocarbons (PAHs) are of great concern due to their mutagenicity and carcinogenicity effect on human health. In this context, identification, quantification and inhalation cancer risk (ICR) assessment due to PM₁₀- and PM_{2.5}-bound PAHs has been carried out at six monitoring stations in a critically polluted Jharia coalfield/Dhanbad City. Identification of pollution sources at study area has been performed by using PCA statistical methods. Air quality index (AQI) and air quality health index (AQHI) were calculated based on the concentration levels of PM₁₀. Location-wise direct comparison between AQI, AQHI and ICR was performed to analyse the risk levels. Consequently, maximum concentration levels of particulate (PM_{2.5} and PM₁₀)-bound total PAHs (400 and 482 ng/m³) were recorded at the monitoring station Lodna Thana, followed by Bank More and Sijua Stadium, respectively. It was also

observed that mine fire-affected station Lodna Thana was exaggerated with presence of PAHs due to wood and open coal burning activities. Moreover, about 1000 and 889 cases of inhalation cancer risk were estimated due to direct exposure of PM₁₀- and PM_{2.5}-bound PAHs in the study area, respectively. Active mine fire-affected station Lodna Thana was recorded with maximum probability of lung tumour due to inhalation cancer risk. This study has reported higher AQHI at station Dugdha Basti, Lodna Thana and Bank More, which results increased number of tumours due to ICR. This result concludes that Jharia coalfield/Dhanbad City are not only critically polluted area but it is also an inhalation cancer prone area due to direct exposure of active mine fire.

Keywords PM₁₀ · PM_{2.5} · PAHs · Health risk analysis · Air pollution · Coal mining

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Introduction

Particulate-bound polycyclic aromatic hydrocarbons (PAHs) are the leading risk factors of the global health concern and also responsible for premature deaths every year worldwide (Lim 2012; WHO 2013). It has been reported that heart failure mortality has direct correlation with PM_{2.5} and PM₁₀ (Chuang et al. 2011; Shah et al. 2013). Moreover, particulate-bound PAHs consist of fused benzene rings and cause serious health effects, i.e., lung cancer and depressed immune function (WHO/IPCS 1998; Kim et al. 2013; Behear 2008). Furthermore, many countries, viz., European union, United States of America (USA), United Kingdom (UK), Italy, China and India, have set their air quality guidelines and determined that among aromatic organic compounds, benzo(a)pyrene is highly carcinogenic and mutagenic (Hien et al. 2007; Ravindra et al. 2008; Sin et al. 2003; WHO 2002;

MEP 2012; CPCB 2015). Recently, numerous studies have been conducted on airborne particulate-bound PAHs at different sources, viz., industry, urban, harbours, farms and residential on global scale (Manti et al. 2005; Zhang et al. 2013; Guo et al. 2003; Li et al. 2009; Fang et al. 2006; Fang et al. 2005; Huang et al. 2006; Liu et al. 2010; Sadegh et al. 2015). However, there is limited study on particulate matter (PM₁₀ and PM_{2.5})-bound PAHs in coal-based industrial area (Wu et al. 2014). Literature survey reveals that there is a need of serious concern for the health issues occurring due to particulate matter (PM)-bound PAHs from natural sources like volcanic eruption, forest fire, mine fire, etc.

So, the present study has some unique approaches, which deals with PM-bound PAHs emphasizing (i) air quality index (AQI), air quality health index (AQHI) and inhalation cancer risk (ICR) assessment in a critically polluted area with the additional impact of active mine fire and (ii) correlation among AQI, AQHI and ICR. This study was conducted in Jharia coalfields (JCF) for better understanding of the harmful impacts of natural mine fire emission on human health. We focus on the elements of the causal chain of health risk sources, exposure and health effects and provide estimates of the source strengths, exposure levels and health risks from air pollution in severely mine fire-affected JCF.

Experiment

Description of the study area and sampling stations

JCF is one of the most important coalfields in India, located in Dhanbad District, between the latitude 23°39' to 23°48' N and longitude 86°11' to 86°27' E, about 260 km north-west of Kolkata, in the heart of Damodar Valley, mainly along the north of the Damodar River. The field is roughly elliptical or sickle shaped, its longer axis running north-west to south-west, covering the area of over 460 km² and extending for a maximum of about 38 km east-west and 19 km north-south. This is the most exploited coalfield because of availability of metallurgical grade coal reserves.

Ambient air quality monitoring and sample collection were carried out at six sampling stations as per the citing criteria (IS:5182 Part XIV, 2000) with special consideration of meteorological data and sources of pollution, apart from security, accessibility and availability of electricity. One reference ambient air quality monitoring stations is established at ISM, Dhanbad, which is also considered as background for this study (CPCB 2015). Location map of the study is shown in Fig. 1 and details of these locations are as follows:

Bank More (BM/A1) has a latitude 23°47'16.9" N, longitude 86°25'07.2" E and elevation (SL) of 236.22 m. This monitoring location was adjacent to the main road of Dhanbad City. Heavy load of air pollution reported here was

due to vehicular traffic and several commercial and construction activities. *Sijua Stadium (SS/A2)* has a latitude 23°47'32.9" N, longitude 86°19'41.2" E and elevation (SL) of 267.31 m. This sampling station has a proximity of various mining-associated activities like Jogta mine fire, Rajhans coke oven plant, and Sendra Bansjora and Nichitpur opencast colliery. This site is situated in between the main road in one side and Sijua stadium on the other. This location receives pollution load from all types of sources, e.g. mining-related activities, commercial works and vehicular pollution as well. *Hero Honda Showroom/Steel Gate (HS/A3)* has a latitude 23°48'54.86" N, Lon-86°27'51.05" E and elevation (SL) of 234.70 m. The location of this sampling site was the main road to Saraidhela/Steel Gate of Dhanbad City. That site had intense effect of commercial activities and vehicular traffic. *Dugdha Basti (DB/A4)* has a latitude 23°45'25.74" N, longitude 86°09'01.76" E and elevation (SL) of 220.98 m. The monitoring station was near Dugdha coal washery, adjacent to railway track and the newly proposed coal washery. Residential activities and vehicular movements are the main sources of pollution. *Lodna Thana (LT/A5)* has a latitude 23°43'41.8" N, longitude 86°25'30.6" E and elevation (SL) of 173.74 m. The sampling station was very close to mines and receives considerable pollution and heavily affected by Lodna active mine fire, Goluckdih opencast mines and Sudamdih coal washery. The main sources of pollution are transportation of coal on unpaved haul roads, mine fire and mining with allied activities (like overburden removal, blasting, mineral haulage, mechanical handling operations, stock piles and site restoration, etc.). *ISM, Dhanbad (ISM/A-6)* has a latitude 23°48'45.3" N, longitude 86°26'23.5" E and elevation (SL) of 250.55 m. It was the reference monitoring station and isolated from the industrial activities. Main pollution is due to ongoing construction activities inside the campus and residential activities.

Respirable particulates PM₁₀ and PM_{2.5} were sampled using fine particulate samplers (Envirotech APM 541 and APM 550). The instruments were kept at 15 m above the ground level. Collection for PM was performed in the year of 2012–2013. The instruments were operated simultaneously at an average flow rate of 0.0167 m³/min at all monitoring stations. Whatman ® PTFE filter papers (47 mm diameter) were used for collection of PM₁₀ and PM_{2.5}. Ambient air quality monitoring has been carried out to assess the impact of 24 h and twice in a week at all monitoring stations concurrently. The field study was planned over a period of 24 days at each monitoring site in each season to represent the variation of air quality during the whole year. Before and after sampling, the filters were kept in a Secador desiccators (Tarson) under controlled temperature (5–30 °C; with automatic controller) and 35–40% relative humidity for at least 24 h to prevent hydration of the filter surface. The filters were

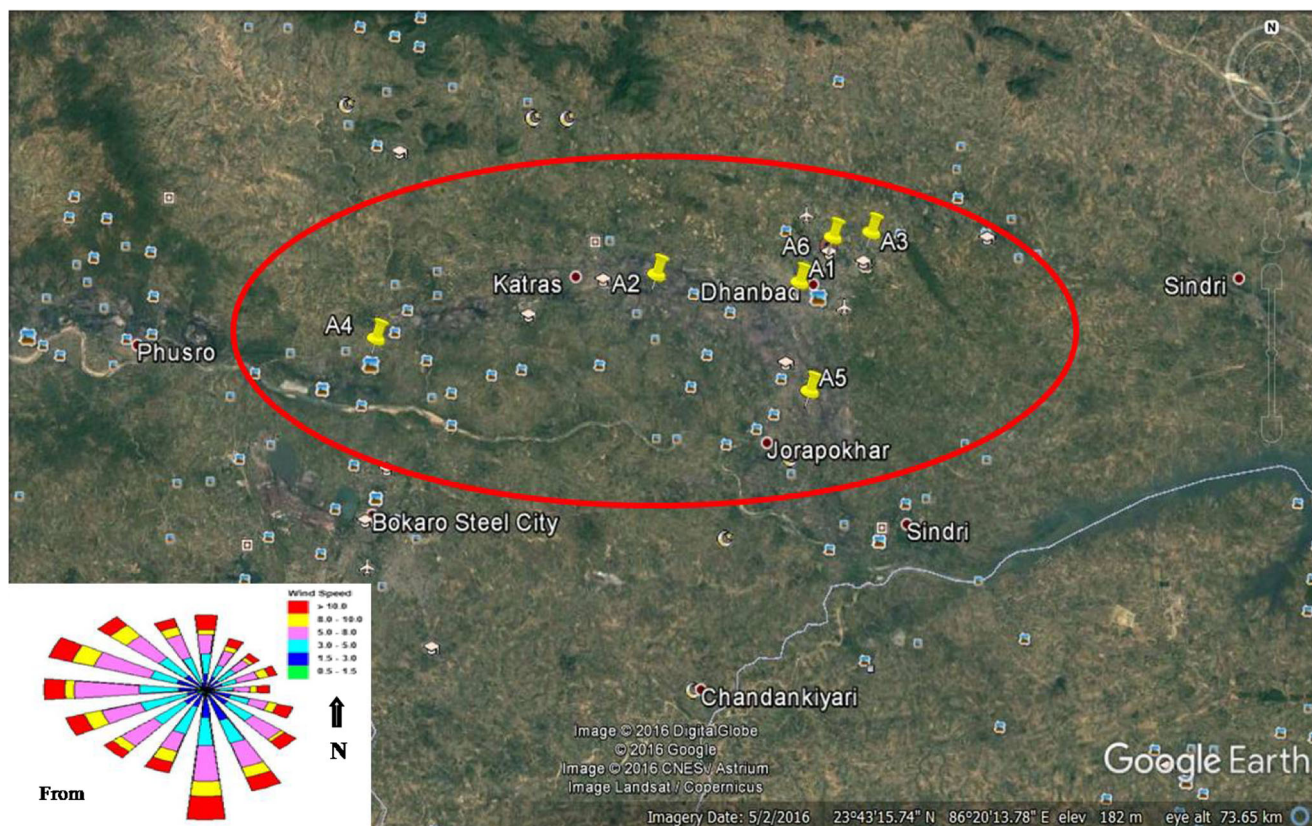


Fig. 1 Monitoring stations in the study area (Source; Google Earth 2016, imagery date 24 October 2012)

weighed twice before and after sampling using four digit microbalance with 0.010 mg sensitivity (Denver Instruments, Model TB-215D) to obtain PM₁₀ and PM_{2.5} concentration, respectively. After weighing, the filter papers were packed in aluminium foil to protect them from sunlight and they were stored under refrigeration (−16 °C) to prevent volatilization of PAHs of lower molecular weight, until extraction and analysis could be completed. The balance room’s relative humidity must be maintained at a mean value range of 45 ± 5% and its air temperature must be maintained at 25.0 °C.

Analytical process

A gas chromatograph-mass spectrometer, Agilent 7890A-5975C, USA, equipped with DB-5MS capillary column (30 m × 0.25 mm × 0.25 mm) from J&W Scientific (Folsom, CA, USA) was used for analytical determination of the particulate PAHs. For extracting the PAHs, exposed PTFE filter paper (both in PM₁₀ and PM_{2.5}) was cut into small stripes and extracted with 30 ml of HPLC grade dichloromethane (DCM) according to solvent extraction process (NIOSH *Manual of Analytical Methods* (NMAM), Fourth Edition, 8/15/94). The extract was kept in an ultrasonic agitator for 30 min of mechanical shaking. Extraction was repeated twice to get maximum extraction. The extract was allowed to evaporate up to small volume (~5 ml) using a rotary evaporator at a

temperature not exceeding 40 °C. Then under flow of dry nitrogen gas, it was further reduced and finally adjusted to exactly 1 ml by DCM after filtering through membrane filter (Sigma-Aldrich PVDF 0.5 µm micro syringe).

Then the PAH sample extract was injected into GC-MS for detailed analysis. The column temperature was held at 80 °C for 2 min, then increased to 140 °C at 20 °C/min, and then to 305 °C at 3 °C/min. One millilitre volume of each sample was injected in the splitless mode and the purge time was 1 min. The ionization was carried out in the electron impact mode at 70 eV and multiplier voltage of 1506 V. Mass spectra were scanned from 33 to 400 amu for 0.4 s in total ion chromatogram (TIC) mode to identify the compounds in the standard mixtures and samples. The temperatures of injector, transfer line and ion source were set at 300, 280 and 230 °C, respectively. Helium (purity >99.999%) was used as carrier gas at a constant flow rate of 1 ml/min.

Each compound was identified by comparison of its mass spectra with that of a standard compound and by comparison to the NIST2011 mass spectral reference library (National Institute of Standards and Technology, Gaithersburg, MD, USA version 2011). The 15 PAH compounds were identified as follows: naphthalene (NaP), acenaphthylene (Ap), acenaphthene (Ac), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyren (Pyr), benzo[a]anthracene (BaA), chrysene (Chr),

benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), Dibenz[a,h]anthracene (DBA), indeno[1,2,3-cd]pyrene (Ind) and benzo[ghi]perylene (BghiP) in the study area.

Quality assurance/quality control

All the glassware used for the PAH analysis were properly washed and sterilized/dried in an oven at 180 °C. Calibrations of the instruments were carried out with the standards of known concentrations. The PAHs were quantified using an external standard method. The standard curve of each PAH was prepared by plotting the peak area against standard concentration (ng/ml), and the amount of each PAH was calculated on the basis of its respective calibration curve. The limits of detection (LOD) and quantitation (LOQ) were calculated by using the signal-to-noise ratio of S/N = 3 and S/N = 10, respectively. The average LOD of 15 PAHs ranged from 0.005 to 0.36 mg/kg. The method recovery was determined by spiking the samples with individual PAH standards at three levels from 10 to 200 ng/ml in triplicate. Linear equations of the calibration curves and recoveries of 15 PAHs are shown in Table TS2. The regression coefficient (R^2) of the PAHs standard calibration curves covering the concentrations from 5 to 200 ng/ml ranged from 0.9897 to 0.9998. Average recoveries of 15 PAHs were in the range of 79–103%. All the measured values reported in the manuscript were blank corrected (Singh et al. 2011a, b; Li et al. 2016).

Data processing

Coefficient of divergence is used to identify the difference of PAH composition PM_{10} and $PM_{2.5}$ at different monitoring stations. Data point of two data sets are measured using self-normalizing parameter. The following equation is used to determine the parameters:

$$CD_{jk} = \sqrt{\frac{1}{P} \sum_{i=1}^P \left(\frac{X_{ij} - X_{jk}}{X_{ij} + X_{jk}} \right)^2} \quad (1)$$

In this equation, X_{jk} and X_{ij} represent the average mass concentration of chemical component i , for both j and k (Zhang and Friedlander 2000; Wongphatarakul et al. 1998). Where x_{ij} represents the average concentration for a chemical component i at the site j , where j and k represent two different sampling sites and p is the number of chemical components. When CD_{jk} approaches to one then PAH composition profiles J and K are different, and if it approaches to zero, then both are similar. CD value 0.269 indicated the similarity between two sites (Wongphatarakul et al. 1998).

Principle component analysis (PCA) method is used to identify the PAHs source in $PM_{2.5}$ and PM_{10} . For PCA, SPSS 21.0 software is applied to reduce the set of variables (factors) and interception of complex systems. Only chemical interaction or emission sources can be identified by these

factors and these factors can also indicate more than one possible sources (Kong et al. 2010). PCA with Varimax normalization rotation technique has been applied in chemical composition of particulate (PM_{10} and $PM_{2.5}$)-bound PAHs at different monitoring stations (Wu et al. 2014), as we know the minimum sample size to apply this method (Callén et al. 2009):

$$n = 30 + \left(\frac{V + 3}{2} \right) \quad (2)$$

where n = number of samples and V = number of variables/elements.

Health risk assessment PAHs at the study area

PAHs are widespread environmental pollutants due to their toxic properties. Health risk due to the presence of particulate-bound PAHs has been analysed using the following techniques:

- (1) Effective accumulated PAH inhalation estimation with respect to BaP_{eq} (ng).
- (2) The increment human health risk assessment due to exposure of PAHs which can be denoted by the number of threatened people.

Toxic equivalent of PAH inhabitants

The magnitude of exposure depends on the exposure duration and concentration of ambient PAHs with the concentration–time unit (Liao et al. 2006).

$$E = \int_{t_1}^{t_2} C(t) dt \quad (3)$$

Here, $C(t)$ is the concentration of PAHs in the ambient air ($\mu\text{g}/\text{m}^3$). Exposure duration (ED) is $(t_2 - t_1)$, Magnitude of exposure is denoted as E in $\text{mg} / \text{m}^3 \cdot \text{d}$.

Effects of all PAH species have been estimated using the factor TEF, which is used to convert the effect of PAH species to the equivalent values measured based on BaP (Yang et al. 2007).

$$BaP_{eqi} = C_i \times TEF_i \quad (4)$$

Total equivalent toxic quantity (TEQ) is indicated by the total BaP_{eqi} for an individual in 1 day (Chen and Liao 2006). It can be expressed as follows:

$$TEQ = \left[\left(\sum_{i=1}^n C_i \times TEF_i \right) \times AF_i \right] \times IR \times ED \quad (5)$$

Here, TEQ is calculated as PAH accumulation from ambient air through 1 day, ng BaP_{eq} .

Inhalation cancer risk for habitants

ICR can be estimated using total equivalent factor (TEF) by following the three steps:

- (1) ICR can be estimated from unit of pollutants inhaled (Lau et al. 2003). Equation 8 is showing the formula for cancer risk assessment:

$$R_i = C_i \times IUR_i \tag{6}$$

R_i is estimated individual life time cancer risk from pollutant i , C_i is the concentration of hazards that can be denoted by inhalation cancer risk of unit of pollutants i ($\mu\text{g}/\text{m}^3$)⁻¹ if the $IUR_i = 2 \times 10^{-6} \mu\text{g}/\text{m}^3$, (USEPA 2005a, b, 1996).

- (2) Life time ICR due to PM₁₀- and PM_{2.5}-bound PAHs is the summation of the cancer risk from individual pollutants (Wu et al. 2011; Liao and Chiang 2006). To estimate the number of cancer cases from the exposure in a city, the total cancer risk can be estimated by multiplying to the total population of the city.

$$\text{ICR} = \sum_{i=1}^n EC_i \times IUR_{\text{BaP}} \tag{7}$$

EC_i is the exposure concentration of chemical in ambient air ($\mu\text{g}/\text{m}^3$). ICR is the population that is affected by cancer risk per 10⁶ people.

- (3) TEF value is made a link with PAH concentration as BaP_{eq}. C_i equivalent concentration can be also expressed as BaP. Then, the term ICR can be defined as follows:

$$\text{ICR} = \left(\sum_{i=1}^n C_i \times \text{TEF}_i \right) \times IUR_{\text{BaP}} \tag{8}$$

In this case, IUR_{BaP} is a slope factor of inhalation risk as BaP (USEPA 2005a, b). ICR can be assessed using a linear model of extrapolation of risk in the low dose region (USEPA 1986, 2000). The slope factor IUR_{BaP} is used to link the linearity between ambient concentration and risk. The value of IUR_{BaP} is $1.1 \times 10^{-3} (\mu\text{g}/\text{m}^3)^{-1}$ is recommended by the California Environmental Protection Agency (CEPA). Relative potency of individual PAHs compared with B[a]P toxic equivalence factor TEFs (Larsen and Larsen 1998).

Air quality health indexing

Determination of AQI and AQHI are the most important to inform the air quality status to the scientific community, government officials, policy makers, and in particular to the general public in a simple and straightforward manner.

AQI may be defined as a single number on reporting the air quality with respect to its effects on the human health. In the most elaborate form, it combines many pollutants in some mathematical expression to arrive at a single number for air quality (CPCB). This approach for the evaluation of air quality is entirely based on National Ambient Air Quality Standards (NAAQS 2009).

In this method, equal importance is given to the pollutants using observed and standards values and accordingly the quality ratings for each pollutants were calculated. The geometric mean of the quality rating gives the AQHI. The existing concentrations of pollutants were compared with ambient air quality standard (with the standard being assumed as reference base line for each pollutant), and accordingly, the quality rating for a particular pollutant was derived as shown below.

$$Q_i = 10 (C_i/S_i) \tag{9}$$

where Q_i = Quality rating for i th pollutant, C_i = Concentration of i th pollutant, S_i = Air quality standard for i th pollutant (NAAQS 2009).

$$\text{AQHI} = (Q_1 \times Q_2 \times \dots \times Q_n)^{1/n} \tag{10}$$

AQHI has been calculated in 0–10+ scale.

where n = number of pollutants considered.

The categorization has been assigned as per guidelines provided by central pollution control board of India. In this study, only PM₁₀ has been considered.

Air quality indexing

AQI has been estimated by using daily average concentration of air pollutants and methodology used by CPCB final report of AQI 2015. The breakpoints aggregation of sub-indices and sub-indices of each pollutant are made according to the Indian National Ambient Air Quality Standard. In order to assess the status of air quality and its effects on human health, the range of index values, applicable to Indian cities, has been taken from earlier study by Nagendra et al. (2007), which reflects the different ranges as ‘Good (0–100)’, ‘Moderate (101–200)’, ‘Poor (201–300)’, ‘Very Poor (301–400)’ and ‘Severe (401–500)’, where the values of PM₁₀ are 0–100, 101–150, 151–350, 351–420 and >420 $\mu\text{g}/\text{m}^3$, respectively.

The AQI formula (USEPA 1999) for four-criteria pollutants RSPM is given as follows:

$$I_p = \left[\frac{(I_{Hi} - I_{Lo})}{(BP_{Hi} - BP_{Lo})} \right] (C_p - BP_{Lo}) + I_{Lo} \quad (11)$$

where

I_p	AQI of the pollutant 'P'
C_p	actual ambient concentration of pollutant 'P'
BP_{Hi}	the brake point given in TS1 that is greater than or equal to C_p
BP_{Lo}	the breakpoint given in TS1 that is less than or equal to C_p
I_{Hi}	the sub-index value corresponding value to BP_{Hi}
I_{Lo}	the sub-index value corresponding to BP_{Lo}

Result and discussion

Mass concentration of PAHs in $PM_{2.5}$ and PM_{10}

PM_{10} - and $PM_{2.5}$ -bound 15 PAH concentration at six monitoring stations of study area are listed in Tables 1 and 2, respectively.

Concentration levels of PAHs in particulates ($PM_{2.5}$ and PM_{10}) are found to be in the range of 0.01–103.5 ng/m^3 to 0.015–124.4 ng/m^3 , respectively, throughout the study area. Concentration of PM_{10} - and $PM_{2.5}$ -bound PAHs like Phe, Flu, Pyr, BaA, Chr, BkF, BaP, Ind, DBA and BghiP dominating almost at all monitoring stations, except the reference station (ISM). Very significant amount of PAHs were observed at BM, SS and LT in both PM_{10} and $PM_{2.5}$ category. Among all the monitoring stations, maximum concentration levels of particulate-bound PAHs (400–482 ng/m^3) were recorded at LT followed by BM (262–315 ng/m^3) and SS (129–136 ng/m^3) for $PM_{2.5}$ and PM_{10} , respectively. Very less amount of PM_{10} - and $PM_{2.5}$ -bound PAHs were estimated (9.7 and 11.31 ng/m^3) at ISM, Dhanbad (background location). These PAHs contributed 97.18 to 99.7% in all monitoring stations in both $PM_{2.5}$ and PM_{10} . These particulate-bound PAHs can be considered as signature element for the pollution sources. The presence of Flu and Pyr was reported at thermal power stations and cement plants where coal was used as a fuel source, whereas iron smelting operations are reported by the presence of Chr and BghiP. BghiP presence also indicates gasoline vehicle emission (Kong et al. 2010). It is also observed that higher concentration levels of Ind and BghiP would be indicators of vehicular emission, which is the dominating source of particulate-bound PAHs (Ravindra et al. 2008; Fang et al. 2006; Bourotte et al. 2005; Sienna et al. 2005). Gasoline and diesel vehicle fuel burning was identified by the presence of

BghiP, Ind, BkF, BaA and Chr (Chen et al. 2013). Emission of PAHs is directly related from coal combustion, coal processing industries and vehicle emissions in this critically polluted coalfield area. Concentration level of PAHs in PM_{10} and $PM_{2.5}$ has been shown in Tables 1 and 2. Peak height concentration level was recorded at the monitoring station LT followed by BM, SS and HS. PAHs concentration at JCF/Dhanbad City was recorded higher than the recent study carried out at Delhi (Khillare et al. 2012). Very few literatures are available on PAHs study in coal mining and mine fire areas. A critical study on PM_{10} - and $PM_{2.5}$ -bound PAHs and its health risk studies have been carried out at five cities of China (Wu et al. 2014). This study reveals that Dhanbad City which is known as a “Coal capital of India” and JCF as “largest and most productive coal fields” is facing more threats from natural source of air pollutants like mine fire and higher concentration levels of particulate-bound PAHs.

Diversity in different function zones of PAHs in $PM_{2.5}$ and PM_{10}

Distribution of PAHs in different functional zone

Monitoring stations were located at coal mining area, mine fire area, Dhanbad City and dense traffic area. It was observed that PM_{10} and $PM_{2.5}$ both exhibit the following trend as LT (mining and mine fire area) > BM (heavy traffic with residential and market site) > SS (coal mining and transportation) > HS (residential and traffic site) > DB (mining and thermal power station) > ISM (sensitive and background site). Pollutant concentration at different sites with respect to PAH concentration was compared with the background site (ISM). From this study, BM/ISM, SS/ISM, HS/ISM, DB/ISM and LT/ISM were shown in Fig. 2. It could be found that LT/ISM exhibit the higher value for all PM_{10} - and $PM_{2.5}$ -bound PAH species (in the range of 2.7–914 for $PM_{2.5}$ and 3–679.7 for PM_{10} , bound PAHs) including the PAH concentration in the atmosphere due to the presence of opencast coal mining and mine fire activities. In case of urban site, city junctions BM/ISM and HS/ISM ratio range in 1.5–310 and 1.5–320 for $PM_{2.5}$ and between 1.75–281.1 and 3.08–300 for PM_{10} as calculated in Fig. (2a, b).

Wu et al. (2014) pointed that city activities nearby coal mining complex may contribute significant amount of PAHs in the atmosphere. Source of PAHs include wood preservation, rubber tire manufacturing, creosote, thermal power station, etc., which contributed a considerable amount of PAHs in the rest of the two study areas. SS/ISM and DB/ISM ratio ranges between 1.77–227.33 and 3.75–247 for $PM_{2.5}$. This ratio has been noticed 2.33–233.3 and 3.08–300 for PM_{10} , respectively. In this case, the ratio of DB/ISM of PAHs is the second largest at DB, which may be due to wood, barren land waste burning and thermal power station (Chandrapura) that are very close to this station. Stack emission from thermal

Table 1 Concentration of PAHs in PM₁₀ at different location

Location PAHs	BM Concentration in ng/m ³	SS Concentration in ng/m ³	HS Concentration in ng/m ³	DB Concentration in ng/m ³	LT Concentration in ng/m ³	ISM Concentration in ng/m ³
NaP	0.4 ± 0.1	0.28 ± 0.1	0.18 ± 0.1	0.97 ± 0.4	0.3 ± 0.1	0.10 ± 0.1
Ac	0.2 ± 0.05	0.09 ± 0.01	0.14 ± 0.02	0.13 ± 0.08	0.9 ± 0.3	0.01 ± 0.1
Ap	0.04 ± 0.02	0.05 ± 0.01	0.08 ± 0.02	0.13 ± 0.08	0.08 ± 0.02	0.02 ± 0.1
Fl	0.14 ± 0.04	0.19 ± 0.05	0.15 ± 0.03	0.8 ± 0.2	0.28 ± 0.1	0.08 ± 0.1
Phe	2.2 ± 0.8	1.3 ± 0.4	1.34 ± 0.64	3 ± 0.9	4.6 ± 1.1	0.01 ± 0.1
Ant	0.32 ± 0.1	0.18 ± 0.06	0.22 ± 0.05	0.3 ± 0.1	0.93 ± 0.4	0.02 ± 0.1
Flu	3.8 ± 0.7	2.13 ± 0.6	3.78 ± 1.5	2.77 ± 0.8	10.97 ± 3.1	0.90 ± 0.1
Pyr	4.4 ± 0.8	2.32 ± 0.5	3.9 ± 1.2	2.9 ± 0.8	11.3 ± 2.1	0.18 ± 0.1
BaA	25.3 ± 2.1	21 ± 4.1	13.19 ± 2.1	5.3 ± 1.1	60.9 ± 4.1	0.09 ± 0.1
Chr	79.4 ± 8.2	38.8 ± 5.1	35.74 ± 8.1	15.19 ± 3.1	124.4 ± 12.5	0.60 ± 0.1
BkF	31.7 ± 5.1	21.36 ± 4.5	16.5 ± 3.1	11.2 ± 1.1	50.2 ± 8.2	1.10 ± 0.1
BaP	24.3 ± 6.5	22.78 ± 5.1	17 ± 4.1	11.7 ± 1.1	55.5 ± 6.11	1.80 ± 0.1
Ind	26.11 ± 6.3	7.64 ± 3.1	10.46 ± 2.1	4.68 ± 1.3	31.2 ± 4.3	1.00 ± 0.1
DBA	58.55 ± 9.5	19.5 ± 3.7	20.74 ± 3.1	12.87 ± 2.1	65.288.1	2.90 ± 0.1
BghiP	58.01 ± 8.4	19.5 ± 3.9	20.74 ± 3.1	12.87 ± 2.1	65.28 ± 0.1	2.50 ± 0.1
LMW	7.1 ± 1.2	4.22 ± 1.4	5.89 ± 1.12	8.1 ± 2.1	18.06 ± 3.1	1.14 ± 0.4
MMW	109.1 ± 12.1	62.12 ± 6.03	52.83 ± 4.1	23.39 ± 3.3	196.6 ± 14.1	0.87 ± 0.2
HMW	198.67 ± 12.1	66.34 ± 5.1	85.44 ± 5.1	53.32 ± 4.2	267.46 ± 15.5	9.3 ± 1.3
COMPAHs	253.02 ± 14.1	135.53 ± 8.1	121.31 ± 8.1	66.61 ± 8.2	409.75 ± 15.2	8.17 ± 1.5
C-PAHs	165.96 ± 12.1	92.28 ± 7.1	77.89 ± 5.1	45.75 ± 5.4	263.08 ± 12.5	6.89 ± 1.2
Total PAHs	314.87 ± 15.1	135.76 ± 9.1	144.16 ± 11.2	84.81 ± 6.3	482.12 ± 15.4	11.31 ± 1.3
	Concentration in µg/m ³					
PM ₁₀	441 ± 45	480 ± 34	165 ± 21	530 ± 48	354 ± 41	76 ± 17

Number of sample (*n*) is 104 for each site

power station may cause PAH pollution at DB location (Ravindra et al. 2008; Kong et al. 2010). Mass concentrations of PAHs from industrial/ commercial and residential combustion were 5.6 and 3.7 times for PM_{2.5} and 5.7 and 3.8 times for PM₁₀. Higher levels of PAH emission were associated with coal fire, coal based power station, coal based power station, factories and using coal as a fuel. Coal burning/coal mine fire emission was indicated by higher concentration of Flu, Pyr and Phe (Liu et al. 2007). The present study observed that the high concentration levels of Phe, Flu and Pyr were recorded at LT and three to four times greater than that of other locations. Higher factor loading of BaP, Chr and BbF is a good factor for wood and coal combustion from stationary emission sources (Sevimoglua and Rogge 2016). For LT, remarkable elevations were recorded in the concentration level of Chr and BaP among the PAHs which were three to eight times than SS, HS and DB. Monitoring station BM also recorded very considerable levels for Bap, Chr and BbF. BM is surrounded by market place, small scale industries and road junctions. Higher concentration level of Chr, BaP and BbF may be the indicators of vehicular emission, paved/unpaved road emission sources. High concentration level of BaP at LT indicates the burning of

wood and mine fire emissions on the earth’s surface (Sevimoglua and Rogge 2016).

Coefficient of divergence analysis

Mass concentration of PAH components, *j* and *k* (as per Eq. 1) of PM_{2.5}- and PM₁₀-bound PAHs are shown in Fig. 3 as scatter diagram plots. The value of CD_{jk} is higher than 0.55, which indicates significant difference in PAH composition. Higher CD_{jk} values 0.239 and 0.238 were observed for LT and SS, respectively. Lower concentrations of CD_{jk} were recorded 0.082, 0.088 and 0.97 for BM, HS and DB, respectively. The background monitoring station (ISM) showed lowest CD_{jk} value.

Source identification

Distribution in different rings

PAH distribution pattern for each fraction was classified based on their number of rings. As per aromatic ring distribution-2-ring NaP; 3-ring Acy, Ace, Fl, Phe and Ant; 4-ring PAHs are

Table 2 Concentration of PAHs in PM_{2.5} at different location

Location PAHs	BM Concentration in ng/m ³	SS Concentration in ng/m ³	HS Concentration in ng/m ³	DB Concentration in n/m ³	LT Concentration in ng/m ³	ISM Concentration in ng/m ³
NaP	0.3 ± 0.1	0.23 ± 0.1	0.15 ± 0.1	0.8 ± 0.1	0.27 ± 0.1	0.10 ± 0.05
Ac	0.19 ± 0.05	0.07 ± 0.03	0.12 ± 0.02	0.12 ± 0.08	0.07 ± 0.01	0.01 ± 0.01
Ap	0.03 ± 0.01	0.04 ± 0.01	0.07 ± 0.02	0.12 ± 0.07	0.06 ± 0.01	0.02 ± 0.01
Fl	0.12 ± 0.06	0.15 ± 0.05	0.12 ± 0.03	0.69 ± 0.12	0.23 ± 0.1	0.08 ± 0.1
Phe	1.8 ± 0.4	1.08 ± 0.4	1.12 ± 0.63	2.47 ± 0.8	3.8 ± 1.1	0.01 ± 0.01
Ant	0.27 ± 0.1	0.15 ± 0.06	0.19 ± 0.05	0.27 ± 0.1	0.77 ± 0.3	0.02 ± 0.01
Flu	3.1 ± 1.1	1.78 ± 0.7	3.2 ± 1.5	2.3 ± 0.8	9.14 ± 3.1	0.01 ± 0.01
Pyr	3.7 ± 1.2	1.9 ± 0.6	3.3 ± 1.2	2.5 ± 0.9	9.4 ± 3.4	0.15 ± 0.05
BaA	21.06 ± 3.1	17.52 ± 3.1	10.99 ± 2.4	4.4 ± 1.1	50.7 ± 12.6	0.08 ± 0.02
Chr	66.16 ± 12.1	32.33 ± 4.7	29.78 ± 4.3	12.65 ± 2.1	103.55 ± 14.6	0.54 ± 0.1
BkF	26.39 ± 5.1	15.7 ± 2.3	13.77 ± 1.4	9.4 ± 1.6	41.86 ± 5.6	1 ± 0.4
BaP	20.25 ± 5.2	18.98 ± 2.1	14.3 ± 1.5	9.7 ± 1.8	46.3 ± 6.2	1.73 ± 1.1
Ind	21.76 ± 4.1	6.37 ± 1.1	8.7 ± 1.3	3.9 ± 0.8	26 ± 5.1	0.96 ± 0.3
DBA	48.38 ± 6.3	16.24 ± 2.8	17.26 ± 4.1	10.7 ± 3.1	54 ± 12.1	2.85 ± 0.8
BghiP	48.38 ± 6.5	16.24 ± 2.5	17.2 ± 04.5	11 ± 3.1	54 ± 12.1	2.45 ± 0.7
LMW	5.81 ± 1.3	3.5 ± 1.5	4.97 ± 1.4	6.77 ± 1.6	14.34 ± 2.3	0.25 ± 0.1
MMW	5.81 ± 1.7	3.5 ± 1.5	4.97 ± 1.6	6.77 ± 1.7	14.34 ± 2.1	0.25 ± 0.1
HMW	165.16 ± 12.5	73.53 ± 7.1	71.23 ± 6.1	44.7 ± 5.1	222.16 ± 14.1	8.919 ± 2.1
COMPAHs	210.8 ± 14.3	110.82 ± 11.5	101.24 ± 8.1	55.85 ± 7.3	340.95 ± 18.2	6.846 ± 2.3
C-PAHs	137.84 ± 12.4	74.81 ± 8.2	65.02 ± 4.1	38.1 ± 4.6	218.86 ± 12.4	6.546 ± 2.1
Total PAHs	261.89 ± 14.6	128.78 ± 12.2	120.27 ± 11.4	71.02 ± 5.2	400.15 ± 19.2	9.936 ± 3.2
	Concentration in µg/m ³					
PM _{2.5}	191 ± 24	164 ± 29	110 ± 12	192 ± 25	118 ± 14	47 ± 06

Number of sample (*n*) is 104 for each site

Flu, Pyr, BaA and Chr; 5-ring including BkF and BaP and 6-ring PAHs are Ind, DBA and BghiP. Lower molecular weight (LMW) contains 2- and 3-ring PAHs, middle molecular weight (MMW) contain 4-ring PAHs and higher molecular weight PAHs (HMW) contains 5- and 6-ring PAHs (Kong et al. 2013). Combustion-derived PAHs (COMPAHs) include Flu, Pyr, Chr, BbF, BkF, BaA, BeP, BaP, IND and BghiP. Carcinogenic PAHs (C-PAHs) include BaA, BbF, BkF, BaP, Ind and DBA. Detailed study of PAHs reflected that 4-, 5- and 6-ring PAHs are dominated in PM₁₀ and PM_{2.5} (RSPM). Concentration levels of PM₁₀-bound PAHs (Fig. 3) were found in the range between 7.08–172 ng/m³, 2.73–88.16 ng/m³ and 66.76–134 ng/m³. PM_{2.5}-bound PAH concentration ranges were found in between 1.77–207.5 ng/m³, 2.90–211.6 ng/m³, 6.4–161.78 ng/m³, total 93.7–98.97% and 93.72–99.36% PAH, respectively. Moreover, it was also observed that concentration levels of PM₁₀- and PM_{2.5}-bound C-PAHs were found maximum at LT (263.08 and 218.86 ng/m³) followed by BM (165.96 and 137.85 ng/m³), SS (92.28 and 74.81 ng/m³), HS (77.89 and 65.02 ng/m³) and DB (45.75 and 38.1 ng/m³) and minimum at ISM (6.89 and 6.5 ng/m³), respectively (Tables 1 and 2). It was also observed that

concentration levels of C-PAHs in PM₁₀ were found 43, 28, 13, 12 and 8 times higher at monitoring stations LT, BM, HS, SS and DB, respectively, from the background station (ISM). On the other hand, concentration levels of PM₁₀-bound total PAHs were found to be 38, 24, 13, 11 and 7 times higher at LT, BM, SS, HS and DB, respectively, from the background station (ISM). For PM_{2.5}, it was also observed that concentration levels of C-PAHs were found 34, 21, 11, 10 and 6 times higher at monitoring stations LT, BM, SS, HS and DB, respectively, from the background station (ISM). In the case of PM_{2.5}-bound, total PAHs were found to be 40, 26, 13, 12 and 7 times higher at LT, BM, SS, HS and DB, respectively, from the background station (ISM). Results indicate that LT and BM can be more in inhalation cancer risk probable area among the all monitoring stations.

At LT, concentration of particulate-bound PAHs was respectively higher among the all monitoring stations in the study area. At first, 3 rings (LMW) of PAHs were found in less concentration, 4 rings (MMW) contributed 55–57% of total PAHs concentration in PM_{2.5} and PM₁₀, respectively. 3-ring and 4-ring PAHs combined recorded 50% of their total PAH concentrations. The presence of 3- and 4-ring PAH in the

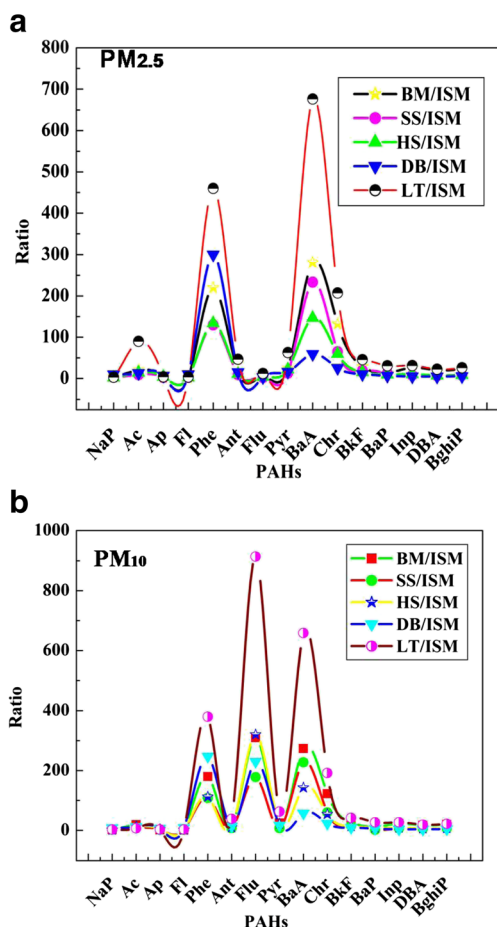
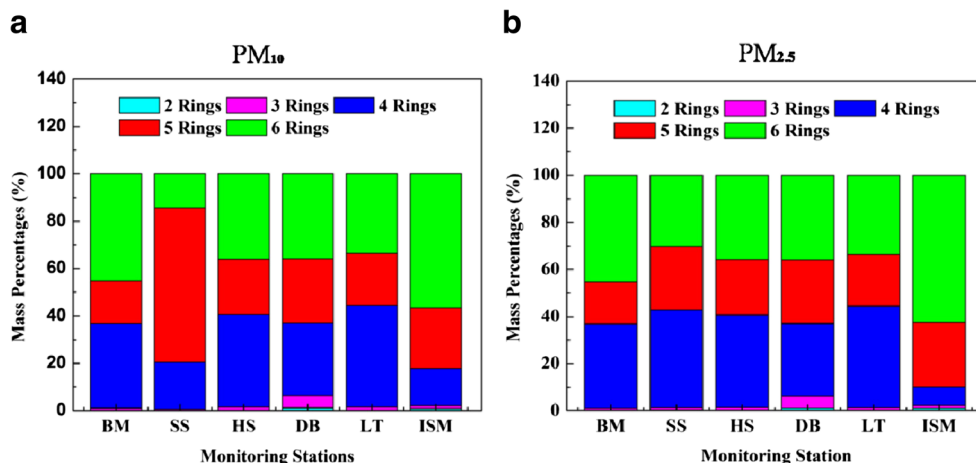


Fig. 2 a, b Comparison of PM₁₀- and PM_{2.5}-bound PAH species in other different function sites with respect to background (ISM, Dhanbad)

particle indicates the source of coal combustion (Ravindra et al. 2006). Fifty percent of 3- to 4-ring PAHs was observed at LT which indicated coal burning as a source. Monitoring stations BM, DB, HS and SS were affected by 5- and 6-ring (HMW) PAHs. The presence of 5–6 ring PAHs are indicator of gasoline-based vehicular emissions (Ravindra et al. 2006). LT is an active mine fire site (Roy et al. 2015) and population

Fig. 3 Ring distribution of PAHs in PM₁₀ (a) and PM_{2.5} (b) at the five sites as mass percentage



density of the area is very less. Negligible human contribution in PAHs concentration level was recorded at LT. Ring distribution of PAHs at background level, 80–90% of 5- and 6-ring (HMW) PAHs were recorded. Mainly vehicular emission can be the net source of pollution level at ISM (background) for PM₁₀ and PM_{2.5}, respectively. Station SS was showing pollution composition of 60–80% due to 5–6 rings for PM_{2.5} and PM₁₀, respectively. Vehicular emissions could be one of the main contributors of particulate-bound PAH pollution levels in this area.

Combustion-derived PAHs

Combustion sources of PAHs can be identified by combustion-derived PAHs (COMPAHs) (Kong et al. 2010). COMPAH concentration covered 80–86 and 78–99.8% for PAHs in PM_{2.5} and PM₁₀ except background site. PM_{2.5}- and PM₁₀-bound PAH concentration levels at background site were 69 and 72%, respectively. Highest mass percentage of COMPAHs were recorded at SS with 86 and 99.8% of PAHs in PM_{2.5} and PM₁₀, respectively. Second highest concentration level of COMPAHs was recorded at LT, and other locations were with 85% in both PM_{2.5} and PM₁₀, respectively. The COMPAHs/∑PAH ratio for catalyst-equipped (0.5) and non-catalyst (0.4) for automobiles and 0.3 heavy-duty diesel trucks was determined by Rogge et al. (1993). On the other hand, the values that range from 0.62 to 0.73 are recorded for Cement Plant and 0.79% for Sinter and Coke Production Plant was obtained (Kong et al. 2013). The present study reflected that except ISM (background station), COMPAHs/∑PAH ratio ranges from 79.5 to 99.8%. So, it can be concluded that coal combustion, mine fire and its associated activities are the dominating source of particle-bound PAHs at JCF, Dhanbad City during the study period (2012–2013). Pre-harvested burning of foliage and coal combustion can be the sources of PAHs in this area (Sevimoglua and Rogge 2016; Zhang et al. 2017). Moreover, diesel, gasoline and fueled vehicles

along with natural gas combustions can be another source of PAHs (Hazarika et al. 2017). Vehicular source contribution may not be ignored at some places in the study area. Comparison of average concentration PAHs in PM_{10} and $PM_{2.5}$ at different stations is shown in Fig. 4.

Diagnostic ratio analysis

Identification of possible sources can be done by diagnostic ratio approaches (Kong et al. 2010). Flu/(Flu + Pyr) ratio more than 0.5 indicates biomass or coal combustion and less than 0.4 for petroleum source (Han et al. 2011). Figure 5 shows that for $PM_{2.5}$, Flu/(Flu + Pyr) are 0.46, 0.49, 0.49, 0.48 and 0.49 for BM, SS, HS, DB and LT, respectively. Flu/(Flu + Pyr) ratio for BM, SS, HS, DB and LT are 0.46, 0.47, 0.49, 0.49 and 0.49, respectively, for PM_{10} . The value of BaA/(BaA + Chr) is the 4-ring PAH indicator. Ratio of BaA/(BaA + Chr) higher than 0.35 indicates the presence of pyrolytic sources and lower value than 0.2 signals towards the petrogenic emission sources (Hu and Jiang 2013). BM which is a residential and high traffic dense site, the average BaA/(BaA + Chr) value is less than 0.35 and higher than 0.2, indicating either presence of pyrolytic sources or petrogenic sources. At stations DB and HS, ratio of BaA/(BaA + Chr) lies in between 0.25 and 0.29, which indicates the presence of either petrogenic or pyrolytic sources. The BaA/(BaA + Chr) value is very close to 0.354 SS and LT. Above result indicates the maximum probability of presence of pyrolytic sources. Ant/(Ant+Phe) less than 0.1 indicates the presence of petroleum combustion sources in the study area (Han et al. 2011).

The ratio of Ant/(Ant + Phe) is 0.09 and 0.08 for $PM_{2.5}$ and PM_{10} , respectively. From these findings, it can be concluded that petroleum source dominates in this area. Other locations BM, SS, HS and LT have Ant/(Ant + Phe) and have values higher than 0.1 and the highest ratio is at LT (0.18). Combustion source was dominated at LT and other locations. Diagnostic ratio analysis output has been shown in Fig. 5.

Above analysed diagnostic ratio and different case studies conclude that monitoring station LT and SS are affected by the combustion and pyrolytic source emission. HS, DB and BM were affected by either petroleum or pyrolytic source emission. Both techniques are very informative and still show some particular confirmative matter needed to be employed for actual identification for sources in particulate-bound PAHs in the study area. Principal component analysis with factor analysis is the most acceptable technique for the pollution source identification.

Principal component analysis

Principal component analysis has been used to identify the actual source of particulate-bound PAHs among the multiple

polluting sources. SPSS 21.0 software has been used for this study and the outputs from SPSS of standardized PAHs concentrations have been presented in Fig. 6.

PCA is performed on $PM_{2.5}$ and PM_{10} datasets for all the five sites together (15 PM_{10} and 15 $PM_{2.5}$ samples) with satisfying minimum criteria as $V = 15$, $n = 104$ (Eq. 2). Factor interpretation has been carried out to consider the variables with higher than 0.54 (due to close plotting) in case of $PM_{2.5}$; three factors explaining up to 97.79 % of the data variance were obtained. Phe, Ant, Flu, Pyr, BaA, Chr, BkF, BaP, DBA, Ind and BghiP plunge on factor 1 with 67.37 % variance. BaA and Pyr are considered for coal combustion (Khalili et al. 1995; Ravindra et al. 2006; Wang et al. 2006; Tian et al. 2009); on the other side, Ind is taken as factor for gasoline vehicle (Khalili et al. 1995; Ravindra et al. 2006). Flu, Pyr and BkF are signature elements for PAHs for diesel vehicles (Bourotte et al. 2005; Ravindra et al. 2006; Hong et al. 2007). Phe, Flu, Pyr (Khalili et al. 1995; Bourotte et al. 2005; Ravindra et al. 2006; Wang et al. 2006) and BaP (Bourotte et al. 2005; Hong et al. 2007) are signature elements for wood combustion. Higher loading on NaP, Acy and Fl is obtained for factor 2 with 21.96 % variance. These are the theoretical PAHs associated with petrogenic sources. Industrial area is affected by the active mine fire (coal burning) and city area was affected by volatile petroleum spilling. NaP and Acy are the significant indicators for combustion source (Park et al. 2002) and Fl reflects diesel vehicle emission source (Yang et al. 1998). Ace obtained as third factor 08.46 % of the variance. Ace is the indicator of cement as a source. On site construction activities may be responsible for cement emission. From the above discussion, it is hard to identify the particulates as a single source of study area. Multiple sources emissions were affected throughout the study area at different levels.

For PM_{10} , two components are identified as a factor with 93.88% of data variance. Ace, Ant, Flu, Pyr, BaA, Chr, BkF, Bap, DBA, Imp and DghiP are accounted for 72.56% of the total data variance. The first factor is showing the same PAHs for PM_{10} and $PM_{2.5}$ only where Phe was absent in the list of PM_{10} -bound PAHs. It can be assumed that the sources of PM_{10} - and $PM_{2.5}$ -bound PAHs were the same during the study period at all monitoring stations. NaP, Fl and Acy are obtained as a factor 2 with 21.32% data variance. Factor 2 is precisely second with the second factor of $PM_{2.5}$ monitoring station that was affected by the same sources as $PM_{2.5}$ for the PM_{10} also.

Health risk and inhalation cancer risk assessment

Health risk assessment of PAHs has been carried out with respect to BaP equivalent concentrations (BaP_{eq}). TEF of different PAHs has been calculated and estimated as a BaP_{eq} equivalent (Nelsen et al. 1996; Chang et al. 2006; Wang et al. 2006; Hong et al. 2007; Akyüz and Çabuk 2008). The total carcinogenic potential of

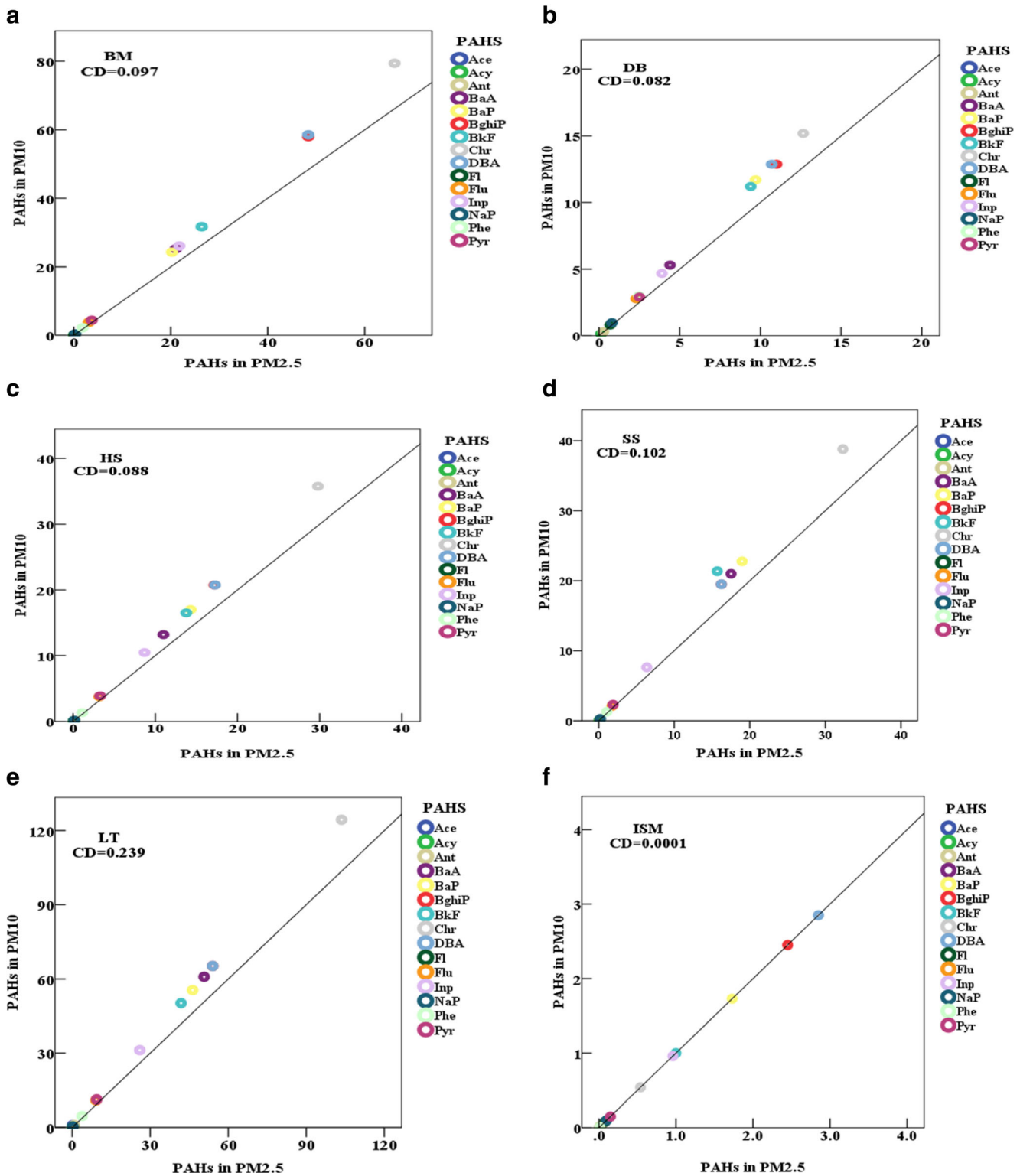


Fig. 4 a–f Comparison of average concentration PAHs in PM₁₀ and PM_{2.5} at different stations

PAHs is the summation of carcinogenic potency of individual PAHs as a BaP_{eq}. The total BaP_{eq} concentration of PM₁₀ is found to be 89.4, 49.7, 40.7, 24.9, 131.9 and 4.8 ng/m³. For BM, SS, HS, DB, LT and ISM, the value of PM₁₀ are found to be 76.7, 39.7, 35.4, 22.4, 113.8 and 4.8 ng/m³, respectively, as shown in Fig. 7.

From these results we concluded that people residing near-by mine fire and coal city are seriously affected by the particulate emission. On the other hand, health risk factor is slightly higher due to PM₁₀ with respect to PM_{2.5} among all the locations.

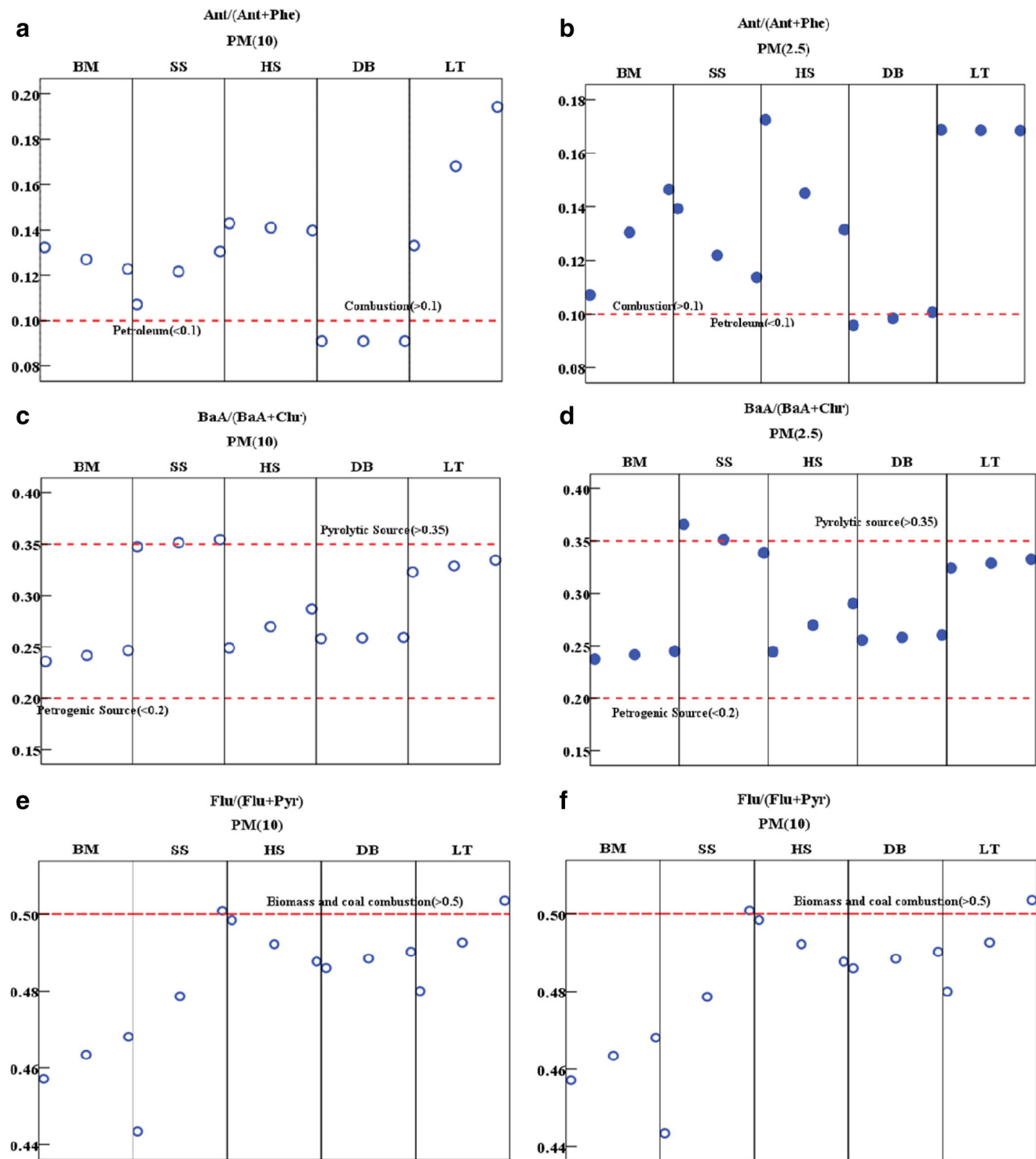


Fig. 5 a–f Diagnostic ratio for Flu/(Flu + Pyr), Ant/(Ant + Phe) and BaA/(BaA + Chr) of the five sites for PM₁₀ and PM_{2.5}

ICR has been calculated for carcinogenic PAHs as BaP_{eq} concentrations varied from 1.73 to 46.3 ng/m³ in this study area. Maximum concentration of BaP was found at monitoring station LT, followed by BM (20.25) and SS (18.98) PM_{2.5} at the study area. BaP concentration was in the range of 1.8 to 55.5 ng/m³ for PM₁₀. Highest concentration of BaP was

recorded at LT (55.5 ng/m³) followed by BM (24.3 ng/m³) and SS (22.78 ng/m³). The concentration of BaP was 56% higher for PM₁₀ and 46% for PM_{2.5}, respectively. As per National Ambient Air Quality Standard (2009), the optimum concentration of BaP is 1 ng/m³ (MoEF 2009). The seven carcinogenic PAHs were identified by USEPA at 2000, i.e.

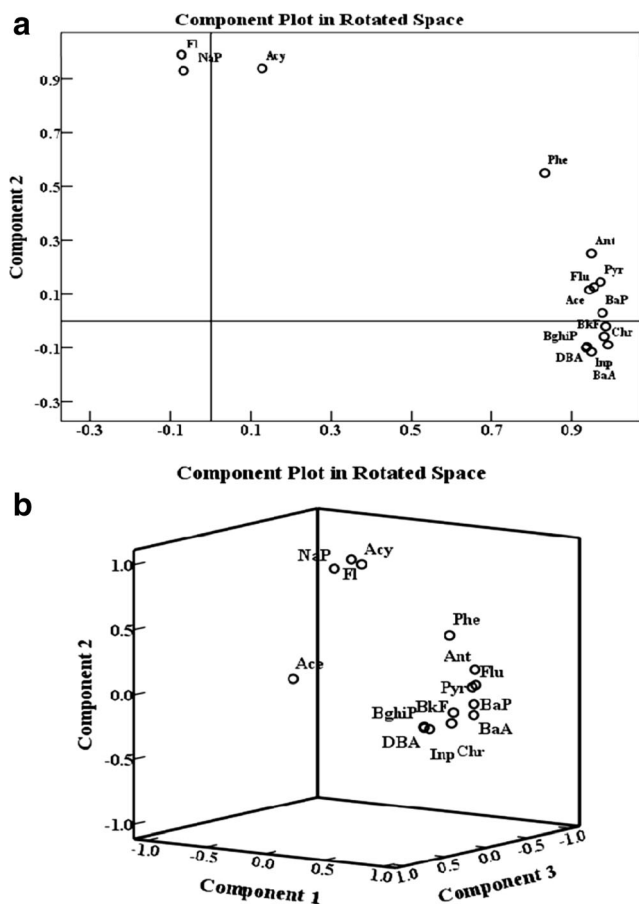
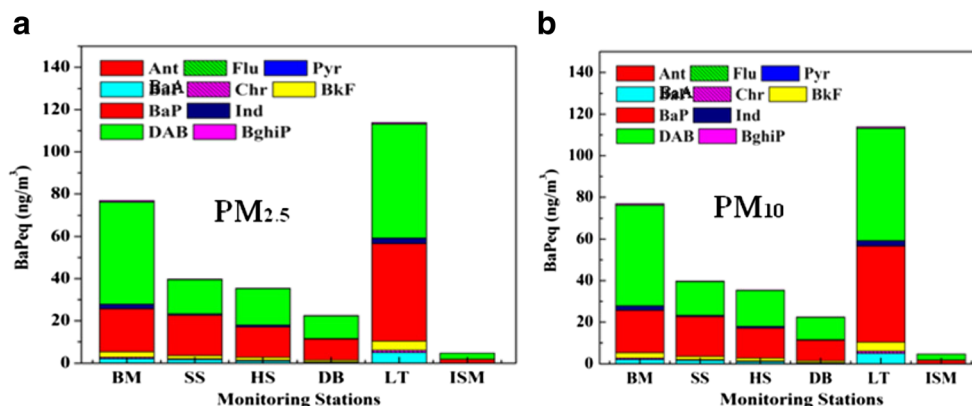


Fig. 6 a, b Principle component analysis for PAH profile of (A) PM₁₀ and (B) PM_{2.5} at the study area

BaP, Chry, Bbk, Bkf, BaP, DAB and IP: \sum_7 PAHs. Carcinogenic PAHs contributed 96, 96, 93, 87, 94 and 88% of \sum_7 PAHs in PM₁₀-bound PAHs at BM, SS, HS, DB, LT and ISM, respectively. PM_{2.5}-bound carcinogenic PAHs emitted 93 to 98% of the total concentration of PAHs at different stations. LT was affected by highest concentration of inhalation cancer creating PAHs in the study area and the concentration of PAHs is 376 and 452 ng/m³ for PM_{2.5} and PM₁₀, respectively. After Eqs. 6, 7 and 8 were taken into

Fig. 7 BaPeq concentrations of PAHs in PM_{2.5} (a) and PM₁₀ (b) at six locations in the study area



consideration at first carcinogenic PAHs, BaP was calculated and then it was multiplied by IUR_i factor. The factor IUR_i is 1.1×10^{-3} for BaP (CEPA 2004). Figure 8 shows output of ICR assessment of the study area.

ICR for PAHs were found to be 9.8×10^{-5} , 5.03×10^{-5} , 4.47×10^{-5} , 2.48×10^{-5} , 14.5×10^{-5} and 0.53×10^{-5} for the locations BM, SS, HS, DB, LT and ISM, respectively, for PM₁₀. For PM_{2.5}, these ICR values were recorded as 8.4×10^{-5} , 4.4×10^{-5} , 3.9×10^{-5} , 2.47×10^{-5} , 12.5×10^{-5} and 0.53×10^{-5} at BM, SS, HS, DB, LT and ISM, respectively.

ICR potential risk was regularly cited in between 10^{-6} to 10^{-4} , and high potential health risk has been indicated, when ICR is larger than 10^{-4} . The ICR value 10^{-6} generally represents zero risk value (Liao and Chiang 2006). Output of ICR analysis reflected that 10 times cancer tumours are found among the 10^{-6} population at BM. For other locations 5, 4, 3, 15 and 5 cancer tumour can be found at SS, HS, DB, LT and ISM for PM₁₀, respectively. For PM_{2.5}, the number of tumours can be found 8, 4, 4, 2, 12 and 5 for BM, SS, HS, DB, LT and ISM, respectively. As per census 2011, population of Dhanbad District was 2,684,487. The social ICR was calculated by multiplying individual ICR by the population of Dhanbad and it was found up to 999 cases for PM₁₀ and 864 cases for PM_{2.5} which may occur at Jharia/Dhanbad City due to lifetime inhalation exposure to PAHs. LT recorded maximum probability of tumours due to inhalation cancer (389–336) followed by BM (264–227) and HS (135–117) for PM₁₀ and PM_{2.5}, respectively.

The formula-based comprehensive assessment of AQI, AQHI and ICR tumours with respect to PM₁₀ at six monitoring stations are shown in Fig. 9. The highest AQI is calculated at BM (470) station, followed by DB (426) and SS (195), respectively. The QAI, AQHI and PM₁₀ concentrations are showing more or less the same trend at all monitoring stations of the study area. On the other hand, ICR tumour number trend is totally different from the trend followed by PM₁₀, AQI, and AQHI. Maximum AQHI is estimated at DB monitoring station (53), followed by BM (44) and LT (36), respectively. In case of ICR tumour values, the estimation showing

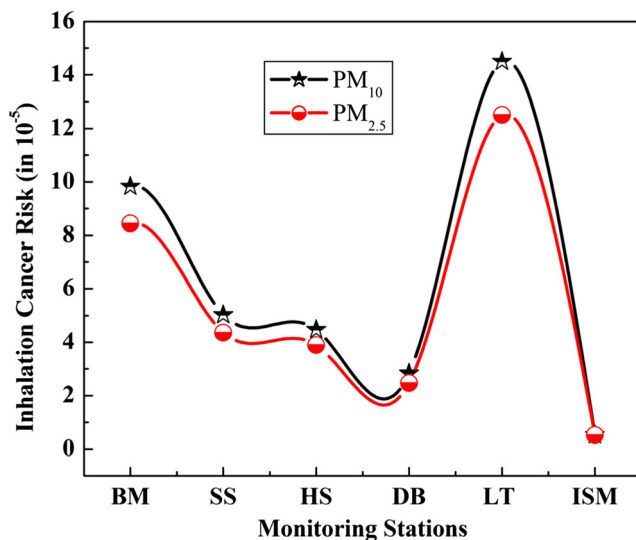


Fig. 8 Inhalation cancer risk at different locations of the study area as ICR in 10^{-5} scale

the maximum ICR tumours count at LT (145), followed by BM (98) and SS (50), respectively. A sharp peak of ICR tumour is obtained at monitoring station LT, because this station is severely affected by the mine fire. Monitoring station BM is identified as the second largest ICR tumour apparent location among the entire study area. Vehicular emission is the most probable carcinogenic source at this location. The results also show that ICR tumours are highly correlated with concentration of PM₁₀-bound carcinogenic PAHs not the concentration of PM₁₀.

The correlation between AQI, AQHI and ICR tumour numbers are shown in Table 3. The positive correlation is observed between AQI and AQHI with the correlation coefficient of 0.74. A negative correlation (0.26) is observed between AQI and ICR tumour numbers. There was no relation found between AQI and ICR tumour numbers. AQHI positively correlated with AQI by a correlation factor of 0.74 and ICR tumour is negatively correlative with the coefficient of

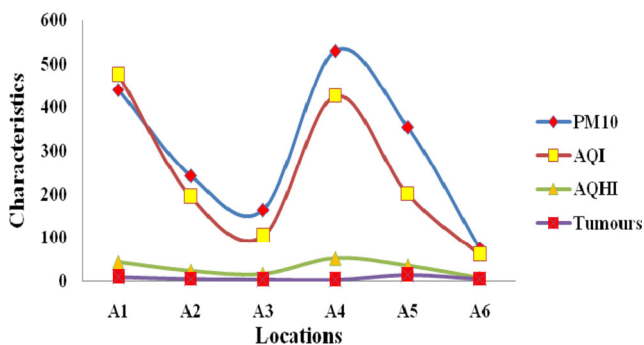


Fig. 9 Location wise plotting of PM₁₀, AQI, AQHI and ICR tumours. [PM₁₀ is showing the concentration value in micrograms per cubic meter; AQI is an indexing which shows the value ranging from 0 to 500 scale; AQHI is another indexing of air quality health which shows the value ranging from 0 to 10+ scale and tumours indicates number of lung cancer tumours risk (ICR in 10^{-5} scale)]

Table 3 Correlation coefficient among AQI, AQHI and ICR Tumours

Correlations				
Control variables		AQI	AQHI	IN_CNC_RISK
PM ₁₀	AQI	1	0.735	-0.261
	AQHI	0.735	1	-0.092
	IN_CNC_RISK	-0.261	-0.092	1

-0.092. The correlation study between AQI, AQHI and ICR tumour number indicates that AQI and AQHI directly correlated with each other. ICR tumours result is totally different from AQI and AQHI. The value of AQI and AQHI is dependent on specific pollutant concentration. On the other hand, ICR tumours depend on chemical species that are carcinogenic in nature. The in and around area of LT is affected by active mine fire which contributed with trace elements that are responsible for cancer to human health.

Conclusion

As JCF/Dhanbad City is critically polluted by coal mining complex with multiple polluting sources raising the pollution level continuously in this area. Concentration of PAHs was found in range of 0.01–103.5 to 0.01–124.4 ng/m³ for PM_{2.5} and PM₁₀, respectively. Maximum concentration levels of PAHs were recorded at LT (400.15–482.12 ng/m³) for PM₁₀ and PM_{2.5}, individually. Among the six locations, one is the background and other locations have been divided into city area, mining area and mine fire-affected area. Identification of the sources at different sections of the study area has been performed by using (1) combustion-derived PAHs identifications, (2) diagnostic ratio analysis and (3) principal component analysis.

Source identification process can conclude that all locations were affected by the combustion sources and especially LT was affected by the coal burning and wood ignition. On the other hand, construction activity and petroleum sources affected stations located at cities (BM, HS). SS and DB were affected by all types of sources. SS locations were facing pollution problem such as coal burning problem nearby Jogta mine fire. LT station is also affected by mine fire. The abovementioned results support the practical data. From health risk analysis, it is estimated that about 1000 and 889 cases of inhalation cancer can be found due to PM₁₀- and PM_{2.5}-bound PAHs. LT was recorded with maximum probability of tumours due to inhalation cancer (389–336), followed by BM (264–227) and HS (135–117) for PM₁₀ and PM_{2.5}, respectively.

AQI with AQHI parameters are considered for the estimation of acute health impacts in the study area due to the presence of PM₁₀, whereas ICR tumour study has been done for

calculating the number of lung cancer tumour cases. Then, the correlation study has been done in between AQI, AQHI and number of ICR tumours among all the study locations. The result reveals that the AQI and AQHI has positive correlation with each other, whereas ICR tumour shows independent relationship with respect to AQI and AQHI. This is only due to the fact that ICR tumour number depends on the concentration of the carcinogenic chemical species present in PM₁₀ in the study area.

This study concludes that Jharia/Dhanbad City is not only a critically polluted area but it is also an inhalation cancer prone area. LT which is very near to the active mine fire is more carcinogenic with respect to the other locations. It would also be concluded that mine fire activity not only causes particulates or gaseous pollutions but it also causes huge amounts of particulate-bound carcinogenic PAH emission.

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