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ORIGINAL PAPER

Polycyclic aromatic hydrocarbons in PM10 of a north-western city, India: distribution, sources, toxicity and health risk assessment

S. Kaur[1] · B. Kumar[2] · P. Chakraborty[3] · V. Kumar[4] · N. C. Kothiyal[5]

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Abstract

The aim of this study was to evaluate possible sources, toxicity and human health risk via inhalation of particulate matter (PM_{10}) bound polycyclic aromatic hydrocarbons (PAHs). During November 2013–January 2014, 54 PM_{10} samples were collected from a tropical city of north-west India and analysed for sixteen priority PAHs ($\sum_{16}$PAHs). The concentration of $\sum_{16}$PAHs ranged between 18 and 164 ng m^{-3} with an average of 96 ± 48 ng m^{-3}. The concentration of seven carcinogenic PAHs (C-PAHs) accounted for 22.68% of $\sum_{16}$PAHs. The carcinogenic potency of 16 PAHs as benzo(a)pyrene equivalent (BaP_{eq}) ranged between 2.49 and 11.37 ng m^{-3}. Composition profile and results of source apportionment indices suggested mixed pyrogenic sources. Back trajectory analysis revealed that the level of PAHs in ambient air (PM_{10}) at Amritsar might have been influenced by long-range atmospheric transport and various local emission sources. The BaP_{eq}-based PAH concentrations are used to estimate daily exposure level through inhalation pathways. The estimated inhalation cancer risk (CR) for human adults (8.5×10^{-9}–6.5×10^{-6}) and children (1.6×10^{-8}–1.2×10^{-5}) was within the stipulated acceptable limit. However, due to limitations in the exposure through inhalation, and lack of data on exposure through diet, total daily intake of PAHs and CR could not be estimated.

Keywords Polycyclic aromatic hydrocarbons · Urban air · Source apportionment · Cancer risk · National ambient air quality standard

Introduction

Polycyclic aromatic hydrocarbons (PAHs) in the environment are mainly associated with the anthropogenic activities of combustion processes (pyrogenic sources) including carbonaceous materials (biomass, coal and petroleum combustion) burning, coke & metal production, forest fires and volcanic eruptions (natural processes) and petroleum products (petrogenic sources) (ATSDR 1995). Among numerous PAHs, 16 compounds [naphthalene (Npt), acenaphthene (Ane), acenaphthylene (Any), fluorene (Fle), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz(a)anthracene (BaA), chrysene (Chr), benzo(b)-fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenzo(a,h)anthracene (DBA), benzo(ghi)perylene (BghiP) and indeno(1,2,3-cd)pyrene (IP)] are classified and listed as the priority pollutants by the United States Environmental Protection Authority (USEPA) (USEPA 2015) and European Community (EC) (EC 2001). Further, some priority PAHs have been included in the Convention on Long-range Transboundary Air Pollution Protocol on Persistent Organic Pollutants by the United Nations Economic Commission for Europe (UNECE 1998).

Based on the carcinogenicity, USEPA and the International Agency for Research on Cancer (IARC) (IARC 2010) classified 16 PAHs into different groups. BaP with sufficient evidence of carcinogenicity in human has been classified

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✉ P. Chakraborty
paromitc@srmist.edu.in

[1] Department of Botanical and Environmental Sciences, Guru Nanak Dev University, Amritsar, Punjab 143005, India

[2] Central Pollution Control Board, East Arjun Nagar, Delhi 110032, India

[3] SRM Research Institute and Department of Civil Engineering, SRM Institute of Science and Technology, Kattankulathur, Tamil Nadu 603203, India

[4] CT Group of Institutions, Jalandhar, Punjab 144001, India

[5] Dr B R Ambedkar National Institute of Technology, Jalandhar, Punjab 144001, India

as a human carcinogen (group I). DBA with limited evidence of carcinogenicity in human and sufficient evidence of carcinogenicity in experimental animals is classified as probable carcinogens (group 2A). BaA, Npt, Chr, BbF, BkF and IP are classified as possible carcinogens (group 2B), since inadequate evidence of carcinogenicity in human and limited evidence of carcinogenicity in experimental animals is available (ATSDR 1995; IARC 2010). Human exposure to PAHs occurs mainly through ingestion, dermal contact and inhalation of ambient air vapour or particles (ATSDR 1995; IARC 2010). Inhaled PAHs by humans can enter into the circulatory system through tracheobronchial and alveolar epithelium (Gurbani et al. 2013; Bostrom et al. 2002). Through oxidation–hydroxylation reactions, PAHs causes DNA adducts and mutations in various physiological systems including respiratory and urinary (Bosetti et al. 2007), digestive (Diggs et al. 2011) and reproductive system (Gaspari et al. 2003).

Once released into the atmosphere, PAHs get partitioned between the suspended particulate matter (SPM) and gaseous phase, depending upon their volatility (Chen et al. 2017; Li et al. 2014). SPM is classified into different aerodynamic fraction sizes of air particles, while SPM with 2.5–10 µm diameters size air particles is commonly known as PM_{10}. Pollutants attached to PM_{10} are associated with increased mortality and morbidity in humans (Samet et al. 2000). Majority of low molecular weight-PAHs (L-PAHs) are associated with the gaseous phase, while high molecular weight-PAHs (H-PAHs) are attached to PM_{10} (Hassan and Khoder 2012; Wang et al. 2009; Baek et al. 1991). Vapour/gaseous phase fraction of PAHs is < 5% of total PAHs; on the other hand, most of the PAHs are attached to particles, particularly with aerodynamic diameter of < 10 µm (Lai et al. 2017). Toxicity potential of PAHs in PM_{10} and health risks to humans through inhalation are reported worldwide including studies from India (Ray et al. 2019; Yunesian et al. 2019; Hazarika et al. 2019; Yadav et al. 2018; Jin et al. 2018; Neupane et al. 2018; Hazarika and Srivastava 2016; Amarillo et al. 2014; Li et al. 2014; Khairy et al. 2013).

Human exposure to PM_{10} bound PAHs and associated risk in cities is high considering the density of population, vehicular traffic, increasing energy demands and unusual dispersion of the atmospheric pollutants (ATSDR 1995). PAHs concentrations in PM_{10} are reported for various urban locations in India like Kolkata (Roy et al. 2017; Ray et al. 2019), Pune (Roy et al. 2019), Raipur (Ramteke et al. 2018), Dhanbad (Roy et al. 2017), Mangalore (Kalaiarasan et al. 2017), Imphal (Devi et al. 2014) and Lucknow (Pandey et al. 2013). But, reports on PAHs in PM_{10} and human health risk are scarce for north-western part of India (Kaur et al. 2013). However, elevated levels of PAHs are reported for other matrices from nearby areas (Garg et al. 2018; Kumar et al. 2014; Kumar and Kothial 2012). Hence, USEPA enlisted 16 priority PAHs in PM_{10} were measured during this study for the identification of possible sources, and assessment of carcinogenic risk to humans in a typical subtropical city, near India–Pakistan border in north-west of India.

Materials and methods

Study area and sampling

The study area was the city of Golden Temple (Amritsar), in the north-western region of India (31.63 °N, 74.87 °E). The area is located 32 km east of Lahore near Pakistan border in India. Amritsar city with urban population~10,16,079 (~7,137 persons km^{-2}) covers 142 km^2 area at an elevation of ~175–200 m. The ambient temperature of the city during the winter season (November to March) ranges from ~4 °C 16 °C and can reach up to 45 °C during the summer season (April–June). The city receives on an average of ~ 600 mm rainfall during monsoon season (July–September). The wind directions are from north-west to south-east. About 70% of the city's population is engaged in ~18,000 small-scale and 8 medium/large-scale industries including pulp and paper, textile, pharmaceutical, agro-products and steel products. As the city is a religious and cultured place, the city is visited by a total number of 36 million tourists per year. The number of registered vehicles has tremendously increased from 4,86,869 (2003–04) to 755,044 (2010–11) (PUDA 2010).

Six sampling locations (Fig. 1) were selected based on characteristic features of places. The samplers were located in places such as near the railway station (RS) with the intersection of the public vehicles and eateries, at Crystal Chowk (CC) near a market with commercial activities, at Bhandari Bridge (BB) near big traffic intersections, at Ram Bag (RB) near small-scale industrial area with bus stand and commercial activities, Garden Colony (GC) in an urban residential area, Ranjit Avenue (RA) in a suburban residential area with low traffic (Table 1). Sampling was carried out during November 2013–January 2014 as per the sampling protocol described elsewhere (Kaur et al. 2013). After collection of each sample, filters were wrapped in aluminium foil, packed in zip-lock bags, transported to the laboratory with ice packs and stored in a freezer for analysis. A total number of 54 samples were collected at the six sites (three samples per day per site). During the sampling period, the average PM_{10} concentrations at six sampling locations were 325 ± 24, 284 ± 65, 275 ± 51, 462 ± 11, 207 ± 20 and 196 ± 13 µg m^{-3}, respectively, at RS, CC, BB, RB, RA and GC locations.

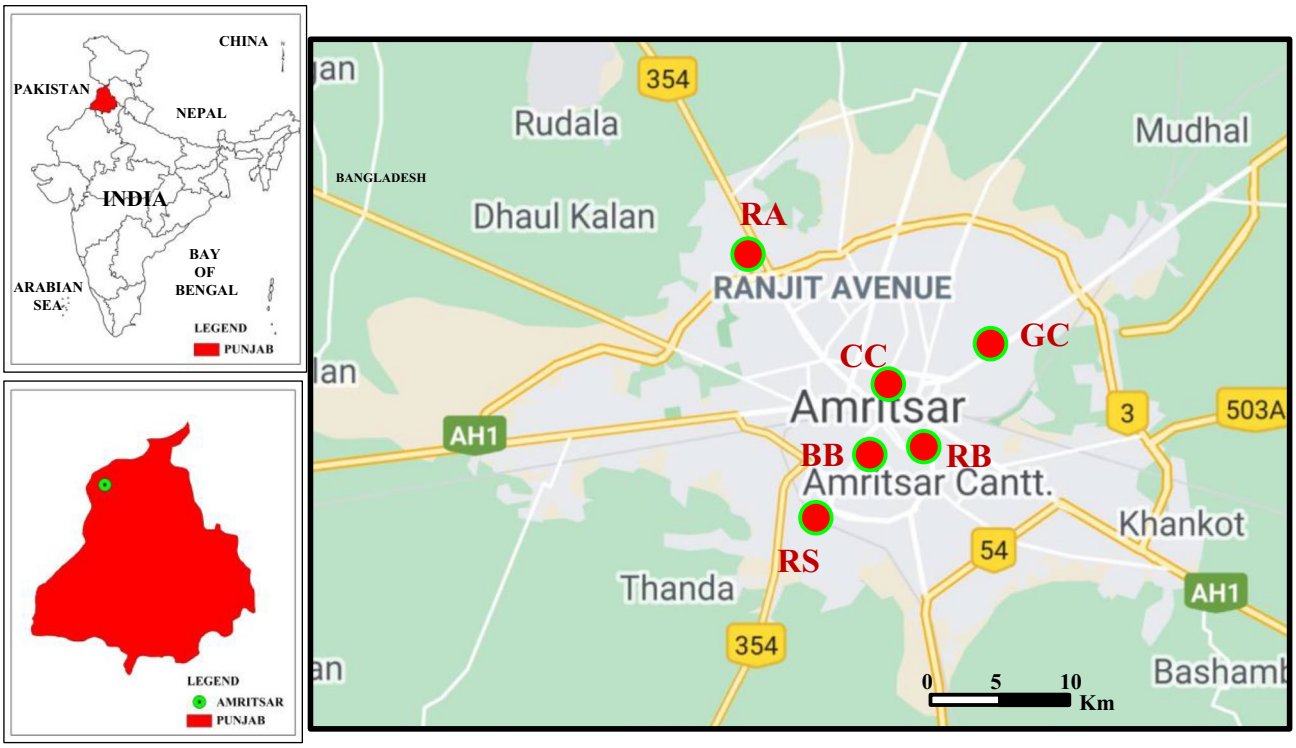


Fig. 1 Map showing study area and sampling locations in Amritsar, India

Analysis of PAHs

USEPA's method 3540C and method 3630C were followed for sample extraction and extract clean-up, respectively (USEPA 1996). Exposed filter paper from each site was cut into small pieces, put into a thimble and inserted into a Soxhlet assembly. The samples were extracted using 100 mL of extraction solvent mixture (*n*-hexane and acetone, 1:1 v/v) for 20–24 h at a temperature of approximately 30–40 °C by setting the rate at three cycles per hour. After extraction, the sample was concentrated to 1 mL at 40 °C water bath using a rotary evaporator (Heidolph, Germany). The concentrated extract was cleaned by column chromatography using 100–200 mesh activated silica (Sigma-Aldrich, USA). The clean fraction was solvent exchanged to HPLC grade acetone and reduced the volume to 1.0 mL under a gentle stream of purified nitrogen gas using Minivap (Supelco, USA), and preserved in the refrigerator at < 4 °C till further analysis. Sixteen PAH compounds were separated on RH-5 capillary column (30 × 0.53 with 3.0 µm coating) and quantified using a gas chromatograph (Nucon, Model 5765) equipped with flame ionization detector (FID) (Kumar et al. 2014).

The external standard solutions were prepared including 16 PAHs: Npt, Ane, Any, Fle, Phe, Ant, Flt, Pyr, BaA, Chr, BbF, BkF, BaP, DBA, BghiP and IP. Individual and mixture of EPA610 (16 PAHs of USEPA listed) were purchased from Supelco (Sigma-Aldrich, USA), and used for instrument calibration and QA/QC analysis. Strict quality assurance and quality control (QA/QC) analysis was performed including procedural blanks (analytes concentrations was < MDL 'method detection limit'), multi-level calibration curves (r^2, 0.994–0.999), calibration verification (< 10%) and matrix spiked recovery. The procedure performance and matrix effects were checked by analysing samples spiked with a known concentration of 16 PAHs and surrogate standard (1-fluoronaphthalene). The recovery for 16 PAHs ranged between 79 and 113%, and 96% for 1-fluoronaphthalene. Each sample was analysed in duplicate (< 1%), and the average value was used in calculations. The instrument was calibrated with every batch of sample analysis. Method detection limits with valid quantifiable peak were estimated by using signal to noise ratio > 3:1 (S/N ratio > 3) and ranged between 0.09 and 0.21 (± 0.03) ng.

Back trajectory analysis

The HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) back trajectories were used to investigate the possible sources of PAHs and to assess the influence of long-range transport of air masses in this study area. HYSPLIT is a comprehensive modelling system developed by the National Oceanic and Atmospheric Administration (NOAA), Air Resource Laboratory (https://www.ready.noaa.gov/HYSPLIT_traj.php) (Stein et al. 2015).

Table 1 Description of sampling locations and Metrological conditions on sampling days

S. No.	Sampling site/code	Geographical coordinates	Date of sampling	PM_{10}(μg m^{-3})		Temperature (°C)		Humidity (%)		Wind speed (Km/h)		Precipitation (mm)	Dew point (°C)	Visibility (Km)
						Max.	Min.	Max.	Min.	Min.	Max.			
1	Railway Station(RS)	31° 38′ 3.57″E 74° 52′ 5.6" N	Nov 8, 2013	345.24	325 ± 24	22	11	100	62	15	2	0.0	15	3.7
			Dec 6, 2013	352.68		25	8	100	28	6	0	0.0	12	1.9
			Jan 7, 2014	276.79		16	0	89	66	15	2	0.0	7	2.1
2	Crystal Chowk (CC)	31° 38′ 8.77″ E 74° 52′ 32.75"N	Nov 5, 2013	400.30	284 ± 65	26	11	100	33	9	1	0.0	14	2
			Dec 5, 2013	276.79		24	6	100	30	9	1	0.0	10	2.2
			Jan 6,2014	174.11		17	0	100	72	11	1	0.0	7	1.1
3	Bhandari Bridge (BB)	31° 37′ 49.44" E 74° 52′ 25.25"N	Nov 11,2013	261.90	275 ± 51	25	9	100	39	15	2	0.0	13	3.1
			Dec 9, 2013	193.45		23	6	100	54	13	2	0.0	12	1.5
			Jan 8, 2014	369.05		12	3	93	70	17	4	0.0	7	2.7
4	Ram Bag (RB)	31° 37′ 50.84" E 74° 52′ 40.66" N	Nov 2,2013	467.26	462 ± 11	27	13	100	29	18	3	0.0	15	2.7
			Dec 2, 2013	440.40		25	9	100	37	13	3	0.0	13	1.7
			Jan 2, 2014	479.17		18	0	100	68	74	5	0.0	7	2.6
5	Ranjit Avenue (RA)	31°39′33.78" E 74° 51′ 18.77"N	Nov 13,2013	226.19	207 ± 20	25	9	100	31	11	2	0.0	12	2.5
			Dec 11, 2013	166.67		22	6	100	44	18	2	0.0	12	2.3
			Jan 10, 2014	229.17		17	0	100	61	83	3	0.0	8	3.3
6	Garden Colony (GC)	31°38′44.15"E 74° 52′ 55.65"N	Nov 15,2013	197.92	196 ± 13	25	7	100	23	13	2	0.0	10	2.6
			Dec 12,2013	217.20		22	7	100	45	22	4	0.0	11	3.4
			Jan 11,2014	174.11		17	0	100	60	15	2	0.0	8	2.4

Toxicity of PAHs and health risk assessment

The carcinogenic potency of each PAH as BaP toxicity equivalent (BaP_{eq}) was calculated by multiplying the concentration with the appropriate toxicity equivalent factors (TEFs). Formerly compiled TEFs (Nisbet and LaGoy 1992) were adopted to calculate the BaP_{eq}. Humans are exposed to pollutants including PAHs through various pathways like ingestion, inhalation or dermal contact. For this study, the inhalation pathway of PAHs was considered for the assessment of cancer risk to humans. Cancer risk (CR) was estimated by calculating the chronic daily intake (CDI) of PAHs following recommended guidelines (ATSDR 2005; USEPA 2019) and input parameters (Table S1) using the following equations.

$$\mathrm{CDI}\left(\mathrm{mg\ kg^{-1}d^{-1}}\right) = (\mathrm{Cs} \times \mathrm{IR} \times \mathrm{CF} \times \mathrm{EF} \times \mathrm{ED})/(\mathrm{BW} \times \mathrm{AT})$$

$$\mathrm{CR} = \mathrm{LADD} \times \mathrm{cancer\ oral\ slope\ factor\ (CSF)}$$

where Cs is the PAH concentration in air particles (ng m^{-3}), IR is the air inhalation rate, CF is the unit conversion factor, EF is exposure frequency (d/yr), ED is the lifetime exposure duration (yr), BW is the body weight (kg), AT is the averaging time for carcinogens (d), and CSF is the cancer oral slope factor (per mg kg^{-1} d^{-1}).

Results and discussion

Concentrations of PAHs

The average concentrations of $\sum_{16}$PAHs in PM_{10} at six sampling sites of Amritsar, viz., RB, BB, RS CC, RA and GC in ng m^{-3}, were 114 ± 17, 134 ± 27, 128 ± 7.8, 132 ± 22, 20 ± 4.1 and 49 ± 6.3, respectively (Table S2). In general, higher concentrations were found in urban sites, with comparatively low concentrations in RA and GC. The suburban and urban residential site, respectively, demonstrated a strong urban–suburban gradient of comparatively less concentration. This indicates the urban impacts of traffic and industrial activities in the study region, which can be explained by the dominant emission activities in Indian urban cities, where biomass burning, coal combustion and industrial activities are the dominant emission sources (Roy et al. 2019; Ray et al 2017; Saxena et al. 2016; Sampath et al. 2015; Devi et al. 2014; Kaur et al. 2013; Sarkar and Khillare 2013). The overall concentration of $\sum_{16}$PAHs in PM_{10} ranged between 18 and 164 ng m^{-3} with an average of 96 ± 48 ng m^{-3}. The Pyr, Npt, Any, Ane, Fle, Phe, Ant, BaA, BkF and BbF were the dominant PAH compounds (Table 2), which indicated mixed pyrogenic sources. Volatile and semi-volatile PAHs, such as Any, Ane, Fle, Phe, Ant, Flt, Pyr,

Table 2 Concentration of 16 PAHs and BaPeq in ambient air (PM_{10})

PAHs	Concentration (ng/m^3)			BaPeq (ng/m^3)		
	Range	Mean ± SD	% of ∑	Range	Mean ± SD	% of ∑
Npt	1.8–21	10 ± 5.7	9.42	0.002–0.021	0.010 ± 0.006	0.15
Any	3.6–35	18 ± 8.6	17.00	0.004–0.035	0.018 ± 0.009	0.26
Ane	1.2–50	17 ± 13	15.87	0.001–0.050	0.017 ± 0.013	0.25
Fle	1.0–8.7	3.5 ± 2.2	3.23	0.001–0.009	0.003 ± 0.002	0.05
Phe	0.7–8.8	3.6 ± 2.0	3.39	0.001–0.009	0.004 ± 0.002	0.05
Ant	1.3–11	4.0 ± 2.7	3.71	0.013–0.105	0.040 ± 0.027	0.57
Flt	0.2–8.6	2.8 ± 3.0	2.56	0.001–0.009	0.003 ± 0.003	0.04
Pyr	2.4–37	18 ± 2.9	17.01	0.002–0.037	0.018 ± 0.013	0.26
BaA	1.3–7.8	3.9 ± 2.4	3.62	0.132–0.783	0.389 ± 0.239	5.60
Chr	2.5–3.3	2.9 ± 0.2	2.68	0.025–0.033	0.029 ± 0.002	0.42
BbF	1.8–9.8	6.2 ± 2.5	5.77	0.183–0.983	0.621 ± 0.248	8.94
BbK	2.0–11	7.4 ± 2.4	6.83	0.205–1.14	0.736 ± 0.236	10.58
BaP	1.2—3.2	1.6 ± 1.3	2.71	1.24–6.25	2.92 ± 1.35	41.99
BghiP	0.1–5.9	1.9 ± 1.7	1.80	0.001–0.059	0.019 ± 0.017	0.28
DBA	0.4–4.4	1.8 ± 1.3	1.70	0.361–4.42	1.83 ± 1.27	26.36
IP	0.6–5.3	2.9 ± 1.4	2.71	0.058–0.532	0.292 ± 0.138	4.20
2–3-ring	4.0–103	50 ± 28	51.56	0.004–0.156	0.079 ± 0.045	1.24
4-ring	2.4–42	25 ± 16	25.70	0.002–0.841	0.299 ± 0.288	4.68
5-ring	8.8–22	16 ± 4.3	16.10	2.34–7.49	4.17 ± 1.35	65.31
6-ring	0.8–12	6.4 ± 3.4	6.65	0.061–4.96	1.84 ± 1.44	28.77
∑16PAHs	18–164	96 ± 48	100	2.49–11.37	6.39 ± 2.47	100
∑C-PAHs	10–43	24 ± 9.0	22.68	2.48–11.20	6.27 ± 2.44	98.14

BaA and Chr, have been reported as dominant indicators for vehicular emissions and biomass combustions (Chen et al. 2005; Dickhut et al. 2000; Marr et al. 1999). Npt has a source of coal and fuel combustion, Ant and Pyr are tracers of coal combustion, Fle and Ane mainly originate from biomass burning, and Phe is produced by combustion of natural gas and vehicle exhaust (Simcik et al. 1999). The dominance of BkF, BbF and BaA is indicative of combustions in high-temperature processes including gasoline engine emissions and vehicular emissions (Jin et al. 2018; Sarkar and Khillare 2013; Yunker et al. 2002; Khalili et al. 1995).

The concentrations of $\sum_{16}$PAHs in PM_{10} obtained during the present study were compared with other studies undertaken in various countries including India (Table 3). The average concentration (96 ± 48 ng m^{-3}) were found comparable with concentrations observed in major cities of India, including Jorhat (Islam et al. 2020), Delhi (Singh et al. 2011), Kolkata (Ray et al. 2017) and Mangalore (Kalaiarasan et al. 2017). In the literature, authors several studies have reported elevated PAHs in PM_{10} for other Indian cities including Pune (Roy et al. 2019), Lucknow (Pandey et al. 2013), Manipur (Devi et al. 2014) and Raipur (Ramteke et al. 2018). However, Kulkarni et al. (2014) reported low concentration of PAHs in Visakhapatnam. Observed concentrations were higher than those reported for Islamabad, Pakistan (Mehmood et al. 2020), Zagreb, Croatia (Pehnec and Jakovljevic, 2018), Aliaga, Turkey (Kaya et al. 2012), Doha, Qatar (Javed et al. 2019) and Cordoba, Argentina (Amarillo et al. 2014), but were much less than those reported for Lahore, Pakistan (Kalim et al. 2015, 2020), Beijing (Liu

Table 3 Comparatives of concentration of priority PAHs in PM_{10} at various locations

Location	Sampling	No. of PAHs	∑PAHs		Reference
			Mean	Range	
Locations in India					
Amritsar, Punjab	2013–14	16	96	18–164	Present study
Jorhat, Assam	2018	14	95	–	Islam et al. (2020)
Kolkata, West Bengal	2017–18	17	95	56–142	Ray et al. (2019)
Pune, Maharashtra	2015–16	13	345	64–814	Roy et al. (2019)
Raipur, Chhattisgarh	2013–14	20	292	0–1049	Ramteke et al. (2018)
Kolkata, West Bengal	2015–16	14	89	38–147	Ray et al. (2017)
Mangalore, Karnataka	2014	7	109	39–252	Kalaiarasan et al. (2017)
Dhanbad, Jharkhand	2012–13	15	11–482	–	Roy et al. (2017)
Vishakapattnam, Andhra Pradesh	2010–11	16	58	23.3–105	Kulkarni et al. 2014
Manipur, Imphal	2009	16	236	42–734	Devi et al. (2014)
Delhi	2008–09	16	105	10–512	Sarkar & Khillare (2013)
Lucknow, Uttar Pradesh	2007–08	9	251	–	Pandey et al. (2013)
Amritsar, Punjab	2011	16	153	37–274	Kaur et al. (2013)
Delhi	2007–08	16	82	–	Singh et al. (2011)
Locations in other countries					
Lahore, Pakistan	–	16	556	189–2121	Kalim et al. (2020)
Islamabad, Pakistan	2017	16	40.07	23.0–88.9	Mehmood et al. (2020)
Doha, Qatar	2015	16	0.721	0.193–2.849	Javed et al. (2019)
Lahore, Pakistan		20	742	141–1642	Kalim et al. (2018)
Zagreb, Croatia	2014	10	15	0.055–124	Pehnec and Jakovljevic (2018)
Tianjin, China	2012–14	16	417	–	Jin et al. (2018)
Kaohsiung, Taiwan	2012	16	158	–	Lai et al. (2017)
Riyadh, Saudi, Arabia	2011–12	16	18.4	0.13–516	Bian et al. (2016)
Beijing, China	2009–10	16	126	0.2—559	Liu et al. (2015)
Lahore, Pakistan	2013–14	16	239	30–784	Kalim et al. (2015)
Cordoba, Argentina	2012	14	9.04	0.7–33	Amarillo et al. (2014)
Makkah, Saudi Arabia	–	16	137	120–166	Habeebullah (2013)
Alexandria, Egypt	2010–11	16	120, 147	27–340, 14–420	Khairy & Lohmann (2013)
Giza, Egypt	2007–08	16	1,430	–	Hassan & Khoder (2012)
Aliaga, Turkey	2009–10	16	89	1.6–838	Kaya et al. (2012)
Baoji, China	2008	17	594	46–1517	Xie et al. (2009)

et al. 2015), Tianjin (Jin et al. 2018) and Baoji (Xie et al. 2009) in China, Makkah, Saudi Arabia (Bian et al. 2016), Kaohsiung, Taiwan (Lai et al. 2017) and Alexandria (Khairy and Lohmann 2013) and Giza (Hassan and Khoder 2012) in Egypt (Table 3). Concentrations observed during the present study were less than previously reported levels from Amritsar (Kaur et al. 2013). The lower concentrations may be attributed to various government initiatives for mitigation of air pollution in India such as improvement of fuel quality and standards, and mandatory use of compressed natural gas (CNG) in urban public transportation (MoEF and CC 2019). Replacement of diesel and petrol with CNG in public transport system has reported a reduction of 51–74% and 58–68% in the concentration of PM_{10} and PAHs, respectively, for Delhi ambient air (Khillare et al. 2008).–

Toxicity of PAHs

BaP is used as an index for toxicity and as an indicator for other 15 priority PAHs (USEPA 2017; IARC 2005). However, various studies on multiple animal species concluded that BaP is carcinogenic at multiple tumour sites (alimentary tract, liver, kidney, respiratory tract, pharynx and skin). Therefore, to assess and estimate the health risk associated with PAHs, the concentrations of PAHs and individual compounds can be compared with regulatory standards established to protect public health. India is regulating BaP in ambient air at 1.0 ng m^{-3} as 'National Ambient Air Quality Standards' (NAAQS) (MoEF and CC 2009). The BaP criterion for WHO is 1.0 ng m^{-3} (WHO 1987), while the target value of BaP in countries of the European Union is 1.0 ng m^{-3} (EC 2004). The observed mean concentration of BaP in PM_{10} (1.6 ± 1.3 ng m^{-3}) in the present study is comparable to NAAQS. The observed concentration of BaP was less than the values reported from Lahore (81.4 ng m^{-3}, Kalim et al. 2020; 62.3 ng m^{-3}, Kalim et al. 2018; 13 ng m^{-3}, Kalim et al. 2015), Islamabad (2.3 ng m^{-3}, Mehmood et al. 2020) in Pakistan; Giza in Egypt (151 ng m^{-3}, Hassan and Khoder 2012*)*; Zagreb in Croatia (0.07–7.66 ng m^{-3}, Pehnec and Jakovljevic 2018); Kanpur (19 ng m^{-3}, Singh and Gupta 2016), Lucknow (51.96 ng m^{-3}, Pandey et al. 2013), Dhanbad (9–46 ng m^{-3}, Roy et al. 2017), Raipur (43–76 ng m^{-3}, Ramteke et al. 2018) and Kolkata (5.3 ng m^{-3}, Ray et al. 2017) in India. BaP concentrations were comparable with Jamshedpur (1.24–4.74 ng m^{-3}, Kumar et al. 2020), Delhi (1.8–3.6 ng m^{-3}, Singh et al. 2012) and Visakhapatnam (1.7 ng m^{-3}, Kulkarni et al. 2014) in India, but were higher than reported for Kaohsiung in Taiwan (0.008–0.42 ng m^{-3}, Lai et al. 2017), Imphal (0.2–1.0 ng m^{-3}, Devi et al. 2014) and Jorhat (0.02 ng m^{-3}, Islam et al. 2020) in India, Makkah in Saudi Arabia (0.10 ng m^{-3}, Habeebullah 2013) and Cordoba in Argentina (0.22 ng m^{-3}, Amarillo et al. 2014).

The concentration of seven carcinogenic PAHs (C-PAHs) (sum of BaP, BaA, Chr, BbF, BkF, DBA and IP) ranged between 10 and 43 ng m^{-3}, with a mean of 24 ± 9.0 ng m^{-3} and accounted for 22.68% of $\sum_{16}$PAHs. The individual contribution of BaA, Chr, BbF, BkF, BaP, DBA and IP was 3.62%, 2.68%, 5.77%, 6.83%, 2.71%, 1.70% and 2.71%, respectively, to $\sum_{16}$PAHs (Table 2). Error plot of carcinogenic PAHs is given in Fig. S1. Contribution of $\sum$C-PAHs accounted for 22%, 24%, 23%, 24%, 62% and 33% of $\sum_{16}$PAHs, respectively, at sites RB, BB, RS CC, RA and GC. The average concentration of $\sum$C-PAHs ranged from 13 ± 4.5 ng m^{-3} (RA, suburban residential site) to 32 ± 3.3 ng m^{-3} (BB, traffic intersection with heavy traffic). (Table S2).

Given the highest carcinogenic potential of BaP, carcinogenic factor for rest of 15 priority PAHs was derived for estimation of relative carcinogenic potential which was expressed in terms of BaP_{eq}. The estimated BaP_{eq} of $\sum$PAHs ranged between 2.49 and 11.37 ng m^{-3}, with an average of 6.39 ± 2.47 ng m^{-3}. The estimated $\sum BaP_{eq}$ at different locations of RB, BB, RS CC, RA and GC was 4.48, 7.78, 7.58, 6.94, 3.56 and 6.90 ng m^{-3}, respectively. The BaP_{eq} contributed by $\sum$C-PAH accounted for more than 98% of $\sum BaP_{eq}$. The major contributors to $\sum BaP_{eq}$ were 5-ring PAHs and 6-ring PAHs including BaA (5.6%), BbF (8.9%), BkF (10.6%), BaP (42%), DBA (26.4%) and IP (4.2%) (Table 2). Contribution from L-PAHs was considerably negligible (< 1.0%) towards $\sum BaP_{eq}$. It indicates the importance of H-PAHs to understand therefore associated carcinogenic potential. The contribution of the sum of 5-ring PAHs and 6-ring PAHs to $\sum BaP_{eq}$ accounted for > 94%. BaP and DBA both with higher TEF (Nisbet and LaGoy 1992) contributed > 68% to $\sum BaP_{eq}$. Observed results for BaP_{eq} are in agreement with results reported for Delhi (Hazarika et al. 2019), Pune (Roy et al. 2019), Chandigarh (Garg et al. 2018) and Zagreb, Croatia (Pehnec and Jakovljevic 2018).

Possible source of PAHs

Composition profiles of PAHs with different aromatic rings

Priority 16 PAHs are classified as 2–3-ring to 6-ring PAHs, and their concentrations are presented in Table 2. The mean concentration of 2–3-ring, 4-ring, 5-ring and 6-ring PAHs observed during present study was 50 ± 28 ng m^{-3}, 25 ± 16 ng m^{-3}, 16 ± 4.3 ng m^{-3} and 6.4 ± 3.4 ng m^{-3}, respectively. The overall contribution of 2–3-ring, 4-ring, 5-ring and 6-ring PAHs to $\sum$PAHs accounted for 51.56%, 25.70%, 16.10% and 6.65%, respectively. The distribution of PAHs homologs with different aromatic rings at different locations is depicted in Fig. 2. The dominant homolog was 2–3-ring PAHs, and ranged between 48 and 59% of total PAHs, except at RA location, where 5-ring PAHs were

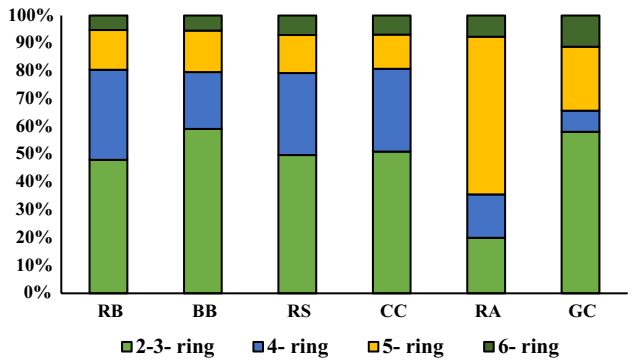


Fig. 2 Per cent contribution of PAHs homolog at different locations

dominant with 56%. Second dominant homolog was 4-ring PAHs with 20–32% at RB, BB, RS and CC locations. However, 2–3-ring and 5-ring PAHs were the second dominant homologs at RA and GC (Fig. 2). The observed homologs pattern of PAHs suggested mixed combustion sources and petrogenic sources from spillage of petroleum products. Mixed origin of sources is also suggested for location RA. Similar composition profiles were reported for biomass combustion and vehicular emission sources in India (Saxena et al. 2016; Ray et al. 2017).

Further, emissions of PAHs based on their molecular weight are associated with different representative sources, e.g. L-PAHs have been associated with petroleum products (Petrogenic sources). Therefore, sources of H-PAHs have been reported from various pyrogenic activities (Yunker et al. 2002; Dickhut et al. 2000; Khalili et al. 1995). Their ratios are used to distinguish pyrolytic sources (ratio < 1) and petrogenic sources (ratio > 1) (Wilcke 2007). The concentrations of L-PAHs and H-PAHs ranged between 4.1 ± 0.1 to 79 ± 22 ng m^{-3} and 16 ± 4.2 to 65 ± 10 ng m^{-3}, respectively. The percentage contribution of L-PAHs at different locations ranged between 35.62% (RA) and 80.84% (CC). On the other hand, the lowest contribution of H-PAHs was 19.16% at CC and highest 64.38% at RA locations (Fig. 2). Therefore, the dominance of L-PAHs over H-PAHs, and their consequent ratios varying between 0.19 and 2.31 (mean, 1.03) (Table 4), indicated mixed combustion sources of PAHs (Roy et al. 2019).

Molecular diagnostic ratios of PAHs

Concentration ratios between some PAHs are often used as a diagnostic tool for identification of possible sources like petrogenic and pyrogenic sources including diesel combustion, gasoline, wood combustion, coal combustion and vehicular emission (Chen et al. 2005; Yunker et al. 2002; Dickhut et al. 2000; Simcik et al. 1999; Khalili et al. 1995). During the present study, the molecular diagnostic ratios of Ant/(Ant + Phe), BaA/(BaA + Chr), BbF/BkF, BaP/BghiP, BaP/(BaP + Chr) and IP/(IP + BghiP) were calculated and used to identify the possible sources of PAHs (Table 4). The

Table 4 Diagnostic ratio of PAHs at various locations for possible sources identification of PAHs

Diagnostic ratios with their reported values for possible sources				This study
PAH ratio	Value	Possible sources	References	
LMW/HMW	< 1	Pyrogenic	Wilcke (2007)	1.03 (0.19–2.31)
	> 1	Petrogenic		
Ant/(Ant + Phe)	< 0.1	Petrogenic	Yunker et al. (2002)	0.46 (0.24–0.65)
	> 0.1	Fossil fuel combustion		
BaA/(BaA + Chr)	< 0.2	Petrogenic	Yunker et al. (2002)	0.53 (0.24–0.74)
	0.2–0.35	Petroleum comb		
	> 0.35	Biomass, coal comb		
BbF/BkF	0.92	Wood comb	Dickhut et al. (2000)	0.91 (0.34–1.80)
	1.07	diesel engine		
	1.30	Vehicular emission	Dickhut et al. (2000)	
	3.7	Coal combustion	Dickhut et al. (2000)	
BaP/BghiP	0.3–0.78	Vehicular emissions	Simcik et al. (1999)	5.90 (0.43–8.98)
	0.9–6.6	Coal combustion		
BaP/(BaP + Chr)	0.07–0.24	Coal combustion	Chen et al. (2005)	0.65 (0.33–1.00)
	0.49	Gasoline	Khalili et al. (1995)	
	0.73	Diesel engine		
IP/(IP + BghiP)	< 0.2	Petrogenic	Dickhut et al. (2000); Yunker et al. (2002)	0.68 (0.33–0.95)
	0.2–0.5	Petroleum comb		
	> 0.5	Biomass, coal comb		

*Range in parenthesis

higher mean value of Ant/(Ant + Phe) than 0.1 indicated fossil fuel combustions sources (Yunker et al. 2002). Previously documented value for BaA/(BaA + Chr) (Yunker et al. 2002) and estimated values indicated petroleum, biomass and coal combustion sources. The ratio of BaA/(BaA + Chr) is also used to distinguish between recent emissions of PAHs (ratio > 0.40) and transportation of aged aerosol (ratio < 0.40) with PAHs (Ding et al. 2007). The observed ratio of BaA/(BaA + Chr) reflects recent emissions in addition to the transportation of air mass with PAHs. The average value of 0.91 (range, 0.34–1.80) for BbF/BkF ratio indicated pyrogenic sources of wood combustions and emissions from vehicles and diesel engine (Dickhut et al. 2000). BaP/BghiP ratio values (mean, 0.590, range, 0.43–8.98) indicated vehicular emissions and coal combustion sources (Simcik et al. 1999). Formerly reported BaP/(BaP + Chr) ratio value of 0.49 and 0.73, respectively, indicated for gasoline and diesel engines (Khalili et al. 1995) supported the pyrogenic sources. The observed mean value of IP/(IP + BghiP) ratio (0.68) suggested petroleum, biomass and coal combustions sources (Yunker et al. 2002). Industrialization, increase in vehicles and energy consumption for the requirement of the rapid growth of population are the major factors for deterioration of air quality in urban and suburban areas of developing countries. Studies reported that the majority of PAHs in urban air are contributed by emissions from vehicles, incineration, coal and gasoline combustions and through via atmospheric depositions (Hassan and Khoder 2012; Wild and Jones 1995). Based on results of molecular diagnostic ratios (Table 4), it is concluded that mixed pyrogenic sources, such as vehicle emissions, diesel engines, biomass and coal combustion, were the main sources of atmospheric PAHs in Amritsar.

Further, the ratios between the sum of the concentrations of the combustion PAHs (Flt, Pyr, BaA, BbF, BkF, BaP, BghiP and IP) and $\sum_{16}$PAH have been used to distinguish between mobile sources (ratio > 0.51) and heavy-duty diesel trucks emission sources (ratio = 0.30) (Singh et al. 2012; Wang et al. 2007). Those ratios in this study ranged between 0.28 and 0.84 with an average value of 0.49, suggesting that substantial emissions of PAHs might have originated from mobile sources such as automobiles along with heavy-duty diesel trucks. Furthermore, higher values of ratios between 4-ring PAHs to the sum of 5-ring PAHs and 6-ring PAHs also indicated long-range atmospheric transport of PAHs and lower ratios indicates local emission sources (Wang et al. 2007). The observed ratios of 4-ring PAHs to sum of 5-ring PAHs and 6-ring PAHs ranged between 0.34 and 5.76 with the mean of 3.16, suggesting that local emission sources coupled with long-range atmospheric transport (LRAT) of PAHs. LRAT may include regional transport of ambient air with elevated PAHs (Kalim et al. 2015, 2018, 2020). The transportation of PAHs could also be possible from crop stubble burning in the north-western region of India (Rishipal 2009). It is reported that crop residue burning is the cause of increased levels of PM_{10} (23.7% to 86.7%) in nearby areas of Punjab (Chanduka 2013), and it can increase the concentrations of PAHs in ambient air by 58% (Chen et al. 2008).

Principal component analysis (PCA)

Site-wise and compound-wise principal component analysis (PCA) was conducted (Fig. 3). PC-1 represented 39% of the total variance and had the presence of both L-PAHs and H-PAHs. L-PAHs, viz., Npt, Ane and Any and H-PAHs, viz., BaA, Chr, BkF, Pyr, BbF, IP and BghiP. Ane, Chr and BghiP were suspected to be sourced mainly from traffic exhausts emission in India (Cheng et al. 2013). PC-2 shows 17% variance and was mainly loaded with Fle, Ant, DBA and Phe. H-PAHs can be significantly emitted from light vehicles, and Flt is an indicator for heavy-duty diesel combustion (Marr et al. 1999). Pyr, BaA, Chr and Flt are markers for coal combustion (Tavakoly Sany et al. 2014) and BaP for biomass burning (Belis et al. 2011). PC-3 depicts 14% variance; this component was weighted with BaP & Flt.

Pearson's coefficient of correlation

Pearson's correlation coefficient analysis was performed to determine relationships between individual PAH to understand their similar source of origin for PAHs. During this study, a significant correlation ($p < 0.01$) was obtained among some compounds of L-PAHs as well as H-PAHs (Table 5). A significant correlation ($p < 0.01$) among 2–3-ring PAHs (L-PAHs) indicated PAHs emission from coal, wood and biomass combustions (Sarkar and Khillare 2013; Khalili et al. 1995). However, a significant correlation

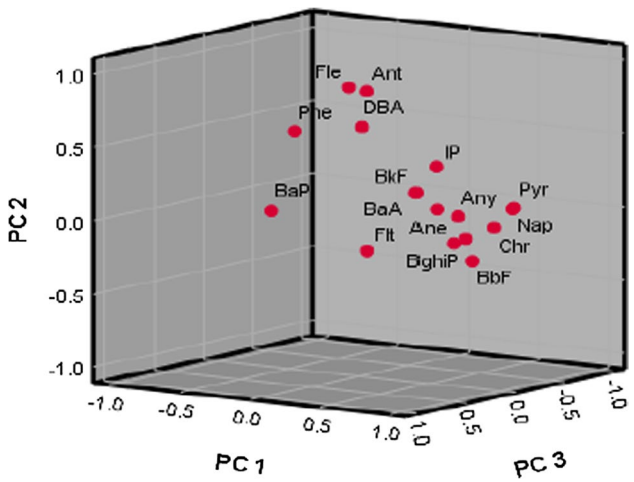


Fig. 3 Compound-wise principal component analysis of the 16PAHs

Table 5 Pearson's correlation coefficient among PAH compounds in ambient air (PM_{10})

PAHs	Any	Ane	Fle	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BaP	BghiP	DBA	IP
	Low molecular weight-PAHs					High molecular weight-PAHs									
	2–3-ring PAHs					4-ring PAHs				5-ring PAHs			6-ring PAHs		
Nap	***0.453***	0.373	−0.460	−0.052	−0.348	−0.569	***0.900***	−0.014	−0.669	0.372	0.358	−0.101	***0.526***	−0.308	***0.584***
Any	1	***0.726***	-0.445	0.024	−0.305	***0.608***	0.324	***0.706***	***0.468***	0.235	0.162	0.259	***0.687***	0.097	0.399
Ane		1	-0.564	−0.066	−0.140	0.397	0.145	***0.442***	0.410	0.129	0.277	−0.132	***0.676***	−0.180	0.238
Fle			1	0.398	***0.572***	−0.203	−0.263	−0.145	−0.189	0.033	−0.507	0.112	−0.228	0.175	−0.126
Phe				1	***0.587***	***0.539***	−0.157	***0.580***	0.320	***0.421***	0.131	***0.544***	−0.018	0.193	0.170
Ant					1	0.020	−0.245	0.093	−0.004	0.030	−0.038	−0.084	−0.212	0.127	−0.105
Flt						1	−0.648	0.313	***0.741***	0.104	−0.055	***0.534***	0.306	0.145	−0.018
Pyr							1	−0.384	−0.803	0.342	0.209	−0.188	***0.513***	−0.369	***0.460***
BaA								1	0.285	−0.136	0.199	***0.510***	0.171	***0.508***	***0.761***
Chr									1	−0.107	−0.167	0.325	0.149	−0.012	−0.081
BbF										1	0.280	−0.084	***0.470***	0.316	0.308
BkF											1	−0.048	0.374	0.194	0.240
BaP												1	−0.051	0.215	0.277
BghiP													1	−0.111	0.361
DBA														1	***0.502***

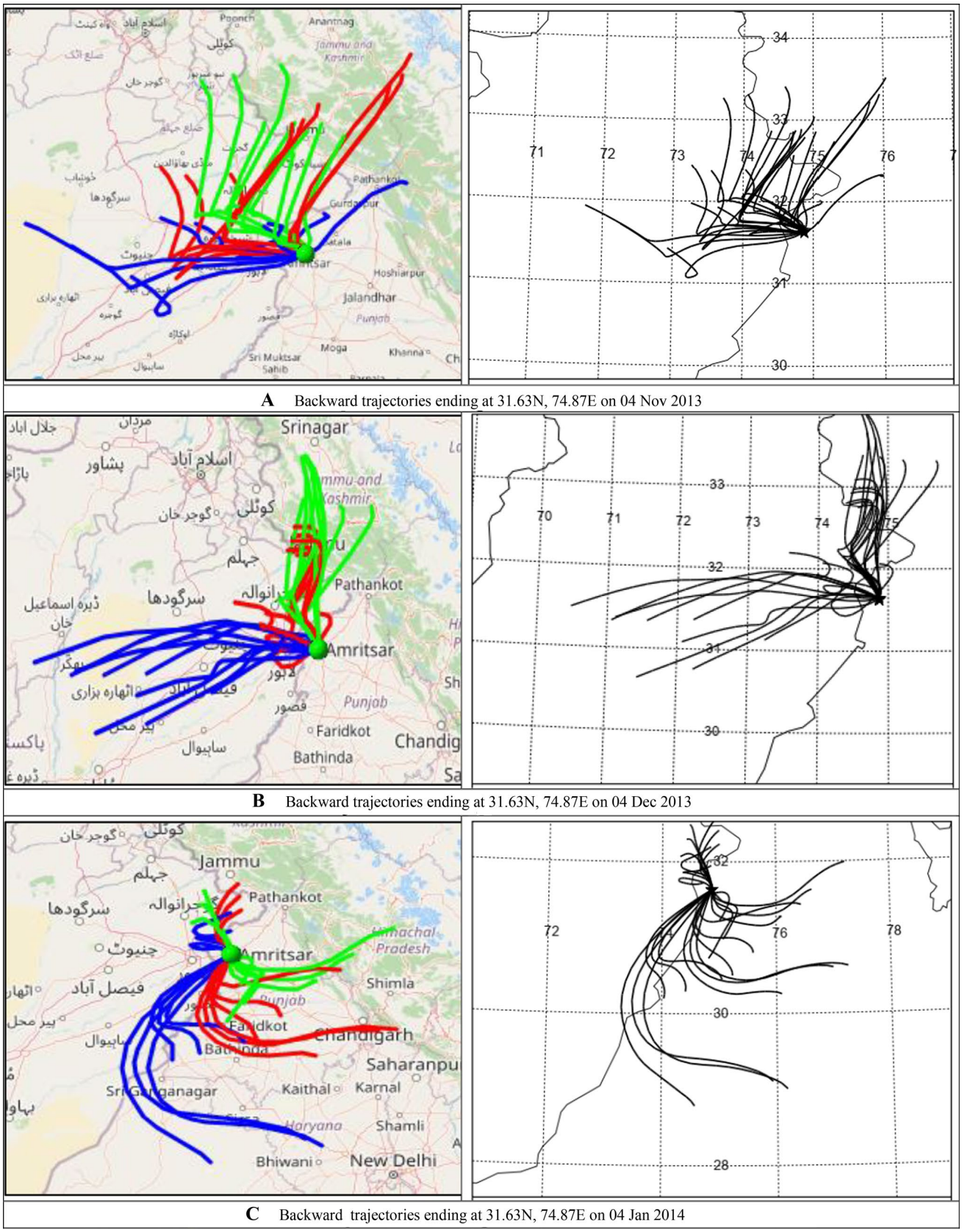
Significant correlations at $p < 0.01$ are in bold

($p < 0.05$) between 4-ring and 6-ring PAHs (H-PAHs) can be attributed to the combustion process at high temperatures, like coal combustion, incinerators, steel industries, diesel-powered vehicles and gasoline (Khalili et al. 1995; Marr et al. 1999). Correlations among Any, Ane, Flt, BaA, Nap, Ane, Fle and BghiP have been associated with emissions from vehicular sources (diesel and gasoline) (Sampath et al. 2015; Li et al. 2014; Sarkar and Khillare 2013; Khalili et al. 1995). Fle and Pyr derived from combustion of L-PAHs at high temperature are considered as pyrogenic products (Yang et al. 1998). A correlation between Pyr and BghiP might be the result of pyrogenic emissions from automobile exhaust and wood-burning (Hazarika et al. 2019). Correlation of Phe with Ant, Flt, BaA and BaP, and IP with Pyr, BaA and DBA is an indication of waste incineration (Ray et al. 2019). A positive significant correlation ($p < 0.01$) of BaA with BaP, DBA and IP suggested emissions from natural gases (Simcik et al. 1999). Any and Fle have been reported from diesel vehicles, and Flt and Chr from gasoline combustions (Yang et al. 1998). Similar correlations amid PAHs were also observed in Indian environment (Kumar et al. 2020; Roy et al. 2019; Sampath et al. 2015; Devi et al. 2014; Kaur et al. 2013; Sarkar and Khillare 2013).

Therefore, this study suggests that pyrolytic activities including vehicular emissions, biomass and coal combustion and diesel engines are the significant contributory sources of H-PAHs, while possible sources of L-PAHs may be due to biomass combustions and petrogenic sources from the spill of petroleum products and automobile workshops.

Back trajectory analysis

The PAH concentrations in ambient air atmospheric PAHs are generally influenced by long-range transport and nearby sources. Back trajectory analysis is often used to analyse the atmospheric transport of PAHs (Islam et al. 2020; Kumar et al. 2020; Chen et al. 2017). Figure 4a–c in Google Earth demonstrates that the back trajectories originate from different sources including from long-distance LRAT and local sources, and ending at the sampling sites during the study period. Figure 4a, b shows that air parcel mass originates from neighbouring regions in Pakistan and Jammu in India, and ending up at in our study area could be a possible reason for long-range atmospheric transport of PAHs. Further, these observations can be related to elevated levels of PAHs in ambient air of neighbouring regions. We suggest this LRAT since high atmospheric PAHs have been reported from those regions (Kalim et al. 2015, 2018, 2020). However, Fig. 4c shows air masses originating from Indian states including Rajasthan, Haryana, Himachal Pradesh and Punjab, and ending up at the present study sites. Similar transport of air masses was also reported in the north-western region (Pawar et al. 2015; and Kuniyal et al. 2015). Hence, the level of PAHs in ambient air (PM_{10}) at Amritsar might



A Backward trajectories ending at 31.63N, 74.87E on 04 Nov 2013

B Backward trajectories ending at 31.63N, 74.87E on 04 Dec 2013

C Backward trajectories ending at 31.63N, 74.87E on 04 Jan 2014

Fig. 4 Bawkward clustered trajectories using HYSPLIT during sampling period over study area

have been influenced by long-range atmospheric transport and various local emission sources.

Cancer risk to humans

The USEPA generated a slope factor of 7.3×10^{-3} per µg BaP kg^{-1} bw d^{-1}, meaning that 1 µg BaP kg^{-1} bw d^{-1} would pose a lifetime cancer risk of 7.3×10^{-3} (USEPA 2017). The World Health Organization (WHO) estimated inhalation unit risk of 8.7×10^{-5} per ng/m^3 BaP, and acceptable excess lifetime CR of 1/10,000, 1/100,000 and 1/1,000,000 or 10^{-6} to 10^{-4}. Under most guidelines for CR assessment, the CR between 10^{-6} and 10^{-4} implicit potential cancer risk, while a potential risk is high at CR > 10^{-4} (WHO 2000). During this study, CR assessment was based on assumption that human adults and children exposed to PAHs through inhalation of ambient air particles (PM_{10}). Accordingly, CR was estimated by calculating the CDI of 16 PAHs. The calculated CDIs at different locations for human adults ranged between 5.0×10^{-7} and 2.3×10^{-6}, and for children, it ranged between $9.3\times10^{-7}$to 4.2×10^{-6} (Table 6). The mean CDI for adults (1.3×10^{-6} mg kg^{-1} d^{-1}) was comparatively higher than children (2.4×10^{-6} mg kg^{-1} d^{-1}) (Fig. 5). These calculated CDIs were less than the recommended dose of 1 µg BaP kg^{-1}bw d^{-1}. Based on CDIs, the estimated CR for human adults ranged between 8.5×10^{-9} to 6.5×10^{-6}, while for children, it ranged between 1.6×10^{-8} and 1.2×10^{-5} (Table 6). The average CR for all locations for human adults and children was 2.3×10^{-6}, and 4.2×10^{-6}, respectively (Fig. 5), and was within the acceptable limit of 10^{-6}–10^{-4} as acknowledged by various agencies. However, limitation to exposure through inhalation, and lack of data on exposure through diet, total daily intake of PAHs CR could not be estimated.

The estimated CDI and CR were less at location RA than other locations with commercial and vehicular activities (Table S3 and Table S4). The observed CR was comparable with other locations around the world like Egypt (1.08×10^{-6}–5.88×10^{-6}, Khairy and Lohmann 2013), Delhi (2.15×10^{-6}, adults, 1.93×10^{-6}, children, Hazarika et al. 2019), Kolkata (1.73×10^{-6}–7.36×10^{-6}, Ray et al. 2019), Jamshedpur (1.65×10^{-6}, adults, 8.6×10^{-6}, children, Kumar et al. 2020), Dhanbad (0.53×10^{-6}–9.8×10^{-5}, Roy et al. 2017) and Nepal (8.95×10^{-7}–1.04×10^{-4}, Neupane et al. 2018). However, CR was less than those reported for Islamabad, Pakistan (98.19×10^{-5}, Mehmood et al. 2020), Nepal (5.2×10^{-5}, Yadav et al. 2018), Delhi (2.96×10^{-5}–2.34×10^{-3} Sarkar and Khillare 2013), Tamil Nadu (17.8×10^{-5}–23.7×10^{-5}, Sampath et al. 2015), Argentina (6.69×10^{-5} to 1.23×10^{-4}, Amarillo et al. 2014) and Amritsar (72.3×10^{-5}, adults, 74.3×10^{-5}, children, Kaur et al. 2013), but higher than Jorhat (2.2×10^{-8}, Islam et al. 2020) and Tianjin, China (9.0×10^{-8}–$1.\times\times10^{-7}$, Jin et al. 2018).

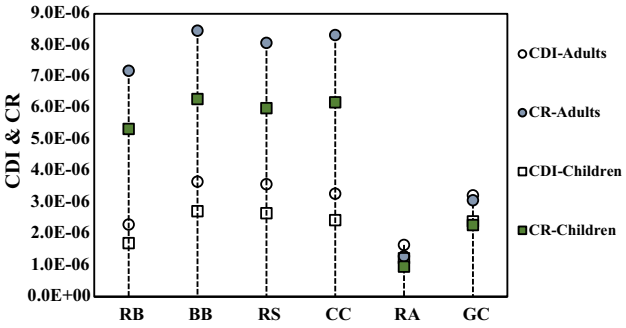


Fig. 5 CDI (mg kg^{-1} d^{-1}) of ∑16PAHs and CR to human adults and children through PM_{10}

Summary and conclusion

PAHs are persistent and toxic organic compounds released into the environment mainly through combustion processes. Sixteen PAHs are classified and listed as the priority pollutants by various agencies. Human exposure to atmospheric PAHs through inhalation causes various health effects including cancer. Various reports on PAHs in ambient air (PM_{10}) and human health risk are available for India, but scarce for north-western part of India. Therefore, present

Table 6 LADD of PAHs and Incremental Lifetime Cancer Risk (ILCR) to human adults and children due to inhalation of ambient air (PM_{10})

PAHs	LADD (mg kg^{-1} d^{-1})				ILCR			
	Adults		Children		Adults		Children	
	Range	Mean	Range	Mean	range	Mean	Range	Mean
2–3-ring	7.9×10^{-10}–3.1×10^{-8}	1.6×10–8	1.5×10^{-9}–5.8×10^{-8}	3.0×10^{-8}	2.5×10^{-6}–1.3×10^{-5}	6.8×10^{-6}	4.6×10^{-6}–2.4×10^{-5}	1.3×10^{-5}
4-ring	4.8×10^{-10}–1.7×10^{-7}	6.0×10–8	8.9×10^{-10}–3.1×10^{-7}	1.1×10^{-7}	2.5×10^{-6}–1.3×10^{-5}	6.8×10^{-6}	4.6×10^{-6}–2.4×10^{-5}	1.3×10^{-5}
5-ring	4.7×10^{-7}–1.5×10^{-6}	8.3×10–7	8.7×10^{-7}–2.8×10^{-6}	1.5×10^{-6}	5.8×10^{-12}–1.6×10^{-9}	5.5×10^{-10}	1.1×10^{-11}–3.0×10^{-9}	1.0×10^{-9}
6-ring	1.2×10^{-8}–9.9×10^{-7}	3.7×10–7	2.3×10^{-8}–1.8×10^{-6}	6.8×10^{-7}	3.5×10^{-12}–1.1×10^{-7}	3.8×10^{-8}	6.5×10^{-12}–2.1×10^{-7}	7.1×10^{-8}
∑C-PAHs	5.0×10^{-7}–2.2×10^{-6}	1.3×10–6	9.2×10^{-7}–4.2×10^{-6}	2.3×10^{-6}	2.0×10^{-6}–9.3×10^{-6}	4.4×10^{-6}	3.7×10^{-6}–1.7×10^{-5}	8.3×10^{-6}
∑16PAHs	5.0×10^{-7}–2.3×10^{-6}	1.3×10–6	9.3×10^{-7}–4.2×10^{-6}	2.4×10^{-6}	8.5×10^{-9}–6.5×10^{-6}	2.3×10^{-6}	1.6×10^{-8}–1.2×10^{-5}	4.2×10^{-6}

study was carried out on measurement of 16 priority PAHs in PM_{10} for the assessment of their possible sources, and carcinogenic risk to humans in a typical tropical city in India. During the study, total 54 PM_{10} samples were collected from six sites within an area of 142 km^2 with prevalence of various anthropogenic activities of Amritsar city located near country's border in the north-western region of India. The collected samples were processed and analysed using GC-FID following USEPA's methods using gas chromatograph equipped with RH-5 capillary column (30 × 0.53 with 3.0 µm) and flame ionization detector (FID). Required analytical quality assurance and quality control (QA/QC) analysis was performed including procedural blanks, multi-level calibration curves, calibration verification, matrix spiked recovery and estimation of method detection limits.

The observed concentration of ∑PAHs in PM_{10} (range, 18–164; mean 96 ± 48 ng m^{-3}) was more or less comparable with global and Indian studies. Among detected PAHs, Pyr, Npt, Any, Ane, Fle, Phe, Ant, BaA, BkF and BbF were the dominant compounds, and their dominance has been reported for mixed pyrogenic sources. BaP, the index for toxicity and as an indicator for other 15 priority PAHs, is considered as carcinogenic. The concentration of BaP in ambient air is regulated by various agencies at 1.0 ng m^{-3} (MoEF and CC 2009; WHO 1987; EC 2004). The observed mean concentration of BaP in PM_{10} (1.6 ± 1.3 ng m^{-3}) in the present study is comparable to the regulatory standard. The concentration of seven carcinogenic PAHs ranged between 10 and 43 ng m^{-3} (mean ± SD, 24 ± 9.0 ng m^{-3}), accounted for 22.68% of ∑PAHs. The estimated average BaP toxicity equivalent (BaP_{eq}) of ∑PAHs was 6.39 ± 2.47 ng m^{-3}, are comparable with other Indian cities. Various diagnostic tools were used for identification of possible sources of PAHs. The outcomes of composition profiles of PAHs with different aromatic rings, molecular ratios of selected PAHs concentration, correlations among PAHs and PCA analysis indicated mixed sources of origin. The suggested mixed sources are pyrolytic activities (vehicular emissions, biomass and coal combustion and diesel engines) and petrogenic sources (from the spill of petroleum products and automobile workshops). The HYSPLIT back trajectories demonstrated that air masses originating from different sources from long distance and local sources impacted the samples before ending at the sampling sites during the study period.

The inhalation of PM_{10} with PAHs was considered for cancer risk assessment (ATSDR 2005; USEPA 2019). Based on estimated low CDI, CR for human adults (2.3×10^{-6}) and children (4.2×10^{-6}) due to inhalation of 16 PAHs through PM_{10} was within acceptable guidelines, suggesting low cancer risk to the human population in the north-western city of India. Due to low vehicular, commercial and industrial impact, comparatively low concentrations $\sum_{16}$PAHs and their corresponding low CR were observed at urban and suburban residential sites.

Due to limitation to exposure through inhalation, and lack of data on exposure through diet, total CDI and CR could not be estimated. Though low risk was observed during the present study, it is recommended to control the PAH emissions from burning of biomass including garbage, leaves, grass and crop residue. However, Government of India has undertaken various initiatives for mitigation of air pollution in India, such as National Clean Air Programme (NCAP), a central sector scheme to reduce 20–30% of PM by 2024 at 2017 base year (MoEF and CC 2019). Other several initiatives have been taken by the Government in the recent past for the improvement of environmental conditions in India by mitigation of air pollution. Those initiatives include phasing out of old vehicles in metropolitan cities, stringent industrial emission standards, closure/shifting of polluting industries in residential areas, metro rail transit system, mandatory use of CNG in urban public transportation, and switched over coal-based power plants to gasoline, reduction of ash content in coal for thermal power plants, benzene in gasoline and sulphur in diesel, and improvement of fuel quality standards (MoEF and CC 2019). As there are scarce reports on pollutants and health risk for the north-west of India, the present study indicates the importance of sixteen priority PAHs to understand their carcinogenic impact potential on human health.

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Author's contribution Dr. Simerpreet Kaur was involved in sampling, analysis, data compilation; Bhupander Kumar helped in data interpretation, graphics and manuscript writing; Dr. Paromita Chakraborty contributed to data and manuscript review and editing; Dr. Vaneet Kumar was involved in sampling and analysis; Dr. Navin Chandra Kothiyal helped in overall planning and guidance.

Declarations

Conflict of interest All authors declare that they have no conflict of interest.

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