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Seasonal and Spatial Occurrence and Distribution of Respirable Particulate-bound Atmospheric Polycyclic Aromatic Hydrocarbons in Hisar City (India) and Their Potential Health-risks

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Abstract: A group of nine PAHs was detected using HPLC in commercial, industrial and residential areas of Hisar city and their health effects were discussed. PAHs have detrimental biological effects, including acute and chronic toxicity, mutagenicity and carcinogenicity. An average of total PAHs in commercial, industrial and residential areas was found to be 1798.52, 2196.81 and 2474.13 µg/g, respectively. For seasonal changes, maximum variations were observed in monsoon followed by post-monsoon season, summer, autumn, and least in winter. Among the nine PAHs analysed, benzo(k)fluoranthene exhibited maximum concentration throughout the period of study. Significantly high variation was observed for benzo[b]fluoranthene followed by benzo(k)fluoranthene, pyrene and benzo(e)pyrene. Benzo[ghi]perylene > anthracene > fluoranthene > phenanthrene > naphthalene represented significantly low variations. Large PAHs were predominantly present and they represented large variations of concentration during the study. The presence of other air pollutants can affect the residence time of PAHs in air and the atmospheric chemical lifetimes of particle-bound PAH with respect to thermal reactions tend to be longer than those of gas-phase PAH.

Key words: PAHs, RSPM, carcinogenicity, health-risk.

Introduction

India has 23 major cities of over one million people and ambient air pollution levels exceed in many of them (Gupta et al., 2002). Pollution in these cities has associated serious to moderate health problems (Kaushik et al., 2006). About 4% to 8% premature deaths on global scale are due to exposure to high levels of particulate matter in ambient air (WHO, 2000). The particulate matter, especially the respirable suspended particulate matter (RSPM), is of major concern as most of the pollutants of air are associated with it and they have high probability to deposit on the respiratory tract (Pope et al., 1995). Heavy metals (Haritash and Kaushik, 2007), pesticides, and polycyclic aromatic hydrocarbons are the chiefly occurring toxic pollutants of air. PAHs are aromatic hydrocarbons with two or more fused benzene rings. They occur as colourless, white or pale yellow solids with low solubility in water, high melting and boiling points and low vapour pressure. With an increase in molecular weight, their solubility in water decrease, melting and boiling point increase and vapour pressure

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decreases (Clar, 1964; Patnaik, 1999). They are formed during the incomplete combustion at high temperature (500-800°C) or subjection of organic material at low temperature (100-300°C) for long periods of time.

The common sources of PAHs in environment include natural as well as anthropogenic. Natural sources include forest and rangeland fires, oil seeps, volcanic eruptions and exudates from trees. Anthropogenic sources of PAH include burning of fossil fuel, coal tar, wood, garbage, refuse, used lubricating oil and oil filters, municipal solid waste incineration and petroleum spills and discharge (Kaushik and Haritash, 2006). Due to their ubiquitous occurrence, recalcitrance, bioaccumulation potential and carcinogenic activity, the PAHs have gathered significant environmental concern. PAHs are widely distributed environmental contaminants that have detrimental biological effects, including acute and chronic toxicity, mutagenicity and carcinogenicity (IARC, 1983). Despite their early identification as environmental carcinogens, PAHs remained unnoticed for a long time. The fate of PAHs in environment includes volatilization, photo-oxidation, chemical oxidation, bioaccumulation, adsorption on soil particles, leaching and microbial degradation (Wild and Jones, 1995). They are present in every component of environment, but are chief pollutants of air. Studies have reported the presence of PAHs in ambient air of Ahmedabad (Raiyani et al., 1993) and Hyderabad (Suneela et al., 2004), water and sediments of Gomti river (Malik et al., 2004), sediments of Kolleru lake wetland (Rao, 2003), and agricultural soils in Delhi (Agarwal et al., 2009). PAHs released into atmosphere have a strong affinity for particles since they have lower vapour pressure and therefore, they can readily condense onto particulate matter in air and they move to distant places with the wind currents (McVeety and Hites, 1988). They occur in two phases in air. The low molecular weight (MW< 200) PAHs occur in vapour phase whereas the heavier PAHs (MW> 250) occur in particulate phase. They partition between the particulate phase and vapour phase depending upon the prevailing atmospheric conditions and the particular physical characteristics (Table 1) of the compounds themselves.

Keeping in view the toxicity associated with PAHs, the site-specific activities and sources, effect of meteorology on levels of PAHs, the present study was undertaken to assess the levels of different PAHs in RSPM in Hisar city of Haryana state.

Materials and Methods

Study Area

The present study was undertaken at Hisar city of Haryana state, India. Hisar is a sub-urban city located at 29°10′ North latitude and 75°46′ East longitude. Hisar has moderate to high industrial activity with a steel plant, a distillery, a number of small and medium scale industries and a fair number of registered vehicles. Most of the industries are located in industrial estate situated in its north-east outskirts. The climate of Hisar is characterized by its dryness and extremes of temperature. The mean temperature is 40°C and 10°C in summer and winter, respectively. On an average, there are 24 rainy days in a year with average annual rainfall of 450 mm. Generally, light winds blow but there occur dust-storms in summer and thunder-storms in monsoon. Weak inversions are commonly observed in winter.

Table 1: Physical and chemical properties of polycyclic aromatic hydrocarbons (PAHs)

S. No.	*Name*	*M.F.*	*CAS# registry No.*	*B.Pt.# (°C)*	*M.Pt.# (°C)*	*V.P.# (Pa at 25 °C)*	*Aqueous solubility (mg/l)**	*IARC@ group*
1	Benzo[k]fluoranthene	$C_{20}H_{12}$	207-08-9	480	215.7	5.2×10^{-8}	—	2B
2	Anthracene	$C_{14}H_{10}$	120-12-7	342	216.4	1×10^{-3}	0.015	3
3	Benzo[b]fluoranthene	$C_{20}H_{12}$	205-99-2	481	168.3	6.7×10^{-5}	—	2B
4	Benzo(e)pyrene	$C_{20}H_{12}$	192-97-2	493	178.7	4×10^{-7}	—	3
5	Fluoranthene	$C_{16}H_{10}$	206-44-0	375	108.8	1.2×10^{-3}	0.25	3
6	Naphthalene	$C_{10}H_8$	91-20-3	218	80.2	11	30	n.e.
7	Phenanthrene	$C_{14}H_{10}$	85-01-8	340	100.5	2×10^{-2}	1-2	3
8	Benzo[ghi]perylene	$C_{22}H_{12}$	191-24-2	500	277	6×10^{-8}	—	3
9	Pyrene	$C_{16}H_{10}$	129-00-0	150.4	393	6.0×10^{-4}	0.12-0.18	3

WGPAH, 2001; * Mackay et al., 1991; @ IARC, 1983

Sample Collection

For the collection of air samples, three sites namely, Industrial Estate (industrial area), Nagori Gate (commercial area) and Sector 15 (residential area) within the city were selected (Figure 1). The samples were collected from July to December, 2002 representing prolonged summer (July), monsoon (August), post-monsoon (September), autumn (October) and winter (December). The first monsoon rain occurred on 22 August, 2002. A group of nine PAHs, namely benzo(k)fluoranthene (B(k)F), pyrene (Py), benzo[b]fluoranthene (B(b)F), benzo(e)pyrene (B(e)P), anthracene (Anth), napthalene (Naph), benzo[ghi]perylene (Bply), phenanthrene (Phe), and fluoranthene (Fluo) was detected using HPLC. The collection of respirable suspended particulate matter (RSPM) was done using Envirotech make APM-460 Respirable Dust Sampler with a provision of separation of PM_{10} particles on glass fibre filter (GFF) sheet. The non-respirable suspended particulate matter falls through the cyclone's conical hopper and gets collected in the cyclonic cup. The fine dust comprising the respirable fraction of TSPM passes through the cyclone and gets collected on GFF. Meteorological data was collected from

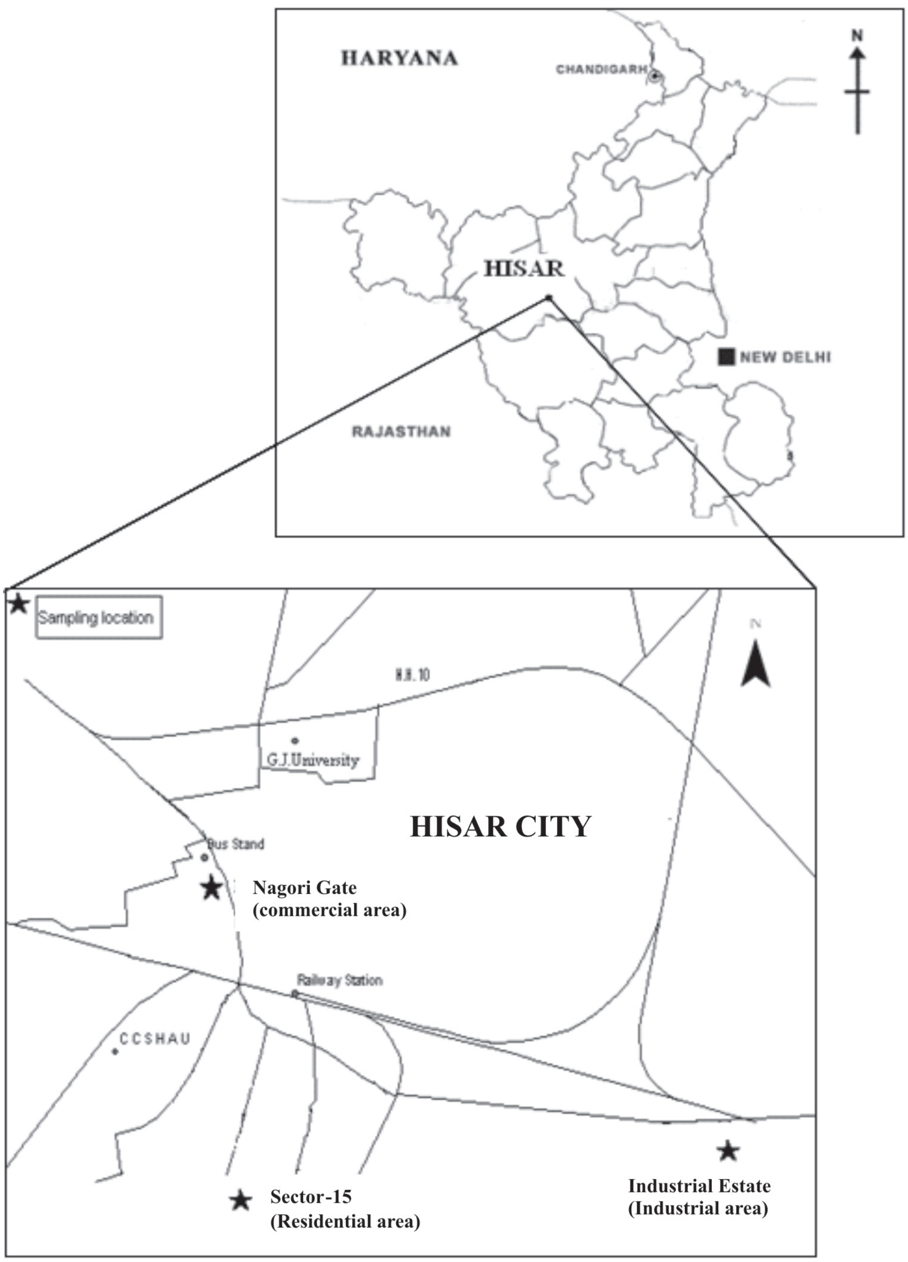

Figure 1: The location of sampling sites in Hisar city.

the Department of Meteorology, CCS Haryana Agriculture University, Hisar.

Sample Extraction and Analysis

The GFF sheet was used for the extraction of PAHs. Exposed filter was cut into four equal sizes and a single quarter was further cut into small pieces. The PAHs from the pieces were extracted in toluene using ultrasonic extraction for 30 minutes. The extracts were eluted through silica-sodium sulphate column and reduced to 5 ml volume using a rotary vacuum evaporator. Details of the method have been reported by Tyagi (1994). The extracts were analysed using High Pressure Liquid Chromatography (HPLC), Shimadzu LC 2010 CHT model with UV detector. The conditions for chromatographic analysis were—injection volume: 20 µL; mobile phase: acetonitrile/water gradient @ 1 ml/min; column: 2 × 250 mm, reverse-phase, 5-µm C18; detector: UV at a wavelength of excitation 254 nm; emission: 400 nm. PAH associated with RSPM was calculated using the following relation

$$\text{PAH in RSPM } (\mu g/g) = \frac{A\ (mg/ml) \times 4}{B\ (gm)}$$

where A is chromatographic PAH concentration (µg/ml) and B is respirable suspended particulate matter (gm).

Results and Discussion

Based on the chromatographic analysis of the extracts, PAHs have been found associated with various fractions of suspended particulate matter in ambient air of Hisar city.

Site-specific Variations

All the sampling locations in Hisar city represented considerable concentration of the nine PAHs. Nagori Gate (commercial-cum-vehicular area) represented an average concentration (µg/g) of 563.43, 191.19, 251.95, 307.92, 171.30, 33.98, 69.14, 94.26 and 115.36 for benzo(k)fluoranthene, pyrene, benzo[b]fluoranthene, benzo(e)pyrene, anthracene, napthalene, benzo[ghi]perylene, phenanthrene, and fluoranthene, respectively. The overall average for total PAHs was found to be 1798.52 µg/g. Higher levels for all PAHs were observed in the month of August except pyrene which was highest in the month of September. Chief sources of PAH in the area are vehicles and the diesel generator sets operating in commercial area during power failure. High vehicular density with frequent traffic jam at the intersection adds to the concentration as accelerating and decelerating vehicles produce more PAHs and RSPM (Kaushik and Haritash, 2006).

Industrial Estate (industrial area) represented average concentration (µg/g) of 570.42, 336.83, 421.09, 357.27, 181.11, 41.92, 88.09, 108.01 and 92.06 for benzo(k)fluoranthene, pyrene, benzo[b]fluoranthene, benzo(e)pyrene, anthracene, napthalene, benzo[ghi]perylene, phenanthrene, and fluoranthene, respectively. An average of total PAHs in industrial area was found to be 2196.81 µg/g. The month of September represented maximum concentration for all the PAHs. A large steel plant, several medium-scale pipe manufacturing units, vegetable oil plants etc. are operating in Industrial Estate and constitute the probable sources. The waste and refuse of small-scale units is often burnt which may add to the concentration (Haritash and Kaushik, 2007).

In Sector 15 (residential area), the average levels observed were 577.71, 384.23, 442.53, 401.88, 202.56, 76.59, 197.49, 69.04 and 122.10 µg/g for benzo(k)fluoranthene, pyrene, benzo[b]fluoranthene, benzo(e)pyrene, anthracene, napthalene, benzo[ghi]perylene, phenanthrene and fluoranthene, respectively. Highest levels were observed in the month of August and an overall average for residential area was observed to be 2474.13 µg/g. The probable sources are vehicles, and burning of wood and coal for space heating in winter. Proximity to the industrial area also adds to the levels. The site-specific variations for PAHs have been reported in Table 2 and Figure 2. It was observed that the average PAH concentration was maximum in residential area followed by industrial area and commercial-cum-vehicular area. There were no significant variations in commercial area but industrial and residential areas reported considerable variations.

Seasonal Variations

During the month of July (prolonged summer), the average concentration of PAHs was 1849.6 µg/g and it increased to about two times in monsoon (3617.23 µg/g). Though there was a slight increase in commercial and industrial area, a sharp hike was observed in residential area. The probable reason could be resuspension of industrial ash from the ash dumping-site owing to high wind-velocity during the season. The average concentration (3455.45 µg/g) dipped a bit during post-monsoon but it exhibited an unusual high in industrial area. The probable reason could be resuspension and/or an unnoticed event of refuge burning on the day of sampling. The average level was 1184.73 µg/g in autumn and it dipped to minimum in winter with an average of 675.43 µg/g. Stable

Table 2: Meteorological parameters and concentration of PAHs in respirable dust of Hisar during July to December, 2002

Date	*Mean Temp. (°C)*		*Wind Speed (km/hr)*	*Wind Direction*	*Relative Humidity (%)*		*Location**	*Concentration (µg/gm)*									
	Max	*Min*			*Morning*	*Evening*		*RSPM (gm)*	*B(K)f*	*Py*	*B(E)Ap*	*B(e)Py*	*Anth*	*Naph*	*Bply*	*Phe*	*Fluo*
July, 18, 2002	41.4	28.5	6.7	NE	64	39	C	0.2026	445.41	135.64	188.75	341.56	192.69	36.53	68.31	120.63	86.48
							I	0.1768	344.34	206.33	352.94	306.79	126.92	39.14	74.43	138.24	95.48
							R	0.0867	394.46	285.58	479.82	577.62	110.27	75.20	142.56	73.82	108.88
Aug., 23, 2002	31.5	26.5	8.4	ESE	90	49	C	0.1104	721.01	271.74	357.61	539.86	312.32	52.54	103.99	193.12	200.72
							I	0.1776	376.13	213.96	230.18	371.62	146.62	30.41	59.46	29.73	78.60
							R	0.0211	1243.60	1042.7	1175.4	1006.6	599.05	244.55	688.15	160.19	401.90
Sept., 28, 2002	35.6	20	3.3	NNE	86	32	C	0.0959	689.89	352.45	469.24	463.82	190.20	50.05	102.61	95.52	182.69
							I	0.0413	1433.41	1099.3	1363.7	934.62	514.29	110.41	241.16	320.58	214.04
							R	0.1299	295.00	432.33	263.59	219.25	152.12	27.71	56.97	49.27	42.19
Oct., 23, 2002	29.6	12.5	3.3	NNW	83	33	C	0.1993	506.37	129.65	193.68	131.46	106.57	19.87	44.76	50.18	72.45
							I	0.1909	334.21	137.45	128.86	117.97	79.83	19.28	49.03	39.81	45.47
							R	0.1184	516.89	118.24	221.28	168.24	121.62	23.99	85.14	54.05	37.84
Dec., 05, 2002	24.6	5.4	0.9	E	91	34	C	0.3612	454.49	66.45	50.50	62.90	54.71	10.92	26.02	11.85	34.44
							I	0.3759	364.03	27.13	29.80	55.33	37.88	10.38	16.39	11.71	26.71
							R	0.3306	438.60	42.35	72.60	37.63	29.76	11.49	14.64	7.86	19.72
Mean ± Standard deviation									570.52 ± 336.19	304.08 ± 331.50	371.86 ± 390.94	355.69 ± 304.05	184.99 ± 167.25	50.83 ± 60.01	118.24 ± 167.64	90.44 ± 85.34	109.84 ± 102.63

*C: Commercial; I: Industrial; R: Residential

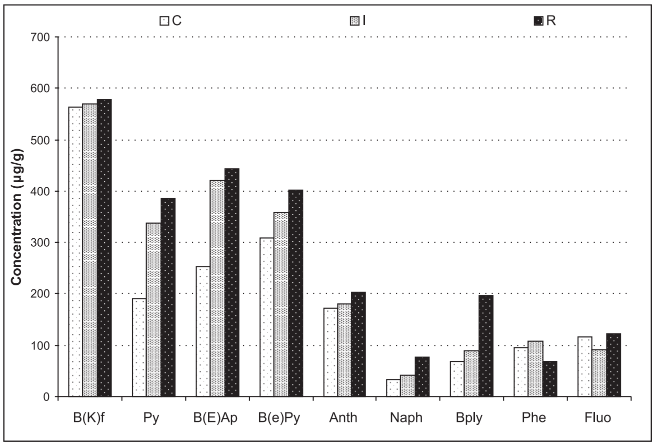

Figure 2: Average concentration PAHs (µg/g) in RSPM at commercial, industrial and residential areas during July to December, 2002.

atmospheric conditions, low-wind velocity, and high moisture content during winter-nights leads to formation of dew and scavenging of pollutants. The seasonal trend for the concentration was Monsoon > Post-monsoon > Summer > Autumn > Winter. Similar trend has been observed for heavy metals in Hisar city (Haritash and Kaushik, 2007). The seasonal variations have been presented in Table 2 and Figure 3. Maximum/significant variations were observed in monsoon followed by post-monsoon season. The variations were marginally low in summer followed by autumn and least in winter.

PAH-specific Variations

Among the nine PAHs analysed, benzo(k)fluoranthene exhibited maximum concentration throughout the period of study with an average of 570.52 µg/g. It constituted about 27% fraction of the total PAHs. It was followed by

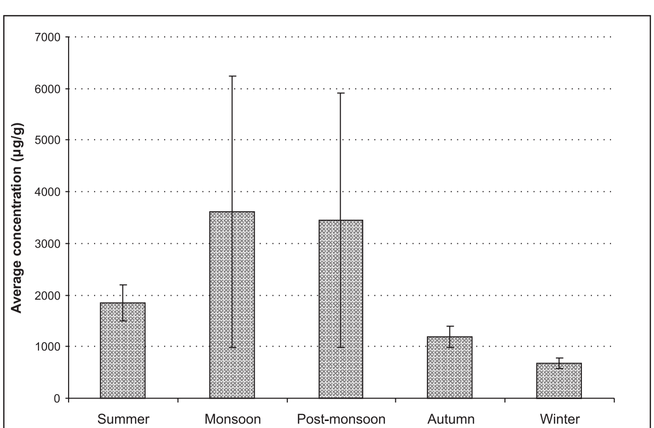

Figure 3: Average concentration of total PAH (µg/g) in RSPM in Hisar during July to December, 2002.

benzo[b]fluoranthene, benzo(e)pyrene, pyrene, anthracene, benzo[ghi]perylene, fluoranthene, phenanthrene and naphthalene with an average concentration (µg/g) of 371.86, 355.69, 304.08, 184.99, 118.24, 109.84, 90.44 and 50.83, respectively. The percent fraction of different PAHs is represented in Figure 4. Significantly high variation was observed for benzo[b]fluoranthene followed by benzo(k)fluoranthene, pyrene and benzo(e)pyrene. Benzo[ghi]perylene > anthracene > fluoranthene > phenanthrene > naphthalene represented significantly low variations.

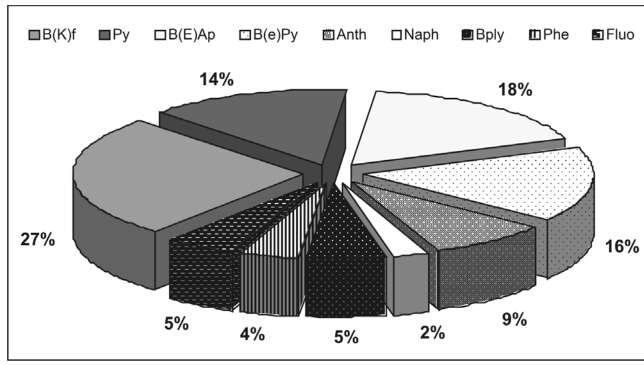

Figure 4: Percent fraction of different PAHs in RSPM in Hisar city during July to December, 2002.

It was observed that the large PAHs, i.e. with 4-6 rings, were predominantly present and they represented large variations of concentration during the study. The association of large PAHs with particulate phase could be reason for a high concentration of these PAHs. The reactions of particle-phase PAH under atmospheric conditions are not well characterized despite particulates being one of the more important vectors for PAH carcinogenicity in humans. Much of the uncertainty arises from the large variation in the composition of the air-borne particulate potentially containing the PAH, and the possible influence the nature of the substrate has on the chemical reaction rates. Most of the small PAHs (2-3 rings) are predominantly present in vapour phase and they are susceptible to photochemical degradation. The tropospheric ozone is an important PAH-regulating factor. Its presence can significantly enhance the representative chemical half-life of vapour-phase PAHs (Atkinson and Arey, 1994; Brubaker and Hites, 1998), and can reduce the half-life of 4-6 ring particle-phase PAHs (PORG, 1997). On the other hand, the thermal (non-photolytic) reaction with oxides of nitrogen can significantly enhance half-life of particle-bound as well as vapour-phase PAHs (Wayne et al., 1991). The atmospheric chemical lifetimes of particle-bound PAH with respect to thermal reactions tend to be longer than those of gas-phase PAH (WGPAH, 2001).

Pearson correlation of the nine PAH compounds revealed that a strong significant positive correlation ($p < 0.01$) exists between them except a weak correlation between phenanthrene and naphthalene (0.580; $p < 0.01$), and non-significant weak correlation between phenanthrene and benzoperylene (0.500). The significant positive correlation between the compounds suggests of a common source of origin.

Health Aspects of PAHs

PAHs were, perhaps, the first recognized environmental carcinogens in 18th century by Sir Percival Pott. He reported the high incidence of scrotal cancer among the chimney-sweepers due to the soot accumulated in their scrotal folds. Another study showed that coke oven workers exposed to high concentration of PAHs had reduced level of serum immunoglobulins. High lung cancer rate has also been associated with smoky coal used (Xu et al., 1994). A positive correlation has also been established for PAH-DNA adducts in placentas from women burning coal without chimney. Air pollution is significantly associated with the amount of PAH bound to DNA in both maternal and infant cord WBCs. Newborns with high PAH-DNA adducts had low birth weight, birth length and head circumference (Perera et al., 2005). Experimental evidences suggest that developing foetus is more susceptible to the effect of PAHs. The factors responsible for it are proliferation rates, detoxification capabilities, and DNA repair capacity. In India, human placenta, umbilical cord blood, maternal blood and breast milk from mothers were analyzed for the presence of PAHs and a relatively high concentration was observed in umbilical cord blood and breast milk samples showing that developing foetus can be exposed to these contaminants in-utero (Madhavan and Naidu, 1995). In another study, analysis of umbilical cord blood of babies revealed that DNA of child was damaged by PAH (Edwards, 1996). Another study on Dutch children revealed a high association between urinary 1-hydroxypyrene (1-HP) concentration and the creatinine adjusted 1-HP concentration and PAH exposure through air and soil (Winjen et al., 1996).

The International Agency for Research on Cancer (IARC) has classified 48 PAH compounds likely to be carcinogenic to humans. According to a public health statement by the Agency for Toxic Substances and Drug Registration (ATSDR), benz(a)anthracene and benzo(a)pyrene are probably carcinogenic to humans and benzo(b)fluoranthene and indeno(1,2,3-c,d)pyrene are possibly carcinogenic to humans. Several epidemiological studies have suggested increased mortality from lung cancer in humans exposed to PAH through coke oven emission, roofing tar emission and cigarette smoke. The estimated risk of lung cancer from human epidemiological data is 1.3×10^{-4} to 2.4×10^{-3} $\mu g/m^3$ (IARC, 1983).

Conclusion

All the sampling locations in Hisar city represented considerable concentration of PAHs. An average of total PAHs in commercial, industrial and residential areas was found to be 1798.52, 2196.81 and 2474.13 µg/g, respectively. There were no significant variations in commercial area but industrial and residential areas reported considerable variations. The major sources in commercial area are vehicles and the diesel generator sets, steel plant and several small and medium units in industrial area, and vehicles, re-suspension of road-side dust, burning of wood etc. in residential area. Proximity to industrial area and wind direction, favours dispersion of pollutants generated in industrial area to the residential which is a cause of concern w.r.t. health. The seasonal trend for the concentration was monsoon > post-monsoon > summer > autumn > winter. The probable reason could be high wind speed in monsoon and post-monsoon, which leads to formation of turbulent conditions leading to re-suspension of industrial ash laden with high concentration of PAHs.

Photochemical reactions also affect the concentration. Stable atmospheric conditions, low-wind velocity and high moisture content during winter-nights lead to formation of dew and scavenging of pollutants. So, meteorology plays a chief role in regulating the levels of atmospheric particle-bound PAHs. Significantly high variation was observed for benzo[b]fluoranthene followed by benzo(k)fluoranthene, pyrene and benzo(e)pyrene because of the association of large PAHs with particulate phase. The longer atmospheric chemical lifetimes of particle-bound PAH than of gas-phase PAH could also be the reason. Most of the PAHs are carcinogens and mutagens and carcinogenicity of 4-7 ring PAHs is more. Reaction of PAHs with oxides of nitrogen, a criteria pollutant, can increase their half-life as well as toxicity. Association of such nitro-PAHs with respirable dust, especially in residential area, bears potential health risks.

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