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Intra-urban variability of particulate matter ($PM_{2.5}$ and PM_{10}) and its relationship with optical properties of aerosols over Delhi, India



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ABSTRACT

Highly time-resolved measurements of particulate matter (PM: PM_{2.5} and PM₁₀) were made at three different sites across Delhi (CCRI: a highly traffic site, IMD: a less traffic site and IITM: an urban background site) from 1st December, 2011 to 30th June, 2013. Also, coarse mode ($PM_{10-2.5}$) mass was estimated as the difference between PM₁₀ and PM_{2.5}. In addition, columnar aerosol optical properties such as aerosol optical depth (AOD) and Angstrom exponent (AE) were studied concurrently over IMD. The mean mass concentrations of PM_{2.5}, $PM_{10-2.5}$ and PM_{10} were 118.3 \pm 81.7, 113.6 \pm 70.4 and 232.1 \pm 131.1 $\mu g \, m^{-3}$, respectively. Among the three sites, relatively higher mass concentrations of PM_{2.5} (~35% and 3%) were observed at CRRI compared to IMD and IITM.PM₁₀ and PM_{10-2.5} were higher at these sites by ~31% and 19%; and 27% and 40%, respectively, compared to CRRI. Coefficients of divergence (COD) and correlation coefficients (r) were calculated between site pairs to assess the spatial and temporal heterogeneity of PM and moderate spatial divergence was found over the three sites. Traffic emission particles (PM_{2.5}) exhibited high spatial heterogeneity as well. The mass concentrations of PM_{2.5} and PM₁₀ were found to be higher during the night compared to the day. The mean $PM_{2.5}/PM_{10}$ ratio was ~51%, indicating generally equal amounts of coarse and fine mode PM in the Delhi urban atmosphere. AOD and PM_{2.5} were positively correlated and a negative correlation was observed between AE and PM_{10-2.5}. PM_{2.5} particles were significantly correlated with AOD during post-monsoon and winter. Because of the lower vehicular emissions on weekends compared to weekdays, PM at CRRI, IMD, and IITM were separated by day of week and large heterogeneities were found. During weekdays, the mass concentrations of PM₁₀ were ~4, 2, and 12% higher than on weekends. However, for PM_{2.5}, weekend values were 5, 7, and 9% higher for CRRI, IMD and IITM, respectively.

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1. Introduction

The concentrations of particulate matter (PM) in the ambient air have increased during recent years because of the rapidly increasing urbanization, industrialization, and the resulting anthropogenic activities. As a result of these activities, large cities of the world are facing serious air quality problems, particularly in the second half of the last century, causing increased air pollutant concentrations and related environmental hazards (Elbayoumi et al., 2013). The air pollutant concentrations are not only an environmental issue within the cities themselves, but also have regional and global environmental effects (Delfino et al., 2005; Obaidullah et al., 2012). Atmospheric PM plays a crucial role

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in atmospheric visibility, precipitation, and air quality. It can be classified into primary and secondary particles based on their emission sources or their formation mechanisms (Poschl, 2005; Tiwari et al., 2014a).

Particles affect the climate directly because of their effects on the solar and atmospheric radiation. Particles can behave as a cooling or warming factor, depending on the process that predominates: light absorption or scattering (Myhre, 2009). They can also have an indirect effect on the climate by modifying physical and radiative properties of clouds (Wang and Penner, 2009). Through this effect they can influence cloud lifetime and precipitation processes. However, at present, knowledge of the net effect of particles on the climate is at a medium to low level of scientific understanding.

To evaluate the climate effects of particles, it is necessary to determine their local dynamics. Since PM can have high spatial and temporal variability (Lagadu et al., 2011; Kumar et al., 2012), it is important to characterize the PM properties at multiple sites. This present study

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has been conducted across Delhi, India located in the northwestern Indo-Gangetic Plains (IGP), encompassing the highly populated Ganga River basin where more than 700 million inhabitants live and are exposed to the extensive pollution from natural and anthropogenic sources (Dey and Tripathi, 2007; Srivastava et al., 2012b). The IGP region extends from north to east and is one of the most poluted regions in northern India with emissions from small scale industries, vehicular traffic and biomass burning (wood-fuel and agriculture waste) (Srivastava et al., 2012b; reference therein). The relative increase in the source strength of anthropogenic emissions, lower boundary layer heights during wintertime, and the Himalayan topology confine the pollutants within IGP (Tiwari et al., 2013a,b). Under favorable meteorological conditions, advective transport from northern India and subsequent dispersal of pollutants to the marine atmospheric boundary layer of the Bay of Bengal has been well documented (Lelieveld et al., 2001; Sudheer and Sarin, 2008).

Aerosol optical depth (AOD) is a parameter describing air quality and atmospheric conditions and is used to estimate the concentrations of PM (Dey et al., 2012; Srivastava et al., 2014). AOD has a strong correlation with the $PM_{2.5}$ and PM_{10} concentrations (Srivastava et al., 2012a; Xin et al., 2014).

Physico-chemical characterization of $PM_{2.5}$ and PM_{10} has been conducted in northern and other parts of India (Das et al., 2009; Kulshrestha et al., 2009; Pipal et al., 2011; Srivastava and Jain, 2009; Tiwari et al., 2009). However, limited work has been done using continuous, simultaneous spatially dispersed measurement of PM ($PM_{2.5}$ and PM_{10}). The main objectives of this study are to (i) characterize the daily, monthly, seasonal, and spatial variability of PM (fine: $PM_{2.5}$, coarse: $PM_{10-2.5}$ and inhalable: PM_{10}), (ii) understand the impact of meteorology on local atmospheric environment, and (iii) study the relationship between optical properties and PM over a mega city "Delhi."

2. Observational site and experiment setup

2.1. Topography and weather conditions at Delhi

This study has been conducted at three locations in Delhi with simultaneous, continuous measurements of $PM_{2.5}$ and PM_{10} from 1st December 2011 to 30th June 2013. The sampling locations are the Central Research Road Institute (CRRI: high traffic), the Indian Institute of Tropical Meteorology (IITM: urban background with high traffic), and the India Meteorological Department (IMD: less traffic). Simultaneous columnar aerosol optical properties were measured with a Sun/sky radiometer installed on the premises of IMD, Lodhi road, New Delhi.

Delhi has many small/large scale industries and three major coal-fired power plants along with a high vehicular density (4.8 million) (Srivastava and Jain, 2009). Delhi is situated between the rain-washed Gangetic plains in the east and semi-arid tracts of the Rajasthan to the southwest, about 1100 km from the coast of the Arabian Sea. This city of ~19 million inhabitants is the fourth most polluted city in the world with respect to suspended particulate matter (Chelani et al., 2010; Goyal and Sidharta, 2002). The climate of Delhi is semi-arid with maximum temperatures of ~45–48 °C during the pre-monsoon and minimum temperatures during winter reaching ~1–2 °C (Tiwari et al., 2009).

To study the seasonal trends, the year was classified into four distinct seasons i.e., pre-monsoon (April–June), monsoon (July–September), post-monsoon (October–November) and winter (December–March). During the pre-monsoon, the area is mostly influenced by dust (Thar Desert), transported from the northwest and west (Pakistan, Afghanistan and Iran) whereas in the pre-monsoon and monsoon seasons, the area is dominated by highly humid, southwesterly winds from the Bay of Bengal and the Arabian Sea. During the post-monsoon and winter seasons, the wind comes mostly from the northwest bringing dry, cool air called western disturbances. In general, winds flow across the Himalayas and move southeasterly across the country. The

cold and dry weather associated with the northwesterly winds coming from the northwest Himalayas is characteristics of the winter season over central IGP (Mishra and Shibata, 2012).

Fig. 1 shows the aerosol optical depth (http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instanceid=MODIS_DAILY_L3:back ground AOD) over the Indian subcontinent during the study period and the sampling locations in Delhi. Higher aerosol loadings are observed over the IGP region compared to other parts of Indian subcontinent. The average AOD over Delhi during study period was nearly 0.7.

Meteorological parameters such as hourly temperature (temp.: °C), relative humidity (RH: %), visibility (VIS: km) and wind speed (WS: m/s) were obtained from the India Meteorological Department's observatory located at the Indian Agricultural Research Centre, 500 m from the IITM monitoring location. Mixing heights (MH: meter) were retrieved from the stability time series data available on the NOAA Air Resources Laboratory web server (http://www.arl.noaa.gov/ready. html). During the study period, the mean values of daily averages of weather parameters and its variation were MH (702 \pm 431 m) varying from 354 (winter) to 1213 m (pre-monsoon); temp. (24.4 \pm 8.2 °C) varied from 16.7 (winter) to 39.5 °C (pre-monsoon); WS (3.08 \pm 0.80 m/s) varied from 2.39 (post-monsoon) to 3.47 m/s (pre-monsoon); RH (60 \pm 21%) varied from 40 (pre-monsoon) to 80% (monsoon) and VIS (2.6 \pm 0.9 km) varied from 1.9 (post-monsoon) to 3.2 km (monsoon).

2.2. Instrumentation

Beta-attenuation monitors (Thermo Andersen, Inc. USA, series FH 62 C14) with temporal resolution of 1 hour were deployed at all three stations for PM mass measurements (i.e. $PM_{2.5}$ and PM_{10}) (Tiwari et al., 2014a). These units use sharp-cut cyclone inlets with particle cut-off diameters $d \le 2.5 \ \mu m$ (for $PM_{2.5}$ sampler) and $d \le 10 \ \mu m$ (for PM_{10} sampler) with 1 m³/h flow rate. The detection limit of these analyzers is 1 μg m $^{-3}$, and they span the range from 0 to 5000 μg m $^{-3}$ (Kenny et al., 2000; Hyvärinen et al., 2009). At each location, the analyzers were housed in a rack inside with air-conditioned hut.

Sun/sky radiometer (Kipp & Zonen model POM-01) measurements were used for columnar aerosol parameters such as aerosol optical depth (AOD). The sky radiometer measures the diffuse radiation at various scattering angles from the sun at different wavelengths in narrow band with a half band width of 3 nm for 340 nm and 10 nm for other wavelengths. The measured sky spectral radiances can be used to obtain the different optical and size properties of the aerosols in the total atmospheric column (Nakajima et al., 1996). Thus, the detected spectral radiant intensities at different measured angles from the sun combined with direct solar radiance can be used to numerically estimate the volume size distribution (Pandithurai et al., 2008).

The Angstrom exponent (AE) was determined at five different wavelengths (340, 400, 500, 670 and 1020 nm) using version 4.2 of the Skyrad Pack radiative transfer model (Nakajima et al., 1996).

3. Results and discussion

3.1. Daily, monthly, and seasonal PM variability

The daily variability of 24-hour particulate matter concentrations (PM_{2.5}, PM₁₀ and PM_{10-2.5}) averaged over the three different locations is depicted in Fig. 2. During this period, the mean PM_{2.5} and PM₁₀ concentrations were 118.3 \pm 81.7 μg m $^{-3}$ (varied from 7.8 to 525 μg m $^{-3}$) and 232.1 \pm 131.1 μg m $^{-3}$ (varied from 30.2 to 885 μg m $^{-3}$), respectively. The mean coarse mode particulate mass concentration (PM_{10-2.5}) was 113.6 \pm 70.4 μg m $^{-3}$ (varied from 13.6 to 630 μg m $^{-3}$) representing about 49% contribution to PM₁₀. The fine mode contribution was 51% over Delhi. Across Delhi, it was observed that ~37%, 42% and 41% of the measurements were higher than the mean mass concentrations of PM_{2.5}, PM₁₀ and PM_{10-2.5}, respectively.

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