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## RECENT TRENDS IN AEROSOL CLIMATOLOGY AND AIR POLLUTION AS INFERRED FROM MULTI-YEAR LIDAR OBSERVATIONS OVER A TROPICAL URBAN STATION

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### ABSTRACT

Regular nighttime monitoring of aerosol and other atmospheric parameters was initiated in 1985 at the Indian Institute of Tropical Meteorology, Pune. This is a tropical urban station (18°32'N, 73°51'E, 559 m AMSL), situated approximately 100 km inland from the west coast of India. The multi-year aerosol vertical profile database, utilized in the present study, consisted of more than 1200 vertical aerosol concentration profiles. These data were collected with a computer-controlled, bistatic, argon-ion lidar system over a 12 year period from October 1986 to September 1998 and have been utilized to study the morphology of the nighttime atmospheric boundary layer and associated air quality. The recent climatological trend in the aerosol loading at the experimental station has also been studied. The study reveals higher pollution potential during late evenings in the winter and a total increase of about 3% in the aerosol loading over the 12 year observational period. This increase can be attributed partly due to the urban heat island effect and due to growing urbanization and industrialization, as well as to the land-usage patterns in proximity to the experimental station. Further, it has been found that the long-term trend in aerosol loading was not uniform, but it changed from year to year depending on meteorological parameters (precipitation, in particular) and local anthropogenic activities. The short-term variations in aerosol loading and their relationship with concurrent meteorological parameters over the observational site are discussed also. Copyright © 2002 Royal Meteorological Society.

KEY WORDS: tropical urban station; lidar; aerosol climatology; long-term trend; air pollution; precipitation

### 1. INTRODUCTION

The study of atmospheric aerosols is important for a variety of reasons, two of which are (i) aerosols influence the climate directly by altering the radiative energy transfer through their optical properties and (ii) space–time variability of aerosol inhomogeneities provides unique information on atmospheric behaviour needed for environmental research and operational programs. Much interest in atmospheric aerosols has been stimulated by the conspicuous presence and observed impact of anthropogenic emissions associated with many urban environments, and hence it is a matter of public concern (IPCC, 1995). The impact of aerosols on global/regional/local weather and climate, satellite remote sensing, air and water quality has been the subject of numerous investigations in recent years (WCP, 1983; IGAP, 1991; Charlson *et al.*, 1992; Harshvardhan, 1993). The previous estimates reveal that the anthropogenic component of the aerosol burden causes a direct negative radiative forcing (whitehouse effect) in shortwave which is comparable in magnitude but of opposite sign to positive longwave forcing (greenhouse effect) of CO<sub>2</sub> and several trace gases associated with industrial, agricultural, vehicular, biomass burning activities and so on (Charlson *et al.*, 1992; Kiehl and Briegleb, 1993; Taylor and Penner, 1994; Penner *et al.*, 1994).

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An additional indirect forcing from aerosols results from their involvement in nucleation and growth of cloud droplets, reducing droplet size and thereby potentially influencing cloud albedo (Ramanathan *et al.*, 1989; Boucher and Anderson, 1995; Jones and Slingo, 1996; Li, 1998; Haywood and Ramaswamy, 1998). These studies have particular significance over the tropics, where the convective and dynamical processes associated with high-altitude thunderstorms greatly affect the vertical distributions of aerosols. As the aerosol characteristics exhibit a high degree of spatial and temporal variability, and as the anthropogenic share of the total aerosol loading is quite substantial over many parts of the world, it is essential to monitor the aerosol features systematically over longer time scales (WCRP, 1988; Wolf and Hidy, 1997). Such observations are very important for understanding the coupling processes that exist between chemical, radiative, dynamical and biological phenomena in the Earth's environment and provide valuable input information for modelling and simulation studies of weather, climate and air quality (Zuev, 1982; Gultepe, 1995).

Although both *in situ* and remote sensing methods have been in use for atmospheric aerosol studies, the latter has certain inherent advantages over the former. As aerosols in the troposphere dominate optical properties of the atmosphere, remote sensing techniques involving optical wavelengths yield valuable information on atmospheric aerosols. Of the optical remote sensing techniques, lidar has been recognized to be a powerful tool for environmental and climate monitoring studies (McCormick *et al.*, 1993 and references cited therein) because of the excellent spatial resolution unattainable by any other remote sensor.

Systematic nocturnal lidar observations of aerosols (using the Mie–Rayleigh scattering techniques) in the lower atmosphere, time–height variations of aerosol layer structure and their dependence on most controlling meteorological parameters have been in progress at the Indian Institute of Tropical Meteorology (IITM), Pune, India, since 1985 (Devara, 1992). Monthly, seasonal and annual effects have also been studied. The extensive observations of aerosol vertical distributions obtained during the 12 year period from October 1986 to September 1998 have been used to study the nighttime atmospheric boundary-layer (NBL) and associated air quality, as well as the recent long-term climatological trends in the aerosol loading at the experimental station. As part of the Lidar In-Space Technology Experiment (LITE) correlative measurement program, IITM conducted special nocturnal observations on the 12 nights of 7–18 September 1994 (all-night observations were conducted on three of these days). These measurements were utilized to study the short-term variations in boundary-layer aerosol loading and their association with concurrent meteorological parameters at Pune, India. In this paper, the results of the above studies, along with a brief description of the lidar equipment and topography of the experimental site, are presented.

## 2. LIDAR AND TOPOGRAPHY OF THE SITE

The computer-controlled lidar system used in these studies consists essentially of a Lexel 95-4 model, tunable (at discrete wavelengths) argon-ion laser as transmitter and a 25 cm diameter, f/7.6, Newtonian telescope as receiver equipped with an optics assembly composed of condensing–collimating lenses, narrow-band interference filter (1 nm FWHM), and Peltier-cooled PMT. The on-line control and digital data system provides real-time acquisition, analysis and display of lidar data. The lidar is operated in bistatic mode with its transmitter horizontally separated from the receiver by 60 m. The vertically transmitted laser beam (at wavelength of 514.5 nm with an average output power of 400 mW) is scanned by the receiver at pre-programmed elevation angles corresponding to different altitudes. The complete lidar system has been installed on the terrace of the IITM building in order to make the system free from city light pollution and from topographic targets such as tall buildings and trees. A detailed description of the lidar experimental parameters and the analysis procedure for retrieving information on the vertical distribution of aerosols from scattered laser signals have been published previously (Devara and Raj, 1987, 1989; Devara *et al.*, 1995a).

The prevailing environment over the experimental station is urban and the aerosol type present over the observation site is assumed to be a mixture of water-soluble, dust- and soot-like aerosols. This assumption is in moderate agreement with the results of multi-year lidar scattering angle and multi-spectral data, which revealed the aerosol size index to be between 3.5 and 5.5, and refractive index to be between 1.4 and 1.6 (Devara *et al.*, 1995b; Pandithurai *et al.*, 1996). The airflow in the lower troposphere is predominantly westerly

during the South–West (SW) monsoon season (June–September), which brings in a large influx of moist air from the Arabian Sea. The westerly flow weakens in the lower troposphere and the easterly flow sets in during the post-monsoon season (October–November). The continental air masses, rich in particles of continental origin, pass over the region during that season. Also, an increase of dry polar continental air in the wake of low-pressure systems (western disturbances) takes place during the winter season (December–January). Thus, the meteorological conditions at the experimental station vary markedly from continental (winter) to maritime (summer) environment.

The lidar site is located at an elevation of about 573 m AMSL, approximately 100 km inland from the west coast and is surrounded by hillocks (some as high as 760 m AMSL) forming a valley-like configuration. The transport and dispersion of pollutants, particularly those in the lower levels of the atmosphere, are believed to be affected by the circulation processes that evolve because of the typical terrain. There are stone quarries (east side) and brick-kilns (west side) situated within 1 km of the lidar site, which are considered to be the major local anthropogenic sources contributing to aerosol observations. In addition, major urban activity from the eastern part of the lidar site contributes significantly. The western side is a sparsely populated area. The recent increase in industrial and urban development, particularly in the north-eastern sector of Pune city, appears to be influencing the aerosol concentration at the lidar site, especially when the air flow over the station comes from the north-easterly quadrant (0–90°).

### 3. OBSERVATIONS AND ANALYSIS

More than 620 weekly spaced vertical profiles (covering the altitude range between 20 and 1380 m) of aerosol number density have been collected during the 12 year period from October 1986 to September 1998 using the argon-ion lidar system. Apart from these routine measurements, almost an equal number of such profiles have been obtained during various special experiments, conducted from time to time, within the same period. Together, these observations formed the database for the results reported here.

Generally, the amount of scattered laser energy at a particular angle from a volume in the atmosphere is due to aerosols and gaseous molecules. At visible wavelengths, the distribution of extinction in the lower troposphere is primarily a function of aerosol loading and composition. It is common to assume that the aerosols are homogeneous spherical particles characterized by a single refractive index (with both real and imaginary components). However, the validity of this assumption has become one of the most interesting problems in atmospheric optics. The lidar scattered intensity profiles obtained at the argon-ion laser wavelength of 514.5 nm have been converted into aerosol number density profiles by employing the inversion routine programs (Devara and Raj, 1987; Devara *et al.*, 1995a) described briefly below.

The aerosol number density profile has been computed from the scattered signal strength (after correcting it for background noise), obtained at different altitudes by sampling the vertically transmitted laser beam at different elevation angles of the receiver, by using the following bistatic lidar equation:

$$P_R = \frac{P_T V T A_R \eta N \sigma(\theta)}{R_1^2 R_2^2 d\omega_1} \quad (1)$$

where  $P_T$  and  $P_R$  are the transmitted and received power respectively;  $V$  is the scattering volume;  $T$  is the atmospheric transmittance along the transmitter–scattering volume–receiver path;  $A_R$  is the collecting area of the receiver;  $\eta$  is the system constant, including the overall efficiencies of the transmitter and receiver optics;  $\sigma(\theta)$  is the differential cross-section at scattering angle  $\theta$ ;  $R_1$  and  $R_2$  are the ranges from the transmitter and receiver respectively to the centre of scattering volume;  $d\omega_1$  is the solid angle of the transmitting beam and  $N$  is the number density.

Since the atmosphere contains both aerosols and molecules whose scattering cross-sections differ greatly because of size and concentration differences, the scattering coefficient term  $N\sigma(\theta)$  takes the form:

$$N\sigma(\theta) = N_a\sigma_a(\theta) + N_m\sigma_m(\theta) \quad (2)$$

where  $N_a$  and  $N_m$  are the concentrations and  $\sigma_a(\theta)$  and  $\sigma_m(\theta)$  are the differential cross-sections at scattering angle  $\theta$  for aerosols and molecules respectively. Mie theory governs the scattering due to aerosol particles, whereas molecular scattering is governed by Rayleigh theory. By estimating the values of  $\sigma_m(\theta)$  and  $N_m$  from Rayleigh computations and from local radiosonde measurements respectively, measured scattering signals were corrected for Rayleigh scatter to yield the particulate matter.

In the present analysis, the aerosols are assumed to be homogeneous spherical particles. The aerosol size distribution, was assumed to follow a modified power-law distribution as suggested by McClatchey *et al.*, (1972), given by:

$$\begin{aligned} \frac{dN_a(r)}{dr} &= C_1 \quad \text{for } 0.02 < r < 0.1 \mu\text{m} \\ \frac{dN_a(r)}{dr} &= C_1 r^{-r} \quad \text{for } 0.1 < r < 10 \mu\text{m} \\ \frac{dN_a(r)}{dr} &= 0 \quad \text{for } r < 0.02 \mu\text{m} \text{ and } r > 10 \mu\text{m} \end{aligned} \quad (3)$$

where  $N_a(r)/dr$  is the number of aerosol particles with radii between  $r$  and  $r + dr$  in unit volume,  $r$  is the size index (or shaping parameter or size distribution) and  $C_1$  is the normalization constant that follows from continuity:

$$\int_0^{\infty} N_a(r) dr = N_{\text{total}} (\text{cm}^{-3}) \quad (4)$$

According to the power-law size distribution, the number of particles decreases monotonically with increase in radius. The total number of aerosol particles,  $N_{\text{total}}$ , is obtained by integrating  $N_a(r)$  over the 0.02–10  $\mu\text{m}$  size range. The aerosol column content has been computed by integrating aerosol number density throughout the height region as explained elsewhere (Sharma, 1994).

The size index and refractive index values of aerosols over the observing station have been estimated simultaneously by applying the ‘inversion by iteration’ method to the bistatic lidar angular scattering measurements in the surface layer (Pandithurai *et al.*, 1996) and are utilized in the retrieval method (outlined above) for computing the aerosol number density profiles. Possible errors resulting from these assumptions for determining the size distribution from light scattering data (and hence in the aerosol concentration or column content) have been discussed (Tanaka *et al.*, 1982; Pandithurai *et al.*, 1996).

The lidar-derived aerosol column content data sets thus obtained have been utilized for studying the long-term trends in the aerosol loading at the experimental station. Following the analysis procedure suggested by Endlich *et al.*, (1979) and Sasano *et al.*, (1982), normalized concentration gradient profiles have been derived from the aerosol vertical profiles and have been used to infer the nocturnal boundary-layer parameters such as mixing depth. Coupling these lidar-estimated mixing depths with the concurrent pilot balloon wind data, ventilation coefficients have been computed (Holzworth, 1967, 1972) for studying the air pollution potential or air quality over Pune. The aerosol lidar observations carried out during 12 nighttime runs (7–18 September 1994), as part of the LITE correlation measurement program, have been used to study the time evolution of aerosol loading and its association with prevailing meteorological parameters at the lidar site.

## 4. RESULTS AND DISCUSSION

### 4.1. Aerosol climatology and long-term trends

The aerosols present in the atmospheric boundary-layer, particularly in the lowest air layers (up to 200 m), contribute greatly to the overall loading at a given site, and thus the boundary-layer aerosol information is of paramount importance.

*4.1.1. Role of surface-generated aerosols.* In an attempt to estimate the contribution from the surrounding man-made activities, which is mostly confined to the lowest air layer (up to 200 m for the present study), to the total aerosol loading up to the boundary-layer, aerosol column content values were computed separately for the air layers extending up to 200 and 1100 m. These values were computed from the lidar-derived aerosol vertical distributions. Figure 1 depicts the climatologies of aerosol loading in the lowest air layer and in the boundary-layer along with the percentage contribution of the former to the latter. It is clear from Figure 1 that the changes in contribution of the air layer close to the ground to the overall loading appear to be smaller up to 1991 (approximately month number 63), after which it continued to increase significantly. This increase is in agreement with visual observations of the growth in urbanization and industrialization in the proximity of the experimental station for the past 4 to 5 years (1994–98). However, these variations in the aerosol loading are found to have a close relationship with the aerosol generation and removal processes prevailing over the observation site. This aspect is discussed further in Section 4.1.2. The 12 year mean values of aerosol column content in the air layers extending up to 200 m and 1100 m were  $73 \times 10^6 \text{ cm}^{-2}$  and  $188 \times 10^6 \text{ cm}^{-2}$  respectively, and the mean contribution of the 200 m air layer to that of 1100 m layer was about 39%.

*4.1.2. Long-term trends in the boundary-layer aerosol variation.* Figure 2(a) illustrates the year by year seasonal variation of aerosol column content during the 12 year period. The seasonal mean variation of

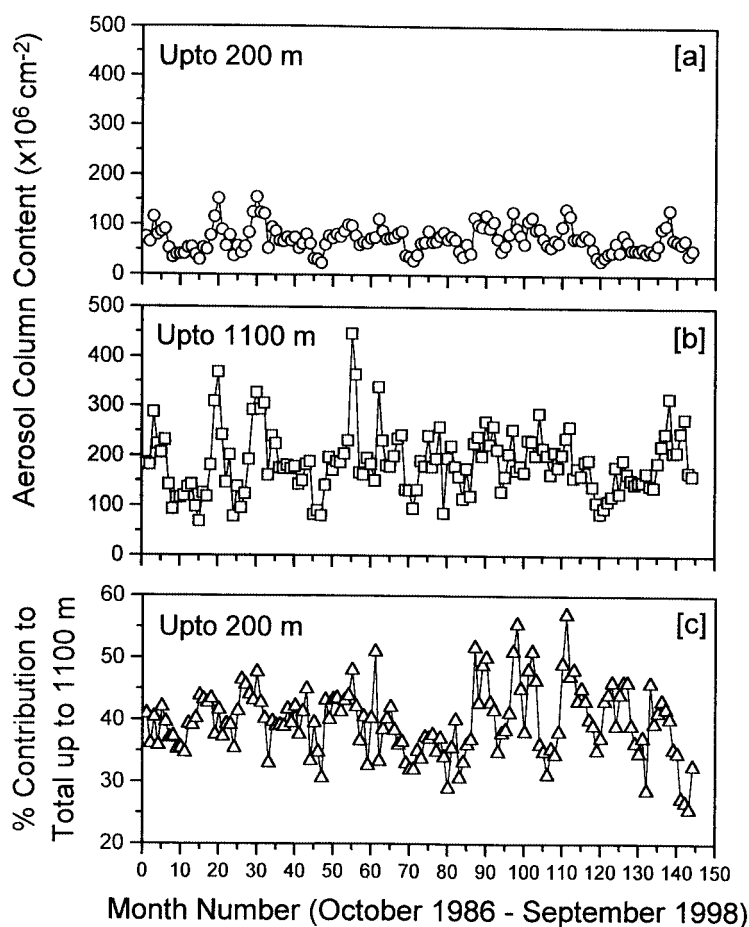


Figure 1. Month-to-month variations in aerosol loading in the air layer up to 200 m (a) and in the boundary-layer, up to 1100 m (b) The percentage contribution of the former to the latter is also shown (c)

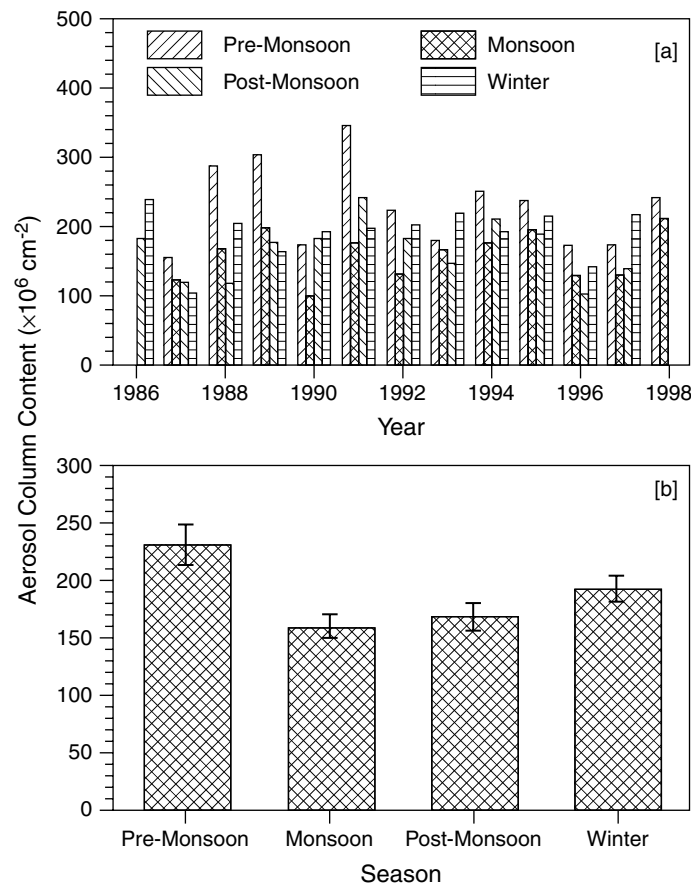


Figure 2. (a) Seasonal variations in aerosol column content during October 1986–September 1998. (b) Mean aerosol column contents

aerosol loading over the experimental station is also shown in Figure 2(b). It can be seen clearly from the figure that the aerosol loading over Pune is at a minimum during the monsoon months, gradually builds up during the post-monsoon and winter months, and finally peaks during the pre-monsoon months in every year. This is ascribed to the predominant convective and dust storm activities during the pre-monsoon and cloud-scavenging/rain washout processes during the monsoon season. It is found that the aerosol loading is higher by about 39% during the pre-monsoon months compared with the monsoon months.

In order to investigate the recent climatological trends in the aerosol particle concentration over Pune, both the weekly and monthly mean values of boundary-layer aerosol column content, computed from the aerosol climatological data archived during October 1986–September 1998, were subjected to polynomial regression analysis. These data, along with their long-term trend, are shown in Figure 3 (thick solid line). The weekly column content data from which the monthly mean values were computed are also depicted in the figure. An examination of Figure 3 reveals an increase of about 3% ( $R = 0.021$  and  $p = 0.803$ ) in the monthly mean aerosol loading at the station over a period of 12 years. This suggests an influence of the monthly, inter-annual and intra-seasonal variability of aerosol particle concentration in modifying the long-term trends in aerosol loading. This increase in aerosol loading over Pune is considered to be due to growth in urbanization, industrialization and changes in land-use pattern in the vicinity of the station in the recent past. Also, it could be due to the urban heat island and associated radiative effects resulting from differences in day and night temperature in Pune city and adjoining sub-urban areas (Woolum, 1964; Stull, 1991).

The deviations of monthly mean aerosol column content from the 12-year mean content ( $188 \times 10^6 \text{ cm}^{-2}$ ) are shown in Figure 4. The positive and negative departures indicate addition and removal of aerosol particles,

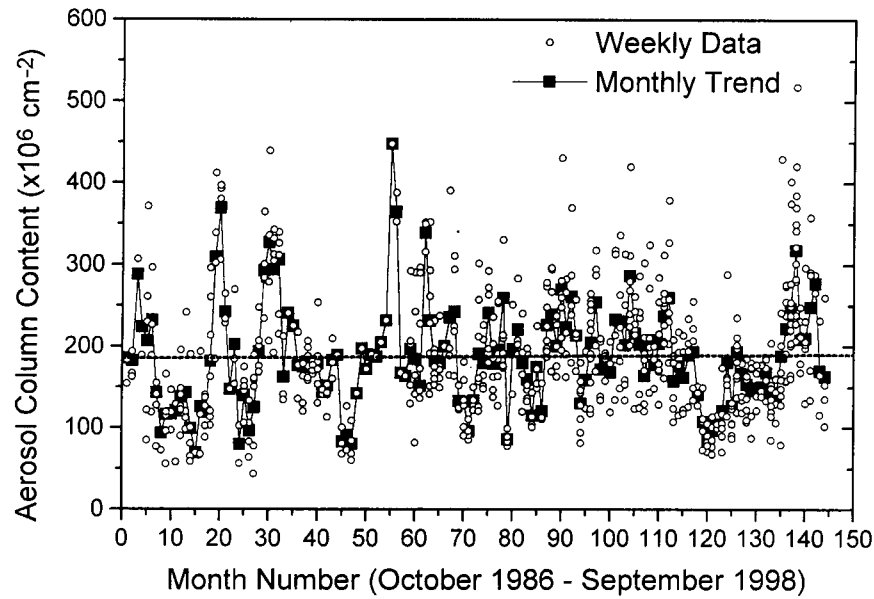


Figure 3. Monthly and weekly variations in lidar-observed aerosol loading at Pune. Thick solid line in the figure represents long-term trend computed from monthly mean aerosol loading

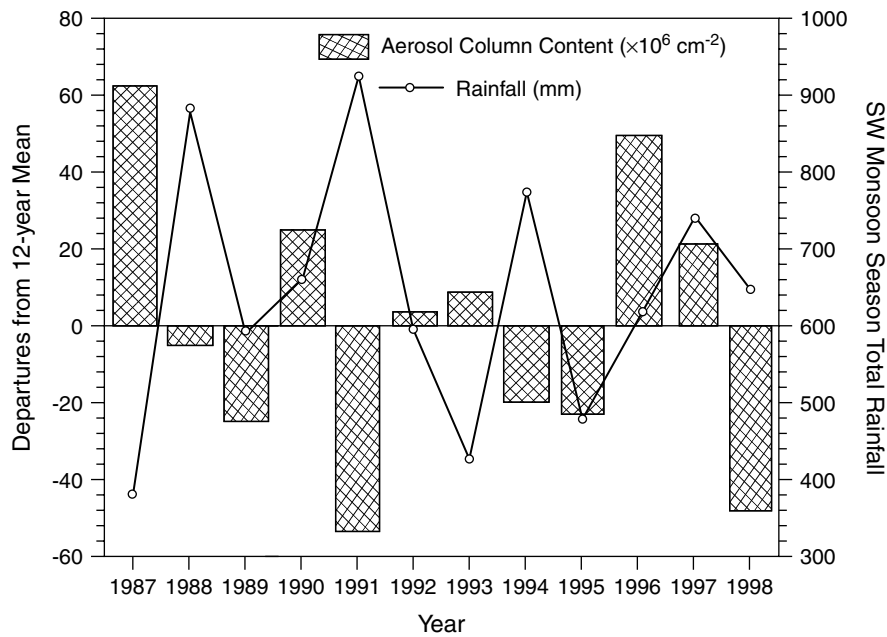


Figure 4. Departures of boundary-layer aerosol column content from their 12 year mean and their relationship with south-west monsoon season total rainfall

mainly by the convective and washout processes respectively. Thus the magnitude and polarity of departures can be attributed to the strength of aerosol sources and sinks, which depend upon the thermodynamical and cloud-microphysical processes induced by aerosols prevailing over the experimental station. It is also clear from Figure 4 that the departures in aerosol column content are not proportional to the rainfall. For example, during 1988 and 1991 (active monsoon years) the departure in aerosol content is less than  $10 \times 10^6 \text{ cm}^{-2}$

for rainfall of 885 mm in 1988, whereas it is around  $55 \times 10^6 \text{ cm}^{-2}$  for rainfall of 924 mm in 1991. This non-proportionality gives rise to a clue about the relative roles of rainout and washout processes. Thus the results show the predominance of the washout process during 1988 and the rainout process during 1991. Another interesting feature is that rainfall values are not very high; hence the departure values are negative instead of positive during 1989 and 1995–97, which is considered to be due to the non-uniform distribution of rainfall during the monsoon season.

**4.1.3. Aerosol–temperature–precipitation relationship.** As explained in the earlier sections, of the two major aerosol production/removal mechanisms, namely, convection during the pre-monsoon season and rainout/washout during the monsoon season, the latter is found to play a dominant role in altering the long-term trend in the aerosol loading at the station. This aspect is shown in Figure 5, which depicts the association between annual pre-monsoon minus post-monsoon aerosol column content, temperature and south–west monsoon season total rainfall during different years under study. It is evident that the variations in temperature and pre-monsoon minus post-monsoon aerosol content almost follow each other, which implies lifting of a greater number of aerosol particles from ground level to higher altitudes during the pre-monsoon season, as expected at the experimental station. This is especially true during the period of dust storms, which normally occur in this season. Another interesting aspect that can be inferred from the co-variations between aerosol column content and rainfall is that pre-monsoon content, in a way, is related to the ensuing south–west monsoon precipitation. This association is, however, noted to deviate during 1990, 1994 and 1997.

**4.1.4. Influence of annual oscillation.** As discussed in Section 4.1.3, the seasonal variation in columnar boundary-layer aerosol content shows a maximum loading during the pre-monsoon season and a minimum during the south–west monsoon season. Thus, it exhibits a dominant regular oscillatory behaviour due to the dust-storm/strong convective activities during the pre-monsoon months and cloud-scavenging/rain washout effects during the monsoon months. It may be considered that the anthropogenic activities in and around Pune city modulate this regular phenomenon. In order to examine the role of this annual oscillation (AO) in the long-term variations in aerosol loading over the experimental station, the regression analysis has been repeated for the aerosol column content data after filtering out this oscillation by following the moving

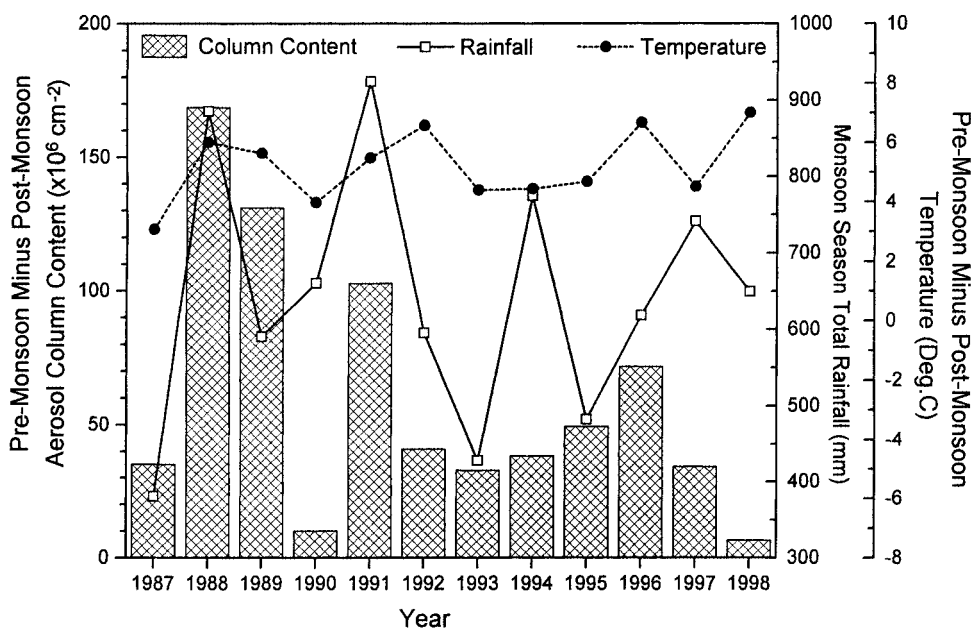


Figure 5. Co-variations between pre-monsoon content minus post-monsoon content and surface-level temperature, and monsoon season total rainfall



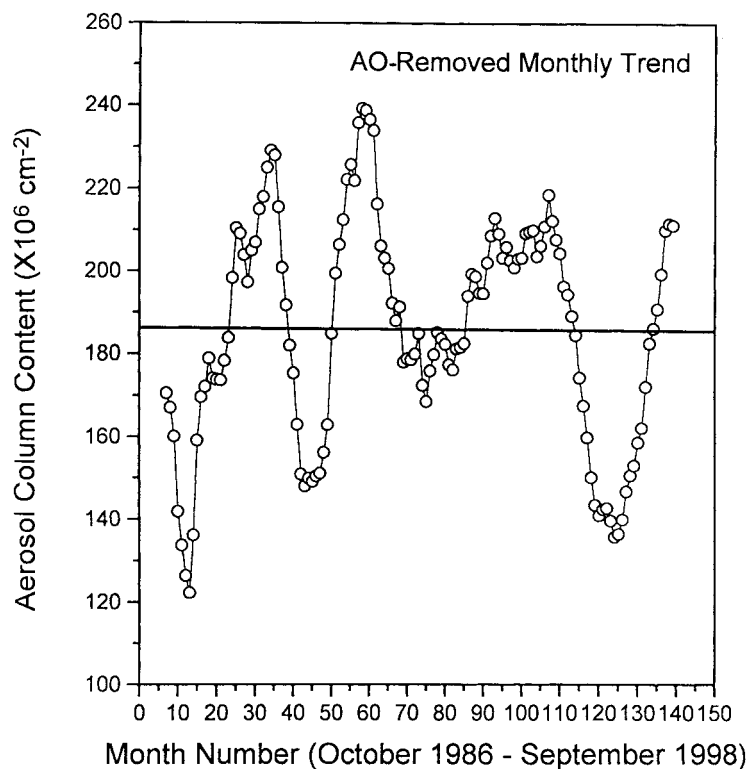


Figure 6. Long-term trend observed in AO-removed monthly mean aerosol column content data for the period from October 1986 to September 1998. Thick solid line indicates long-term trend

average method. Figure 6 shows long-term trend in the AO-removed monthly mean aerosol content data. The observed increasing trend of about 1% in the monthly mean data over the 12 year period clearly indicates the AO's influence on the long-term aerosol loading variations, apart from the increase in aerosol concentration due to human-related activities in and around Pune city. Furthermore, comparison between the long-term trends observed, before and after removal of the AO, clearly indicates the dominance of aerosol production processes (mostly due to anthropogenic activities) over the aerosol removal (rainout/washout) processes at the experimental station.

#### 4.2. Nocturnal evolution of aerosol content and its relationship with meteorological parameters

The aerosols that are lifted due to daytime convective activity would be suspended for a considerable amount of time in the lowest layers of the atmosphere, and those generated particularly near the surface would be confined to lower levels during the night. During the night, the depth of cooling increases with time, whereas the depth of the turbulence remains constant or decreases with time, which leads to the stratification of aerosol layers (Devara *et al.*, 1994, 1997). The stratified turbulence in the nighttime stable boundary-layer arises mainly due to wind shear (Mahrt, 1985). Horizontal mixing takes place in the turbulent layers and the regions in between the turbulent layers favour the formation of aerosol layers. Thus, the surface radiation inversions and turbulence mixing mainly control the nighttime aerosol column content over a place. Furthermore, other meteorological parameters, such as relative humidity, also greatly influence the physico-chemical characteristics of aerosols in a most complex way in the optical remote sensing method. Many investigators have demonstrated the influence of meteorological parameters on the formation, scattering, aging, transport and diffusion of atmospheric aerosols (Shettle and Fenn, 1979; Nilsson 1990; Horvath and Dellago, 1993).

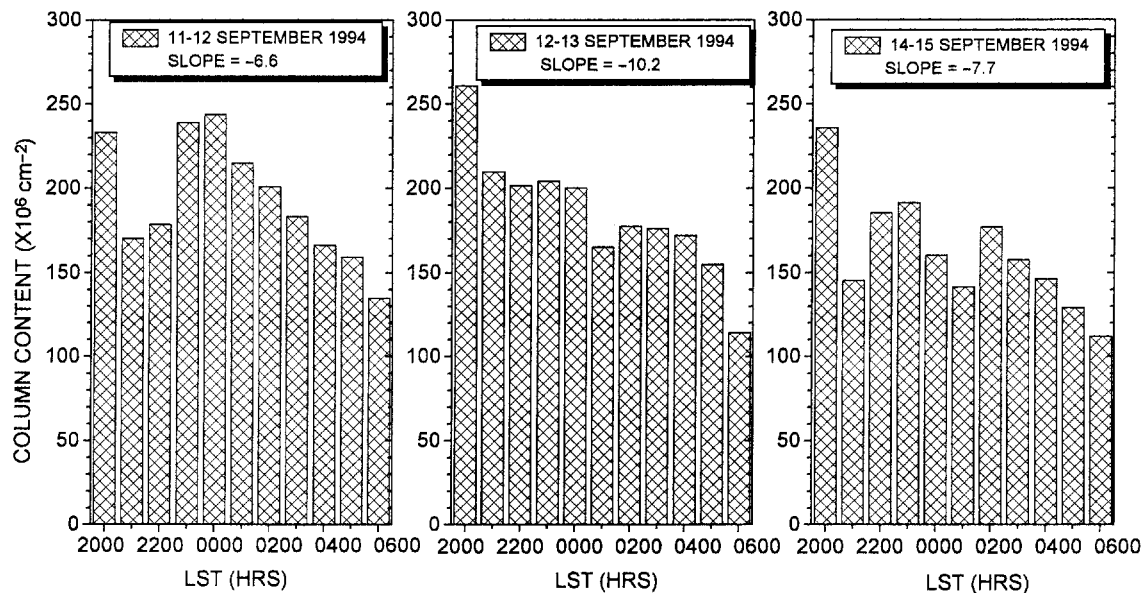


Figure 7. Nocturnal evolution of boundary-layer aerosol column content observed from all-night data collected in synchronization with the LITE mission. Slope of the time variation of aerosol content observed on each experimental day is shown in the frames

Table I. Evaluation of nighttime boundary-layer aerosol column content variations recorded at Pune, India, on three days during the LITE correlative measurement program

Date	Correlation coefficient	Regression coefficient		Standard deviation
		A	B	
11–12 September 1994	-0.61	14.89	-6.6	30.05
12–13 September 1994	-0.91	20.48	-10.2	16.55
14–15 September 1994	-0.75	14.77	-7.7	23.67

Of the 12 days' regular lidar observations, carried out over the experimental site during the LITE correlative measurement program, the data archived during the three experiments that were performed on the nights of the 11–12, 12–13 and 14–15 September 1994 were utilized to study the nocturnal evolution of aerosol column content and its association with some selected meteorological parameters. Figure 7 depicts the nocturnal evolution of boundary-layer aerosol content derived from a series of lidar profile data collected throughout the nights of 11–12, 12–13 and 14–15 September 1994. The slopes of the time variation of the aerosol content are also indicated in the plot. Some of the statistical parameters of the data analysis that describe the time variations in the aerosol column content are given in Table I. The common feature that can be seen on all observation days is that the aerosol column contents are high during the post-sunset period and low during the pre-sunrise period, with intermittent fluctuations that could be ascribed to the variations in the meteorological parameters, such as relative humidity, temperature and wind speed. The higher content during the late-evening hours and lower content during the early-morning hours are possibly due to settling of particles caused by radiation cooling at night.

In order to examine the above variations further, the horizontal gradients of aerosol column content and con-current relative humidity, temperature and wind were computed and are plotted in Figures 8, 9 and 10 respectively. A reasonably good agreement between the variations can be seen from the figures.

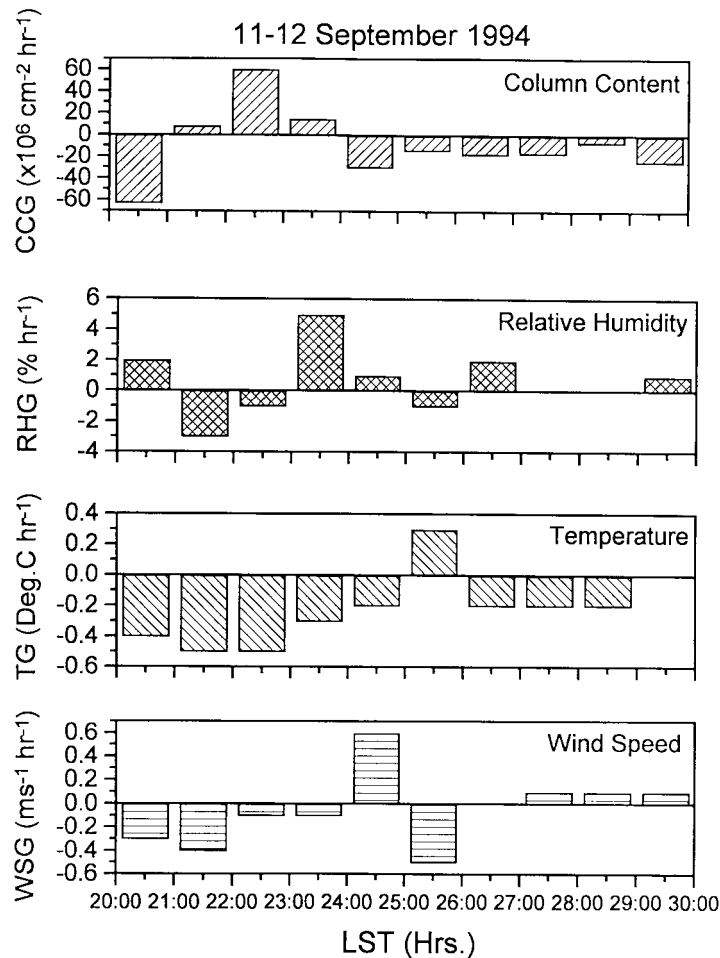


Figure 8. Correspondence between the horizontal gradients in aerosol column content, temperature, relative humidity and wind speed observed on 11–12 September 1994

Apart from the weak relative humidity gradients, the wind and temperature gradients are considered to be responsible for the short-term variations noticed in the nocturnal boundary-layer aerosol column content.

#### 4.3. Aerosol pollution and air quality

The ventilation coefficient, which represents the rate at which air within the mixed-layer (or mixing depth) is transported away from a region of interest, is an index of air pollution/quality over that region (Holzworth, 1967, 1972). The higher the coefficient, the greater is the atmosphere's ability to disperse pollutants. It is usually obtained by multiplying the mixed-layer height and the wind speed averaged through the mixed-layer, and is expressed in  $\text{m}^2 \text{s}^{-1}$ . Ventilation coefficients determined from the lidar-derived mixing depths as the height corresponding to the maximum negative gradient (in the height variation of aerosol concentration) close to the surface and pilot balloon wind data, obtained during the 12 year period from October 1986 to September 1998, are displayed in Figure 11. A significant seasonal variation is evident from the figure, with relatively higher coefficients during the pre-monsoon and lower values during winter and monsoon months. Though the ventilation coefficients are low during monsoon months, the effect of aerosol pollution is considered to be mitigated due to cloud scavenging and rain washout effects. However, the low coefficients noticed during the non-monsoon months of December and January

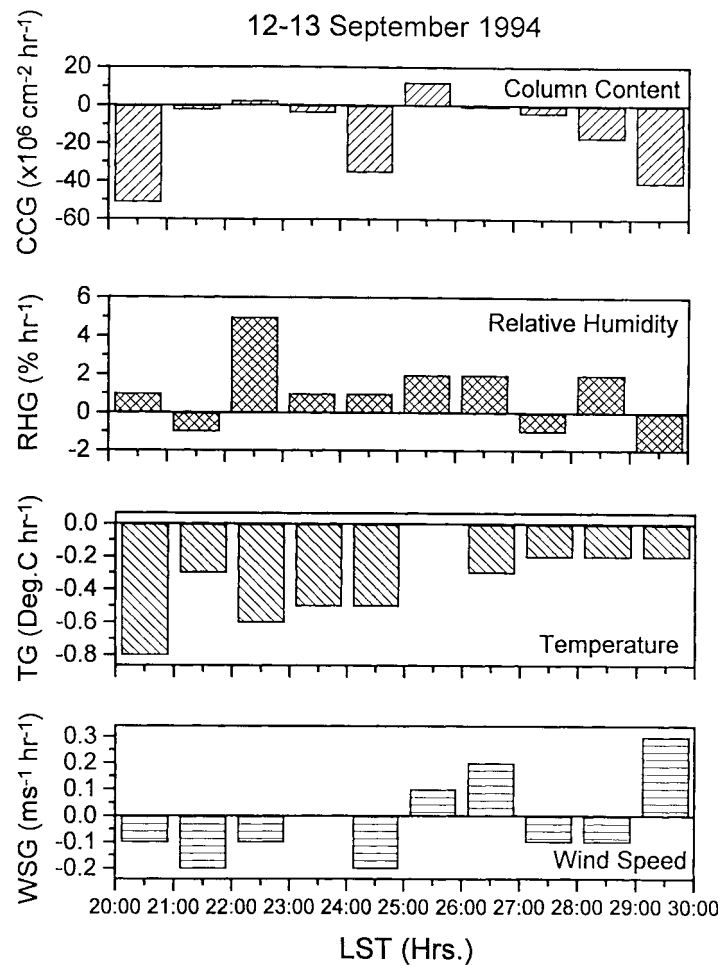


Figure 9. Same as Figure 8, but observed on 12–13 September 1994

clearly suggest that early winter evenings in Pune, India, tend to have higher pollution potential or poorer air quality.

The present results concerning the trends in mixing depths and associated ventilation coefficients are consistent with those observed in the coordinated experiments conducted with the collocated lidar and sodar systems at this Institute (Devara *et al.*, 1995c). In other words, the present study indicates that the air quality is poor over the station due to the recent increase in urbanization and industrialization around it. The study also reveals that the maximum amount of air pollutants is removed by precipitation during the monsoon months.

## 5. CONCLUSIONS

An extensive aerosol vertical profile database has been used for evaluating the recent climatological trends in urban aerosol loading and in air quality over a tropical station in India. The data were acquired with a computer-controlled, bistatic argon-ion lidar, and observations were carried out for a 12 year period from October 1986 to September 1998. In addition, observations of lidar-derived aerosol column content, as part of the LITE correlative measurement program, have been utilized to study the short-term variations

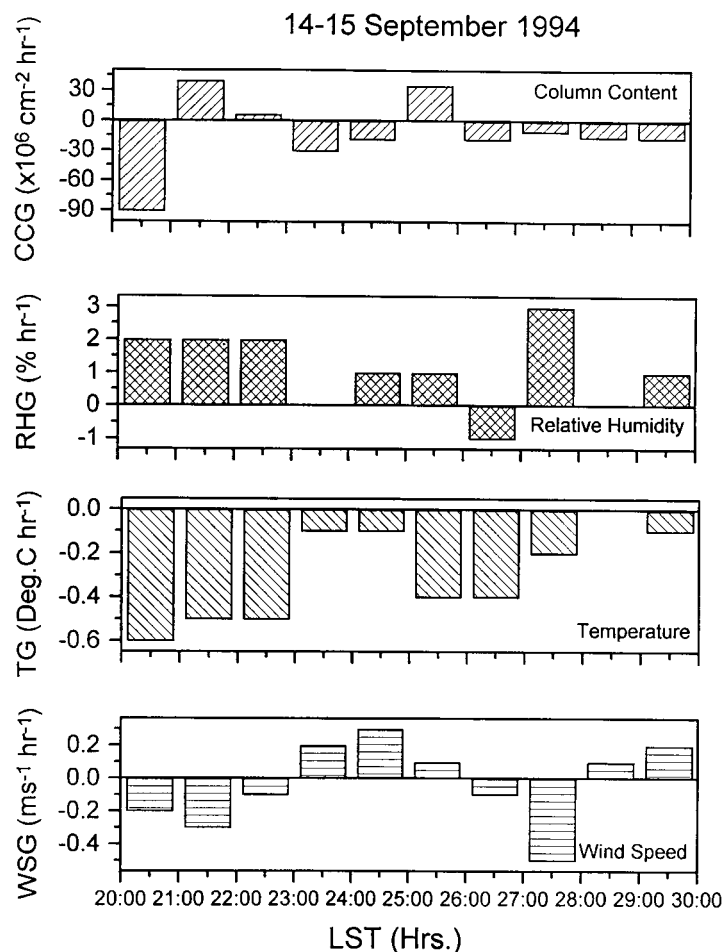


Figure 10. Same as Figure 8, but observed on 14–15 September 1994

in the nocturnal evolution of boundary-layer aerosol column content and its correspondence with concurrent meteorological parameters.

The study has indicated the following results, most of which are specific to this site in Pune, India:

- (i) over a 39% contribution from the air layers close to the ground (up to 200 m) to the overall aerosol loading (up to 1100 m) at the station;
- (ii) a maximum aerosol loading during the pre-monsoon and minimum loading during the monsoon season, and the decrease in loading during the monsoon varies between 34.89 and 36.12% (average being about 35%) from that of the loading during the pre-monsoon months;
- (iii) an increase of about 3% in the aerosol loading over the experimental station during the 12 year period, October 1986–September 1998;
- (iv) a close association between long-term trend in aerosol loading, pre-monsoon minus post-monsoon aerosol column content, temperature, and south-west monsoon season total rainfall;
- (v) an influence of the AO resulting from the combined thermal and dynamical effects and summer monsoon precipitation, on the long-term trends in urban aerosol loading over the experimental station;
- (vi) a close association between nocturnal evolution of boundary-layer aerosol content and concurrent meteorological parameters; and
- (vii) higher pollution potential (or poorer air quality) during the winter late evenings.

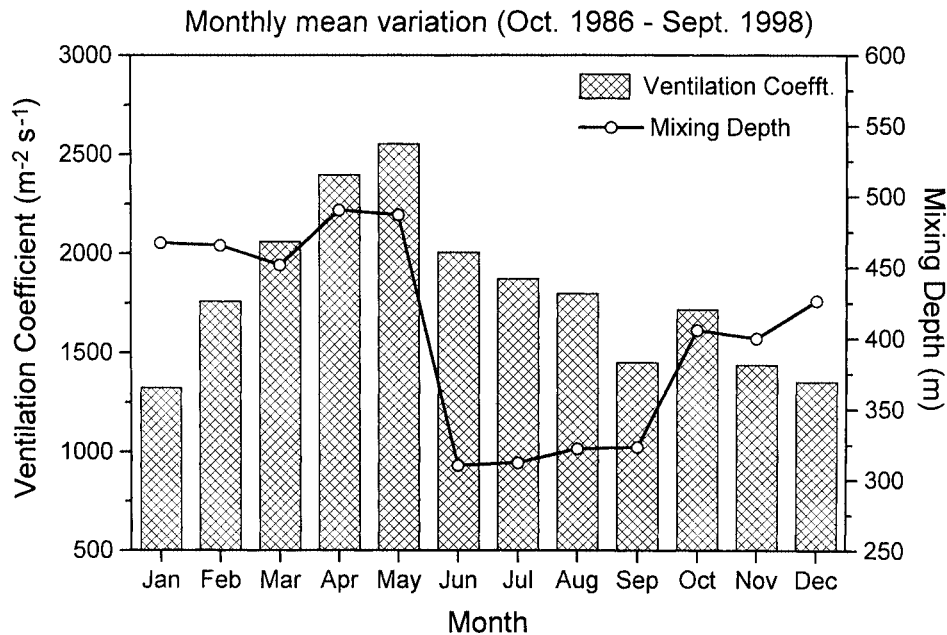


Figure 11. Monthly mean mixing depth and ventilation coefficient derived from the lidar vertical profile observations carried out from October 1986 to September 1998

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#### REFERENCES

- Boucher O, Anderson TL. 1995. GCM assessment of the sensitivity of direct climate forcing by anthropogenic sulfate aerosols to aerosol size and chemistry. *Journal of Geophysical Research* **100**: 26 117–26 134.
- Charlson RJ, Schwartz SE, Hales JM, Cess RD, Coakley Jr JA, Hansen JE, Hoffman DJ. 1992. Climate forcing by anthropogenic aerosols. *Science* **255**: 423–430.
- Devara PCS. 1992. Lidar measurements of atmospheric aerosols. In *Lidar Studies of the Atmosphere*, Scientific Report, Department of Science and Technology: Government of India; 15–26.
- Devara PCS, Raj PE. 1987. A bistatic lidar for aerosol studies. *Institute of Electronics and Telecommunication Engineers, Technical Review* **4**: 412–415.
- Devara PCS, Raj PE. 1989. Remote sounding of aerosols in the lower atmosphere using a bistatic, CW helium–neon lidar. *Journal of Aerosol Science* **20**: 37–44.
- Devara PCS, Raj PE, Sharma S. 1994. Remote sensing of atmospheric aerosol in the nocturnal boundary layer using lidar. *Environmental Pollution* **85**: 97–102.
- Devara PCS, Raj PE, Sharma S, Pandithurai G. 1995a. Real-time monitoring of atmospheric aerosols using a computer-controlled lidar. *Atmospheric Environment* **29**: 2205–2215.
- Devara PCS, Raj PE, Pandithurai G. 1995b. Aerosol profile measurements in the lower troposphere with four-wavelength bistatic argon-ion laser. *Applied Optics* **34**: 4416–4425.
- Devara PCS, Raj PE, Murthy BS, Pandithurai G, Sharma S, Vernekar KG. 1995c. Intercomparison of nocturnal lower-atmospheric structure observed with lidar and sodar techniques at Pune, India. *Journal of Applied Meteorology* **34**: 1375–1383.
- Devara PCS, Raj PE, Pandithurai G, Naresh P. 1997. Atmospheric stability effects on aerosol structures and stratification. *Journal of Meteorology* **22**: 176–187.
- Endlich RM, Luding FL, Uthe EE. 1979. An automatic method for determining the mixing depth from lidar observations. *Atmospheric Environment* **13**: 1051–1056.
- Gultepe I. 1995. Physical, radiative and dynamical processes within a nighttime marine stratus cloud. *Pageoph* **144**: 321–350.
- Hashvardhan. 1993. Aerosol–climate interactions. In *Aerosol-Cloud-Climate Interactions*, Hobbs PV (ed.). International Geophysics Series vol. 54. 75–95.

- Haywood JM, Ramaswamy V. 1998. Global sensitivity studies of the direct radiative forcing due to anthropogenic sulfate and black carbon aerosols. *Journal of Geophysical Research* **103**: 6043–6058.
- Holzworth GC. 1967. Mixing depths, wind speeds and pollution potential for selected locations in the USA. *Journal of Applied Meteorology* **6**: 1034–1044.
- Holzworth GC. 1972. Mixing heights, wind speeds and potential for urban air pollution throughout the contiguous United States. USEPA, Office of Air Programs, Publication no. 101, North Carolina; 118 pp.
- Horvath H, Dellago C. 1993. On the accuracy of the size distribution information obtained from light extinction and scattering measurements — (ii) case studies. *Journal of Aerosol Science* **24**: 143–154.
- IGAP (The International Global Aerosol Program) Plan. 1991. In *Report of the Experts Meeting on Space Observations of Tropospheric Aerosols and Complementary Measurements* Stowe LL, Hitztenberger R, Deepak A (eds). 51 pp.
- IPCC (Inter-Governmental Panel for Climate Change). 1995. *The Science Climate Change*, Houghton JT, Meira Filho LG, Callander BA, Harris N, Kattenberg A, Maskell K (eds). Cambridge University Press; 572 pp.
- Jones A, Slingo A. 1996. Predicting cloud-droplet effective radius and indirect sulfate aerosol forcing using general circulation model. *Quarterly Journal of the Royal Meteorological Society* **122**: 1573–1595.
- Kiehl JT, Briegleb BP. 1993. The relative roles of sulfate aerosols and greenhouse gases in climate forcing. *Science* **260**: 311–314.
- Li Z. 1998. Influence of absorbing aerosols on the inference of solar surface radiation budget and cloud absorption. *Journal of Climate* **11**: 5–17.
- Mahrt L. 1985. Vertical structure and turbulence in the very stable boundary layer. *Journal of the Atmospheric Sciences* **42**: 2332–2349.
- McClatchey RA, Fenn RW, Selby JEA, Volz FE, Garing JS. 1972. Optical properties of the atmosphere. AFCRL-72-0497, Air Force Cambridge Research Laboratories, Bedford.
- McCormick MP, Winker DM, Browell EV, Coakley JA, Gardner CS, Hoff RM, Kent GS, Melfi SH, Menzies RT, Platt CMR, Randall DA, Reagan JA. 1993. Scientific investigations planned for the Lidar In-Space Technology Experiment (LITE). *Bulletin of the American Meteorological Society* **74**: 205–214.
- Nilsson B. 1990. Correlation between atmospheric aerosol extinction and meteorological parameters. In *Aerosols, Science, Industry, Health and Environment*, vol. 1, Masuda S, Takahashi K (eds). Pergamon Press: Oxford; 299–302.
- Pandithurai G, Devara PCS, Raj PE, Sharma S. 1996. Aerosol size distribution and refractive index from bistatic lidar angular scattering measurements in the surface layer. *Remote Sensing of the Environment* **56**: 87–96.
- Penner JE, Charlson RJ, Hales JM, Laulainen NS, Leifer R, Novakov T, Ogren J, Radke LF, Schwartz SE, Travis L. 1994. Quantifying and minimizing uncertainty of climate forcing by anthropogenic aerosols. *Bulletin of the American Meteorological Society* **75**: 375–400.
- Ramanathan V, Cess RD, Harrison EF, Minnis P, Barkstrom BR, Ahmad E, Hartmann D. 1989. Cloud-radiative forcing and climate change: results from the Earth Radiation budget Experiment. *Science* **243**: 57–63.
- Sasano Y, Sigematsu A, Shimizu A, Takuchi N, Okuda M. 1982. On the relationship between the aerosol layer height and the mixed layer height determined by laser radar and low-level radiosonde observations. *Journal of the Meteorological Society of Japan* **60**: 889–895.
- Sharma S. 1994. *Remote sensing of the atmosphere using the lidar technique*. Ph.D. Thesis, University of Pune; 167 pp.
- Shettle EP, Fenn RW. 1979. Models for the aerosols of the lower atmosphere and the effects of humidity variations on their optical properties. Report AFGL-TR-79-0214, Air Force Geophysical Laboratory, Hanscom, MA.
- Stull RB. 1991. *An Introduction to Boundary Layer Meteorology*. Kluwer Academic Publishers: Chapter 14.
- Tanaka M, Nakajima J, Takamura T. 1982. Simultaneous determination of complex refractive index and size distribution of airborne and water-soluble particles from light scattering measurements. *Journal of the Meteorological Society of Japan* **60**: 1255–1272.
- Taylor K, Penner JE. 1994. Response of the climate system to atmospheric aerosols and greenhouse gases. *Nature* **369**: 734–737.
- WCP (World Climate Program). 1983. In *Report of the Experts Meeting on Aerosols and Their Climatic Effects*, Deepak A, Gubei HE (eds). WMO: Geneva; 107 pp.
- WCRP (World Climate Research Program). 1988. Aerosols, clouds and the climatically important parameters: lidar applications and networks (Final Report, meeting of experts), WMO/TD-No. 233, Geneva, Switzerland.
- Wolf ME, Hidy GM. 1997. Aerosols and climate: anthropogenic emissions and trends for 50 years. *Journal of Geophysical Research* **102**: 11 113–11 121.
- Woolum CA. 1964. Notes from a study of a micrometeorology of the Washington, DC area for the winter and spring seasons. *Weatherwise* **17**.
- Zuev VE. 1982. *Laser Beams in the Atmosphere*. Plenum Publishing: New York; 504 pp.