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Short communication

Preliminary chemical characterization of particle-phase organic compounds in New Delhi, India

Dhruv N. Sharma^a, Aniket A. Sawant^b, R. Uma^c, David R. Cocker III^{b,*}

^a Department of Civil Engineering, Indian Institute of Technology Delhi, New Delhi 110016, India

^b Bourns College of Engineering—Center for Environmental Research and Technology, University of California, Riverside CA 92521, USA

^c The Energy and Resources Institute, New Delhi 110003, India

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Abstract

This work describes a pilot study for the investigation of the chemical composition of organic species present in PM₁₀ collected at a residential site in New Delhi during December 2001–March 2002, and in TSP emissions from in-use two-stroke vehicles. Both ambient and emissions samples were analyzed using gas chromatography-mass spectrometry (GC-MS), with an emphasis on quantification of non-volatile polycyclic aromatic hydrocarbons (PAHs) and marker compounds in ambient PM₁₀. Preliminary findings suggest that vehicular emissions and biomass and/or refuse burning are significant contributors to the organic fraction of PM₁₀ in the New Delhi atmosphere.

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1. Introduction

Atmospheric particulate matter is known to contain a number of organic species, including alkanes, alkenes, carboxylic acids, carbonyl compounds, and aromatic compounds (Schauer et al., 1996). Polycyclic aromatic hydrocarbons (PAHs) are a class of aromatic compounds found in ambient particulate matter and shown by several investigators to be carcinogenic and/or mutagenic (Eisenstadt and Gold, 1978; Horikawa et al., 1991; Larsen and Larsen, 1998).

Mass concentrations of PAHs have been reported for Indian cities including Mumbai, Ahmedabad, and Nagpur (Kulkarni and Venkataraman, 2000, and references therein; Mohan Rao et al., 1983; Aggarwal et al., 1982; Venkataraman et al., 1999). In this note, we report the results of chemical analysis of the organic

fraction of ambient particulate matter of aerodynamic diameter < 10 μm (PM₁₀), for a residential site in New Delhi, India, with emphasis on non-volatile PAHs. We also describe a preliminary chemical analysis of TSP emissions from in-use two-wheelers with two-stroke gasoline engines.

The city of New Delhi (latitude 28°35'N, longitude 77°12'E) is the administrative capital of India and is located within the Delhi metropolitan region. Delhi lies in northern India and is roughly equidistant from the Great Indian Desert to the west and the Indo-Gangetic Plains to the east and southeast. The Delhi metropolitan region has a population of more than 14 million. It has an in-use vehicle population of more than 2.7 million (CPCB, 2001a), the largest in India. This is an estimate only, since vehicles in Delhi are not deregistered. Delhi also has a number of medium- to small-scale industries. Aneja et al. (2001) reported TSP mass concentrations in the range of 250–550 μg m⁻³ at a monitoring site in downtown New Delhi. Although it is commonly

*Corresponding author. Fax: +1-909-781-5790.

E-mail address: dcocker@engr.ucr.edu (D.R. Cocker III).

believed that emissions from vehicles (67%), coal-based power plants (13%), industrial units (12%) and domestic sources (8%) are the main contributors to poor air quality in Delhi (CPCB, 2001b), source contributions with respect to a specific type of pollutant (e.g., PM_{10}) have not been reported in the literature.

2. Experimental methods

2.1. Ambient sampling

The substrates for ambient sampling were obtained from The Energy and Resources Institute (formerly the Tata Energy Research Institute, TERI, Lodhi Road, New Delhi). The samples were taken at a permanent monitoring site located at the India Habitat Centre (IHC), Lodhi Road, in the southern part of New Delhi. IHC is a multi-use complex containing a cultural center, offices and dining areas. There are no industries located within the IHC complex. Lodhi Road is a four-lane east-west thoroughfare with a motorized vehicle count of 5333 vehicles h^{-1} (Tiwari, 2002). IHC may be considered representative of a typical Delhi residential area, considering the traffic flows and nature of pollutant sources (Fig. 1). The average fleet composition in New Delhi, estimated for July 2002, approximately comprised 55% two-stroke gasoline vehicles, 37% four-stroke gasoline vehicles, 6% diesel vehicles and 2% compressed natural gas (CNG) vehicles (CPCB, 2001a; Tiwari, 2002).

Samples at IHC were taken using an Envirotech (New Delhi, India) APM 460 high-volume sampler having a

nominal particle cutoff diameter of $10\mu m$ (PM_{10}). Sampling was conducted at a height of 3 m above ground level. Ambient PM_{10} samples were collected on $8'' \times 10''$ Whatman GF/A glass fiber filters having a pore size of $1.6\mu m$ (Whatman, Kent, UK). Each PM_{10} sample was collected for 8 h at a nominal flow rate of $1 m^3 min^{-1}$. The flow rates through the high-volume sampler were monitored once per hour. The sampler flow meter was calibrated once a month. Mass concentrations of the filters were determined by weighing on an electronic balance of $10\mu g$ precision. Field blanks were used to ensure accuracy of weights. Sampling details and measured PM_{10} mass concentrations are presented in Table 1.

2.2. Emissions source sampling

Five two-stroke gasoline vehicles were sampled at a parking lot near the security cabin of the Indian Institute of Technology Delhi. The sampler used was an in-house design, based on a commercially available portable vacuum device. Vehicle exhaust was collected on 47 mm diameter Millipore AP40 binder-free glass fiber filters (Millipore Corporation, Bedford, MA, USA). Raw vehicle exhaust was pulled 4–5 cm from the end of the tailpipe. Engines were allowed to warm up prior to testing. Tests were conducted under idling conditions. The list of vehicles sampled is given in Table 1.

2.3. Methods of analysis

All filters were analyzed in the Analytical Laboratory at the Bourns College of Engineering—Center for

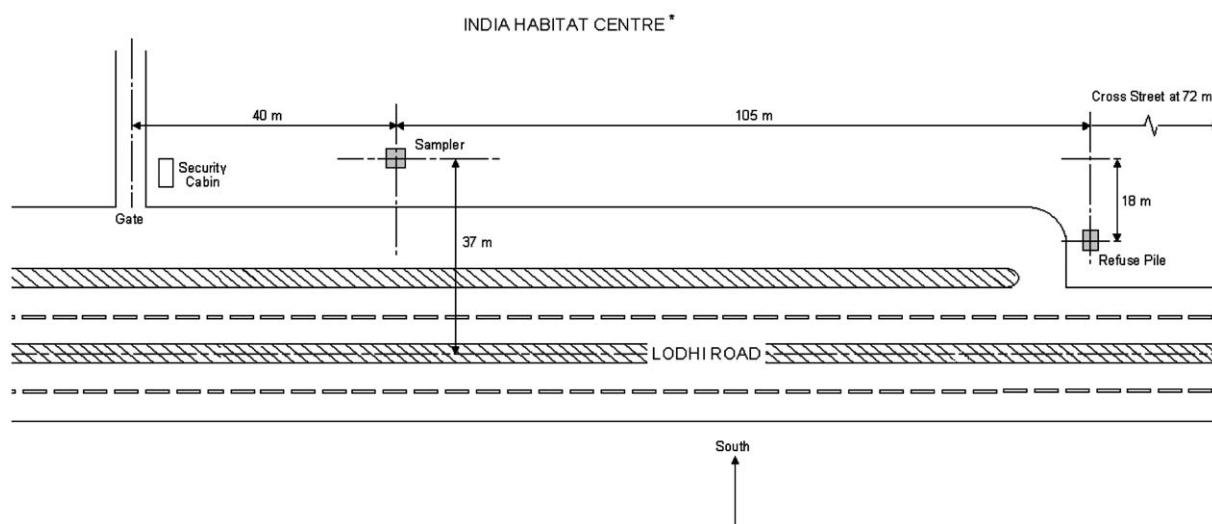


Fig. 1. Schematic of monitoring site. (*Note: The IHC is a multi-use complex having a cultural center, offices and dining areas. There are no known significant stationary sources of PM_{10} within the IHC.)

Table 1
Sampling details

Filter ID	Date	Time of day (h)	Initial flow rate ($\text{m}^3 \text{min}^{-1}$)	Final flow rate ($\text{m}^3 \text{min}^{-1}$)	Duration (min)	PM ₁₀ mass concentration ($\mu\text{g m}^{-3}$)
(a) Ambient PM ₁₀ samples						
1	12/04/2001	2200–0600	1.13	1.00	484	262
2	12/18/2001	2200–0600	1.20	1.10	478	261
3	01/01/2002	1400–2200	1.18	0.90	448	578
4	01/31/2002	0600–1400	1.28	1.13	477	231
5	02/02/2002	1400–2200	1.15	1.05	475	248
6	02/17/2002	0600–1400	1.10	0.90	458	311
7	03/20/2002	0600–1400	1.18	1.10	479	296
8	03/23/2002	1400–2200	1.10	1.00	489	146
9	03/25/2002	0600–1400	1.10	0.90	481	169
10	03/31/2002	2200–0600	0.95	0.95	478	173

Note: Location for all samples is: India Habitat Centre (IHC), Lodhi Road, New Delhi. Sampling was carried out for 24 h in 8-h intervals.

Vehicle ID	Year	Make	Model	Odometer (km)	Sampling time (min:s)
(b) Two-stroke TSP samples					
A	1984	LML	(SC/2T)	63,882	2:30
B	1993	Bajaj	Chetak (SC/2T)	~220,000	2:30
C	1996	LML	(SC/2T)	27,153	2:30
D	1989	Bajaj	Super (SC/2T)	147,662	2:30
E	1994	Yamaha	RX100 (MC/2T)	~80,000	10:00

Note: SC: scooter; MC: motorcycle; 2T: two-stroke.

Environmental Research and Technology (CE-CERT), University of California, Riverside (UCR). Sample extraction with dichloromethane (CH_2Cl_2) was carried out at 2000 psi and 100°C for approximately 25 min using a Dionex ASE 200 Accelerated Solvent Extractor (Dionex, Sunnyvale, CA, USA). Vehicular samples were extracted into 11 ml of CH_2Cl_2 , while the ambient samples were extracted into 22 ml of CH_2Cl_2 . Vehicular samples were concentrated to 1 ml with a stream of helium gas, while ambient samples were not concentrated. Both vehicular and ambient samples were injected into an Agilent 6890N gas chromatograph (Agilent, Palo Alto, CA, USA) with an Agilent 5973N mass selective detector equipped with a 30 m, $0.25 \mu\text{m}$ DB-5 column. The oven temperature program was as follows: the temperature was held at 50°C for 2 min; then increased at 8°C min^{-1} to 320°C and finally held constant at that temperature for 5 min (EPA TO-13A method; US EPA, 1999). Identification and quantification of compounds was carried out using multiple standards of PAHs, *n*-alkanes, and other organics. Compounds were quantified based on ratios of area counts of compounds in the sample to those of the corresponding standards of known concentration, accounting for the total extracted volume and the sampling flow rate.

3. Results and discussion

3.1. Ambient concentration of PAHs at the monitoring site

In the present study, an attempt has been made at quantification of the ambient concentrations of seven PAHs, namely, benzo[fluoranthene]s (i.e., all 3 isomers (b)+(j)+(k)) (BFa), benzo(e)pyrene (BeP), benzo(a)pyrene (BaP), indeno[1,2,3-cd]pyrene (IPY), dibenz[a, h]anthracene (DBa), benzo[ghi]perylene (BgP), and coronene (COR). It should be noted that although lower molecular weight PAHs such as fluoranthene, pyrene, chrysene and benz(a)anthracene were also quantifiable, they were excluded from the analysis due to significant volatilization losses expected during high-volume sampling using glass-fiber filters (Coutant et al., 1988). The analysis was therefore limited to higher molecular weight PAHs with 5 rings or more, which are almost entirely particle-bound (Grosjean, 1983; Cauntreels and Van Cauwenbergh, 1978).

The sum of measured particle-phase PAH concentrations (5 rings or more; Table 2) at the monitoring site were found to range from 35.82 to 162.76 ng m^{-3} . These values are comparable to those reported by TERI (2001) and fall within the range of particle-phase PAH

Table 2
Quantification and comparison

Filter ID	1	2	3	4	5	6	7	8	9	10	Compd. ID ^a
<i>(a) Organic species in ambient PM₁₀ measured at IHC monitoring station, Lodhi Road (ng m⁻³)</i>											
Date	12/04/01	12/18/01	01/01/02	01/31/02	02/02/02	02/17/02	03/20/02	03/23/02	03/25/02	03/31/02	
Start time-end time (h)	2200–0600	2200–0600	1400–2200	0600–1400	1400–2200	0600–1400	0600–1400	1400–2200	0600–1400	2200–0600	
PAHs											
Benzo(bjk)fluoranthene (BFa)	22.78	30.29	25.97	18.67	23.60	24.06	13.98	3.31	15.17	9.40	A
Benzo(e)pyrene (BeP)	5.68	9.84	7.88	5.37	6.91	8.08	4.66	1.15	7.98	3.03	A
Benzo(a)pyrene (BaP)	26.26	48.00	27.99	14.27	15.59	31.30	21.64	18.44	17.87	16.58	A
Indeno[1,2,3-cd]pyrene (IPY)	21.48	26.85	18.09	26.26	16.50	21.07	6.38	5.00	7.45	4.96	A
Dibenz[a,h]anthracene (DBa)	BDL ^b	BDL ^b	BDL ^b	BDL ^b	BDL ^b	BDL ^b	BDL ^b	BDL ^b	BDL ^b	BDL ^b	A
Benzo[ghi]perylene (BgP)	34.49	44.02	18.90	15.07	23.77	30.97	10.14	7.92	16.95	7.90	A
Coronene (COR)	5.78	3.75	6.20	1.77	5.73	2.71	0.51	BDL ^b	9.77	BDL ^b	A
Total PAHs	116.48	162.76	105.05	81.40	92.09	118.19	57.32	35.82	75.19	41.88	
<i>n</i>-Alkanes											
<i>n</i> -Heneicosane	25.46	28.31	24.00	11.25	27.63	14.33	14.59	6.68	10.22	5.84	B
<i>n</i> -Docosane	48.65	50.65	49.65	24.95	52.64	21.49	26.58	10.03	15.74	7.95	A
<i>n</i> -Tricosane	66.66	64.77	60.18	35.37	67.53	26.14	31.74	12.26	18.28	9.87	B
<i>n</i> -Tetracosane	78.73	61.78	58.65	37.79	66.65	25.86	30.01	15.97	19.53	10.53	A
<i>n</i> -Pentacosane	67.27	62.94	69.13	34.12	61.80	28.79	28.84	16.04	21.80	14.90	B
<i>n</i> -Hexacosane	57.20	55.70	61.23	31.23	52.24	30.37	30.49	14.53	24.24	13.13	A
<i>n</i> -Heptacosane	54.83	66.45	69.14	35.32	48.97	41.60	35.48	20.74	38.47	20.16	B
<i>n</i> -Octacosane	39.50	51.59	43.95	23.66	35.69	19.85	23.82	8.89	18.59	9.64	A
<i>n</i> -Nonacosane	59.02	83.96	82.80	51.23	54.93	57.53	60.92	38.75	70.14	33.79	B
<i>n</i> -Triacontane	29.05	23.11	32.55	16.34	24.35	11.15	14.49	7.36	8.13	3.69	B
<i>n</i> -Hentriacontane	35.04	48.76	36.45	18.31	27.12	14.06	23.48	15.47	10.26	8.37	B
Total <i>n</i> -alkanes	561.42	598.03	587.74	319.57	519.55	291.17	320.46	166.72	255.41	137.87	
Other marker compounds											
Syringaldehyde	30.35	44.10	57.29	34.45	35.24	18.62	31.45	1.83	27.73	11.52	A
Propionyl syringol	23.34	26.48	36.95	31.02	32.78	16.05	17.78	2.04	15.88	2.29	A
28-Nor-17α/β-H-hopane	186.71	205.73	181.27	88.45	140.70	121.51	60.20	74.90	63.00	35.43	B
Levogluconan	1324.04	1581.73	1557.80	235.06	131.73	248.91	37.79	305.89	354.45	294.91	B
β-sitosterol	181.51	206.27	222.55	83.41	141.17	33.15	21.76	17.26	11.79	12.56	A

PAH	New Delhi ^c	Mumbai ^d	Algiers ^e	Santiago de Chile ^f	Birmingham ^g
<i>(b) Comparison of ambient PAH concentrations in different cities (ng m⁻³)</i>					
Benzo(b)kfluoranthene	3.3–30.3	7.7–9.9	0.7–5.3	0.1–3.1	0.5–3.0
Benzo(e)pyrene	1.2–9.8	NA ^h	0.2–3.1	0.0–0.9	NA ^h
Benzo(a)pyrene	14.3–48.0	1.7–1.9	0.1–2.1	0.0–0.7	0.2–0.7
Indeno[1,2,3-cd]pyrene	5.0–26.9	1.9–2.1	0.0–4.9	0.0–0.3	0.4–2.0
Dibenz[a,h]anthracene	BDL ^b	NA ^h	0.0–0.3	NA ^h	0.1–0.8
Benzo[ghi]perylene	7.9–44.0	NA ^h	0.2–4.8	0.1–1.3	0.8–1.9
Coronene	BDL ^b –9.8	NA ^h	NA ^h	0.1–0.7	0.3–1.0

^aCompound ID—Used for compound identification (Adapted from Rogge et al., 1993). A—positive, when the sample mass spectrum was identical to that of an authentic standard plus the library spectrum and also the retention time data for the sample and standard agreed quite well; B—probable, same as above except that no authentic standards were available.

^bBDL—below detection limit (<0.4 ng m⁻³).

^cRange of ambient concentrations from this study.

^dKulkarni and Venkataraman (2000). Range between residential and traffic sites in Mumbai for November–December 1996. Does not include benzo(j)fluoranthene.

^eYassaa et al. (2001). Range between May and September 1998 in downtown Algiers.

^fKavouras et al. (1999).

^gHarrison et al. (1996). Range between summer and winter 1992, Birmingham University campus, Edgbaston, UK. Does not include benzo(j)fluoranthene.

^hNA—data not available.

concentrations (23–190 ng m⁻³) reported for various Indian cities (Kulkarni and Venkataraman, 2000, and references therein). Comparisons of PAH values in this study with those reported for other major cities are reported in Table 2.

3.2. Ambient concentrations of organic markers

Total average *n*-alkane concentrations during the study period ranged from 137.87 to 598.03 ng m⁻³. In general the *n*-alkane concentrations in New Delhi were higher than those measured in Barcelona (198–314 ng m⁻³; Gogou et al., 1994) and Los Angeles (20–146 ng m⁻³; Rogge, 1993) but lower than those measured in Santiago de Chile (452.30–1439.12 ng m⁻³; Kavouras et al., 1999).

The high level of 28-nor-17alpha/beta-H-hopane in the ambient samples is an indicator of unburned petroleum emissions from vehicular traffic (Rogge et al., 1993). The significantly high levels of levoglucosan and the major contribution of benzo(a)pyrene over all other PAHs indicate the presence of wood smoke, which can be attributed to biomass burning (Hornig et al., 1983; Raiyani et al., 1993). The high concentrations of β -sitosterol (Fine et al., 2002), phthalates (probably from plastic burning; ARB, 1997) and a number of other compounds characteristic of wood burning like syringaldehyde and propionyl syringol (Nolte et al., 2001) are all indicators of biomass and/or refuse burning at this site. The relative abundance of PAHs and other marker compounds indicate that biomass and/or refuse burning and emissions from motor vehicles dominate the PAH loading at the Lodhi Road site.

3.3. Source profile of two-stroke gasoline vehicles

TSP samples collected from two-stroke gasoline vehicles were analyzed using the same instrumentation and techniques used for ambient PM₁₀ sample analysis. The ratios of PAHs to benzo(e)pyrene (Fig. 2) have been calculated for five in-use two-stroke gasoline vehicles in New Delhi. Benzo(e)pyrene was chosen as the reference compound because of its relatively higher stability and because it is found almost exclusively in the particle phase (Daisey et al., 1986). PAHs such as benzo[ghi]perylene, indeno[1,2,3-cd]pyrene and coronene, are seen to be present in substantial quantities, a characteristic of spark ignition engines (Rogge et al., 1993; Harrison et al., 1996; Miguel et al., 1998).

4. Conclusion

A number of organic compounds present in ambient PM₁₀ in a residential area in New Delhi were quantified in this study. Qualitative analysis using marker

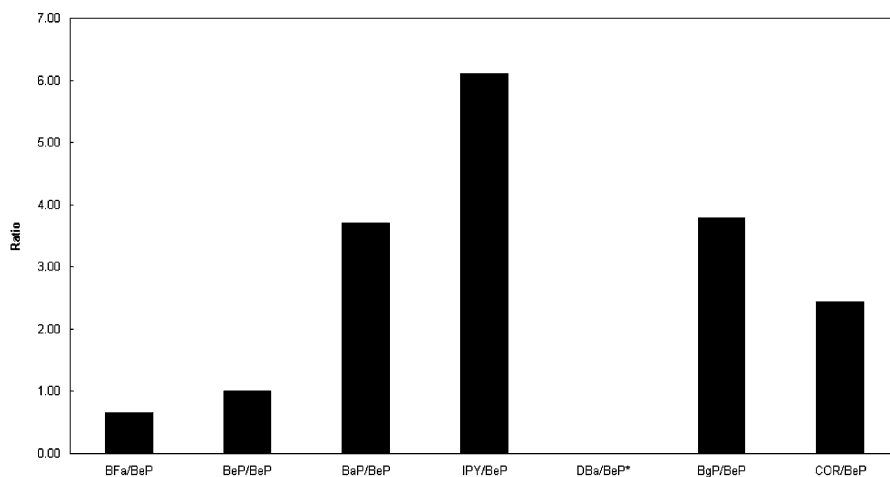


Fig. 2. Average ratios of selected PAHs to benzo(e)pyrene for two-stroke gasoline vehicles. (*Note: The measured concentrations of Dibenz[a,h]anthracene were below detection limit ($<0.4 \text{ ng m}^{-3}$).

compounds suggested that biomass and/or refuse burning and motor vehicle exhaust emissions were significant contributors to the organic fraction of ambient PM_{10} in this area. It should be noted that this was a preliminary study, carried out with the ultimate objective of conducting a large-scale source apportionment study for the Delhi area. Comprehensive long-term measurements of organic and inorganic ambient aerosol constituents and local source signatures are required in order to gain a more complete picture of the particulate air quality in this urban airshed.

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