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Speciation of ambient fine organic carbon particles and source apportionment of $PM_{2.5}$ in Indian cities

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[1] Fine particle organic carbon in Delhi, Mumbai, Kolkata, and Chandigarh is speciated to quantify sources contributing to fine particle pollution. Gas chromatography/mass spectrometry of 29 particle-phase organic compounds, including *n*-alkanes, polycyclic aromatic hydrocarbons (PAHs), hopanes, steranes, and levoglucosan along with quantification of silicon, aluminum, and elemental carbon are used in a molecular-marker based source apportionment model to quantify the primary source contributions to the $PM_{2.5}$ mass concentrations for four seasons in three sites and for the summer in Chandigarh. Five primary sources are identified and quantified: diesel engine exhaust, gasoline engine exhaust, road dust, coal combustion, and biomass combustion. Important trends in the seasonal and spatial patterns of the impact of these five sources are observed. On average, primary emissions from fossil fuel combustion (coal, diesel, and gasoline) are responsible for about 25–33% of $PM_{2.5}$ mass in Delhi, 21–36% in Mumbai, 37–57% in Kolkata, and 28% in Chandigarh. These figures can be compared to the biomass combustion contributions to ambient $PM_{2.5}$ of 7–20% for Delhi, 7–20% for Mumbai, 13–18% for Kolkata, and 8% for Chandigarh. These measurements provide important information about the seasonal and spatial distribution of fine particle phase organic compounds in Indian cities as well as quantifying source contributions leading to the fine particle air pollution in those cities.

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1. Introduction

[2] Urban areas in India experience very high concentrations of airborne fine particulate matter (PM) (i.e., PM with an aerodynamic diameter less than 2.5 μm, PM_{Fine}, or $PM_{2.5}$), [*United Nations Development Programme and World Bank Energy Sector Management and Assistance Program*, 2004] and studies have shown that at even much lower levels, particulate matter contributes to visibility problems and are likely responsible for respiratory and cardiopulmonary diseases like asthma, bronchitis, and heart disease [*Dockery et al.*, 1993; *Pope et al.*, 2002; *Agarwal et al.*, 2002; *Chhabra et al.*, 2001; *Yu et al.*, 2001]. Effective strategies to mitigate such a problem rely on characterizing and identifying the sources of the PM that cause the highest exposure to humans [*Smith*, 1993]. Over the past several decades, a series of methods have been developed for diagnosing the relative importance of the various emissions sources leading to fine particle air pollution problems in urban areas. Source-oriented methods of analysis exist that rely on atmospheric transport models driven by detailed emissions inventories. However, this approach has been inhibited by the lack of chemically speciated emission inventories. A different approach to identifying source impacts is to use receptor-based techniques which rely on observations at sampling sites (receptors) and key tracer species. This method uses the differences in chemical composition of particulate matter emitted from different sources to identify the presence of particles from specific sources. The receptor-based method has been widely used worldwide [e.g., *Chow et al.*, 1992; *Zeng and Hopke*, 1989] and are particularly attractive for application in regions that have not been studied in detail because they are able to yield rapid insights into the causes of a local air pollution problem before the completion of an accurate emissions inventory. In the Indian subcontinent, such a receptor-based source apportionment has been conducted for Bangladesh and in India using inorganic elemental analysis of $PM_{2.5}$ [*Begum et*

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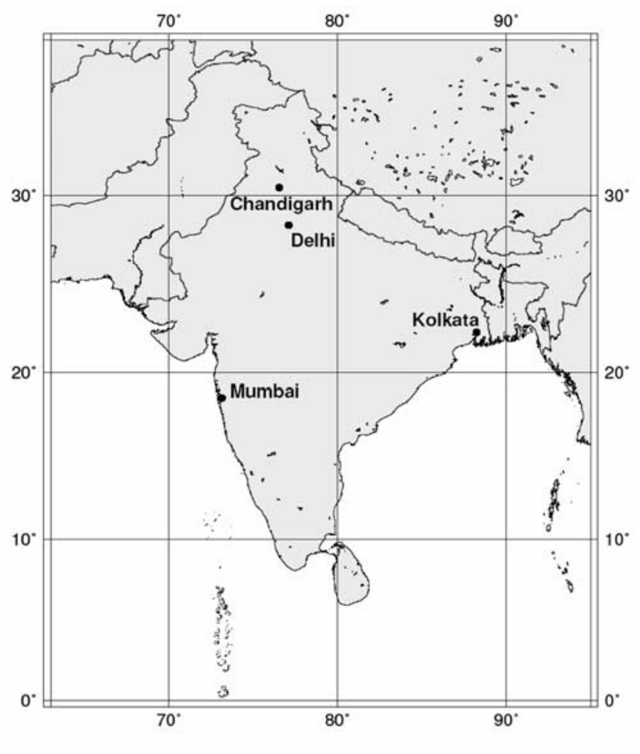

Figure 1. Location of the four sampling sites: Delhi, Mumbai, Kolkata, and Chandigarh.

al., 2004; *Kulkarni*, 2006]; however, no receptor-based source apportionment work using organic compounds as tracers has been conducted in this region. A significant fraction of the fine particulate mass in the urban atmosphere is organic and organic compounds present in fine particles emitted from burning wood, combustion of automotive fuels, and combustion of coal are very different and can be exploited to determine their respective contributions to the atmospheric concentrations of fine particulate matter, as demonstrated in Los Angeles by *Schauer et al.* [1996] and in the southeast United States by *Zheng et al.* [2002]. Here, this receptor-based organic tracer method has been used to quantify the sources that contribute to $PM_{2.5}$ at four cities in India.

2. Experimental Setup

[3] Ambient sampling over a 1-year period was conducted in Delhi, Mumbai, and Kolkata (Figure 1), three megacities located in India. To understand the seasonal pattern observed in the region, samples were taken during March/April (spring), June/July (summer), October/November (autumn), and December/January (winter). For each of the four seasons sampled, samples were taken every 6th day until at least five to six samples were obtained for that season in that site. After analyzing 5 years of backwind trajectories from NOAA [*Draxler and Rolph*, 2003; *Rolph*, 2003], Chandigarh, upwind of Delhi, was chosen as a fourth site representing background characteristics. It is a smaller city with a population of 809,000 (considered small in the Indian context), located north of Delhi. Five samples during the summer season were obtained at Chandigarh. There are very few data sets in these large cities that represent significant exposures to many people, and thus even small data sets like this can be useful in characterizing air pollution.

[4] Sites at Delhi, Mumbai, and Kolkata were selected to avoid undue influence of emissions coming from nearby city traffic or industries, yet were located within the metropolitan area (see Table 1). In each location, a four-channel, $PM_{2.5}$ filter sampler (see Figure 2) was placed either on a rooftop or in the middle of open field to ensure that the sampler inlet was able to sample particles coming from all directions. The Chandigarh site was located outside of the town. Sampling commenced on 4 March 2001 and continued until 16 January 2002. Samples were collected at ambient temperatures and relative humidities for 24 hours starting at midnight local time every 6th day for each of the months sampled, leading to 21 samples for Delhi, 25 for Mumbai, 20 for Kolkata, and 5 for Chandigarh.

[5] Fine particulate matter was collected on one quartz fiber filter (Pallflex, 2500 QAO, 47 mm diameter), two prewashed Nylon Filters (Gelman Sciences, Nylasorb, 47 mm diameter), and on two PTFE filters (Gelman Sciences, Teflo, 1.0 μm pore size). Ambient air was drawn at a rate of approximately 22.5 lpm through an acid-washed Pyrex glass inlet line to a Teflon-coated Air and Industrial Hygiene

Table 1. Description of the Sampling Sites

City	Site Address in India	Location Type	Site Description	Source of Pollution
Mumbai	NEERI Zonal Lab 89/B, Dr. Annie Basen Rd. Worli, Mumbai 400018	Urban Residential	Sampler placed 3 m above ground on a rooftop. A four-story building and slum area nearby.	City traffic typically seen in residential and business areas, and cooking by slum dwellers.
Delhi	National Physical Lab Dr. K. S. Krishnan Marg New Delhi 110012	Urban Residential	Sampler placed 5 m above ground on rooftop of office building in NPL campus. Unobstructed space around.	City traffic typically seen in residential and business areas, and cooking by slum dwellers.
Kolkata	NEERI Zonal Lab I-8, Sect-C, East Kolkata P.O. Box Haltu Kolkata 700078	Urban Residential	Sampler on a 2 m platform located in open field. Ruby General Hospital and a diesel truck garage nearby.	City traffic typically seen in residential and business areas, cooking by slum dwellers, and some emission from combustion by diesel trucks parked in nearby garage.
Chandigarh	Postgraduate Institute of Medical Education and Research Sector_12 Chandigarh 160012	Background	Sampler on rooftop located on fourth floor. Building located in middle of campus away from streets.	Road dust, gasoline vehicle traffic seen in planned town settings.

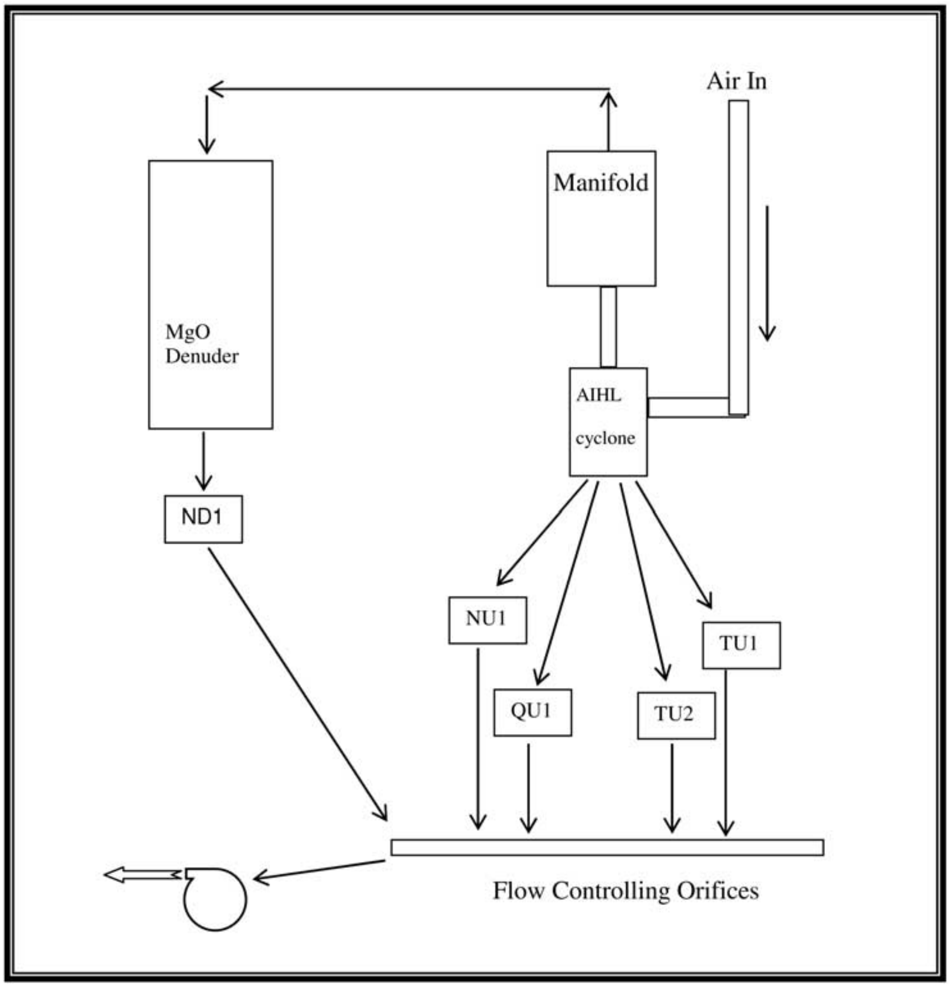

Figure 2. Schematic diagram of the sampling unit. TU1 and TU2 are teflon filters, NU1 and ND1 are undenuded nylon filter and denuded nylon filter, respectively, and QU1 is a quartz fiber filter.

Laboratory (AIHL)-design cyclone separator [*John and Reischl*, 1980], which removed large particles with a collection efficiency curve having a 50% aerodynamic cutoff diameter at 2.5 μm before the air passed through the filters. The nylon filter located downstream of the MgO-coated diffusion denuder was used in conjunction with the nylon filter downstream of the cyclone alone to measure gas-phase nitric acid, hydrochloric acid, and fine particle nitrate by the denuder difference method. The air flow rate through each filter was measured before and after each 24-hour sampling period with a calibrated rotameter.

[6] One of each pair of PTFE filter samples was analyzed by ion chromatography (Dionex Corp, Model 2020i) for the anions NO_3^-, SO_4^{2-}, and Cl^- [*Mulik et al.*, 1976] and by an indophenol colorimetric procedure for NH_4^+ [*Bolleter et al.*, 1961] using an Alpkem rapid flow analyzer (Model RFA-300). The second set of each of these sample sets was analyzed for trace elements using X-Ray Fluorescence (XRF) by the Desert Research Institute (DRI). Quartz fiber substrates were analyzed for elemental and organic carbon content using the thermal-optical carbon analysis method of *Huntzicker et al.* [1982] as modified by *Birch and Cary* [1996]. In the thermal evolution and combustion method of *Birch and Cary* [1996], elemental carbon is defined as carbon that resists volatilization up to a temperature of 900°C in an inert atmosphere. In this paper we will use the term elemental carbon (EC) to define the carbon detected by this method. Variations in elemental carbon values between alternative methods can arise due to differences in the way that alternative methods correct for charring of the samples during analysis.

[7] Extraction of particle-phase organic compounds collected on quartz fiber filters was based on the methods described by *Mazurek et al.* [1987] and further refined by *Schauer et al.* [1996] and *Zheng et al.* [2002]. Samples were combined by season and extracted in annealed glass jars with Teflon-lined lids. Mumbai summertime samples did not contain enough organic carbon (OC) for acceptable GC/MS analysis and thus were not analyzed. In the combined sample, approximately 0.5 mg of OC is desired for accurate GCMS analysis, but summertime Mumbai total OC was much less. In addition, filter blanks, as well as lab blanks, were analyzed. Filter blanks were prepared, stored, shipped in the same manner as the samples, and lab blanks were used to identify possible contaminants from handling samples in the laboratory. Both field and lab blanks were

Table 2. Average Seasonal Concentrations of 29 Organic Compounds Used in the CMB Model, ng-m^{-3}

Organic Compounds	Delhi Spring	Delhi Summer	Delhi Autumn	Delhi Winter	Kolkata Spring	Kolkata Summer	Kolkata Autumn	Kolkata Winter	Mumbai Spring	Mumbai Autumn	Mumbai Winter	Chandigarh Summer
n-Pentacosane	32.12	4.47	37.55	102.43	18.78	10.92	23.69	117.92	5.84	18.70	21.59	23.31
n-Hexacosane	32.14	6.19	36.17	85.92	14.54	19.89	14.11	90.70	7.48	20.48	20.68	14.30
n-Heptacosane	49.19	9.62	54.32	105.10	22.32	15.81	24.71	103.57	12.04	22.71	21.89	24.64
n-Octacosane	37.23	7.01	43.45	76.67	13.68	17.70	19.13	73.75	8.50	24.31	16.43	11.64
n-Nonacosane	86.17	16.79	77.38	150.32	27.37	18.84	45.29	86.32	24.13	31.64	23.58	13.03
n-Triacontane	16.60	3.63	23.50	33.44	12.19	16.69	29.11	31.71	3.73	17.88	7.72	3.95
n-Hentriacontane	39.36	7.04	43.02	71.70	12.56	8.59	10.31	61.99	8.08	32.95	12.92	6.92
n-Dotricontane	31.92	8.38	53.27	94.62	16.09	27.10	24.06	81.76	14.07	9.97	20.02	78.34
n-Tritriacontane	47.12	10.89	74.32	121.76	12.26	14.23	29.36	105.14	11.61	13.94	23.22	5.08
20S&R-abb-Cholestanes	0.81	0.24	1.08	2.99	1.51	0.50	2.43	2.98	0.29	0.49	0.77	1.97
20R-aaa-Cholestane	0.72	0.19	0.66	2.09	0.92	0.70	2.31	5.34	0.18	0.28	0.61	0.50
20S&R-abb-Ergostanes	1.95	0.32	1.04	4.30	1.22	0.65	2.10	9.19	0.41	0.98	2.41	1.40
20S&R-abb-Sitostanes	3.23	0.73	3.80	8.05	1.51	1.22	4.30	9.39	1.04	1.80	2.84	3.66
22, 29, 30-Trisnorneohopane	0.92	0.53	2.19	5.35	1.72	0.45	2.29	8.97	0.30	0.03	1.10	3.28
17a, 21b-29-Norhopane	4.75	1.56	9.89	17.37	5.43	3.28	5.30	21.97	1.20	2.59	5.06	8.93
17a, 21b-Hopane	3.83	1.65	7.37	15.42	6.74	4.01	8.52	19.98	0.99	1.42	3.66	11.05
Isopimaric Acid	2.93	1.12	7.36	28.67	4.03	2.06	6.43	21.06	3.58	3.23	5.59	5.74
Hexadecanamide	17.40	4.14	18.96	44.58	7.62	2.93	6.90	36.59	4.32	2.76	6.97	5.02
Octadecanamide	6.87	1.35	7.55	15.32	2.31	1.13	3.98	9.36	1.30	1.18	2.15	1.65
Benzo[b]Fluoranthene	7.11	1.26	9.97	30.62	4.48	1.67	6.07	53.59	1.25	3.95	6.71	0.68
Benzo[k]Fluoranthene	6.94	1.13	8.10	28.39	4.36	1.31	5.10	40.86	1.25	3.48	5.59	0.53
Benzo[e]Pyrene	6.39	0.82	7.54	25.77	4.19	0.88	5.01	39.38	0.45	2.13	2.69	0.40
Indeno[1,2,3-cd] Fluoranthene	3.51	0.92	5.07	16.58	2.44	1.08	3.36	23.41	0.81	2.26	3.26	0.43
Indeno[1,2,3-cd]Pyrene	3.90	1.06	6.57	18.49	2.85	1.61	3.85	26.13	0.95	2.32	3.58	0.49
Picene	1.26	0.28	1.54	5.11	0.66	0.27	0.82	7.12	0.14	0.48	0.93	0.16
Coronene	6.51	1.64	11.16	21.60	3.58	1.70	3.95	35.95	0.76	2.43	5.13	0.65
Stigmasterol	77.26	29.20	164.40	295.86	49.81	21.79	36.85	301.28	10.33	142.54	34.61	60.05
Levoglucosan	1026.60	210.46	1773.64	5258.30	336.45	75.12	474.38	5491.95	74.52	392.43	907.96	140.30

analyzed and levels from field blanks were subtracted from the sample, whereas the lab blanks were used to see if there were any interferences. In total, 23 of the combined samples were analyzed, including field and laboratory quality assurance/quality control (QA/QC) blanks.

[8] Samples were first spiked with an internal standard mix containing 16 deuterated compounds. Deuterated internal standards were dodecane-*d*26, hexadecane-*d*34, eicosane-*d*42, octacosane-*d*58, hexatriacontane-*d*74, benzaldehyde-*d*6, decanoic acid-*d*19, heptadecanoic acid-*d*33, phthalic acid-3,4,5,6-*d*4, acenaphthene-*d*10, 4,4′-dimethoxybenzophenone-*d*8, dibenz(ah)anthracene-*d*14, chrysene-*d*12, $\alpha\alpha\alpha$-20R-cholestane-*d*4, cholesterol-2,2,3,4,4,6-*d*6, and levoglucosan-$^{13}C_6$ (carbon-13 uniform-labeled compound). These standards provided internal quantification references for the key particle phase organic compounds covering their range of mass spectral fragmentations, polarity, and reactivitiy with derivatization reagents. About 250 μL of the internal standard mix was spiked per milligram of OC. The amount of spiked internal standard mix was proportional to the amount of the OC present in the sample. Samples were extracted using mild sonication (20 min) twice with hexane (Fisher Optima Grade), followed by three successive extractions using a 2:1 mixture of benzene and isopropanal (benzene: HPLC Grade, E&M Scientific Omnisolv; isopropanal: Fisher Optima Grade). Benzene was further distilled in the laboratory to remove the small fraction of impurities and tested for purity by GC/MS prior to use. The extract was filtered to remove loose filter materials, and the volume was reduced to about 5 mL using a rotary evaporator. Finally, it was blown down to the volume of injected internal standard using ultrapure N_2. The extract was split into two fractions. One fraction was derivatized with diazomethane to convert organic acids to their methyl ester analogues for improved quantification.

[9] Following *Zheng et al.* [2002], a Hewlett-Packard GC/MSD (6890 GC and 5973MSD) equipped with a 30-m length × 0.25-mm i.d. × 0.25-μm film thickness HP5 MS capillary column was used. Operating conditions were isothermal hold at 65°C for 2 min, temperature ramp of 10°C min^{-1} to 300°C, isothermal hold at 300°C for 22 min, GC/MS interface temperature 300°C. The flow of the carrier gas, He, was 1 mL min^{-1}. Injection volume was 1 μL for each sample. Scan range was 50–500 amu, and the sample was analyzed under electron ionization mode (70 eV). Estimated measurement uncertainties of the organic compounds were ±20% (1 sigma) [*Schauer et al.*, 1999a].

[10] Chemical Mass Balance (CMB 8.0) [*Watson et al.*, 1998] modeling was used to apportion $PM_{2.5}$ particles to sources. The CMB model combines chemical and physical characteristics of particles or gases measured at the sources and the receptors to quantify the source contributions to the receptor [*Miller et al.*, 1972]. In the present study, CMB was conducted using organic compounds as molecular markers. An important aspect of molecular marker source apportionment is the selection of organic compounds that can be properly used as tracer species in the model. Here 29 organic compounds, along with Al, Si, and elemental carbon (EC) were selected for use as tracer species in the CMB model (see Table 2). These species do not form, do not significantly react, nor have other selective removal processes (i.e., volatilization) in the atmosphere [*Schauer*

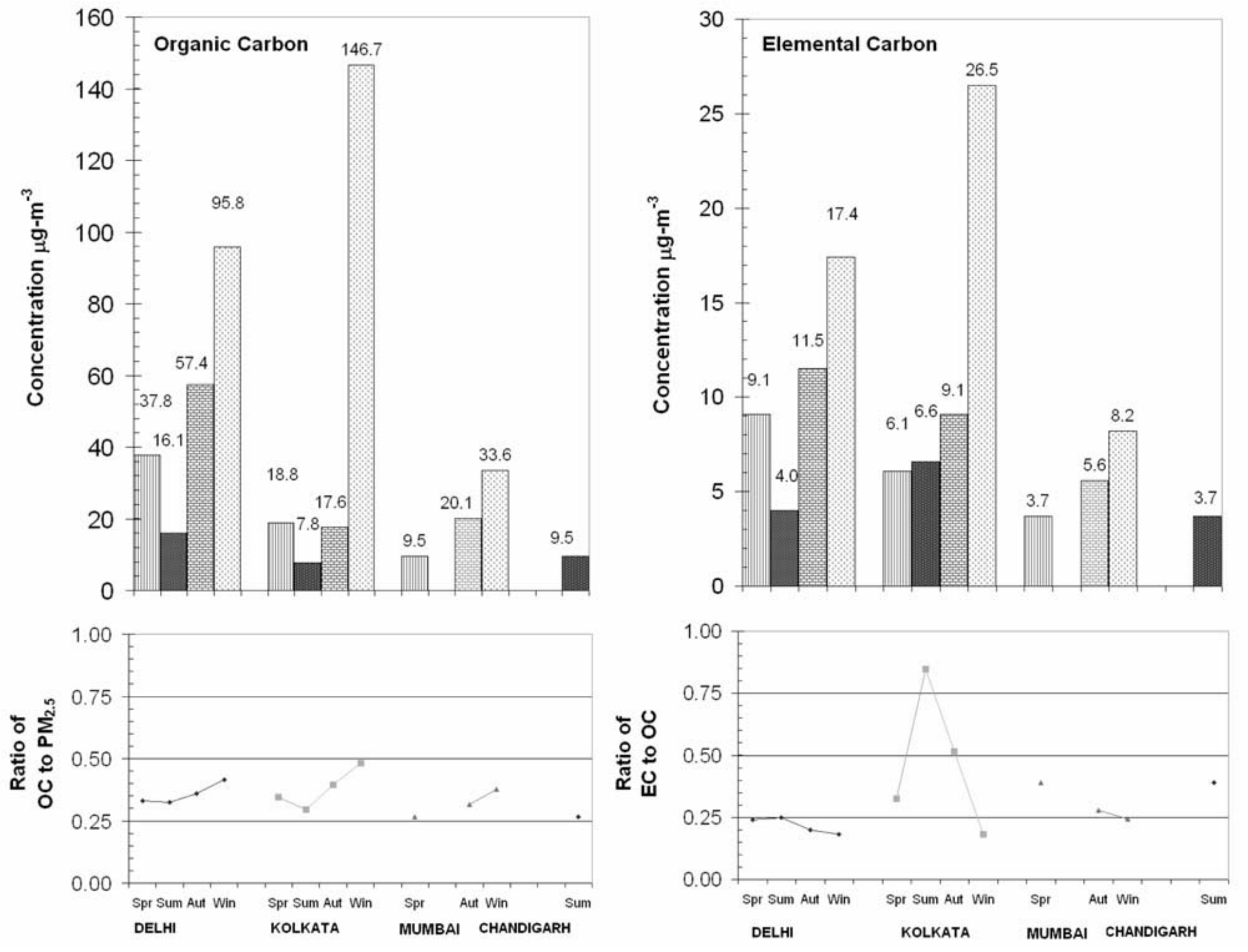

Figure 3a. Seasonal variations of elemental carbon and organic carbon for Delhi, Kolkata, Mumbai, and Chandigarh.

et al., 1996]. Eight source profiles (see Table S1 and Figures S1a and S1b in auxiliary material) were selected for use in this study.[1] These source profiles were developed using the same laboratory procedures described previously for atmospheric samples. Whenever possible, source profiles from the Indian subcontinent were selected. Five source profiles were from Bangladesh (combustion of coconut leaves, rice straw, cow dung, jackfruit wood, and biomass briquette) [*Sheesley et al.*, 2003]. Given the similarity of these profiles, leading to colinearity problems, a combined biomass burning source profile was developed using equal amounts from four of the five (coconut leaves, rice straw, jackfruit wood, and biomass briquette), while the cow dung source profile, which was slightly different from the other four, was kept as a separate source profile. Source profiles for diesel engine exhaust, gasoline engine exhaust, fuel oil, and road dust were obtained from previous studies in North America [*Hildemann et al.*, 1991; *Rogge et al.*, 1993a, 1993b, 1993c, 1997; *Schauer et al.*, 1999b, 2001, 2002b]. The coal source profile was obtained from the analysis of fine particulate matter emitted from the burning of Datong coal in China [*Zheng et al.*, 2005]. While not all of the source profiles have been directly measured in India, the chosen tracer species are more specific to the sources than to the locations.

3. Results and Discussion

3.1. Organic Speciation Results

[11] Samples from the four cities show a seasonally varying distribution of organic marker compounds (see Table 2, Figures 3a and 3b and also Figures S2a–2c in the auxiliary material), indicating a similarly varying set of sources. Hopanes and steranes are organic markers that are present in heavy petroleum distillates such as lubricating oil [*Simoneit*, 1985, 1999]. In the southern California atmosphere, these compounds have been shown to be predominately from the gasoline exhaust and diesel-powered motor vehicles, resulting from the presence of lubricating oil [*Rogge et al*, 1993a, 1993b, 1996; *Schauer et al.*, 1996; *Schauer et al.*, 2002a]. Diesel vehicles are important sources of both elemental carbon and hopanes and steranes, while gasoline-powered vehicles are smaller contributors to elemental carbon concentrations. On the other hand, levoglucosan is a major component of wood smoke aerosol and has been shown to be a good tracer for biomass burning

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[1] Auxiliary material data sets are available at ftp://ftp.agu.org/apend/jd/2007jd008386. Other auxiliary material files are in the HTML.

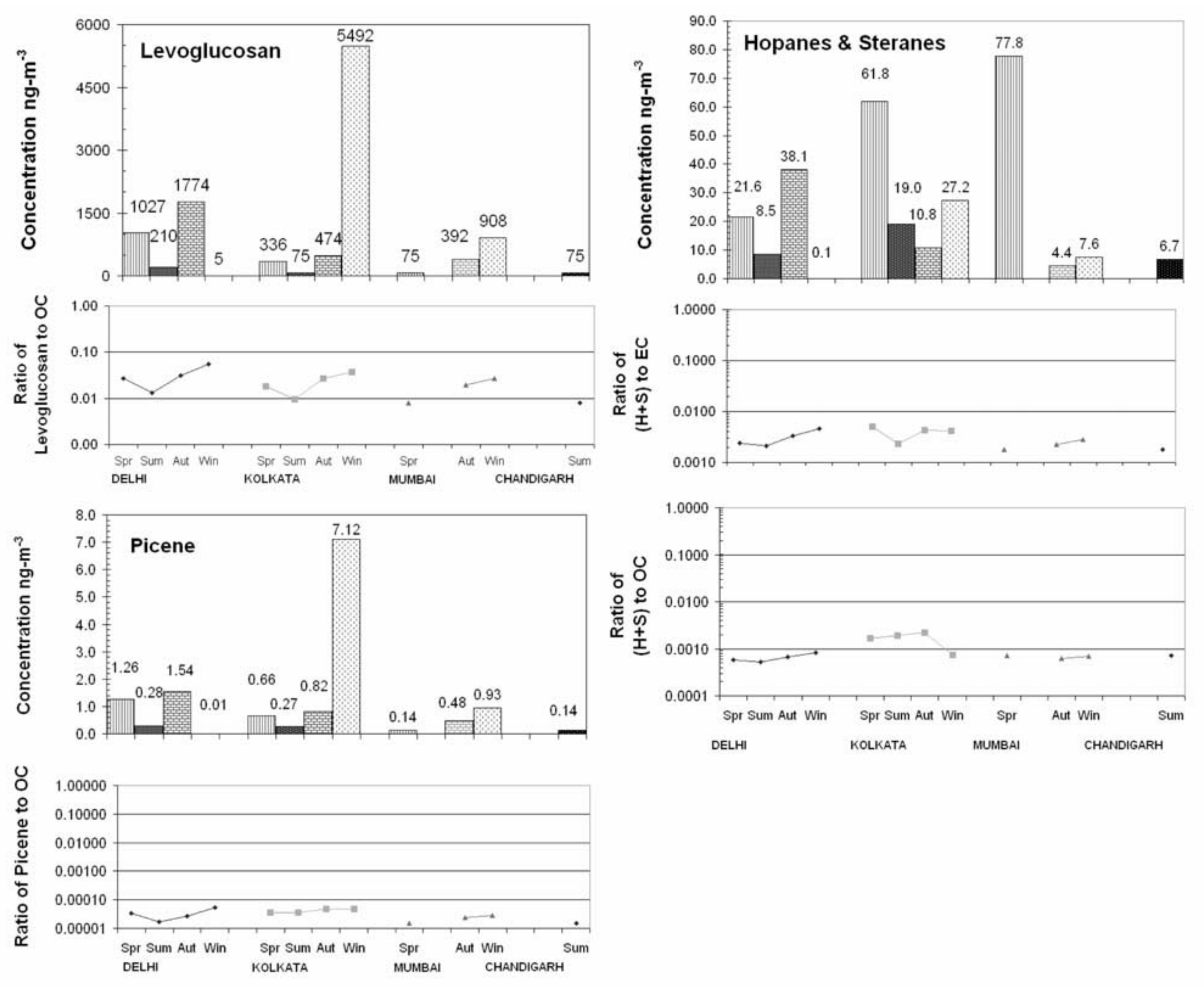
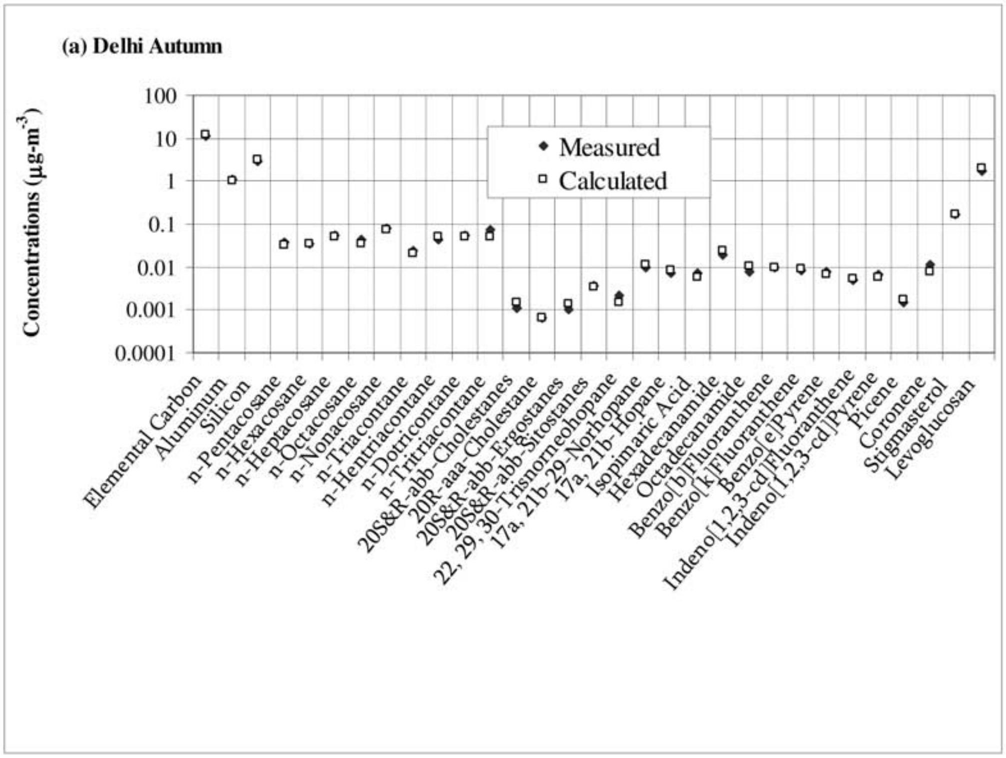


Figure 3b. Seasonal variations of levoglucosan, picene, hopanes and steranes for Delhi, Kolkata, Mumbai, and Chandigarh. The sum of hopanes and steranes is designated by (H + S).

[*Schauer et al.*, 1996; *Simoneit et al.*, 1999] but does not contain hopanes and steranes. Silicon and aluminum are markers for road dust. Picene is used as a tracer for coal combustion [*Oros and Simoneit*, 2000], and stigmasterol combined with the amides (Hexadecanamide and Octadecanamide) are suggested as tracers for cow dung combustion [*Sheesley et al.*, 2003].

[12] Summertime levoglucosan concentrations for Delhi, Kolkata, and Chandigarh were 210 ± 40 ng/m^3, 75 ± 15 ng/m^3, and 140 ± 30 ng/m^3, respectively, whereas wintertime levoglucosan concentrations for the Delhi, Kolkata, and Mumbai are 5300 ± 1100 ng/m^3, 5500 ± 1100 ng/m^3, and 910 ± 180 ng/m^3, respectively. As explained earlier, summertime Mumbai measurements did not meet the criteria for analysis by GC/MS; on the other hand, wintertime Chandigarh measurements were not taken in this study. Ratios for levoglucosan to the sum of hopanes and steranes follow a seasonal trend: ratios are at least three to 10 times higher during the colder months compared to the warmer months for all the cities in this study. This seasonal trend in levoglucosan may suggest increased biomass use for home heating during winter or rice straw burning in the field in the fall. Stigmasterol as well as Hexadecanamide and Octadecanamide, used to identify cow dung smoke, have been detected in all cities. Low-income households use cow dung patties along with tree leaves and branches to cook and heat. No food cooking source profile applicable for this region was available and it is possible that some of the stigmasterol may be attributed from food cooking operations.

[13] Picene concentrations in the summer in Delhi, Kolkata, and Chandigarh were 0.30 ± 0.06 ng/m^3, 0.30 ± 0.06 ng/m^3, and 0.20 ± 0.04 ng/m^3, respectively, and wintertime concentrations in Delhi, Kolkata, and Mumbai were 5.1 ± 1.0 ng/m^3, 7.1 ± 1.4 ng/m^3, and 0.9 ± 0.2 ng/m^3, respectively. Concentrations of picene increased during the winter, likely because of an increase in the use of coal for residential heating. Three thermal power plant stations are present in Delhi: Indraprastha (284 MW capacity, burning 1,150,000 MT/yr of pulverized coal with 39.4% ash and 0.36% sulfur content in the coal), Rajghat (135 MW capacity, burning 876,000 MT/yr of pulverized coal with


(a) Delhi Autumn

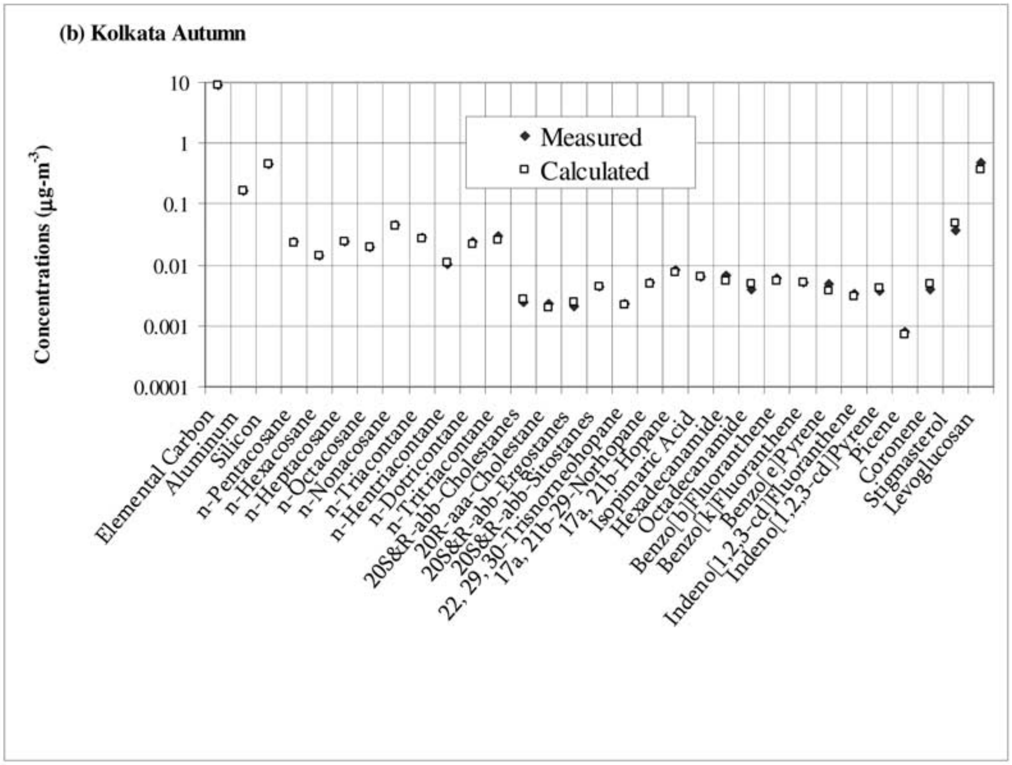

(b) Kolkata Autumn

Figure 4. Comparison of the calculated and measured ambient concentrations of the mass balance species used in the CMB model in (a) Delhi and (b) Kolkata during autumn 2001.

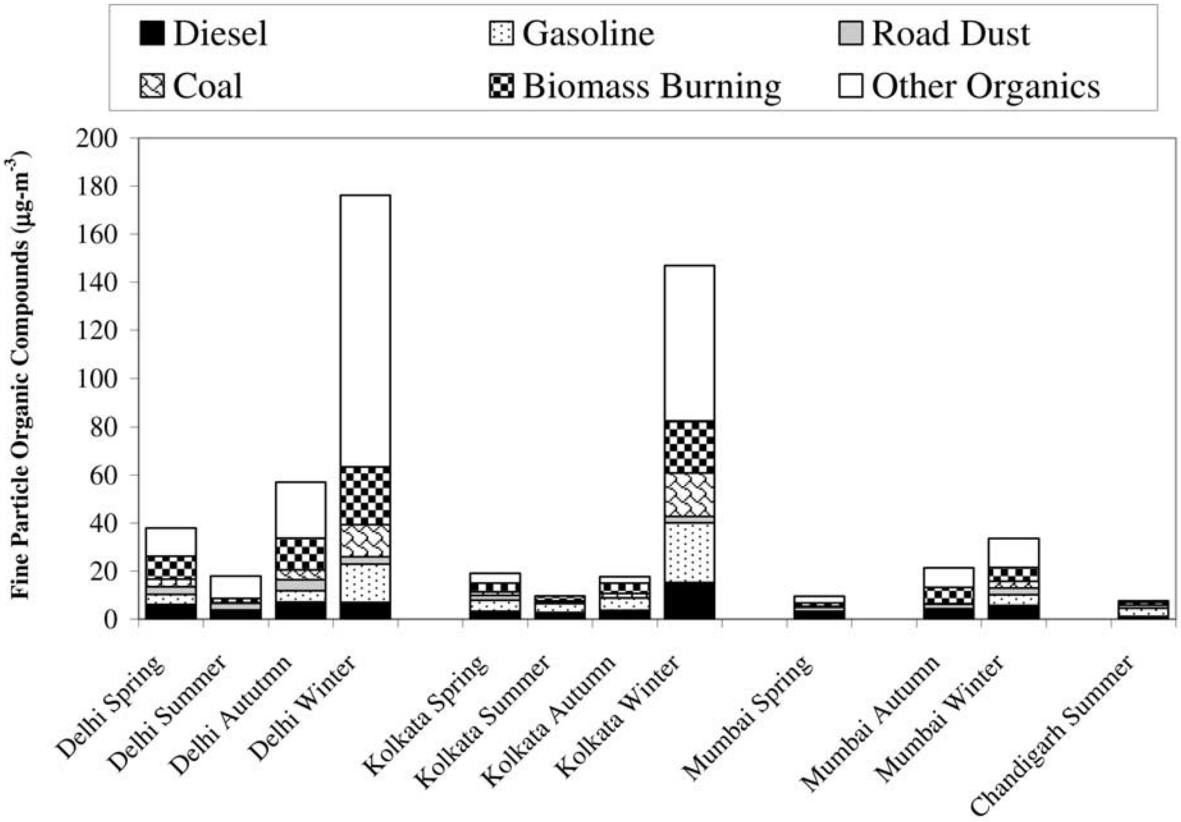


Figure 5. Seasonal source contribution to organic carbon in $PM_{2.5}$ in Delhi, Kolkata, Mumbai, and Chandigarh during 2001.

a 35–42% ash and 0.50% sulfur content in the coal), and Badarpur (720 MW capacity, burning 3,940,000 MT/yr of pulverized coal with a 28–32% ash and 0.35% sulfur content) [*Central Board of Irrigation and Power*, 1997]. Although electrostatic precipitators (ESPs) are present in each of these power generating facilities, poor maintenance has often been blamed for high emissions of fly ash [*Aggarwal et al.*, 1999]. Cholesterol concentrations were below the instrument detection limit in almost all the samples, suggesting that the amount of meat cooking is minimal, consistent with a large fraction of Indians being vegetarian.

[14] We see reasonably good agreement when comparing our $PM_{2.5}$ Delhi average wintertime organic carbon speciation results (Table 2) with the PM_{10} wintertime organic speciation results from the pilot work conducted at the India Habitat Center (IHC) in New Delhi by *Sharma et al.* [2003]. The concentration found at our site is 3–52% higher for n-alkanes, relatively close for the PAHs, and three times higher for levoglucosan.

3.2. CMB Results

[15] The source contribution to organic carbon can be computed by CMB modeling method as described by *Schauer et al.* [2002a] and *Zheng et al.* [2002]. The CMB results presented in this study are statistically significant with r^2 ranging from 0.82 to 0.95. As seen in Figures 4a and 4b, there is good agreement between the calculated concentrations for each of the CMB fitting species (29 organic species along with EC, Al, and Si) and the measured concentrations for the same species. Figure 5 shows the five identified sources (diesel, gasoline, coal, biomass, and road dust) that contribute to the organic carbon in $PM_{2.5}$. The "other organics" category listed in the figure represents the residual difference between the measured total organic carbon concentrations and the sum of the apportioned contributions of the five identified sources as quantified from the receptor model.

[16] Since the ratio of the emissions of fine organic carbon to fine particle mass is known for each of the sources in the model, source contributions to fine particle mass concentrations can be computed. Figure 6 and Table 3 show the fine particle mass contributions from the primary sources plus the unapportioned material as well as secondary sulfate, nitrate, and ammonium ion concentrations. From Figure 6 we can see that there is no single dominant source of $PM_{2.5}$. Gasoline combustion is primarily from mobile sources, but the diesel contribution is likely from both stationary and mobile sources. It is not possible to attribute secondary sulfates, nitrates, and ammonium to specific primary sources using CMB, although sulfates can likely be linked to the sulfur in fossil fuels. Secondary sulfates, nitrates, and particle-phase organics formation comprised approximately one-tenth to one-fifth of $PM_{2.5}$. Broadly, mobile sources and biomass combustion appear to contribute substantially and in several cases approximately in equal proportions, while road dust dominates at times.

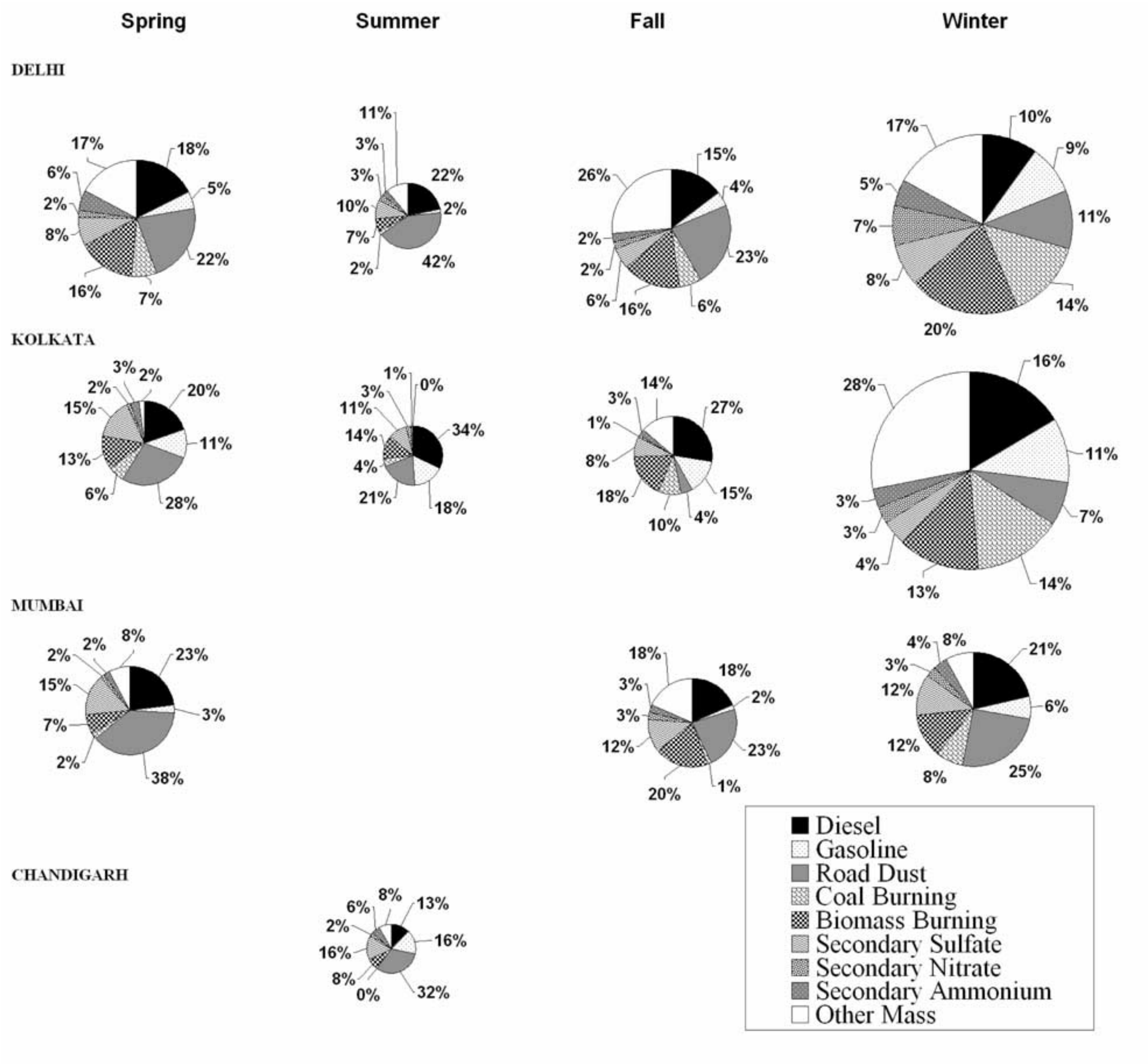


Figure 6. Source contribution to the ambient fine particles in Delhi, Kolkata, Mumbai, and Chandigarh. The sizes of the pie charts are proportional to the average seasonal $PM_{2.5}$ concentration.

Table 3. Calculated Concentrations and Uncertainties, mg-m^{-3}, of Sources and Performance of CMB Analysis by Season and Site

	Delhi				Kolkata				Mumbai			Chandigarh
	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Autumn	Winter	Summer
Diesel	20.00 ± 2.14	10.66 ± 1.28	23.39 ± 2.80	23.03 ± 17.17	10.82 ± 1.45	5.10 ± 1.58	12.27 ± 8.26	50.16 ± 5.63	8.32 ± 0.99	11.78 ± 1.41	18.95 ± 1.94	3.68 ± 0.43
Gasoline	5.48 ± 0.60	0.90 ± 0.19	6.19 ± 0.73	20.68 ± 2.35	6.09 ± 0.71	4.88 ± 0.35	6.54 ± 1.31	32.15 ± 3.49	0.99 ± 0.17	0.96 ± 0.25	5.71 ± 0.58	4.61 ± 0.61
Road Dust	25.38 ± 1.19	20.77 ± 0.95	36.68 ± 1.35	24.43 ± 0.95	15.12 ± 0.72	5.57 ± 0.40	1.99 ± 0.72	21.80 ± 1.27	13.69 ± 0.64	14.72 ± 0.95	22.60 ± 0.72	9.23 ± 0.48
Coal	7.73 ± 0.93	0.83 ± 0.20	9.58 ± 1.34	32.57 ± 4.08	3.42 ± 0.59	1.17 ± 0.22	4.30 ± 0.76	44.01 ± 5.77	0.78 ± 0.15	0.86 ± 0.56	6.87 ± 0.73	0.00 ± 0.00
Biomass Burning	18.34 ± 3.77	3.22 ± 0.91	25.35 ± 4.95	45.87 ± 9.64	7.09 ± 1.60	3.73 ± 0.91	8.11 ± 1.83	40.78 ± 9.39	2.63 ± 0.67	12.74 ± 2.50	11.05 ± 1.85	2.32 ± 0.21
Secondary Sulfate	8.98 ± 0.14	4.80 ± 0.12	9.22 ± 0.22	17.84 ± 0.65	8.36 ± 0.14	2.86 ± 0.12	3.76 ± 0.10	12.00 ± 0.31	5.33 ± 0.12	7.54 ± 0.16	10.37 ± 0.26	4.58 ± 0.13
Secondary Nitrate	2.29 ± 0.16	1.38 ± 0.14	3.35 ± 0.26	16.51 ± 0.76	0.95 ± 0.12	0.78 ± 0.14	0.45 ± 0.12	8.75 ± 0.36	0.59 ± 0.11	1.76 ± 0.14	2.69 ± 0.16	0.66 ± 0.15
Secondary Ammonium	6.39 ± 1.89	1.41 ± 0.05	3.27 ± 0.10	11.06 ± 0.31	1.72 ± 0.05	0.34 ± 0.05	1.13 ± 0.05	9.06 ± 0.14	0.81 ± 0.04	1.84 ± 0.06	3.46 ± 0.06	1.71 ± 0.06
Other Mass	19.60 ± 5.72	5.53 ± 1.98	42.06 ± 6.09	38.91 ± 20.36	1.12 ± 2.95	2.07 ± 1.97	6.15 ± 8.64	85.78 ± 12.98	2.77 ± 2.71	11.70 ± 3.91	7.20 ± 2.99	2.43 ± 1.17
Total Calculated Mass	94.60 ± 5.01	43.97 ± 1.87	117.04 ± 6.05	191.99 ± 20.30	53.58 ± 2.46	24.43 ± 1.92	38.55 ± 8.62	218.72 ± 12.93	33.13 ± 1.38	52.20 ± 3.09	81.70 ± 2.94	26.79 ± 0.93
Total Measured Mass	114.2 ± 2.76	49.5 ± 0.64	159.1 ± 0.63	230.9 ± 1.6	54.7 ± 1.63	26.5 ± 0.45	44.7 ± 0.56	304.5 ± 1.13	35.9 ± 2.33	63.9 ± 2.4	88.9 ± 0.54	29.22 ± 0.7
r^2	0.93	0.94	0.93	0.92	0.95	0.92	0.92	0.92	0.92	0.90	0.92	0.82
χ^2	3.03	2.7	2.91	3.45	1.99	3.19	2.92	3.39	3.38	4.45	3.64	6.03

Predictably, the contributions of biomass and coal, presumably used for heating, are high in winter in Delhi and Kolkata.

[17] During INDOEX high EC to OC ratios were observed in the small island of Kaashidhoo where the intensive field experiment occurred [*Chowdhury et al.*, 2001] and over the Indian Ocean from ship cruises [*Ramanathan et al.*, 2001] suggesting a strong heating of the atmosphere from aerosol. The high EC concentrations have been related through trajectory analysis to the source regions in the Indian subcontinent [*Chowdhury et al.*, 2001; *Ramanathan et al.*, 2001]. The current study conducted upwind of the INDOEX region confirms these high EC to OC ratios. Moreover, the combustion of solid biomass, such as wood, agricultural waste, and dried animal manure in cooking stoves, is the largest source of black carbon emissions in India [*Venkataraman et al.*, 2005]. EC/OC ratios of 0.11 to 3.53 in Indian biomass aerosol were reported by *Venkataraman et al.* [2005] which are much higher than previous reported ratios. Kolkata, located in the eastern part of India where most of the rural people use biomass and cow dung for cooking and heating, shows high EC to OC ratios.

[18] Unattributed mass ("Other Mass" category) was also substantial in some of the samples. This fraction is due, in part, to unidentified organics and water. A fraction of the unidentified OC can be due to secondary formation. $PM_{2.5}$ diesel exceeds gasoline in almost all cases, which is not surprising, given the relatively higher emission rates for diesel compared to gasoline and the higher consumption of diesel compared to gasoline in India. The source profile for diesel is from medium-duty diesel trucks, and it is hard to distinguish between mobile and stationary diesel combustion. The use of diesel in small power generators is not insignificant in the Indian cities studied because of frequent power outage, so it is likely that not all diesel-derived $PM_{2.5}$ is from mobile sources. Gasoline, in contrast, is used almost exclusively in vehicles, and can be attributed to mobile sources with little error. By summing the contributions from diesel, gasoline, and coal, it can be seen that in most cases fossil fuel combustion exceeds biomass combustion. The reconstructed mass from identified sources from the CMB results for summer in Kolkata is 107% of the measured fine particle mass. Uncertainties in the reconstructed mass, as seen in Table 3, and uncertainties in the measured fine particle mass can explain this discrepancy. The precision of the mass measurement is on the order of 1 to 2 $\mu g/m^3$. Also, higher levels can be due to larger ratios of total $PM_{2.5}$ to OC from source emissions than are actually present, and propagation of the various uncertainties involved in this process [*Zheng et al.*, 2002], or the use of an organic matter to organic carbon ratio (a ratio of 1.7 was used in here to convert OC to organic matter) higher than actually present. Total mass concentrations from identified sources should equal approximately 100%, although values ranging from 80 to 120% are acceptable [*Watson et al.*, 1990].

3.3. Fuel-Based Particulate Matter Emissions

[19] An initial confirmation of the CMB-based source apportionment was conducted by comparing the results here for fuel oil, diesel, and gasoline emissions to the results obtained from National Environmental Engineering Research Institute's (NEERI) yearly fuel usage and fuel-based

Table 4. First-Order Approximation of Particulate Emission From Mumbai Using a Fuel-Based Approach[a]

	Yearly Consumption	Emission Factor	Density	Fuel-Based PM Emission	CMB PM Emission	Reddy and Venkataraman
	10^6 l	g/kg	kg/l	Kg/Day	% of Fine	Entire India
Furnace Oil	424	0.65	0.95	620	Negligible	Negligible
Diesel (automotive + industrial)	1140	4.2	0.85	11,000	21%	10%
Gasoline	565	0.6	0.75	700	3.6%	Negligible

[a] The estimate for gasoline is sensitive to the assumed fraction of fuel use by motorcycles and the emissions factor for that source.

emissions factors in Mumbai for the year 2001. Emission factors were obtained from *Reddy and Venkataraman* [2002]. The emission factor for gasoline vehicles is a weighted average between the emission factor for non-catalytic vehicles using unleaded gasoline [*Reddy and Venkataraman*, 2002] and for motorcycles [*Environmental Protection Agency*, 2002]. Diesel contribution to fine particle emissions in Mumbai is an order of magnitude higher than gasoline and fuel oil (Table 4). This is comparable to our results obtained from CMB. In addition, gasoline and diesel emissions from vehicular activity are directly emitted at ground level and thus have a proportionally greater effect on urban air quality. *Reddy and Venkataraman* [2002] find that utility coal burning has the largest emissions nationally in India. Such emissions are not concentrated as much in cities as are motor vehicle emissions. Further, those emissions, along with the emissions from burning fuel oil, are often injected into the atmosphere well above the mixed layer. Thus one expects fuel oil and coal sources to have a smaller impact, relative to their total emissions rate, on urban, ground level particulate measurements, as found here.

[20] It is also of interest to conduct an order of magnitude approximation of the expected levels of PM from the various sources. Using an approximate size of greater Mumbai of 50 × 50 km^2, an average mixing height of 200 m, and an average wind speed of 2 m/s, and assuming instant mixing in to the whole airshed, the above emissions estimates would suggest PM levels from the three sources would be about 5 $\mu g/m^3$ for diesels, 0.3 $\mu g/m^3$ for gasoline fueled vehicles, and 0.2 $\mu g/m^3$ for furnace oil. While all of these values appear low (due to the approximations in the calculation and possible underestimates in the emissions factors for the various sources), it does suggest that it is not surprising that our source apportionment does not find significant levels of PM coming from furnace oil, kerosene, and other liquid and gaseous fuel used in industrial, external combustion boilers. Again, gasoline fueled vehicles emit near the ground, similar to where the monitors are sampling air, and will have a greater impact, particularly at night when the mixing depths are much lower.

4. Uncertainties

[21] Receptor modeling of the type performed here is open to uncertainties, though the use of organic molecular markers provides significant extra information. Further, the agreement with other approaches of estimating sources of $PM_{2.5}$ conducted above is encouraging, as is the agreement with other studies [*Reddy and Venkataraman*, 2002]. When possible, we use source profiles specific to the region, recognizing that a source in one location can have a different profile than ostensibly the same source in another location, for example, diesel trucks. The source profile used, in that case, was developed in the United States. The profile for coal was taken from a Chinese coal. While a comprehensive formal error analysis is beyond the scope of this work, Table 3 does provide calculated uncertainties based on data provided in the CMB analysis. S. Lee and A. Russell (manuscript in preparation, 2007) conducted Monte Carlo analysis with Latin hypercube sampling to better understand the uncertainties of the source contributions apportioned by CMB using inorganic species. The objective was to evaluate the source contribution uncertainties and to identify uncertainty contributors due to uncertainties in ambient measurement and source profiles. They found that the uncertainties in the source profiles contribute more to the source contribution uncertainties as compared to the uncertainties in ambient measurement data. *Zheng et al.* [2006] conducted sensitivity tests for biomass and road dust by replacing the key organic tracers' concentrations and uncertainties with the concentrations and uncertainties found in different profiles for the same source while keeping all other species the same. They show that the biomass relative contribution changed 63–130% and road dust relative contribution changed 8–19%. In the present paper, sensitivity analysis was conducted on wintertime Delhi CMB results by changing the uncertainties of the key tracers for each of the source profiles used in the CMB work. This approach provides an assessment of the sensitivity of the results to the uncertainty choice used, but likely gives less variation than if one uses a range of source profiles [e.g., *Zheng et al.*, 2006]. Uncertainties of the key tracers for one profile were first increased by 25%, then by 50%, and finally by 100% while keeping the uncertainties of all remaining species and all other source profiles constant (see Table 5). This was repeated for all source profiles and the CMB work was conducted and compared with the base scenario. Increasing the uncertainties usually had a small effect on the calculated uncertainties and on the final source apportionment results (average CoV of about 4%). The diesel and gasoline contribution appeared to be the only two sources being impacted significantly when changing the uncertainties of the hopanes and steranes. For other sources, the uncertainties in both the apportioned mass and the CMB-derived uncertainties are small, suggesting that the uncertainties and the results are robust.

5. Conclusions

[22] Chemically detailed particulate matter characterization, including organic speciation, and detailed source

Table 5. Sensitivity Analysis on Wintertime Delhi Source Apportionment (Total Measured Mass 230.9 $\mu g\ m^{-3}$) by Multiplying the Uncertainties of Key Tracers by 1.25 (25% Increase), 1.5 (50% Increase), and 2 (100% Increase)[a]

	Diesel	Gasoline	Road Dust	Coal	Biomass Burning	Sec. Sulfate	Sec. Nitrate	Sec. Ammonium	Others	Total Calculated Mass Conc.
Base Case	23.03 ± 17.17	20.68 ± 2.35	24.43 ± 0.95	32.57 ± 4.08	45.87 ± 9.64	17.84 ± 0.65	16.51 ± 0.76	11.06 ± 0.31	38.91 ± 20.36	191.99 ± 20.30
Diesel × 1.25	23.45 ± 17.17	20.09 ± 2.35	24.51 ± 0.95	32.68 ± 4.08	45.25 ± 9.64	17.15 ± 0.12	16.52 ± 0.14	11.06 ± 0.05	40.19 ± 20.33	190.71 ± 20.27
Diesel × 1.5	23.51 ± 17.20	20.08 ± 2.34	24.51 ± 1.67	32.68 ± 4.08	45.25 ± 9.64	17.15 ± 0.22	16.52 ± 0.16	11.06 ± 0.10	40.14 ± 20.41	190.76 ± 20.34
Diesel × 2	23.64 ± 17.27	20.08 ± 2.34	24.51 ± 1.67	32.68 ± 4.08	45.25 ± 9.64	17.15 ± 0.65	16.52 ± 0.26	11.06 ± 0.31	40.01 ± 20.49	190.89 ± 20.42
Gasoline × 1.25	17.78 ± 16.48	21.52 ± 2.70	24.12 ± 1.59	32.41 ± 4.08	45.27 ± 9.66	17.21 ± 0.14	16.53 ± 0.12	11.11 ± 0.05	44.95 ± 19.85	185.95 ± 19.78
Gasoline × 1.5	14.91 ± 16.18	22.70 ± 3.10	23.88 ± 1.67	32.21 ± 4.08	45.25 ± 9.66	17.24 ± 0.12	16.54 ± 0.14	11.14 ± 0.05	47.03 ± 19.67	183.87 ± 19.60
Gasoline × 2	11.69 ± 15.99	24.46 ± 3.94	23.56 ± 1.67	31.92 ± 4.08	45.23 ± 9.66	17.27 ± 0.10	16.55 ± 0.12	11.18 ± 0.05	49.04 ± 19.66	181.86 ± 19.56
Road Dust × 1.25	25.39 ± 17.96	19.96 ± 2.35	24.59 ± 1.75	32.70 ± 4.08	45.23 ± 9.64	17.13 ± 0.31	16.52 ± 0.36	11.05 ± 0.14	38.33 ± 21.06	192.57 ± 21.00
Road Dust × 1.5	25.98 ± 5.16	19.92 ± 2.17	24.59 ± 0.72	32.73 ± 4.08	45.19 ± 9.60	17.13 ± 0.12	16.52 ± 0.11	11.02 ± 0.04	37.82 ± 11.97	193.08 ± 11.86
Road Dust × 2	25.98 ± 5.16	19.92 ± 2.17	24.59 ± 0.80	32.78 ± 4.08	45.17 ± 9.58	17.13 ± 0.16	16.52 ± 0.14	11.02 ± 0.06	37.79 ± 11.96	193.11 ± 11.85
Coal × 1.25	23.75 ± 16.97	20.26 ± 2.35	24.44 ± 1.67	34.14 ± 4.38	45.46 ± 9.73	17.17 ± 0.26	16.53 ± 0.16	11.02 ± 0.06	38.13 ± 20.32	192.77 ± 20.26
Coal × 1.5	25.72 ± 17.47	20.11 ± 2.35	24.51 ± 1.67	31.01 ± 4.01	46.77 ± 10.08	17.13 ± 0.13	16.52 ± 0.15	11.06 ± 0.06	38.07 ± 20.82	192.83 ± 20.76
Coal × 2	23.45 ± 17.17	20.09 ± 2.35	24.51 ± 0.95	32.68 ± 4.08	45.25 ± 9.64	17.15 ± 0.13	16.52 ± 0.15	11.06 ± 0.06	40.19 ± 20.33	190.71 ± 20.27
Cowdung × 1.25	22.99 ± 17.11	20.09 ± 2.35	24.44 ± 1.67	32.78 ± 4.08	45.08 ± 9.75	17.16 ± 0.13	16.52 ± 0.15	11.06 ± 0.06	40.78 ± 20.38	190.12 ± 20.32
Cowdung × 1.5	22.63 ± 17.07	20.09 ± 2.35	24.44 ± 1.67	32.85 ± 4.08	44.94 ± 9.85	17.16 ± 0.13	16.53 ± 0.15	11.06 ± 0.06	41.20 ± 20.40	189.70 ± 20.33
Cowdung × 2	22.10 ± 16.97	20.09 ± 2.35	24.36 ± 1.67	32.95 ± 4.08	44.79 ± 9.94	17.17 ± 0.13	16.53 ± 0.15	11.06 ± 0.06	41.04 ± 20.36	189.86 ± 20.30
Biomass × 1.25	23.45 ± 17.17	20.09 ± 2.35	24.51 ± 1.67	32.68 ± 4.08	45.25 ± 9.64	17.15 ± 0.13	16.52 ± 0.15	11.06 ± 0.06	40.19 ± 20.38	190.71 ± 20.32
Biomass × 1.5	23.45 ± 17.17	20.09 ± 2.35	24.51 ± 1.67	32.68 ± 4.08	45.25 ± 9.64	17.15 ± 0.13	16.52 ± 0.15	11.06 ± 0.06	40.19 ± 20.38	190.71 ± 20.32
Biomass × 2	23.45 ± 17.17	20.09 ± 2.35	24.51 ± 1.67	32.68 ± 4.08	45.25 ± 9.64	17.15 ± 0.13	16.52 ± 0.15	11.06 ± 0.06	40.19 ± 20.38	190.71 ± 20.32
Avg Contribution	22.44 ± 3.72	20.59 ± 1.16	24.40 ± 0.27	32.62 ± 0.57	45.32 ± 0.41	17.20 ± 0.16	16.52 ± 0.01	11.07 ± 0.04	40.75 ± 3.07	190.15 ± 3.07
Avg Uncertainty	15.79 ± 3.77	2.47 ± 0.41	1.46 ± 0.36	4.09 ± 0.07	9.70 ± 0.13	0.20 ± 0.17	0.19 ± 0.15	0.09 ± 0.08	19.45 ± 2.66	19.38 ± 2.67
COV Contrib.	0.17	0.06	0.01	0.02	0.01	0.01	0.00	0.00	0.08	0.02

[a] Here r^2 was 0.92–0.93 in all cases and χ^2 was 3.4 ± 0.1.

apportionment for Delhi, Mumbai, Kolkata, and Chandigarh were conducted using receptor-based chemical mass balance modeling. Five major sources of primary $PM_{2.5}$ were quantified: diesel exhaust, gasoline exhaust, road dust, coal combustion, and biomass combustion. Important trends in the seasonal and spatial patterns of the impact of these five sources were observed. Primary emissions from fossil fuel combustion (coal, diesel, and gasoline) contributed 25–33% of the $PM_{2.5}$ in Delhi, 21–36% in Mumbai, 37–57% in Kolkata, and 28% in Chandigarh. These figures can be compared to the biomass combustion contributing 7–20% in Delhi, 7–20% in Mumbai, 13–18% in Kolkata, and 8% in Chandigarh. Road dust was also significant. These results are generally consistent with calculations of $PM_{2.5}$ emissions, and an order of magnitude calculation of source contributions. Analyses suggest that the split between light and heavy duty vehicles is somewhat uncertain, but the total fossil fuel impacts and those of other sources are more robust.

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