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Seasonal trends in coarse and fine particle sources in Delhi by the chemical mass balance receptor model

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Abstract

A study of the source contribution of atmospheric particulate matter and associated heavy metal concentrations using chemical mass balance model Version 8 (CMB8) in coarse and fine size mode has been carried out for the city of Delhi. Urban particles were collected using a five-stage impactor at six sites in three different seasons, viz. winter, summer and monsoon in the year 2001. Five samples from each site in each season were collected. The results obtained indicate the dominance of vehicular pollutants in fine size mode, whilst the contribution in coarse mode to some extent is site specific but largely due to vehicular pollution and, soil and crustal dust. Seasons also play an important role but in coarse size fraction only.

Keywords: Chemical mass balance; Source apportionment; Coarse particles; Fine particle; Aerosols; Delhi

1. Introduction

Delhi, the capital city of India with over 14 million populations is facing great risk from various pollutants, especially suspended particulate matters. There are many different sources of SPM like road side dust, vehicles, industries, trans-boundary migrations, power plants, solid waste and local sources. Particulate matters from these sources may contain hazardous pollutants and can have carcinogenic and mutagenic effects. Thus, identification of the sources and estimating their contributions become paramount.

There are many methods to estimate the source contribution like principle component analysis (PCA or factor analysis), multiple linear regression analysis (MLR) and the chemical mass balance (CMB) receptor model. Among these the CMB models are the fundamental receptor models and most trusted for the coarse and fine particle source apportionment [1]. The CMB models estimates source contributions by determining the best-fit combination of emission source chemical composition profiles needed to reconstruct the chemical composition of ambient samples [2,3]. Determination of source contribution relies on the ability to characterize and distinguish differences in the chemical composition of different source types [4]. They are thus independent of source emission data and are particularly useful for the source apportionment of aerosols which generally have many fugitive and distributed sources. However, receptor models also have some limitations, for instance, they cannot be used to find the impact of any specific point source and also they cannot resolve co-linearity among sources having similar chemical and physical compositions [5,6].

Chemical mass balance receptor models have been used to apportion different fractions of SPM and metals in various parts of world [3,7–26]. Of late some important studies have been carried out using CMB receptor models: Breed et al. [27] determined the possible sources of PM10 in Prince George (Canada) by morphology and in situ chemical composition of particulates. Samara et al. [28] applied CMB source apportionment of PM10 in industrialized urban area of Northern Greece. Again Samara [29] identified sources of TSP by CMB in a lignite burning area of Macedonia, Greece. Zheng et al. [30] studied the seasonal trend of sources of PM2.5 in Beijing.

However, in Indian context the studies on source apportionment by CMB are rather limited. Sharma and Patil [6]

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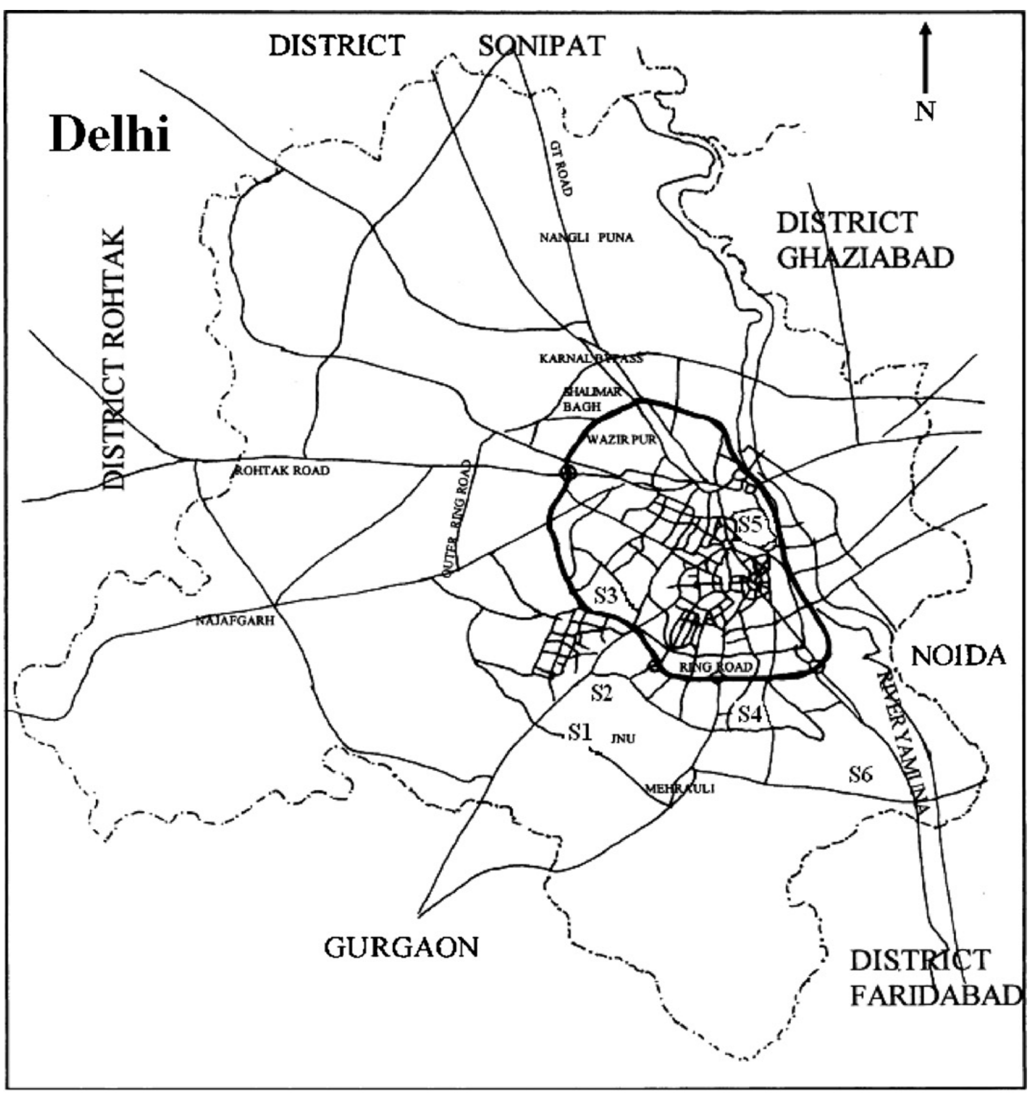
used the CMB for source apportionment of Aerosols in Bombay. In the recent past, Srivastava [31] and Srivastava et al. [32] carried out the source apportionment of ambient VOCs in Mumbai and Delhi, respectively, using CMB8. As far as source apportionment of fine and coarse particles and associated metals along with their seasonal variations are concerned no comprehensive study so far has been done for Delhi, one of the most polluted cities in world. In this paper an attempt has been made to have a fair assessment of the various sources of fine and coarse aerosols in different seasons in Delhi.

2. Experimental

2.1. Area description

Sampling was carried out at six different sites in Delhi (Fig. 1). Delhi, the capital city of India, has over 14 million inhabitants, 3.5 millions of registered vehicles, three coal based thermal power plants and 125,000 industrial units [33]. It lies in the subtropical belt between 76°50′E–77°23′E and 28°12′N–28°53′N. Its climate is semi-arid and consists of summer (March–June), monsoon (July–October) and winter



Sampling Sites:

S1 Jawaharlal Nehru University
S2 Vasant Vihar
S3 Dhaula Kuan
S4 Hauz Khas
S5 Cannaught Place
S6 Okhla

Fig. 1. Locations of sampling sites.

Table 1
Sampling details and categorization of sites

Site	Site character	Duration of sampling (h)	No. of samples in each season
Jawaharlal Nehru University (JNU)	Residential cum sensitive site	24	Five
Hauz Khas (HK)	Residential cum sensitive site	24	Five
Dhaula Kuan (DK)	Heavy traffic site	24	Five
Vasant Vihar (VV)	Heavy traffic site	24	Five
Okhla (OK)	Industrial cum commercial site	24	Five
Cannaught Place (CP)	Industrial cum commercial site	24	Five

(November–February) seasons. It experiences a maximum temperature of ∼45–48 °C in June during summer and minimum of ∼1–2 °C in January during winter. It has a normal annual rainfall of 611 mm. The air over Delhi is dry during the greater part of the year. Humidity is high in the monsoon season while April and May are the driest months. Yearly mean wind speed varies in the range of 0.9–2.0 m/s. Summer months witness the highest frequency of thunderstorms and dust storms. These storms are generally dry but some are accompanied with heavy rains.

2.2. Ambient sampling

To determine the source contributions of aerosols, sampling was performed from January to August 2001 at aforementioned sites in three different seasons. The details of the number of samples and the duration at each category of sites are given in Table 1. A five-stage cascade particle separator (Kimoto Electric Co. Ltd., Japan) was used at an average flow rate of $600 \pm 0.5\,\mathrm{l\,m^{-1}}$. To collect sufficient amount of aerosols 24 h samples were taken continuously for 5 days at each site in three seasons. Various meteorological parameters during three different sampling periods have been provided in Table 2 [34]. Wind rose plots along with the average wind speed during sampling periods, viz. winter, summer and monsoon have also been illustrated in Figs. 2–4, respectively [34].

The samples were collected on pre-weighed Whatman GF/A glass fiber filters. Each set of samples consists of five different filters for various size ranges. These size ranges are as follows—Stage 1: maximum – 10.9 μm; Stage 2: 10.9–5.4 μm; Stage 3: 5.4–1.6, 1.6–0.7 μm; 0.7 μm – minimum. The filters were kept in vacuum desiccators for 24 h to remove any moisture content before mounting them on the air sampler. After the sampling the filter papers were immediately transferred to

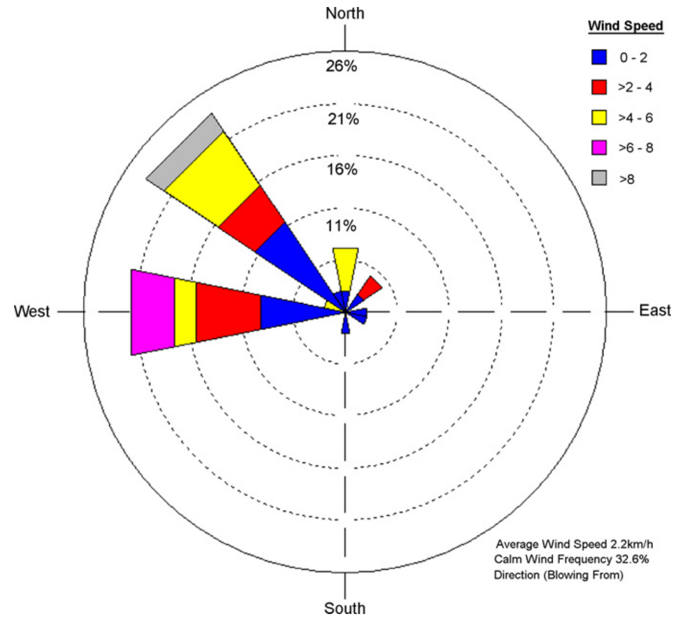


Fig. 2. Wind rose pattern of Delhi during winter sampling (1 January 2001–15 February 2001).

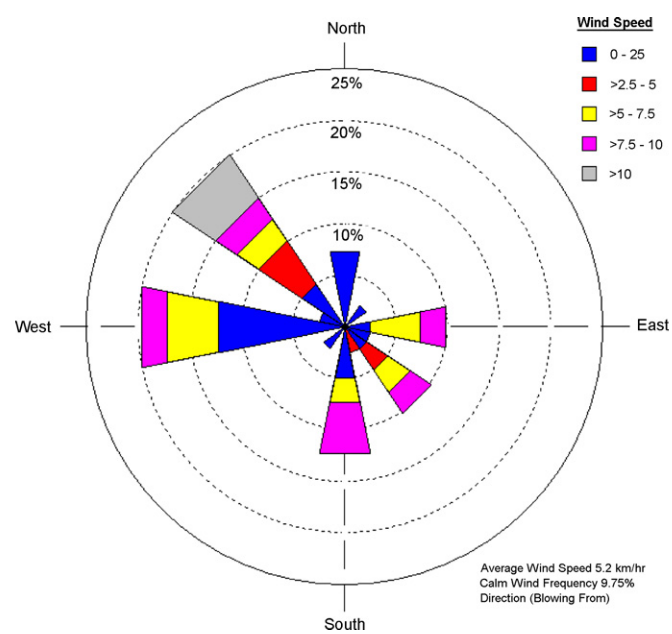


Fig. 3. Wind rose pattern of Delhi during summer sampling (1 May 2001–15 June 2001).

Table 2
Meteorological parameters during sampling period

Period of sampling	Average maximum temperature (°C)	Average minimum temperature (°C)	Average maximum relative humidity (%)	Average minimum relative humidity (%)
1 January 2001–15 February 2001	20.2	5.8	84.2	47.0
1 May 2001–15 June 2001	37.5	25.3	68.5	43.0
1 July 2001–15 August 2001	34.0	26.8	85.3	69.9

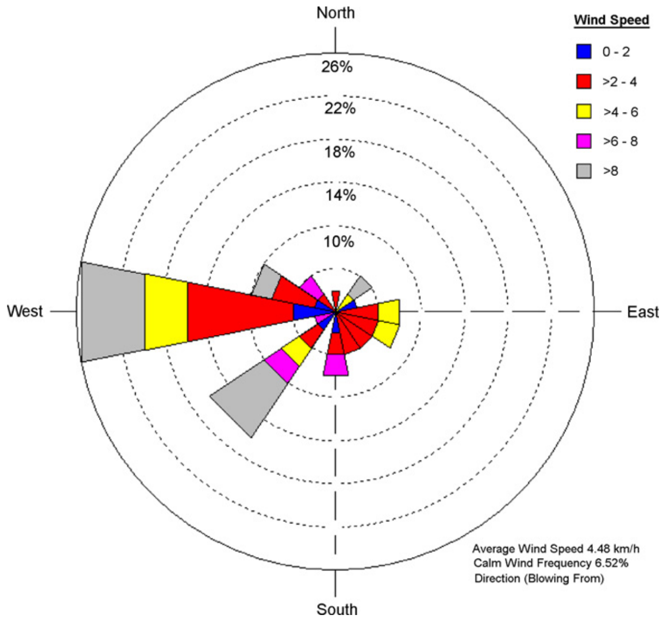


Fig. 4. Wind rose pattern of Delhi during monsoon sampling (1 July 2001–15 August 2001).

vacuum desiccators to again demoisturise them in the same manner. Subsequently these five size range particles were divided in to two broad categories, viz. coarse and fine size particles. Coarse size particles represented the sum total of particles of size ranges—maximum – 10.9, 10.9–5.4 and 5.4–1.6 μm while the fine particles represented the sum total of particles of size ranges: 1.6–0.7 and 0.7 μm – minimum.

2.3. Analysis of samples

Acid digestion, required for the metal estimation by AAS, was carried out according to the standard procedure [35]. The reagents used were HNO_3 70% (S.G. 1.41), HCl 36% (S.G. 1.18) and HF 40% (S.G. 1.13). Acid digestion was performed following these steps. Step 1: samples (dry filters) after taking a small piece out of it, were dissolved in 3 ml HF, 6 ml HNO_3 and 1.5 ml HCl in Teflon bombs and kept at 120 °C for 1 h; Step 2: samples were evaporated to dryness at 70 °C for 1/2 h; Step 3: residue was re-dissolved in 10 ml of 10M HNO_3 and evaporated to dryness. Step 3 was repeated until it was fully dissolved. A series of blanks were prepared using the same digestion method to avoid the matrix effect. Standard solutions of metals were prepared as described in EPA manual [36] and AAS manual [37].

3. Source apportionment methodology

3.1. Chemical mass balance model

In this study a chemical mass balance receptor model, CMB8.0 [1,38,39] was used to apportion the sources contributing to the ambient coarse and fine particles in Delhi. The basic principle of the receptor model may be expressed by an empirical relationship given in Eq. (1). This represents the relationship between the concentrations of the chemical species measured at the receptor site to those emitted form the source:

$$C_i = \sum_{J=1}^{P} F_{ij} S_j \quad (1)$$

where C_i is the ambient concentrations of the species i, measured at the receptor site, P the number of sources that contributes, F_{ij} the fraction of the emissions of the species i starting from the source j, and S_j is the ambient contribution of the source j.

The model fit is considered 'good' if the values of the following statistical parameters lie within the acceptable range given along with them:

(i) correlation coefficient (R^2) more than 0.6,
(ii) chi-square (χ^2, a weighted sum of squares of the difference between calculate and measured fitting species concentration) less than 4,
(iii) degree of freedom (DF) greater than 5,
(iv) percent of aerosol mass explained by sources between 80 and 100,
(v) ratio of calculated to measured concentration (C/M ratio) between 0.5 and 2 and
(vi) absolute value of ratio of residual to uncertainty(R/U) less than 2.

3.2. Source profiles

A total of 13 species, viz. Mn, Cr, Cd, Cu, Co, Pb, Ni, Fe, Zn, Na, K, Mg and Ca for both coarse and fine suspended particles were considered. It is imperative to mention that the source profiles for Indian cities have not been compiled as yet. Thus, the metals source profiles used were taken from Speciate 3.2 [40] data base and some of the previous works carried out in Delhi [41–51]. The emission sources considered are explained in the following paragraphs.

3.2.1. Soil and crustal dust

In Delhi fugitive dust from road side and other unpaved areas with vehicular activities are unlimited reservoirs of dust loading especially when the vehicles are moving. Main constituent of this, are dust loadings tracked out from unpaved areas (such as construction areas, unpaved roads, parking lots, etc.), by transport of dirt collected on vehicle undercarriages, by wear of vehicle components (such as tires, brakes, clutches, and exhaust system components), by water and wind erosion from adjacent areas and the trans-boundary migrations (this is very much significant in summer season). Apart form above mentioned, the pollutants generated from various other sources also become the part of this in course of time [41,45,48].

3.2.2. Paved road dusts

Particulate emissions from paved roads are due to direct emissions from vehicles in the form of exhaust, brake wear and tire wear emissions and re-suspension of loose material on the road surface (i.e., the surface loading). In turn, that surface loading is continuously replenished by other sources. Various field studies

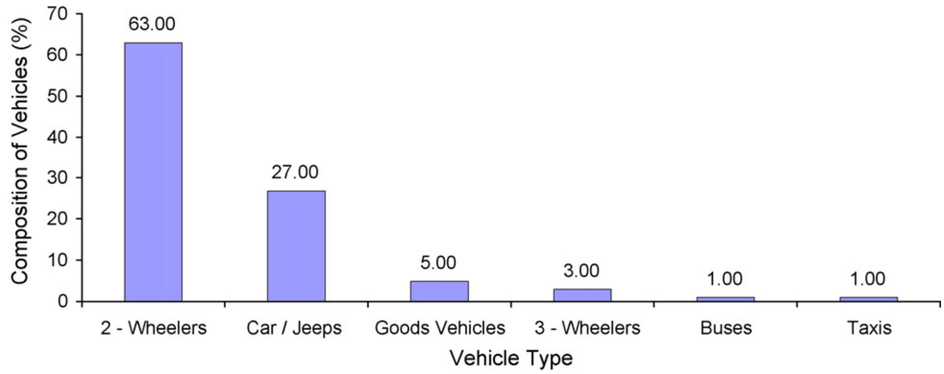


Fig. 5. Composition of registered vehicles in year 2001.

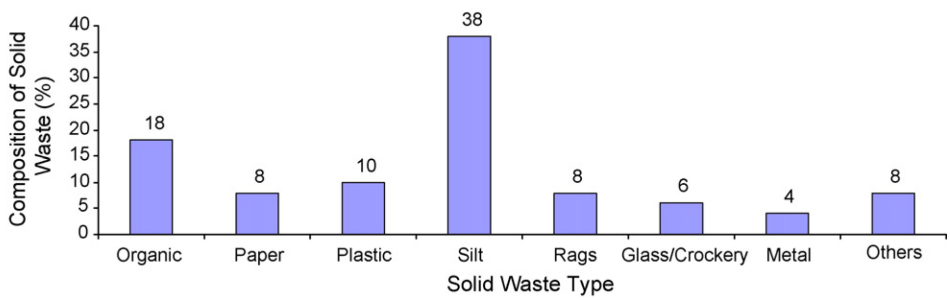


Fig. 6. Composition of various solid wastes for the year 2001.

have found that public streets and highways, as well as roadways at industrial facilities, can be major sources of the atmospheric particulate matter within an area [52].

3.2.3. Composite vehicular emissions

Over 3.5 millions of registered vehicles ply on the roads of Delhi [33]. Their breakup is presented in Fig. 5. It is clear that over 60% of the total vehicles are gasoline fueled two wheelers. Almost 90% is the private vehicles while remaining 10% comprises of goods vehicles and public transport. Here, it is pertinent to mention that most of the private and many of the public transport vehicles are gasoline fueled. Due to concerns over the ability of the model to accurately separate the contribution of diesel and gasoline vehicle particulate matter emissions in Delhi using US-EPA Speciate 3.2 [40] source profiles, the sum of diesel and gasoline exhaust is reported here.

3.2.4. Power plants

There are three coal based thermal power plants in Delhi, at Indraprastha, Rajghat and Badarpur. The total quantity of fly ash from the three power plants is about 6000 tonnes per day (Badarpur 3500–4000, Indraprastha 1200–1500 and Rajghat 600–800 tonnes per day) [33]. None of the thermal power plants in Delhi has an action program for mass scale utilization of fly ash.

3.2.5. Solid wastes

About 5000 metric tonnes of municipal solid waste is generated every day in Delhi. Disposal is mainly in landfills. According to a recent study by the National Environment Engineering Research Institute, Nagpur, the expected quantity of solid waste generated in Delhi would be about 12,750 metric tonnes per day by 2015 [33]. The composition of various solid wastes is given in Fig. 6.

3.2.6. Industrial and other combustible sources

According to a survey by the Delhi Government, out of a total of 125,000 industries, there are 98,000 industries in

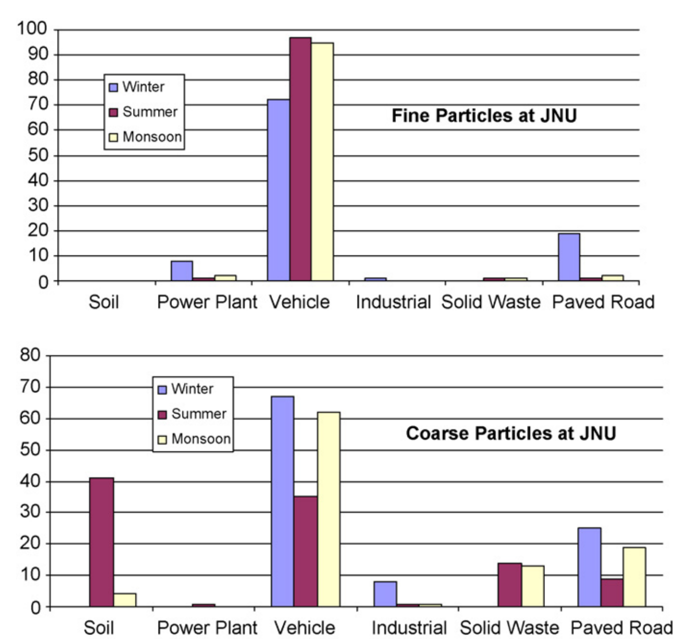


Fig. 7. Source contribution to SPM in coarse and fine size mode at JNU.

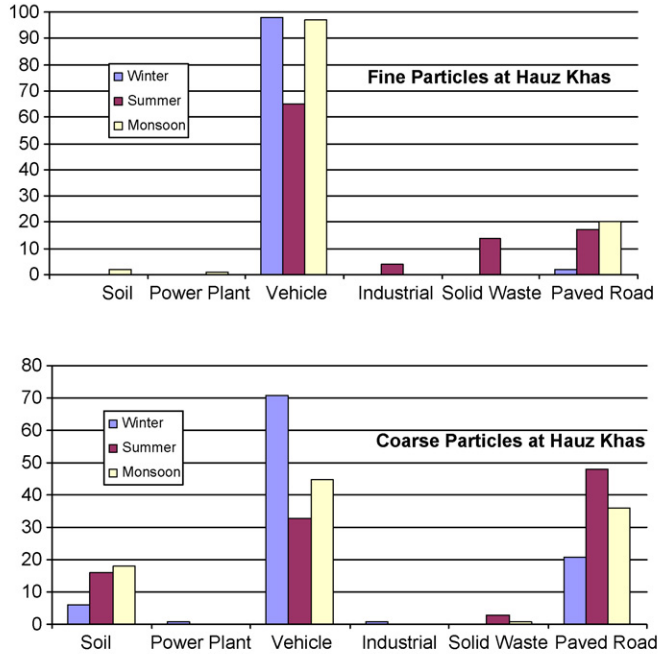


Fig. 8. Source contribution to SPM in coarse and fine size mode at Hauz Khas.

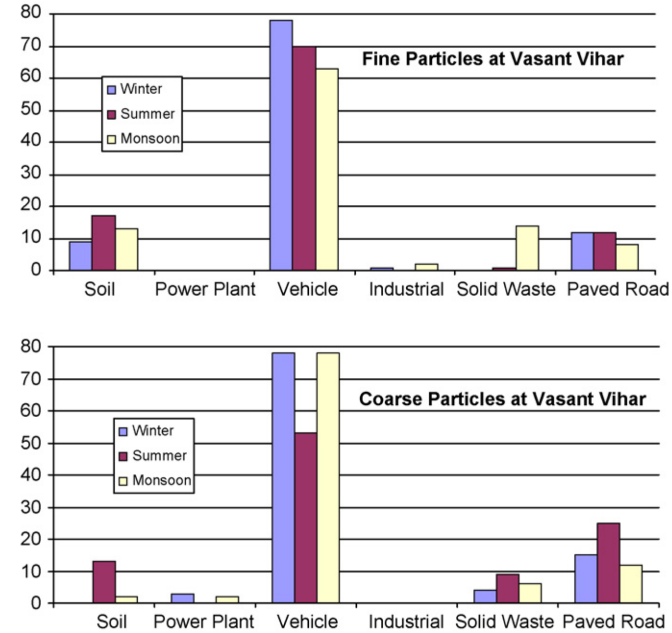


Fig. 10. Source contribution to SPM in coarse and fine size mode at Vasant Vihar.

non-conforming areas as per the Master Plan of Delhi. Non-conforming industries are located in unauthorized colonies, "*lal-dora*" villages (unauthorized settlements), resettlement colonies, the walled city and other residential pockets [33].

4. Results and discussion

Source contributions from various sources are provided in Figs. 7–12 of various sites in all three seasons. It can be inferred that in the fine size range vehicular emission dominates among all other sources, while in case of coarse fraction apart from vehicles, paved road and, soil and crustal dusts have also contributed significantly. Table 3 gives the correlation coefficient R^2 values along with the range of ratio R/U and χ^2 values. As mentioned in Section 2.2 all the six sites depending upon their nature were classified in three broad categories and their details are provided in Table 1. Results obtained at all six sites have been discussed in three different following paragraphs.

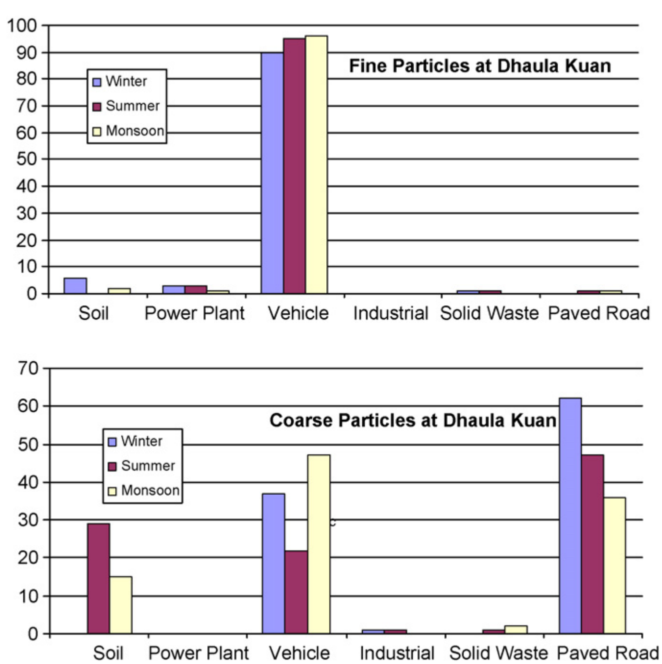


Fig. 9. Source contribution to SPM in coarse and fine size mode at Dhaula Kuan.

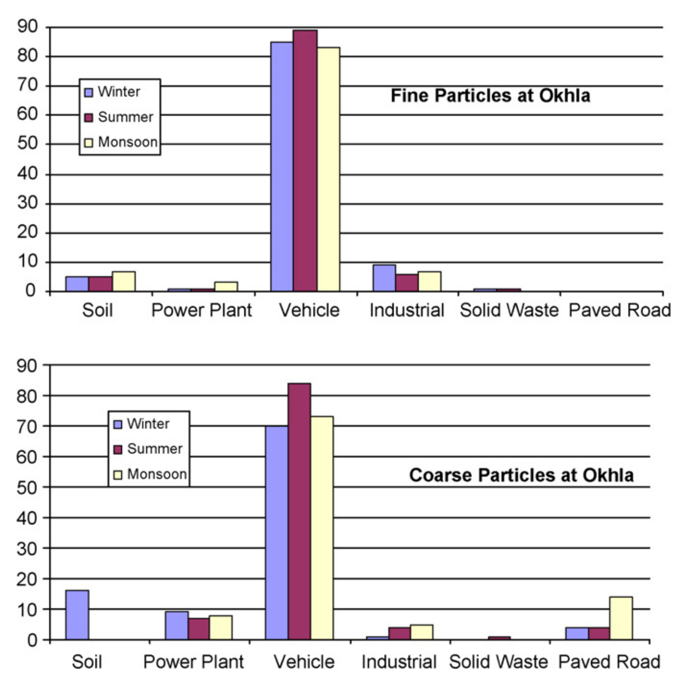


Fig. 11. Source contribution to SPM in coarse and fine size mode at Okhla.

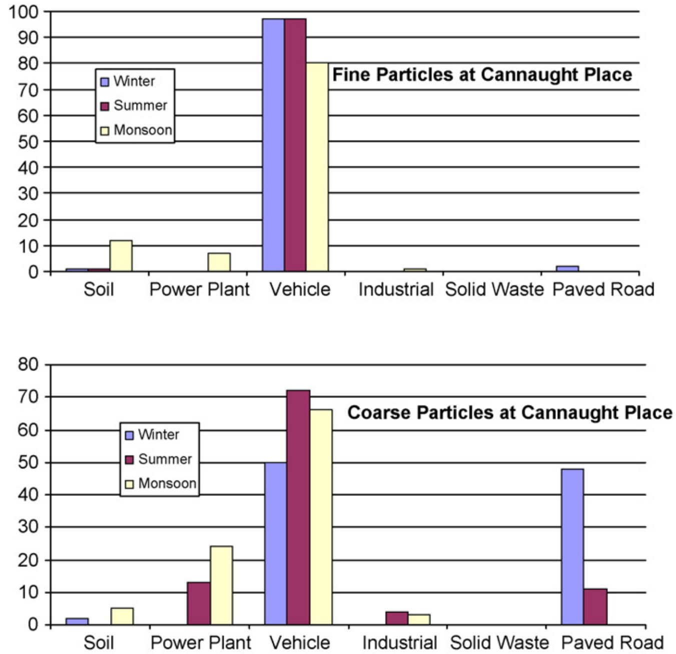


Fig. 12. Source contribution to SPM in coarse and fine size mode at Cannaught Place.

4.1. Residential and sensitive site

It can be observed from Figs. 7 and 8 that in the fine size mode, vehicular pollution completely dominates among all the sources in all the seasons. It contributes almost 90% of total sources. The only exceptions are in winter season at JNU (70%), when paved road dust and fly ash have also contributed significantly, and in summer season at Hauz Khas (Fig. 8) when vehicular contribution has reduced to approximately 60% with a significant increase in paved road dust and solid waste. The reason for increased contribution of fly ash at JNU can be due to the fact that it is highly vegetated and during winter season people use wooden blocks as bonfire in the open spaces to keep themselves warm, resulting in significant quantities of ashes at many places. Whilst the increased contribution of solid waste and paved road dust in the summer season at Hauz Khas is because the sampling site was not far away from a heavy traffic junction (approximately 500–600 m away) and is surrounded by commercial complexes (few hundreds to a km). Here, it is also pertinent to mention that the sampling point at Hauz Khas was around 400–500 m away from a couple of public schools and a leading engineering institute namely Indian Institute of Technology (IIT) Delhi. Therefore, although Hauz Khas is primarily a residential area, its close proximity to an educational institute was the reason behind categorizing Hauz Khas as a residential and sensitive site.

In case of coarse size fraction apart from vehicles, paved road dust and, soil and crustal dust also contribute significantly. Soil and crustal dust contribute over 40% in summer season at JNU (Fig. 7). The possible reason for this is due to its presence on the Aravali hills range. This is the oldest mountain of India and contains large amount of loosely bound Ferro genus quartzite. Its composition to a large extent is similar to upper layer of soil. Since the summer season in India is characterized by heavy windy conditions, causing these loosely bound quartzite to become the part of ambient air. Again the reason for increased contribution from paved road dust, and soil and crustal dust at Hauz Khas (Fig. 8) could be due to its presence in the vicinity of heavy traffic junction and commercial complexes.

4.2. Heavy traffic site

In case of fine particulate source apportionment at Dhaula Kuan almost total contribution is from vehicular pollution in all the seasons (Fig. 9). This is not unexpected because this is the heaviest traffic junction of Delhi and during the course of sampling, construction of a network of flyovers had just begun. This had reduced the vehicles flow speed to almost 5–10 km/h. Fig. 10 reveals that in case of Vasant Vihar the contribution from paved road and, soil and crustal dust is also significant and varies between 10 and 20% in all the seasons. Alarmingly, solid waste's contribution has gone to approximately 15% in

Table 3
Some of the CMB performance statistics

Site	Size	Winter			Summer			Monsoon		
		R^2	R/U	χ^2	R^2	R/U	χ^2	R^2	R/U	χ^2
JNU	Coarse	0.64	0.83	4.45	0.77	0.52	2.15	0.64	0.61	3.94
	Fine	0.91	0.84	1.33	0.87	0.41	1.50	0.84	0.66	3.09
Vasant Vihar	Coarse	0.67	0.82	4.46	0.69	0.66	4.95	0.63	1.08	3.2
	Fine	0.63	0.12	4.73	0.69	1.38	4.3	0.82	0.56	4.73
Dhaula Kuan	Coarse	0.66	0.78	2.1	0.66	1.08	2.1	0.60	1.3	1.4
	Fine	0.68	0.68	3.22	0.90	0.31	2.76	0.84	0.34	1.80
Hauz Khas	Coarse	0.60	0.48	4.04	0.66	0.72	3.91	0.66	0.76	4.47
	Fine	0.96	0.51	0.30	0.66	0.24	3.6	0.60	0.89	3.52
Okhla	Coarse	0.89	0.88	2.59	0.60	0.76	4.28	0.79	0.56	2.84
	Fine	0.66	0.5	1.13	0.63	1.94	2.2	0.94	0.12	1.67
Canaught Place	Coarse	0.96	0.51	0.32	0.61	0.76	2.4	0.62	0.64	3.51
	Fine	0.62	0.69	2.68	0.77	0.47	5.06	0.67	0.53	3.56

the monsoon season. The only plausible reason for this can be attributed to its presence in the close proximity of a large open field where hundreds of slums are present. These slum dwellers dispose off their wastes in self made potholes. During monsoon season these potholes get water logged causing the wastes to come on the roads and other open spaces through the water streams. This in course of time becomes the part of ambient air after getting dried up.

In case of coarse particles at Vasant Vihar (Fig. 10) paved road dust contributes between 10 and 30% in all the seasons, but still the largest contributor is vehicular pollution. However, the situation is quite different at Dhaula Kuan (Fig. 9), where the paved road dust has exceeded the vehicular pollution. This has been explained in the preceding paragraph. Here, it is important to mention that the vehicular density at the Dhaula Kuan is many-many times than that at Vasant Vihar. At Dhaula Kuan in summers soil and crustal dust's contribution has shot up to 30%. This is because the junction was open from all the sides and not far away from agricultural lands.

4.3. Commercial and industrial site

From Fig. 11 it can be inferred that at Okhla in the fine size range again the contribution of vehicular pollution is over 80% in all the seasons. At this site the industrial contribution has also become significant and is approximately 10% in all the seasons. It is imperative to mention that, Okhla is a kind of industrial site where the emanation of smoke is very less but other activities like iron work, polishing, battery manufacture, etc. are maximum. Here, the contribution of soil and crustal dust is also good (between 5 and 10%), because some of the roads are unpaved, causing the soil and crustal dust to be incorporated with the ambient air. At Cannaught Place again the maximum contribution in the fine size mode is from vehicles (over 90%), except in the monsoon season when the contribution of soil and crustal dust, and power plant have also become significant, i.e. approximately 10% each (Fig. 12).

In case of coarse particle fractions the trend is almost similar as in the case of fine size fraction except that the contributions from paved road dust and power plants have also increased significantly (Figs. 11 and 12). This is due to the fact that their locations are in the close vicinity of power plants. Okhla is closer to Badarpur thermal power plant while Cannaught Place is closer to Indraprastha and Rajghat thermal Power Plants. The contribution of Power Plant is more in case of Cannaught Place than Okhla, because Cannaught Place accommodates hundreds of offices, where thousands of office goers use outside eateries. Most of these eateries use coals and other combustible goods (cow dung cake, firewood, wooden blocks and kerosene oil) to cook food.

5. Summary and conclusions

Ambient coarse and fine particles were obtained from six receptor sites of urban environment of Delhi. Chemical source profiles were obtained form Speciate 3.2 data base of US EPA. It was observed that in the fine size mode vehicular pollution completely dominates among all the sources at all sites and in all the seasons. Its contribution varies between 60 and 90%. In coarse mode particles apart from vehicular pollution, contribution from other sources, viz. soil and crustal dust, fly ash, solid waste and paved road dusts have also become significant. Seasons and nature of sites also play an important role but in coarse mode particles only.

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