



## Variations in Mass of the PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> during the Monsoon and the Winter at New Delhi

S. Tiwari, D.M. Chate\*, P. Pragya, Kaushar Ali, Deewan Singh Bisht

*Indian Institute of Tropical Meteorology, Pune - 411 008, India*

### ABSTRACT

PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> mass concentrations have been measured at Delhi (28°35'N; 77°12'E) during the August to December 2007. The running mean of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> data shows large variations. The PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> were ranged from 20 to 180 µg/m<sup>3</sup> during the monsoon and from 100 to 500 µg/m<sup>3</sup> during the winter (up to 1200 µg/m<sup>3</sup> in November due to Deepavali fireworks). For the same running mean cycles, higher mass concentrations in the PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> were corresponded with peaks in the relative humidity and lower levels linked to peaks in the ambient temperature. The evolutions of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> concentrations after the elapsed times are simulated with mean mass scavenging coefficients. These evolution patterns clearly show the difference in washout of PM<sub>10</sub> with impaction scavenging relative to those for PM<sub>2.5</sub> and PM<sub>1</sub> particles over different rainfall durations. Air-mass pathways traced with HYSPLIT model over the study area illustrates the nature of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> levels with monsoon and winter air-mass circulations over Delhi.

**Keywords:** Particulate matters; Visibility; Washout pattern; Time-series variations; Mean mass scavenging coefficient.

### INTRODUCTION

Atmospheric particulate matter (PM) can be classified as PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> by size with mass median aerodynamic diameter less than 10 µm, 2.5 µm and 1 µm respectively. PM plays pivotal role in the climate change, cloud dynamics, health impact, fog formation and visibility through a variety of atmospheric processes (Pillai *et al.*, 2002; Pope *et al.*, 2002; Das *et al.*, 2009). High concentrations in the PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> can cause human health problems, related to both short-term and long-term exposure to these particles (Schwartz *et al.*, 1996; Massey *et al.*, 2009; Chate, 2010). Based on an epidemiological data, WHO states that an increase in total PM by 10 µg/m<sup>3</sup> per year results in a 6% increase in mortality and a short-term increase in levels of the PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> for several days causes more coughing, respiratory problems, bronchodilator use and even mortality (WHO, 2000, 2003). Since health impacts of PM<sub>2.5</sub> and PM<sub>1</sub> in air are more adverse than larger particles (PM<sub>10</sub>), measuring them together with PM<sub>10</sub> is highly important (Wilson and Suh, 1997). During a dust storm, Choi and Choi, (2008) reported high concentrations in the variation of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> owing to circulation patterns and

boundary layer heights in the Korean mountainous coast. Li *et al.* (2009) have presented PM<sub>2.5</sub> particles at an urban, industrial and coastal site in Tianjin, China. Also, recently Sabbagh-Kupelwieser *et al.* (2010) have presented PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> at Vienna, Austria.

In Delhi, India, measurements of aerosol number size distributions have already been performed for air-quality monitoring and aerosol formation purposes. For instance, high concentrations of ultrafine particles (diameter < 0.1 µm) were frequently recorded in the urban atmosphere of Delhi (Monkkonen *et al.*, 2004). With the rapid urbanization and corresponding increase in the traffic and energy consumption, there has been growing evidence that ambient concentration levels of PM<sub>2.5</sub> and PM<sub>1</sub> are also high in Delhi (Gupta *et al.* 2007). The major source of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> are referred as windblown dust, secondary aerosol, coal combustion, traffic exhausts and biomass burning, etc. (Tiwari *et al.* 2009). Furthermore, PM<sub>2.5</sub> and PM<sub>1</sub> remain air-borne through nonlinear processes for days-to-weeks during monsoon months as washout processes are least efficient for cleansing particles in these size bins. Since background number concentrations of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> particles are very high in megacities (e.g. Delhi) their formation and removal processes by rainfall are not clearly understood.

Aerosol distributions presented by taking seasonal or annual simple averages of data can suppress the peaks owing to local effects and also by variations with rain scavenging over very short durations (Chate *et al.*, 2005). Therefore, time series distributions of PM<sub>10</sub>, PM<sub>2.5</sub> and

\* Corresponding author. Tel: +91-20-25904257;  
Fax: +91-20-25893825  
E-mail address: chate@tropmet.res.in

PM<sub>1</sub> can be presented by performing running mean on raw data in order to address environmental and rain scavenging processes in those size regimes. The National Ambient Air Quality Standards ([http://cpcb.nic.in/National\\_Ambient\\_Air\\_Quality\\_Standards.php](http://cpcb.nic.in/National_Ambient_Air_Quality_Standards.php)) for PM<sub>10</sub> and PM<sub>2.5</sub> are set at averages over 24 hours (1440 minutes). The main advantage in time series presentation of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> mass concentrations is that a mean can be performed around every point along a continuous raw data. Furthermore, data points with very high peaks for random spikes due to electronic glitches are to be removed from the raw data.

The mass concentration of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> have been measured continuously at Delhi (28°35'N; 77°12'E) for days-to-weeks period in a month during August–December, 2007. We present 1440 minutes running mean variations in the PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> concentration during the monsoon (August–September) and the winter period (October–December). Differential behavior of PM<sub>10</sub> as compared to PM<sub>2.5</sub> or PM<sub>1</sub> for washout patterns is discussed to understand removal mechanisms in these size regimes. We discuss relative humidity and temperature variations for the available data of these parameters for very short period during August. Also, HYSPLIT (Draxler and Rolph, 2003, <http://www.arl.noaa.gov/ready/hysplit4.html>) backward trajectories aid us in tracing air-mass pathways for likely pollutant source regions. Variations in PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> concentration with 1440 minutes running mean cycles and influences of rain, relative humidity, temperature etc. on these variations are useful in formulating control measures of particulate matter air-quality in Delhi. Also, PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> concentration changes with short and long term atmospheric processes are significant from the environmental point of view due to differential exposure of ambient aerosols to local population.

### Observational Site

The observational site, IITM, Delhi (28°35'N; 77°12'E) is located in urbanized central part of Delhi at about 218 m above mean sea level. The transport and dispersion of pollutants, particularly those in the lower levels of the atmosphere, are believed to be affected by the circulation pattern associated with the surrounded area. The site represents a typical residential area free from major pollutant sources, which is suitable for background air pollution studies. The long-term atmospheric measurements of cloud microphysics and air-quality parameters are available in the literature. Delhi experiences a severe cold and foggy weather during winter. The prevailing winds throughout the year are easterly, northerly and northwesterly. The entire northern part of India, especially the Indo-Gangetic Plain, experiences a thick foggy weather during winter with low boundary layer height. The low winds and reduced boundary layer heights results in very low ventilation factors which determine the dispersion of pollutants. During such conditions, pollutants could not be dispersed or mixed with free troposphere (Srivastava *et al.*, 2005). The impact of such conditions is poor visibility and high levels of pollutants in this region. The daily minimum temperature falls rapidly by the end of October and continental air masses, rich in

pollutants of continental origin, pass over the experimental site during winter.

### METHOD

The sampling of aerosols for this study was carried out at about 15 m above the ground level, on the rooftop of an IITM Building (Delhi) situated in the urbanized central part of Delhi. The area is primarily a residential area, and no large pollutant source exists nearby which could have influenced the sampling site directly.

The GRIMM Model 1.108 (OPC, GRIMM Inc.) is a portable particle analyzer and is specifically designed for PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> ambient air analysis using dual technology consisting of both optical and gravimetric analysis. This technology enables the Model 1.108 to make precise cut off diameters for all three PM sizes. This system allows collecting all three PM fractions simultaneously without changing sampling heads. Coarse particles (PM<sub>10</sub>) and fine particulates (PM<sub>2.5</sub> and PM<sub>1</sub>) have been monitored with the GRIMM particles sampler. The GRIMM particle counter was operated continuously during August 2007 to December 2007. A constant flow rate ~1.2 L/min is maintained throughout the measurements. The GRIMM particles measuring system is equipped with GRIMM 1174 Software for data acquisition. It was set to collect data at 1 minute intervals and store them in memory to be downloaded to a PC and analyzed further for time-series during August to December 2007. The data of relative humidity and ambient temperature during August, 2007 were collected through an automatic weather station. The variations in relative humidity (RH) with the temperature over a very short period at an experimental site during 29–31 August, 2007 are presented in Fig. 1(a). During monsoon month of August, RH varied between 50% and 90% with the temperature variations. As seen in this Figure, at lower temperature ~27°C, the RH found to be at its maxima (90%) and for higher temperature ~35°C, the RH drops below 50% and vice versa.

### RESULTS AND DISCUSSION

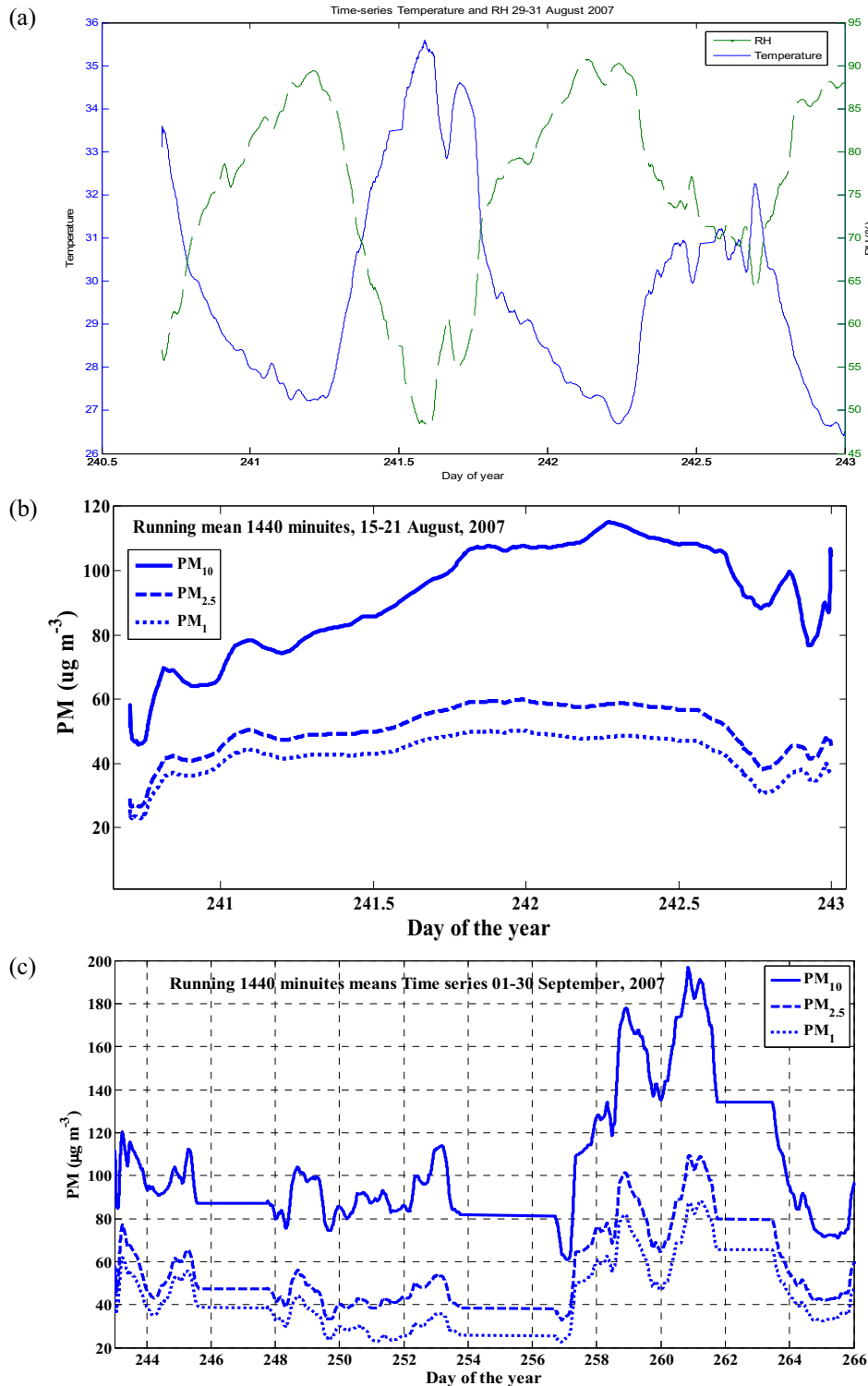
We have performed a mean around every point along a continuous raw data of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> mass concentrations obtained during August–December 2007. The running mean is performed with a time constant of 1440 minutes, or 1440 data points calculating the mean of the first 1440 points, then subtracting the value of first point, and adding the value of 1441<sup>st</sup> point for the next mean value. Eq. (1), computes running means in time-series data of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> mass concentrations over several days.

$$y_{j+1} = y_j + \frac{(x_{j+1} - y_j)}{j+1} \quad (1)$$

where  $x$  – data point in original time series,  $y$  – data point in running mean time series and  $j$  – position of the data point. The time constant of 1440 minutes found to be suitable for the running 1440 minutes PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> mass

concentrations means, as the original 1 minute time-series data are of very high fluctuations owing to random spikes by electronic/electrical glitches. The National Ambient Air Quality Standards for  $PM_{10}$  and  $PM_{2.5}$  are set at  $100 \mu\text{g}/\text{m}^3$  and  $60 \mu\text{g}/\text{m}^3$  respectively over 1440 minutes average.

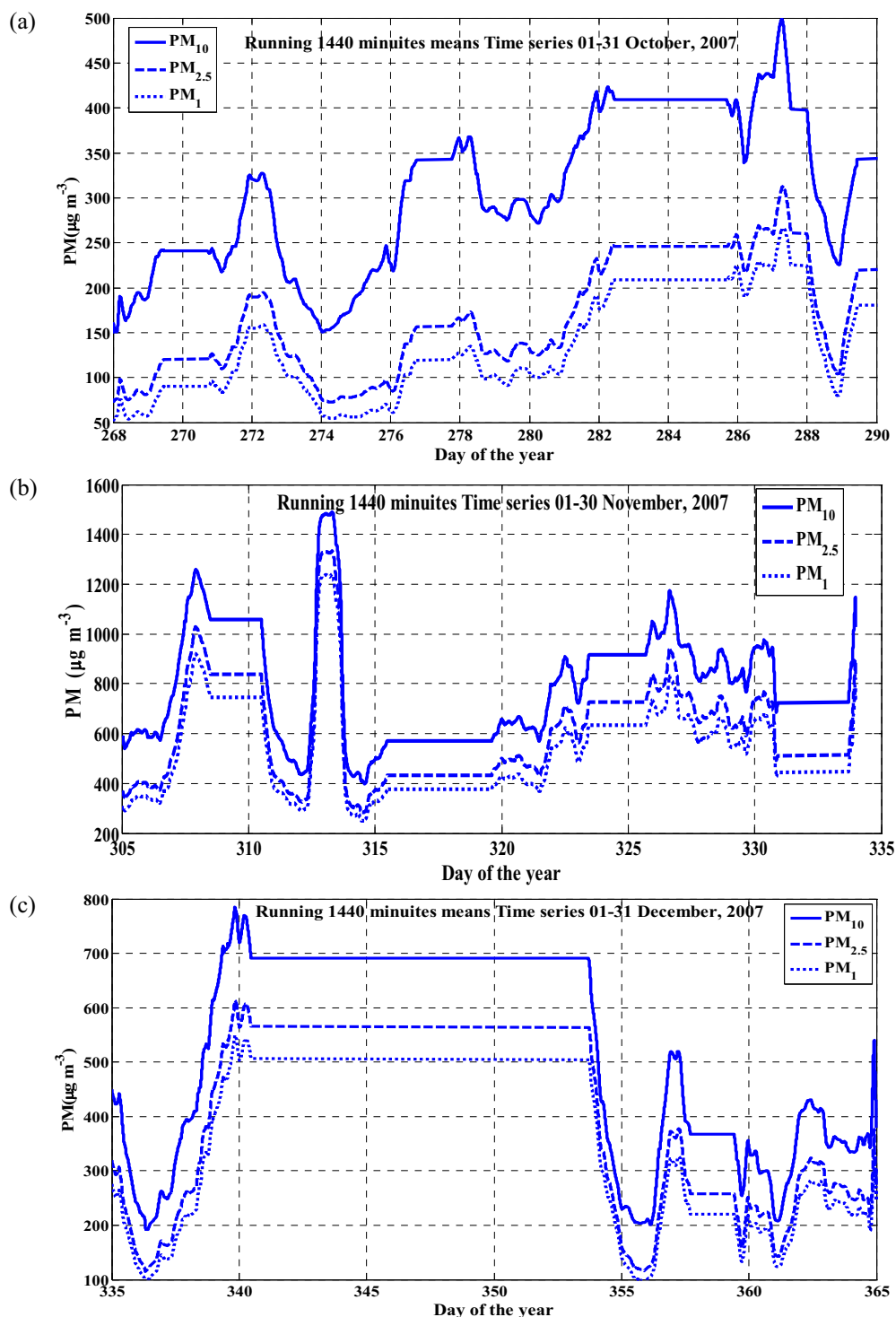
Accordingly, the 1440 minutes running mean variation in  $PM_{10}$ ,  $PM_{2.5}$  and  $PM_1$  mass concentrations for the rainy months (August and September) are shown in Figs. 1(b) and (c) respectively. The winter months (October, November and December) time-series for  $PM_{10}$ ,  $PM_{2.5}$  and  $PM_1$  mass



**Fig. 1.** (a) Relative humidity with ambient temperature during 29–31 August, 2007, (b) Time-series for  $PM_{10}$ ,  $PM_{2.5}$  and  $PM_1$  mass concentrations during 15–21 August, 2007 at 1440 minutes running mean, (c) Same as (b) during 01–30 September, 2007.

concentrations are shown in Figs. 2(a–c) respectively. Higher levels of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> mass concentrations (1200–1400 µg/m<sup>3</sup>) were observed on Deepawali days (8<sup>th</sup>–9<sup>th</sup> November 2007) owing to large emissions by fireworks displays (Fig. 2(b)). In the month of December, the ABL height remains shallow (50 m) and almost no winds till 0900 hour results in too low ventilation factors over

continental areas as reported elsewhere (Murugavel and Chate, 2011). Lower boundary layer heights should confine ambient aerosols at the Earth surface and manifest as an increase in their levels. Low PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> mass concentrations were observed during 15–21 August (20–110 µg/m<sup>3</sup>) and 01–30 September (20–120/180 µg/m<sup>3</sup>) as against those observed during December due to the washout of these particles by



**Fig. 2.** (a) Time-series for PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> mass concentrations during 01–24 October, 2007, (b) Same as (a) during 01–30 November, 2007, (c) Same as (a) and (b) during 01–31 December, 2007.

rainfall (Figs. 1(b) and (c)). The mass concentration is reduced considerably in August, due to washout effect during precipitation (Fig. 1 (b)). It can also be seen from Fig. 2(a–c) that during the months of continental air-mass (winter), levels of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> lie close by, whereas during the monsoon months, these curves are well resolved, with PM<sub>10</sub> much higher than that of PM<sub>2.5</sub> and PM<sub>1</sub> (Figs. 1(b) and (c)). It is worth to note that mass concentration trends in the monsoon months resulted in an order of difference in mass concentrations between PM<sub>10</sub> and PM<sub>1</sub> or PM<sub>2.5</sub> due to the extensive monsoon rain, replenishment of PM<sub>10</sub> aerosols by stronger monsoonal winds and superimpositions of aerosols by splashing of raindrops on the Earth surface (Chate and Kamra, 1993). Variations in PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> display distinct winter and monsoon months' peaks. Winter months' peaks are attributed to enhancement in continental aerosols and monsoon months' peaks upon impositions of aerosols with splashing of raindrops on the Earth surface.

#### **Role of Rain Scavenging in Distributions of PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> over Delhi**

PM<sub>2.5</sub> and PM<sub>1</sub> particles falls in the accumulation size bins and thus they are air-borne for days-to-weeks even during monsoon months as washout processes are least efficient for cleansing these particles. Also, background number concentrations of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> particles are very high in Delhi in turn their washout patterns by rainfall are not clearly understood. Therefore to understand washout processes, a typical rain scavenging patterns for PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> are simulated for August for assumed rainfall rates of 5, 25 and 50 mm/h for PM<sub>10</sub> and 25 and 50 mm/h for PM<sub>1</sub> and PM<sub>2.5</sub>. Terms related to Brownian diffusion, directional interception, inertial impaction, thermo-diffusion-phoresis and electro-scavenging with dimensionless parameters used for simulations are published elsewhere with appropriate references (Chate and Devara, 2005). Overall collision efficiency  $E(D, D_p)$  computations are performed for PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> particles ( $D_p < 10 \mu\text{m}$ ,  $D_p < 2.5 \mu\text{m}$ ,  $D_p < 1 \mu\text{m}$ ) and raindrops of size ( $D$ ) for electro-scavenging, Brownian diffusion, inertial impaction, interception, and thermo-diffusion-phoresis.

For a spatially homogeneous system, assuming rain scavenging is the only sink for PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub>, changes in mass or number concentrations for non-interacting and non-growing particles  $N(D_p)$  with diameters between  $D_p$  and  $D_p + dD_p$  at time  $t$  after scavenging is related to the concentration  $N_0(D_p)$  at  $t = 0$ , as

$$N(D_p) = N_0(D_p) \exp[-A(D_p)t] \quad (2)$$

where  $A(D_p)$  is the scavenging coefficient (Seinfeld and Pandis, 1998) and expressed as,

$$A(D_p) = \int_0^{\infty} (\pi/4) D^2 E(D_p, D) V_r(D) N(D) dD \quad (3)$$

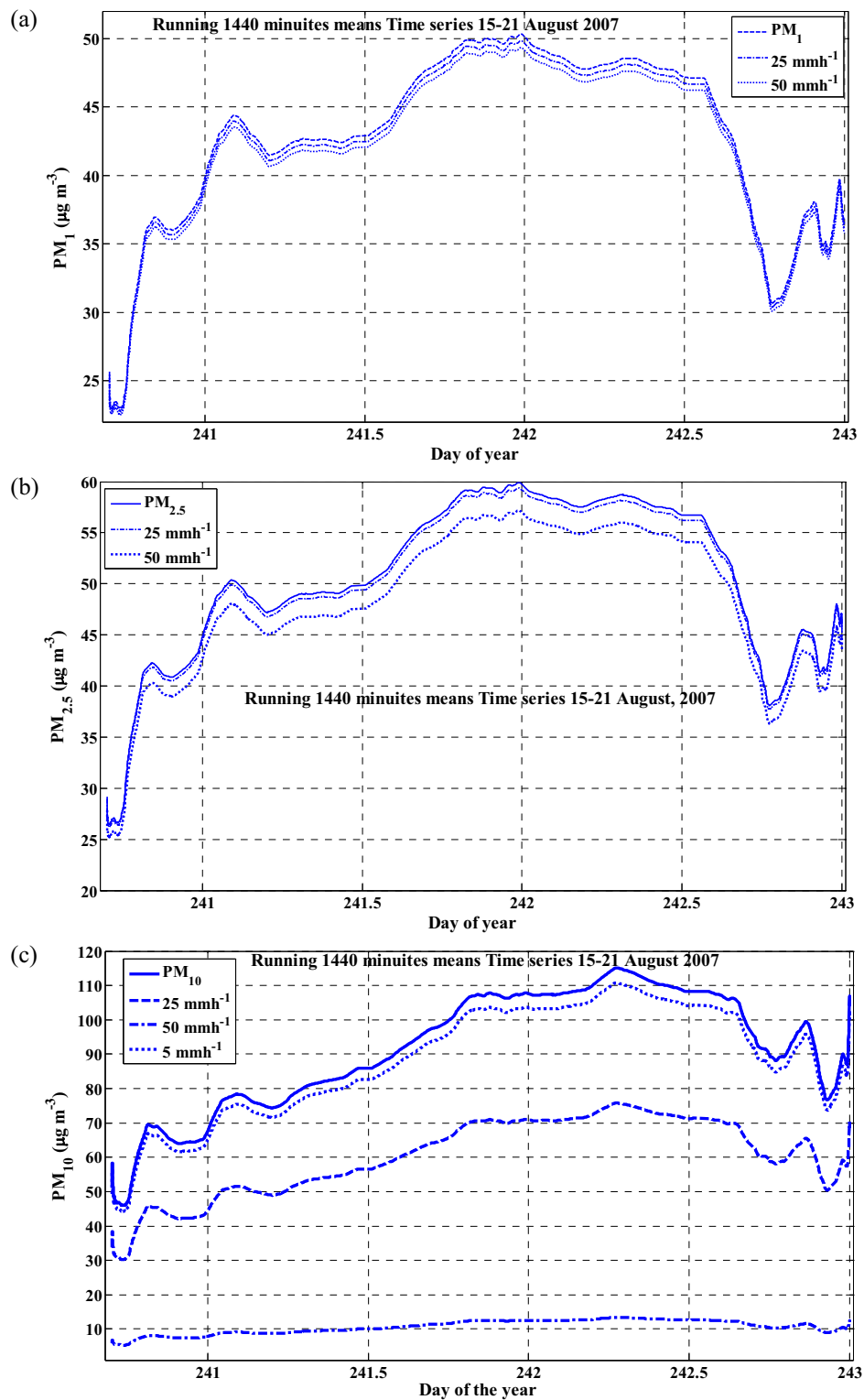
where,  $N(D)$  is raindrop size distribution adopted from classical Marshall–Palmer (1948) drop size distribution.  $V_r(D)$

terminal velocities of raindrop computed using Beard's formulae (1976). Mean mass scavenging coefficient  $A_m$  is calculated for all the three particle size regimes (PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub>) at rainfall rates of 5, 25 and 50 mm/h with equation given below,

$$A_m = \frac{\int_0^{\infty} A(D_p) D_p^3 N(D_p) dD_p}{\int_0^{\infty} N(D_p) D_p^3 dD_p} \quad (4)$$

Figs. 3(a) and (b) show washout pattern at rainfall intensity of 25 and 50 mm/h after elapsed time of 5 hours rain for PM<sub>1</sub> particles and 3 hours rain for PM<sub>2.5</sub> particles. Also, Fig. 3(c) shows the washout pattern for PM<sub>10</sub> particles at rainfall intensity of 5, 25 and 50 mm/h after elapsed time of 1 hour (rainfall duration). Since large mass fractions of PM<sub>1</sub> and PM<sub>2.5</sub> particles resides in the accumulation mode, they are too large to have sufficient Brownian diffusivity and too small to get collected effectively by falling raindrops due to inertial impaction mechanism (see Seinfeld and Pandis, 1998 for wet deposition). The evolutions of PM<sub>1</sub> and PM<sub>2.5</sub> particles in Figs. 3(a) and (b) show very small fractions of initial mass concentrations of these particles' depletion by rain intensity of 25 and 50 mm/h even after elapsed time of rain over 5 hours and 3 hours respectively. As significant fractions of mass concentrations of PM<sub>10</sub> particles are in the size range of coarser mode, they are effectively washed out by rain over 1 hour of elapsed time at intensity of 5, 25 and 50 mm/h due to their inertia (Fig. 3(c)) higher than that of PM<sub>1</sub> or PM<sub>2.5</sub> particles. The evolution of PM<sub>10</sub> particles in Fig. 3(c) clearly shows the dominance of inertial impaction in depleting significant amount of initial mass of these particles by rain over 1 hour at intensity of 5, 25 and 50 mm/h respectively. However, the evolutions of PM<sub>1</sub> and PM<sub>2.5</sub> particles indicate that, impaction scavenging or Brownian diffusivity is insignificant collection mechanism of raindrops for depleting initial mass of these particles by heavy rainfalls of intensity 25 and 50 mm/h over longer durations (5 and 3 hours for PM<sub>1</sub> and PM<sub>2.5</sub> particles).

Air-mass pathways are traced to assess possible sources during the monsoon (August–September) and during the winter (December). Air-mass backward trajectories that reached Delhi at 50, 100 and 500m above ground level were traced with Hybrid Single-Particle Lagrangian Trajectory (HYSPLIT, NOAA/ARL) (Draxler, 2004). The global meteorological data from the National Centers for Environmental Prediction's Global Data Assimilation System (GDAS) were used for the trajectory calculation. Back trajectories are traced using a 168-hour (01–07 December) ending at 1900 UTC [(0030 hrs local time (LT))] and 144 hour (01–06 December) ending at 1400 UTC (1930 LT) backward trajectory with every six hour interval. The air-mass pathways for high PM days (6–7 December, 2007) and also relatively low events (3–4 December, 2007) during December are analyzed as shown in Figs. 4(a–f). During winter, air-mass pathways arriving at Delhi are affected by



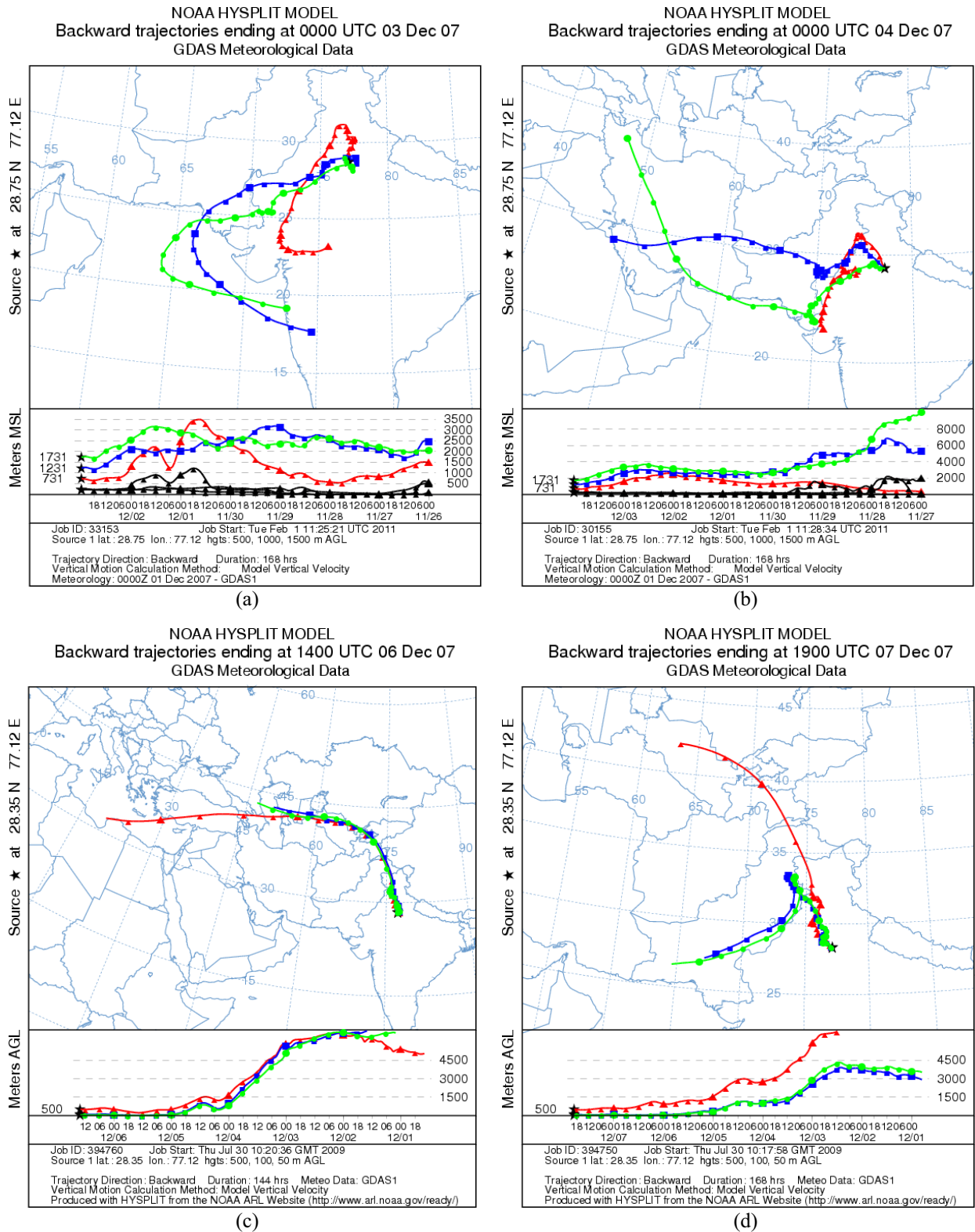
**Fig. 3.** (a) Washout pattern for  $\text{PM}_{10}$  particles at rain intensity of 25 and 50  $\text{mm/h}$  over 5 hours of rainfall, (b) Same as (a) for  $\text{PM}_{2.5}$  particles for 3 hours of rain, (c) The evolution of  $\text{PM}_{10}$  particles by rain over duration 1 hour at intensity, 5, 25 and 50  $\text{mm/h}$ .

anthropogenic continental and local aerosols during 01-07 December 2007 as seen in Fig. 4(a–b). The anthropogenic aerosols from continental air-mass favor the increase in fine particle levels ( $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ ). However, during monsoon

months, air-mass loaded with natural marine aerosols with salt contents were passed over Delhi during 29–31 August and during 25–30 September, which enhanced coarser particles ( $\text{PM}_{10}$ ) (Figs. 4(c)–(f)). In addition to these synoptic

air-mass patterns, atmospheric conditions over the Delhi site were affected by episodic events involving continuous heavy precipitation during monsoon (29–31 August) and dense foggy condition during winter (December).

Running mean in  $PM_{10}$ ,  $PM_{2.5}$  and  $PM_1$  mass concentrations during the monsoon and the winter shows much wide variations at Delhi and could have significant adverse impacts on visibility and health owing to short-term local effects. In



**Fig. 4.** (a–h) Back-trajectories for air-mass pathways for 03–04 (Fig. 4(a) and (b)), 07–06 (Fig. 4(c) and (d)), December, 2007, 30–28 (Fig. 4(e) and (f)), September, and 29–31 August (Fig. 4(g) and (h)).

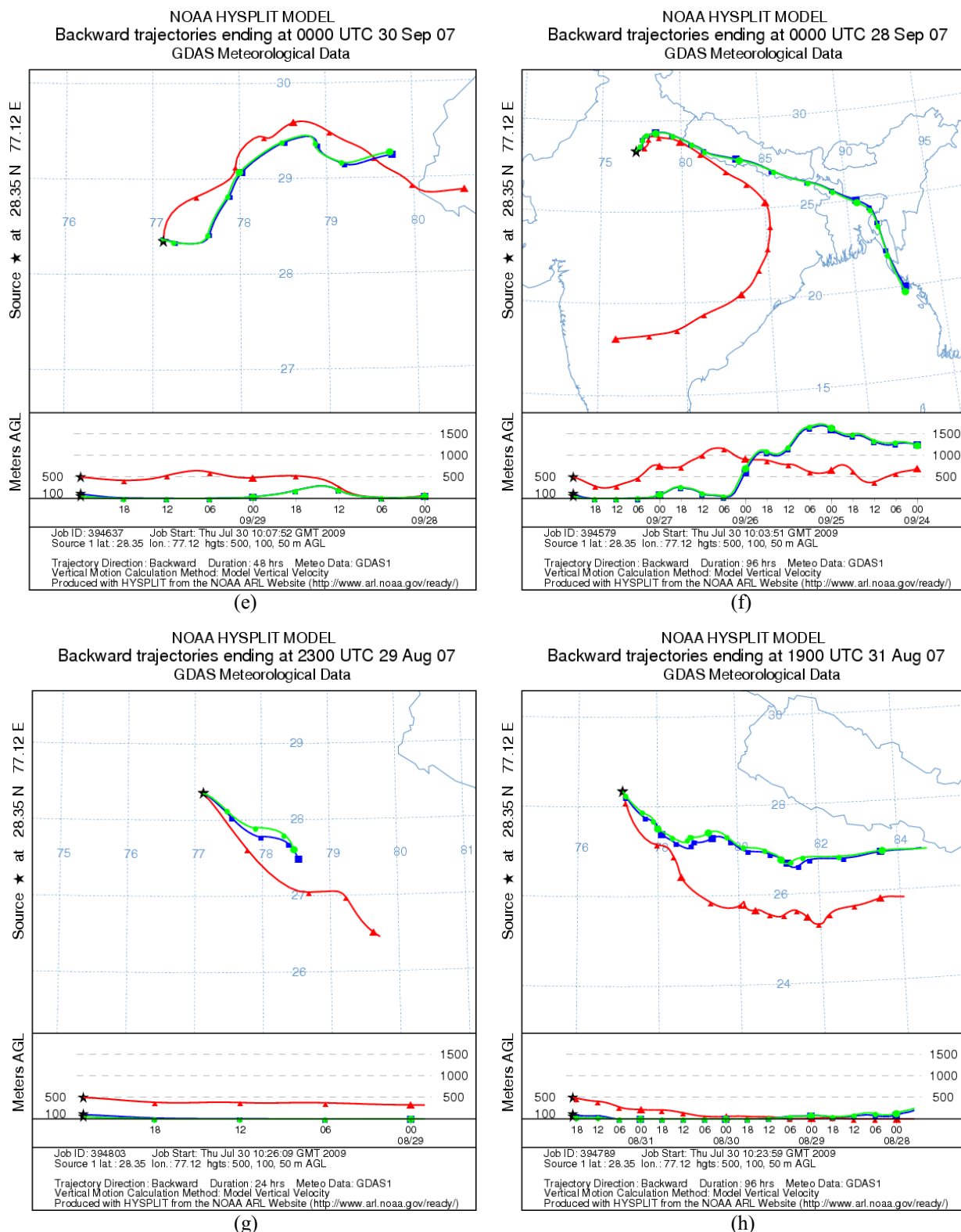


Fig. 4. (continued).

comparison, annual mean  $PM_{10}$  and  $PM_{2.5}$  concentration ( $\pm$  standard deviation) reported as  $219 (\pm 84)$  and  $97 (\pm 56) \mu\text{g}/\text{m}^3$  respectively during January to December, 2007, are about twice to prescribed Indian National Ambient Air Quality Standards (Tiwari et al., 2009). TSP,  $PM_{10}$  and

$PM_{2.5}$  at Erzurum (Turkey) urban atmosphere were measured on an average of 129, 31 and  $13 \mu\text{g}/\text{m}^3$ , respectively for February 2005 to February 2006 (Hanefi et al., 2008). At an urban, industrial and coastal site in Tianjin, China, Li et al. (2009) observed daily average concentrations of  $PM_{2.5}$  ranged



from 61.5–566.8  $\mu\text{g}/\text{m}^3$  with an average of 223.0  $\mu\text{g}/\text{m}^3$ . However, reducing  $\text{PM}_{2.5}$  and  $\text{PM}_1$  emissions and so to decreasing their negative effects on health, visibility etc., is a difficult task especially where they are mainly accumulated through nonlinear atmospheric processes.

## CONCLUSIONS

In the nature of variation in  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and  $\text{PM}_1$ , low concentration occurred during the monsoon (August–September) due to washout of these particles, whereas, higher levels observed during the winter (October–December), when lower boundary heights prevails over this site. During the monsoon, since wet removal processes are most effective,  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and  $\text{PM}_1$  concentrations reaches at their annual minimum. After the monsoon, scenario reverses as air-mass shifts to continental followed by reduction in rainfall and cloudiness and thus  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and  $\text{PM}_1$  concentration gradually increases. The winter peak is attributed to aerosols in the  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and  $\text{PM}_1$  levels due to mixing of continental aerosols with background local particles. On the other hand, monsoon peaks are due to the superposition of transported aerosols from marine environment on local continental aerosols and on aerosols due to splashing of raindrops on the Earth surface during rain. The information presented on running mean variations in  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and  $\text{PM}_1$  mass concentrations are not enough to take steps on controlling fine particulate pollution because of lack of long-term and continuous observations for the  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and  $\text{PM}_1$  in such a rapidly developing region.

## ACKNOWLEDGEMENTS

Authors express sincere gratitude to Prof. B.N. Goswami, Director, Indian Institute of Tropical Meteorology, Pune (India) for encouragements. Indian Institute of Tropical Meteorology, Pune is supported by the Ministry of Earth Sciences, Government of India New Delhi. The authors thank the anonymous reviewers for their suggestions and comments which have helped to improve the manuscript.

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*Received for review, June 3, 2011*

*Accepted, September 6, 2011*