Source Apportionment of Total Suspended Particulate Matter in Coarse and Fine Size Ranges Over Delhi

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Abstract

Source apportionment of total suspended particulate matter (TSPM) and associated heavy metals has been carried out for the city of Delhi using the Chemical Mass Balance Model, Version 8 (CMB8), as well as principle component analysis (PCA) of SPSS (Varimax Rotated Factor Matrix method) in coarse- and fine-size mode. Urban particles were collected using a five-stage impactor at six sites in the winter of 2005-06. The impactor segregates the TSPM into five different size ranges (viz. > 10.9, 10.9–5.4, 5.4–1.6, 1.6–0.7 and < 0.7 μm). Four samples were collected from six different sites every 24 hours. Samples were analyzed in five size ranges gravimetrically and chemically for the estimation of SPM and metals. The five different size ranges were divided into two broad categories: coarse (1.6 to > 10.9 μm) and fine (< 1.6 μm). The CMB8 and PCA were executed separately for both coarse and fine size ranges. Results obtained by CMB8 indicate the dominance of vehicular pollutants (62%), followed by crustal dust (35%) in the fine size range; while in the coarse size range crustal dust dominated (64%) over vehicular pollution (29%). Little contribution from paved-road dust and industrial sources was observed. Results of PCA (or factor analysis) reveal two major sources (vehicular and crustal re-suspension) in both coarse and fine size ranges. The correlations of factors (sources) with the metals show that in the coarse size range the dominant source is crustal re-suspension (68%) followed by vehicular pollution (23%). However, this is reversed in the case of the fine size range factor analysis where vehicular pollution (86%) dominated over crustal re-suspension (10%).

Keywords: Source apportionment; Chemical mass balance; Principle component analysis; Coarse particles; Fine particles; Delhi.

INTRODUCTION

Delhi, the capital of India is one of the 10 most polluted cities in world. It is the second largest Indian city with a population of over 14 million, with 1.3% of India's population. The

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annual average growth rate of Delhi's population is 3.85%, but the annual average vehicular growth rate is 5.85% (Economic Survey of Delhi, 2006). The transportation network in Delhi is predominantly road based with 1284 km of road per 100 km^2. Over 4.8 million vehicles travel Delhi's roads today. The number of vehicles is expected to reach 26 million in 2011. It is estimated that over 3000 metric tons of air pollutants are emitted in Delhi daily (MOEF, 2002).

This alarming vehicular growth rate, frequent traffic jams and not much improvement in the number of roads has resulted in a significant rise in the TSPM (total suspended particulate matter) level of Delhi. However, although vehicles continue to be the biggest contributor to the Delhi's ambient TSPM level, significant contributions from other pollution sources, such as roadside dust, industries, trans-boundary migrations, power plants, solid waste and local sources, have also been observed (Srivastava and Jain, 2006; Srivastava and Jain, 2007). Particulate matter from these sources may contain hazardous pollutants and can have carcinogenic and mutagenic effects. Thus, identification of the sources and estimates of their contributions are important.

Several methods are used to estimate source contributions, including principle component analysis (PCA or Factor Analysis), multiple linear regression analysis (MLR) and the Chemical Mass Balance (CMB) Receptor Model. In the present investigation, we have applied both CMB and PCA for the source apportionment of TSPM in the ambient air of Delhi. CMB models are fundamental receptor models and very useful for coarse and fine particle source apportionment (CMB8, 1997). CMB models estimate source contributions by determining the best-fit combination of emission source chemical composition profiles needed to reconstruct the chemical composition of ambient samples (Watson *et al*., 1991; Watson *et al*., 1994; Schauer *et al*., 1996). While the PCA method focuses on cleaning up the factors, it also produces factors that have high correlations with one smaller set of variables and little or no correlation with another set of variables (Stevens, 1996).

Many studies have been conducted on the source apportionment of different fractions of TSPM and metals in various parts of the world using different techniques (Artaxo *et al*., 1990; Kashara *et al*., 1990; Watson *et al*., 1994; Anderson *et al*., 1995; Hidy *et al*., 1996; Vega *et al*., 1997; Adgate *et al*., 1998; Liu *et al*., 1998; Biegalski *et al*., 1998; Batterman *et al*., 1988; Cheng *et al*., 1989; Ames *et al*., 2000; Bhave *et al*., 2001; Billheimer, 2001; Chen *et al*., 2001; Chow *et al*.,2002; Christensen *et al*., 2004; Salma *et al.,* 2005; Arditsoglou and Samara 2005; Gupta *et al.*, 2006). Recently, some important studies have been carried out using CMB receptor models and the PCA method: Breed *et al*. (2002) determined the possible sources of PM_{10} in Prince George (Canada) based on morphology and in situ chemical composition of the particulate matter. Samara *et al*. (2003) applied CMB source apportionment of PM_{10} in an industrialized urban area of northern Greece. Samara (2005) identified sources of TSP using CMB in a lignite burning area of Macedonia, Greece.

Zheng (2005) studied the seasonal trend of sources of $PM_{2.5}$ in Beijing.

However, in India studies on source apportionment by CMB are rather limited. Sharma and Patil (1992, 1994) used CMB and PCA respectively for source apportionment of aerosols in Bombay. Srivastava (2004) and Srivastava *et al*. (2005) carried out source apportionment of ambient VOCs in Mumbai and Delhi, respectively, using CMB8. Srivastava and Jain (2007a,c) carried out source apportionment studies using CMB and PCA, respectively, on data collected in 2001. In the present study, we assessed the air quality of Delhi at a time when almost all the public transport is compressed natural gas (CNG) fueled and metro rails have been introduced on some fixed routes. In this paper, an attempt has been made to assess the various sources of fine and coarse particulates in Delhi using CMB8 and PCA methods.

MATERIALS AND METHODS

Area descriptions

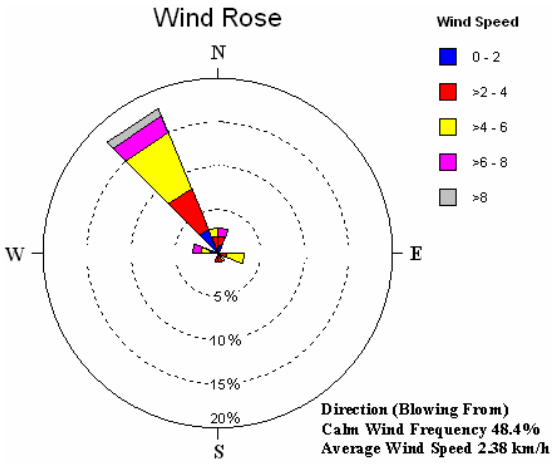

Fig. 1. Wind rose pattern of Delhi during sampling (Dec 10, 05 – Jan 20, 06).

Sampling was carried out at six different sites in Delhi. Delhi has three coal-based thermal power plants and over 125,000 industrial units (Government of India, 2001). It lies in the subtropical belt, characterized by a semi-arid climate, which consists of summer (March–May), monsoon (June–Aug), post-monsoon (Sep–Nov) and winter (Dec–Feb) seasons. It experiences a maximum temperature of ~45–48℃ in June during summer and a minimum of ~1–2℃ in January during winter. It has a normal annual rainfall of 611 mm. The air over Delhi is dry most of the year. Humidity is high in the monsoon season, while April and May are the driest months. Winds are predominantly westerly or northwesterly and tend to be more northerly in the afternoons, except in the monsoon season, when it is easterly and northeasterly. Yearly mean wind speed varies in the range of 0.9–2.0 m/s. The April to June period witnesses the highest frequency of thunderstorms and dust storms. These storms are generally dry, but some are accompanied by heavy rains. During the course of sampling, max, min temperature and max, min relative humidity were 21.0, 4.9^{O}C and 90.4, 45.0%, respectively. The wind rose pattern of Delhi is shown in Fig. 1.

Sampling sites

Sampling was performed from December 10, 2005 to January 20, 2006 at six different sites. Their locations are indicated in Fig. 2. The details of each category of sites and number of samples along with the sampling duration are given in Table 1.

Table 1. Sampling details.

Site	Site character	Sampling period	Number of samples
Jawaharlal Nehru University	Sensitive site	24 h	Four
Uttam Nagar	Residential site	24 h	Four
Nizamuddin	Sensitive site	24 h	Four
ITO	Heavy traffic site	24 h	Four
Okhla	Industrial site	24 h	Four
Cannaught Place	Commercial site	24 h	Four

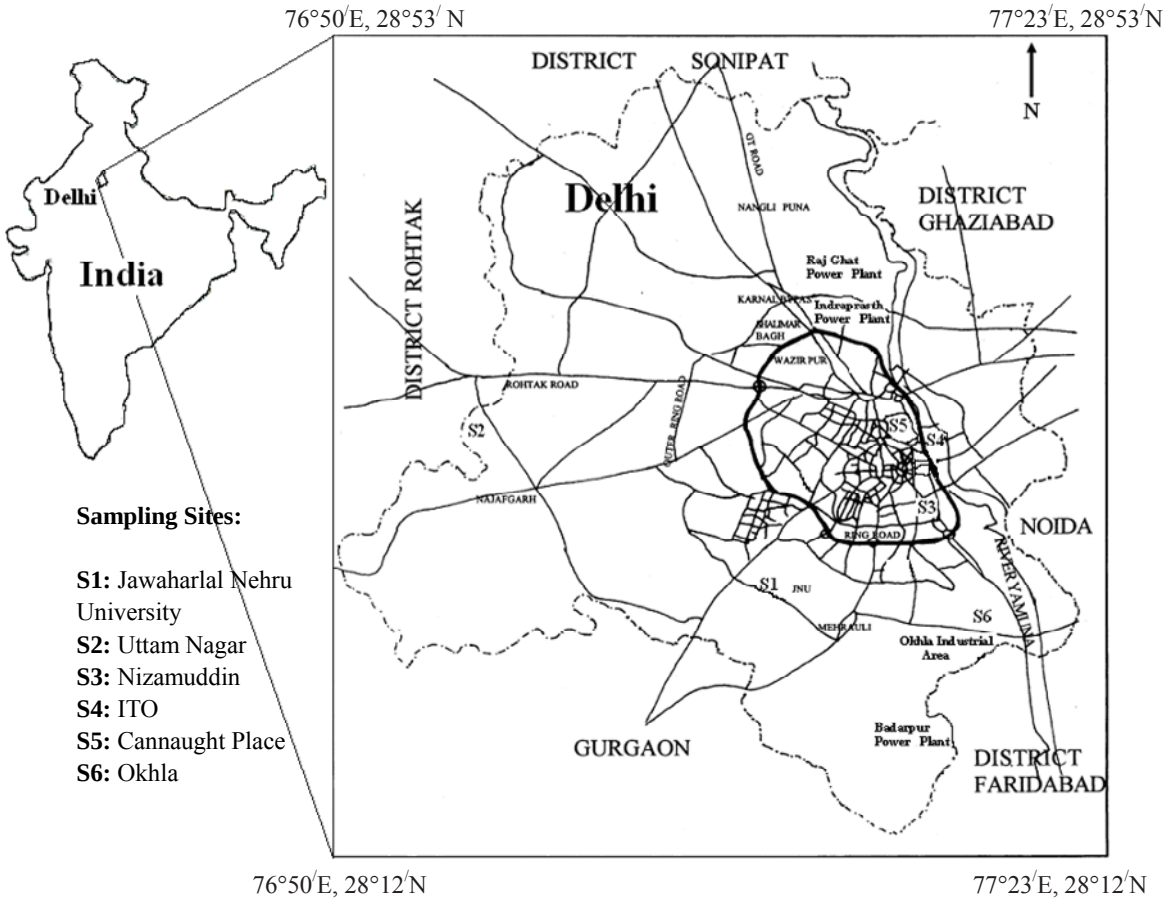

Fig. 2. Locations of sampling sites (not to scale).

Sampling procedure

Sampling was done with a five-stage cascade particulate separator (Kimoto Electric Co. Ltd. Japan). It was operated at an average flow rate

Table 2. Description of the different sizes of particles and filters.

Stage	Size range of particles	Shape of filter	Measurement of filter (mm)
1	> 10.9 μm	Doughnut	197.5 (outer diameter), 171.6 (inner diameter)
2	10.9–5.4 μm	Doughnut	177.5 (outer diameter), 147.5 (inner diameter)
3	5.4–1.6 μm	Doughnut	149.5 (outer diameter), 120.6 (inner diameter)
4	1.6–0.7 μm	Doughnut	132.0 (outer diameter), 97.6 (inner diameter)
5	< 0.7	Rectangular	203 × 254

of 600 L/min with 24 h continuous samples obtained from each site for four days. The samples were collected on EPM-2000, Whatman GF/A glass fiber filters. Each set of samples consisted of five different sized filters for various size ranges. The size ranges of the particles and the sizes of corresponding filters used for each size are provided in Table 2. The filters were kept in vacuum desiccators for 24 h to remove any moisture content, and then weighed by a precision micro balance (Mettler AE 50). Afterwards, they were mounted on the air sampler. After sampling, the filters were immediately transferred to vacuum desiccators to again de-moisturize in the same manner and then again weighed. The difference in pre- and post-sampling weights were used for the estimation of TSPM.

Analysis

Acid digestion, required for the metals determination by AAS, was carried out according to a standard procedure (Katz, 1977). Acid digestion was performed in Teflon bombs following these steps. Step 1: Sub samples of dry filters were dissolved in 3 mL HF, 6 mL HNO_3 and 1.5 mL HCl in Teflon bombs at 120℃ for 1 h; Step 2: Digestates were evaporated to dryness at 70℃ for 1/2 h; Step 3: Residues were re-dissolved in 10 mL of 10 M HNO_3 and evaporated to dryness. Step 3 was repeated until residues were fully dissolved. A series of blanks were prepared using the same digestion method. Standard solutions of metals were prepared as described in the USEPA manual (1983) and AAS manual (1983). Metals and reagents used for standard solutions were of AR grade. The reagents used were HNO_3 70% (S.G. 1.41), HCl 36% (S.G. 1.18) and HF 40% (S.G. 1.13).

Source apportionment methodology

To carry out source apportionment in two different size ranges the samples collected were broadly divided in two categories: coarse and fine. Coarse size particles represented the sum total of particles of size ranges: > 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. To gain a better understanding of the dominant sources in Delhi, two different source apportionment techniques were applied: Chemical Mass Balance Receptor Model, Version 8 (CMB8) and the Principle Component Analysis (PCA).

Chemical mass balance model

Table 3. Some of the CMB performance statistics.

Size	R^2	R/U	χ^2
Coarse	0.98	0.99	3.84
Fine	0.96	3.80	1.49

In this study, a chemical mass balance receptor model, CMB8 (CMB, 1997; USEPA, 1997; Watson *et al*., 1997) 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 from the source.

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

Where C_i is the ambient concentrations of the species *i*, measured at the receptor site, *P* is the number of sources that contribute, F_{ij} is the fraction of the emissions of the species *i* starting from the source *j*, and S_j 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 (Watson *et al*., 1997):

(i) Fraction of variance in measured concentrations, explained by the variation in calculated concentrations (R^2) more than 0.8, Weighted sum of squares of the difference between calculated and measured fitting species concentration (χ^2) less than 4,

(ii)Degree of freedom (DF) greater than 5,

(iii) Percent of aerosol mass explained by sources between 80-120,

(iv) Ratio of calculated to measured concentration (C/M ratio) between 0.5 – 2, and

(v) Absolute value of ratio of residual to uncertainty (R/U) less than 4.

Source profiles

The sources of 11 metals (Mn, Cr, Cd, Cu, Co, Pb, Ni, Fe, Zn, Mg and Ca) for both coarse and fine suspended particles were determined. It is important to note that the source profiles for Indian cities have not been compiled. Thus, the metals source profiles used were taken from the USEPA (Speciate 3.2, 2002) database and some of the previous work carried out in Delhi (Khemani *et al*., 1985; Mehra *et al*., 1998; Balachandran *et al*., 2000; Gadi *et al*., 2000; Anju and Banerjee, 2003; Srivastava and Jain 2003; Khillare *et al*., 2004; Monkkonen *et al*., 2004; Srivastava and Jain 2005; Yadav and Rajamani 2006; Srivastava and Jain 2007a,,b,c). The emission sources considered were (i) Soil and crustal dust, (ii) Paved road dusts, (ii) Vehicular emissions, and (iv) Industrial sources.

Principle component analysis

To identify and estimate the possible sources of coarse and fine size particles, a PCA, or factor analysis, method was applied. PCA was performed by the Varimax Rotated Factor Matrix method, based on the orthogonal

rotation criterion which maximizes the variance of the squared elements in the column of a factor matrix, using a statistical package SPSS (Version 9). This method focuses on cleaning up the factors. It produces factors that have high correlations with one smaller set of variables and little or no correlation with another set of variables (Stevens, 1996).

RESULTS AND DISCUSSION

Source apportionment was carried out for both coarse and fine size ranges of TSPM using CMB8 and PCA methods. The results of their analyses are discussed below.

Source apportionment using cmb8 receptor model

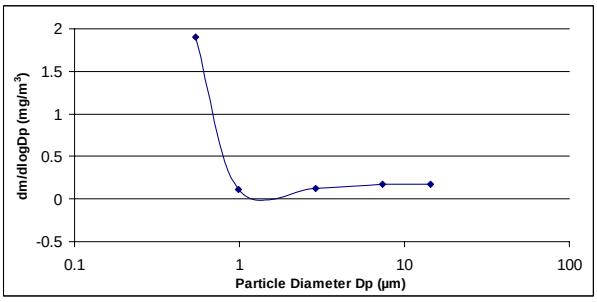

Fig. 3. Mean mass size distribution of daily (24 h) average TSPM

Source contributions from various sources in the coarse and fine size ranges are provided in Fig. 3. Table 2 gives the R^2 values along with the range of ratio R/U and χ^2 values. In the fine size range, vehicular emissions are the dominant source. In the case of the coarse fraction, crustal dust is most dominant. The industrial contribution is more or less the same

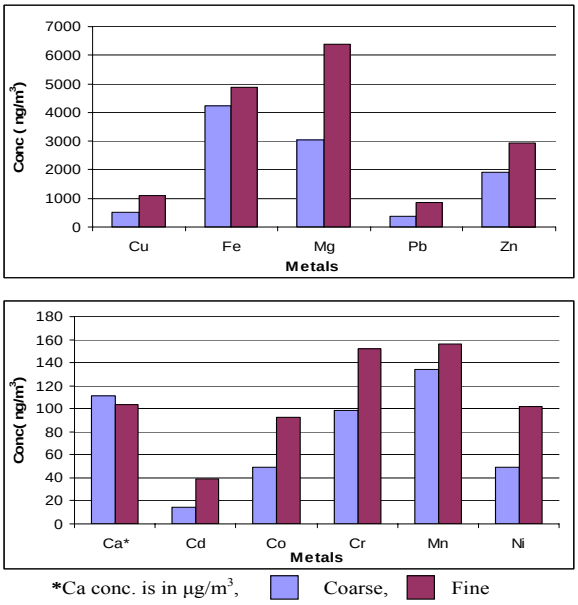

Fig. 4. Daily (24 h) average concentration of different of metals.

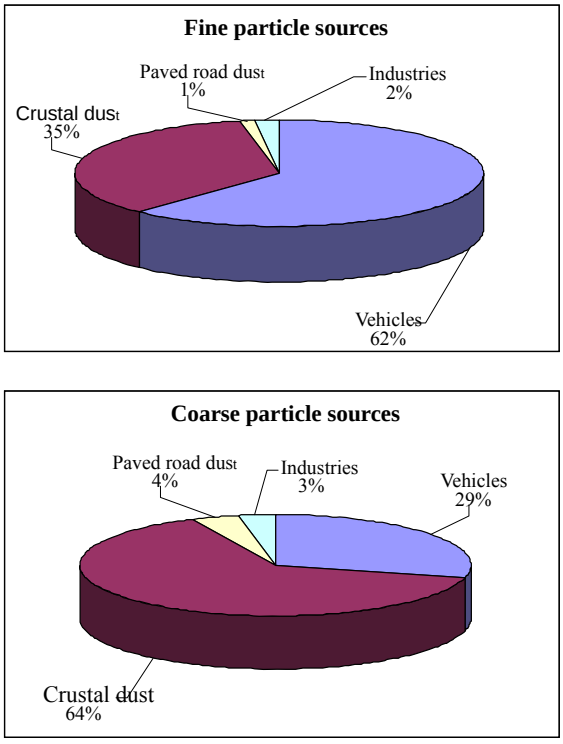

Fig. 5. Source contribution to TSPM in coarse and fine particle range by CMB 8.

in both coarse and fine size fractions; i.e., 3 and 2%, respectively. The contribution from paved road dust is somewhat more for coarse particles (4%) than fine particles (1%). The results obtained in this study are not in agreement with those from the previous study by Srivastava and Jain (2007a) in which it was observed that vehicular pollution dominates in both coarse and fine size ranges. This study was carried out before the implementation of compressed natural gas vehicles (CNG). Therefore, we can conclude that after the implementation of CNG, although vehicles continue to be the largest polluters of Delhi's ambient air, their over-contribution is now confined to fine particles only, while soil and crustal dust are now the predominant contributors to coarse particles.

Source apportionment by PCA

To validate the results obtained by CMB, a PCA was performed by the Varimax Rotated Factor Matrix method of SPSS for both coarse and fine size fractions. Sites were assumed to be repetitive of data, while metals were treated as the variable. A total of nine components were obtained, out of which two were extracted as principle components (Eigenvalue > 1) for both coarse and fine size ranges. The detailed results are given in Table 4. In case of coarse particles, two PCs were obtained, first and second PCs contribute about 68 and 23%, respectively. Considering a correlation coefficient of ≥ 0.5, the metals correlated with factor 1 points it to be the crustal re-suspension source and factor 2 as the vehicular source. It is important to note that the metals generated from various sources also become part of crustal dust over time (Khemani *et al*., 1985; Balachandran *et al.,* 2000; Anju and Banerjee, 2003; Monkkonen *et al*., 2004; Khillare *et al*., 2004). The condition is reversed in the case of fine particulate source apportionment. Again, two PCs were obtained, contributing about 86 and

Table 4. Varimax rotated factor loading matrix.

Size Range	Variable	Factor 1	Factor 2
Fine Particles	Cu	0.405	
	Pb		
	Fe		
	Cd		
	Cr	0.499	
	Zn	0.355	
	Ni	0.328	0.428
	Mg		0.992
	Co		
	Mn		
	Ca	0.487	0.874
Eigenvalues		96.853	11.286
% of Variance		85.609	9.975
Cumulative %		85.609	95.585
Possible Source		Vehicular	Crustal re-suspension
Coarse Particles	Cu		0.990
	Pb		
	Fe	0.426	
	Cd	0.669	
	Cr	0.551	
	Zn	0.780	
	Ni	0.356	0.675
	Mg	0.644	
	Co		0.675
	Mn	0.395	
	Ca	0.961	
Eigenvalues		21.674	7.242
% of Variance		67.936	22.700
Cumulative %		67.936	90.636
Possible Source		Crustal re-suspension	Vehicular

Table 5. Source contributions by both CMB 8 and PCA models.

Size range	Model used	Source contribution (%)			
		Vehicles	Crustal dust	Industries	Paved-road dust
Fine	CMB8	62 %	35 %	2 %	1 %
	PCA	86 %	10 %	Not clear	Not clear
Coarse	CMB8	29 %	64 %	3 %	4 %
	PCA	23 %	68 %	Not clear	Not clear

10% respectively. The correlation between metals and factors suggests factor 1 as vehicular source and factor 2 as crustal re-suspension. The differentiation between vehicular source and crustal re-suspension was made keeping in mind the association of Ca and Mg, because these metals are not associated with vehicular pollution. It was also confirmed from the enrichment factor discussed previously that Ca and Mg at various sites are mainly of crustal origin.

CONCLUSIONS

Source apportionment carried out by both CMB8 and PCA identified two major sources: vehicular and crustal dust (crustal re-suspension for PCA). Results of the CMB8 and uses also revealed the contributions of paved road dust and industrial pollutants to be small in both the coarse and fine size ranges. It has also been observed that the over-dominance of vehicular pollutants are now (2005-06) confined to fine particles only, while they dominated both coarse and fine particles during pre-CNG implementation era (2001).

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