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Spatial distribution in aerosol mass and size characteristics between Delhi and Hyderabad during land campaign in February 2004

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Mass and size distribution of aerosols was studied using different measurement techniques, along Delhi-Hyderabad land corridor, to understand the spatial variability of aerosols over north central Indian region, from Delhi to Hyderabad and back, in the month of February 2004. Aerosol mass loading of Quartz Crystal Microbalance (QCM) derived up to PM25, PM10, PM2.5, PM1 size fractions, were found to be in the range of 17-56, 16-54, 8-44 and 5-37 μg m^{-3}, respectively. The QCM derived up to PM10, PM2.5 and PM1 with respect to total aerosol mass (PM25) are 90%, 80% and 70%, respectively, which indicate the dominance of finer particles. The PM10 aerosol concentrations measured by High Volume Respirable Dust Sampler (HV-RDS) had an average value of 48 μg m^{-3} with the range of 27-75 μg m^{-3}. Aerosol average concentrations by HV-RDS were observed to be 54 ± 5 μg m^{-3} near urban areas, 48 ± 2 μg m^{-3} in forest area and 44 ± 22 μg m^{-3} in rural areas along the land corridor.

Keywords: Particulate matter; High volume sampler; Anderson cascade impactor; Aerosol size distribution; Quartz crystal microbalance

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1 Introduction

Mass and size characteristics of aerosols are important parameters in determining their radiative behaviour[1]. Aerosol size distribution is influenced by the dynamics of aerosol population[2], their production and removal processes, size transformation; lifetime[3,4] and influences their optical properties and radiative effects[5-7]. The fine and accumulation range particles affect visibility, produce health effects and are important in radiative interactions. Accumulation mode particles (PM < 1 μm) may remain in the atmosphere for days-to-weeks, since atmospheric removal processes are least efficient in this size range. Characterization of size distribution and spatial distribution of atmospheric aerosols is important[8-9], which may indicate about the origin of particulates[2,7,10] and their impact.

A land based mobile campaign covering a distance between Delhi-Hyderabad-Delhi of about 3200 km, for measurement of aerosols using different technique, was carried out to study their concentration and distribution as part of Indian Space Research Organization-Geosphere Biosphere Programme (ISRO-GBP) during February 2004. The spatial distance covered was between 17.3 and 28.6° N in latitude and 77.2 and 78.2° E in longitude (Table 1). These studies were first of their kind and have provided baseline data to understand the radiative forcing of aerosols over north central Indian peninsula.

2 Experimental methodology

2.1 Study locations and their background

Sampling sites (Table 1) were chosen along the land corridor to provide representative location of different ecosystems in the route passing through the states of Delhi, Uttar Pradesh (UP), Rajasthan, Madhya Pradesh (MP), Maharashtra and Andhra Pradesh (AP). The aerosol measurements were done using quartz crystal microbalance (QCM), Anderson cascade impactor (ACI) and high volume respirable dust sampler (HV-RDS), for 6-8 h at each site, from 0900 to 1700 hrs IST approximately, on alternate days and at some places QCM observations for 1-2 h during forenoon (FN) and afternoon (AN) in the onward and return journey.

During the campaign, observation sites covered were urban (U), semi-urban (SU), rural (R) and forest (F) regions (Table 1). The New Delhi sampling site was at Central Road Research Institute (CRRI)

Table 1—Description of measurement sites along Delhi-Hyderabad-Delhi land corridor

S.No.	Date	Name of Measurements Location	District / State	Geographical Location Latitude–Longitude	Equipments used*	Ecosystem*
1	1 Feb. 2004	CRRI Delhi	Delhi	28°35'N-77°12'E	QCM	U
2	2 Feb. 2004	Kosi Kalan	Mathura (UP)	27°49'N-77°25'E	QCM & RDS	SU
3	3 Feb. 2004	Garrhi Nohbar	Agra (UP)	25°30'N-77'E	QCM	R
4	4 Feb. 2004	Orcha	Tikamgarh (MP)	24°18'N-77°37'E	QCM & RDS	F
5	5 Feb. 2004	Orcha	Tikamgarh(MP)	24°18'N-77°37'E	QCM & RDS	F
6	6 Feb. 2004	Satravasa	Lalitpur(MP)	24°42' N- 78°25'E	QCM	R
7	7 Feb. 2004	Sanuda	Sagar(MP)	23°53'N-78°54'E	QCM & RDS	R
8	8 Feb. 2004	Murgakheda	Jabalpur (MP)	23°22'N-80°04'E	QCM	F
9	9 Feb. 2004	Tikaria	Jabalpur MP)	23°22'N-80°04'E	QCM & RDS	F
10	11 Feb. 2004	Kurari	Seoni (MP)	21°46'N-79°31'E	QCM & RDS	F
11	13 Feb. 2004	Bhivapur	Nagpur (Mah)	21°5'N-79°2'E	QCM	F
12	14 Feb. 2004	Bussapur	Nirmal (AP)	18°57'N-78°23'E	QCM & RDS	R
13	16 Feb. 2004	Shaadnagar	Hyderabad (AP)	17°1.7N-78°11.3'E	QCM, RDS & ACI	SU
14	17 Feb. 2004	Shaadnagar	Hyderabad (AP)	17°1.7N-78°11.3'E	QCM, RDS & ACI	SU
15	18 Feb. 2004	Shaadnagar	Hyderabad (AP)	17°1.7N-78°11.3'E	QCM, RDS & ACI	SU
16	19 Feb. 2004	Shaadnagar	Hyderabad (AP)	17°1.7N-78°11.3'E	QCM, RDS & ACI	SU
17	20 Feb. 2004	Shaadnagar	Hyderabad (AP)	17°1.7N-78°11.3'E	QCM, RDS & ACI	SU
18	21 Feb. 2004	Shaadnagar	Hyderabad (AP)	17°1.7N-78°11.3'E	QCM, RDS & ACI	SU
19	22 Feb. 2004	Ramampeth	Hyderabad (AP)	18°07'N-78°27'E	QCM & RDS	SU
20	23 Feb. 2004	Pandher Kavra	Nagpur (Mah)	20°05'N-78°34'E	QCM	SU
21	23 Feb. 2004	NEERI	Nagpur (Mah)	21,07N - 79,04E	QCM, RDS & ACI	U
22	24 Feb. 2004	NEERI	Nagpur (Mah)	21°07'N-79°04'E	QCM, RDS & ACI	U
23	25 Feb. 2004	Jhiria Tola	Jabalpur (MP)	21°54'N-79°31'E	QCM	F
24	26 Feb. 2004	Mehar	Satna (MP)	24°17'N-80°46'E	QCM	F
25	27 Feb. 2004	Khajuraho	Chhatapur (MP)	24°52'N-79°56'E	QCM & RDS	SU
26	29 Feb. 2004	Dholpur	Rajasthan	26°40'N-77°54'E	QCM & RDS	SU

***** U-Urban, SU-Semi Urban, R- Rural and F-Forest
*QCM- Quartz Crystal Microbalance, RDS- Respirable Dust Sampler & ACI -Anderson Cascade Impactor

campus at about 100 m away from Delhi-Mathura road. Kosi site lies about 2 km from NH-2. The Agra sampling site surrounded by rich vegetation was about 500 m from the main road and 15 km away from the city. Orchha site was covered by thick forest. Khajuraho represented a semi-urban environment. Dholpur was semi-urban and one of the arid regions in Rajasthan and locality was dusty. Kurai site was inside a forest 4 km away from the main road on NH-7, situated on a narrow, North-South section of Satpura plateau. Nirmal site, on NH-7 was surrounded by rich vegetation, and was 2-3 km away from the main road. In Sagar, observations were taken about 15 km away from the city. Hills and some villages surrounded Jabalpur site. Nagpur site was near residential buildings at National Environmental and Engineering Research Institute (NEERI). In Ramampeth, agricultural and sugarcane trash burning activities were prominent in nearby rural areas. Shadnagar site was located about 80 km south of Hyderabad city. At Shadnagar, some nighttime observations were also taken from 16 to 21 Feb. 2004.

2.2 High volume respirable dust sampler (HV-RDS)

The sampling of PM-10 was carried out by HV-RDS (Netel Chromatographs, HVS/R) technique, operated at a flow rate of 1.1-1.5 m^3 min^{-1} for 6-8 h. Aerosol samples were collected on Whatman quartz micro-fiber (QM-A) filters and Whatman-41 cellulose filters. Quartz filters were pre-heated and desiccated before sampling and cellulose filters were desiccated. The mass concentrations of the aerosols were determined by gravimetric method after sample desiccation for 48 h.

2.3 Anderson cascade impactor (ACI)

The aerosol sampling of size fractions (ranges in μm viz. ≥ 9.0, 5.8-9.0, 4.7-5.8, 3.3-4.7, 2.1-3.3, 1.1-2.1, 0.7-1.1, 0.4-0.7 and 0-0.4) was carried out by eight-stage ACI (California Measurements Inc., USA)

technique, operated for 48-72 h at air flow rate of 28 l min^{-1}. Three ACI samplings were done from 16-19 Feb. 2004 (68 h) and 19-21 Feb. 2004 (58 h) at Shadnagar on quartz QM-A filters and from 23-24 Feb. 2004 (26 h) at Nagpur on cellulose Whatman-41 filter. The mass concentration of aerosol was determined gravimetrically.

2.4 Quartz crystal microbalance (QCM)

The mass and size distributions of aerosols were measured using the QCM cascade impactor (model PC-2, California Measurements Inc., USA). This instrument samples the ambient air and segregates the aerosols in accordance with the aerodynamic diameter (0.05-25 μm) into ten size bins. The instrument was operated at about 2 m above ground and at the ambient relative humidity (RH) of around 50 %. The QCM day-data with a sampling duration of 1 min at every 15-min interval for a total of 4-6 h day^{-1} was collected for 26 days during 1-29 Feb. 2004. Total mass (M_t) were segregated[11] into sub-micron (M_a) and super-micron (M_c), where M_a is the mass concentration in the accumulation size range (0.05-0.4 μm) and M_c in the coarse size range (0.8-12.5 μm), such that, $M_t = M_a + M_c$. Aerosol number density was estimated from the aerosol (assuming spherical particles) size segregated volume concentration as

$$n_{ci} = \frac{v_{ci}}{\frac{4}{3}\Pi r_i^3}$$

where v_{ci} is the volume concentration ($v_{ci} = m_{ci}/\rho$; ρ is assumed as 2.0 g cm^{-3})[11] and r_i the radii of aerosol particle. The total number density is estimated from the size segregated number density as

$$N_t = \sum_{i=2}^{10} n_{ci}$$

From n_{ci} the number size distribution is deduced as

$$\frac{dn}{dr} = \frac{n_{ci}}{\Delta r_i}$$

In calculation, the aerosol particle size up to 25, 12.5, 3.2 and 0.8 μm are considered as PM25, PM10, PM2.5 and PM1, respectively[11].

3 Results and discussion

Spatial variations in mass of sub-micron (M_a; 0.05-0.4 μm) and super-micron (M_c; 0.8-12.5 μm) size particulate measured by QCM and the data on meteorological observations of ambient temperature, pressure, relative humidity, wind speed and direction, at different sites are shown in Fig. 1. The sampling period was nearly same for all sites. The difference between M_a and M_c is more at Shadnagar (Hyderabad) site and in most sites M_a dominates over M_c, indicating the anthropogenic influence (Fig. 1). Value of M_a had mass concentrations around 23±10 μg m^{-3}, and was higher than M_c (10±4 μg m^{-3}) at all sites, except Delhi, Dholpur and Kurai. The enhancement in M_c and M_a concentrations at Delhi may be due to anthropogenic sources, in addition to low temperatures/ winter inversion and high humidity, increasing the growth or conversion of fine into coarse particles. Comparatively more M_c at Dholpur and at Kurai, was due to local dust from arid region. In campaign, QCM derived average mass loading of up to PM25, PM10, PM2.5, PM1 aerosol size fractions were 30, 28, 23, 20 μg m^{-3} respectively, with their ranges 17-56, 16-54, 8-44 and 5-37 μg m^{-3} respectively (Fig. 2).

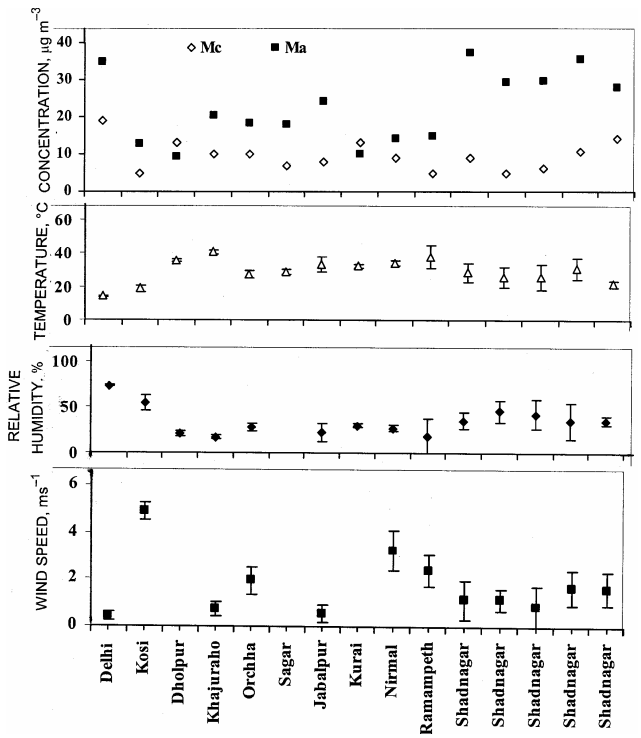

Fig. 1—Variations in M_a and M_c aerosols fractions as measured by QCM and the meteorological observations

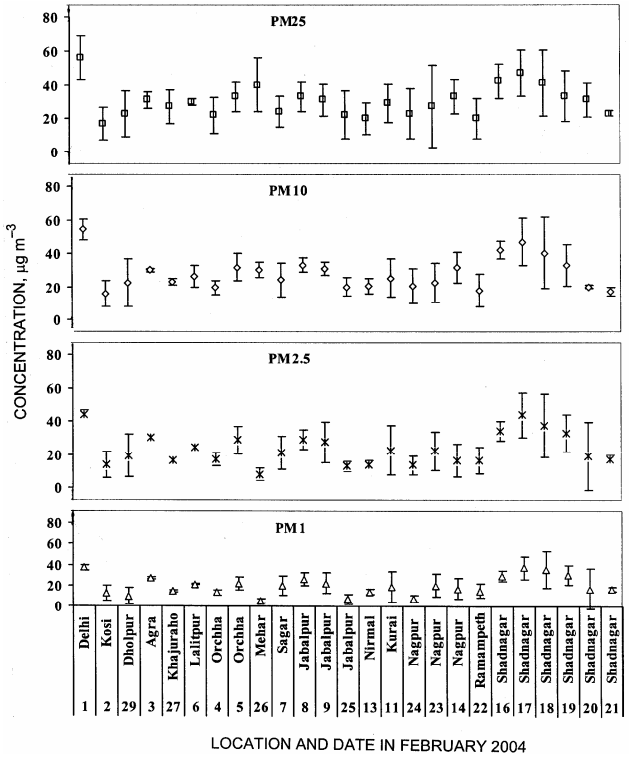

Fig. 2—Aerosol average mass concentrations of PM1, PM2.5, PM10 and PM25 as measured by QCM

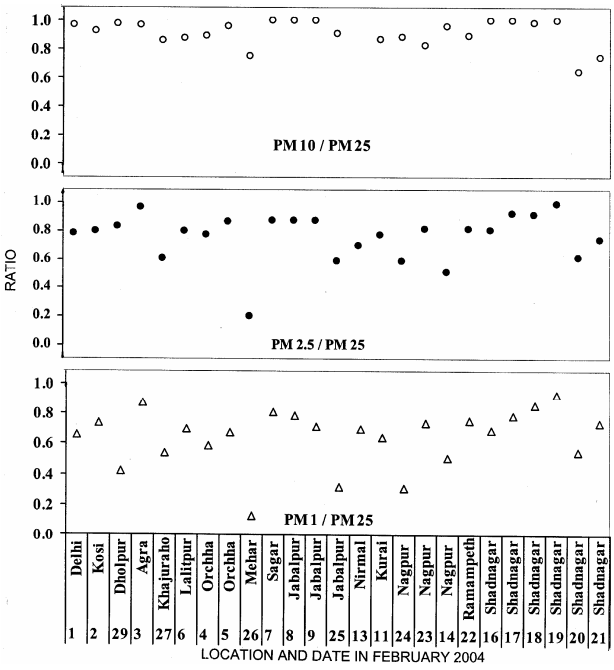

Fig. 3—Variations in PM1, PM2.5 and PM10 mass fractions with respect to PM25 as measured by QCM

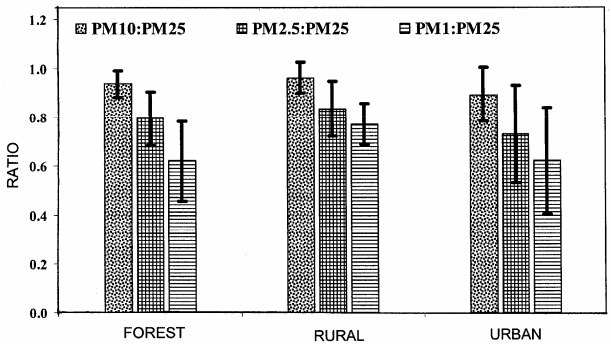

Fig. 4—Aerosol average PM1, PM2.5 and PM10 mass fraction characteristics to QCM total mass in urban, rural and forest regions

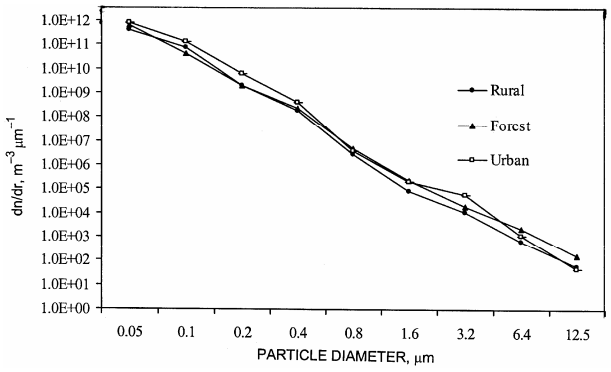

Fig. 5—Average aerosol number size distributions in urban, rural and forest regions as measured by QCM

Average QCM derived particulate mass (PM25) concentration in rural regions along the campaign route was 20±10 µg m^{-3}. The large spread in QCM mass concentrations for different aerosol sizes at most of the sites indicates the large variability as well as contributions from different sources like soil, vegetation, domestic, vehicular traffic, etc. and also effect of local meteorological conditions (Fig. 1). The ratios of average aerosol mass concentrations for sizes up to PM10, PM2.5 and PM1 with respect to total mass PM25 was found in the range of 0.80-0.95, 0.70-0.85 and 0.50-0.70, respectively (Fig. 3). These ratios were higher for rural sites followed by forest and urban sites, respectively (Fig. 4). Aerosols mass variations were more in the accumulation and coarse range however, for some sites like Mehar (nearly 10 km from a cement factory), Kurai (forest region), Nagpur (near urban), and Dholpur (arid region) had most of the mass fraction coming from coarse particles and which was higher than that from accumulation mode (Fig. 5).

The distribution in aerosol number density in different sizes, are shown in Fig. 5, which indicates that the number-size distribution was multi-modal,

wherein accumulation mode ($d_i \leq$ ~0.8 μm or PM1) aerosols were less dominant in rural and forest regions as compared to urban. Coarse mode ($d_i \leq$ ~12.5 μm or PM10) aerosols had shown the reverse trend. The number size particle concentration in the higher radii or coarse aerosol is generally insensitive to the urban characteristics, however a small enhancement was observed (Fig. 5). This suggests that most of the fine particles are of anthropogenic origin.

The HV-RDS technique has given an average PM10 loading for the rural background of about 48±2 μg m^{-3} (Fig. 6) with a range of PM10 from 27 to 75 μg m^{-3} except at Dholpur site, which has shown high value of 232 μg m^{-3}. The background PM10 concentrations attributed to urban, forest and rural regions were 54 ± 5, 48 ± 2 and 44 ± 22 μg m^{-3} respectively. Three observations by ACI or cascade impactor technique at Shadnagar and Nagpur sites have given average PM10 (sum of all 8 stages mass) aerosol mass concentrations of 42, 118 and 29 μg m^{-3}, respectively (Fig. 7).

The higher value in PM10 at Shadnagar is in all particle size ranges and may be due to urban Hyderabad and local influence. Semi-urban site at Shadnagar, during 16-19 Feb. 2004 has indicated high particle concentration in the size fractions of ≥ 9.0, 1.1-2.1 and 0.0-0.4 μm in PM10 mass as compared to Nagpur. Measurement made by QCM (low volume of 250 cc min^{-1} air flow), HV-RDS (high volume of > 1 m^3 air flow) and ACI (moderate 32 l min^{-1} air flow) are three different techniques representing aerosol mass data integration on near real-time (minutes), for few hours (6-8 h) and for many hours (48-72 h), respectively. This study thus also indicates influence on derived aerosol mass concentration values, due to variability of different integration times from different sampling techniques. The results for coarse particles are comparable to the other reported sites of land campaign of other routes[12]. In rural areas larger size particles dominated, indicating the contribution of soil originated particles. Fine particulates dominated in urban areas due to vehicular traffic and other urban sources[13].

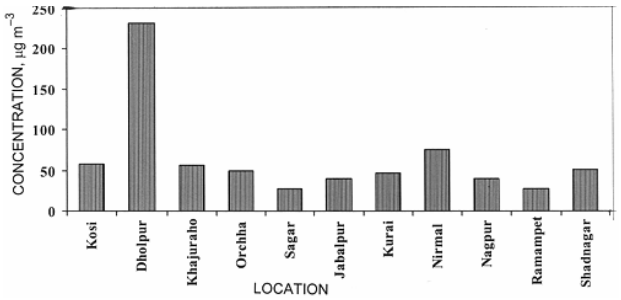

Fig. 6—Spatial distribution of PM10 mass concentrations along Delhi-Hyderabad-Delhi land corridor by RDS

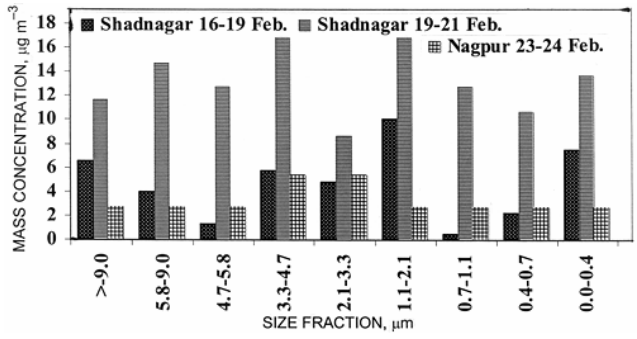

Fig. 7—Aerosol mass-size distribution by cascade impactor at Shadnagar and Nagpur sites

4 Conclusions

Aerosol mass and size measurements were made at 20 locations in February 2004, between Delhi and Hyderabad land corridor, including on inter-comparison days at Shadnagar. The Delhi–Hyderabad–Delhi road route had shown aerosol background concentration for rural ecosystem of about 20 μg m^{-3} by QCM with a range of 10-30 μg m^{-3}. Aerosol number size distributions for rural, forest, and near urban regions had shown values between 10^2 and 10^{11} m^{-3} μm^{-1}. Aerosols mass-size distribution in the rural and semi-urban regions of the study area was dominated by surface-derived dust particles, whereas in the urban regions, there was an overall dominance of fine particles. Aerosol loading in rural, urban, semi urban and forest areas were different under influence of local activities. Study also indicated influence on derived aerosol mass concentration values, due to variability of different integration times from different sampling techniques.

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