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Abstract. This paper essentially addresses the structure and stratification of the nocturnal boundary layer (NBL) derived from the vertical profile measurements of aerosol concentration made with a computer-controlled Argon ion lidar system at the Indian Institute of Tropical Meteorology (IITM), Pune, India. Comparisons are made between the lidar observations during clear night-sky conditions and concurrent aerometric observations carried out on some selected experimental days. The results show multiple stratified aerosol layer structures in the nocturnal lower atmosphere, which drift either upward or downward depending on atmospheric stability conditions prevailing at different altitudes. The normalized aerosol concentration gradient (NCG) profiles indicate the variations in the nocturnal mixing depth from 200 to 426 m and that in the stable layer height from 325 to 725 m during the period of observations. The importance of such observations in the context of monitoring and/or assessment of airborne particulate pollutants over the urban environments associated with non-uniform terrain is discussed.

INTRODUCTION

The vertical distributions of naturally occurring and anthropogenic aerosols, which vary greatly depending on environmental conditions as well as upon local sources, are needed to evaluate their impact on climate (WCRP, 1988). Recent studies show that anthropogenic component of this aerosol burden causes a direct negative radiative forcing in the shortwave, which is comparable but of opposite sign to positive longwave forcing of several trace (greenhouse) gases including CO₂ associated with industrial and agricultural activities (HARSHVARDHAN, 1993). Tracking of aerosol inhomogenities also provides new insight into the dynamic processes of the planetary boundary layer (PBL) and helps to identify the sources/sinks implicated in long-range transport of pollutants (RUSSELL and UTHE, 1978; SASANO et al., 1980). Excellent spatial resolution and sensitivity suggest lidars (laser radars) as natural sensors best suitable for such studies (ENDLICH et al., 1979; SASANO et al., 1980; 1982; STULL and ELORANTA, 1984; HASHMONAY et al., 1991; COOPER and EICHINGER, 1994). Furthermore, when the laser energy propagates through the atmosphere, it is scattered by aerosols as well as by molecules. The retrieved aerosol concentration profiles often exhibit layer structures depending on atmospheric stability conditions (SASANO et al., 1982). Generally, there is sharp gradient of aerosol scattering associated with the PBL top, with high aerosol scattering within it, and very low scattering from the free atmosphere above. It is this gradient in aerosol scattering which is used to visualise the structure of the PBL (ENDLICH et al., 1979).

Monitoring of atmospheric aerosols and trace gases using the lidar techniques has been in progress at the Indian Institute of Tropical Meteorology (IITM), Pune (18° 32' N, 73° 51' E, 559 m AMSL), India since 1985. The Argon ion lidar facility, being fully computer-controlled offers all the benefits of on-line acquisition and processing of data while retaining the very valuable graphical representation in the familiar two- and three-dimensional views of aerosol and trace gas characteristics. In this paper, we present the real-time observations of the height-time variations of aerosol characteristics in a terrain-induced NBL obtained with this lidar facility. The influence of atmospheric stability, derived from the simultaneous aerometric (temperature, wind and relative humidity) observations; the experimental terrain and local pollution sources on such variations are discussed.

ABOUT THE EXPERIMENTAL STATION

The experimental station, Pune is about 100 km inland from the west coast of India and is located on the lee-side of the Western Ghats. The environment in the immediate vicinity of the station is urban with several industries nearby and possible aerosol type present over the station is a mixture of water-soluble, dust-like and soot-like aerosols. Soil dust is the major source of aerosols present over the experimental station. Formation of aerosols in the accumulation mode is considered to be due to gas-to-particle conversion processes while coarse aerosols are attributed mainly to wind-blown dust. Continental airmasses, rich in nuclei of continental origin pass over the station during pre-winter season. Fair weather conditions with clear skies and very low relative humidity exist during winter season. Low-level inversions during morning and evening hours, and dust haze during morning hours, occur during this season.

The experimental site is located at an elevation of about 570 m AMSL and is surrounded by the hillocks having elevations as high as 760 m AMSL, majority with elevation greater than 610 m MSL. The stone quarries (east side) and brick-kilns (west side) situated about 1 km from the site are the local anthropogenic sources influencing the observations (particularly in the lower layers of the atmosphere). In addition, there is major urban activity to the east of the site; to the west is a sparsely populated area. The above typical terrain (valley-like) is believed to induce complex wind field-caused turbulence.

LIDAR AND AEROMETRIC OBSERVATIONS - ANALYSIS TECHNIQUES

The complete description, recent significant improvements and present status of the lidar system used in the experiment have been presented elsewhere (DEVARA, 1992; DEVARA et al., 1994). The lidar setup featured a commercial Argon ion laser system (Lexel 95-4) as transmitter and a 25-cm-diameter, f 7.6 Newtonian telescope as receiver tailored with an optics assembly composed of condensing-collimating lenses, narrowband interference filters (1nm FWHM) and Peltier-cooled PMT. As the information on aerosols in the lowest layers of the atmosphere can be obtained most conveniently with the help of bistatic systems, the lidar in the present experiment has been operated in the bistatic mode with a horizontal separation of about 60 m between its transmitter and receiver. This bistatic experimental arrangement also provides

information on the angular scattering of aerosols of different sizes present in the atmosphere for determining the size distribution.

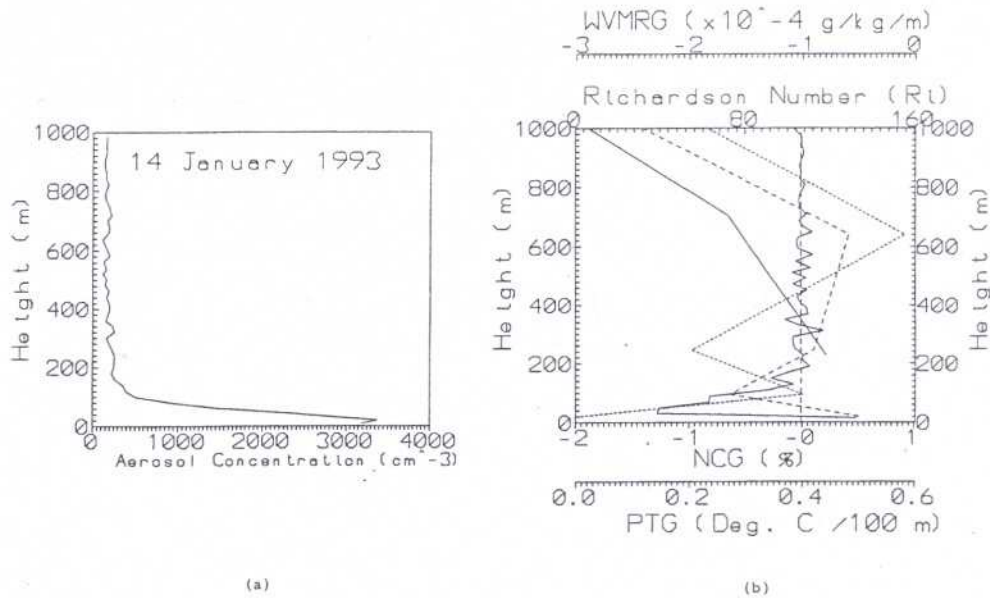


Fig 1. Vertical distributions of [a] aerosol number concentration and [b] relationship among the height gradients of aerosol number concentration (NCG), potential temperature (PTG) and water vapour mixing ratio (WVMRG), and Richardson number (Ri) observed on 14 January 1993.

The scattered signal strength profiles were obtained by scanning the vertically sent Argon ion laser beam (at wavelength of 514.5 nm and of about 400 mW power) with the receiver at pre-selected elevation angles corresponding to different altitudes. As the scattered intensity from atmospheric aerosols at different scattering angles depends on the size distribution and refractive index parameters, we conduct multi-scattering angle and multi-wavelength experiments in every month to determine these parameters (RAJ and DEVARA, 1995) and utilize them in the inversion method (DEVARA and RAJ, 1987; 1989) which is briefly described in the paragraphs to follow.

Generally, the amount of scattering of 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 commonly assumed that the aerosols are of homogeneous spherical particles characterized by 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. Another basic assumption involved in the lidar measurement of atmospheric aerosols is that aerosol

characteristics do not change appreciably during the period of observations and as such these observations need to be collected as quickly as possible.

From the scattered signal strength profiles, the aerosol concentration (N) at each scattering angle was computed by using the following bistatic lidar equation.

$$N = N_s [T\eta\sigma(\theta_{\text{scat}})]^{-1}$$

where N_s is normalized signal strength computed by considering the system and geometrical parameters; T is atmospheric transmittance along the transmitter-scattering volume-receiver path η is the system constant including all the overall efficiencies of the transmitter and receiver optics; $\sigma(\theta_{\text{scat}})$ is differential Mie scattering cross-sections at scattering angle (θ_{scat}). Amongst the different aerosol size distributions available in the literature (JUNGE, 1955; DEIRMENDJIAN, 1969; SHETTLE and FENN, 1979), the following modified power law distribution as given by McCLATCHEY et al (1972) which is found representative of the conditions of the experimental station has been used in the measurements reported in this paper.

$$\begin{aligned} N(r) = dN(r)/dr &= C 10^{\nu} \text{ for } 0.02 \mu\text{m} < r < 0.1 \mu\text{m} \\ &= C r^{-\nu} \text{ for } 0.1 \mu\text{m} < r < 10 \mu\text{m} \\ &= 0 \text{ for } r < 0.02 \mu\text{m} \text{ and } r > 10 \mu\text{m} \end{aligned}$$

where ν is defined as the size index (also called as shaping or size distribution parameter) and C is the normalization constant that follows from continuity.

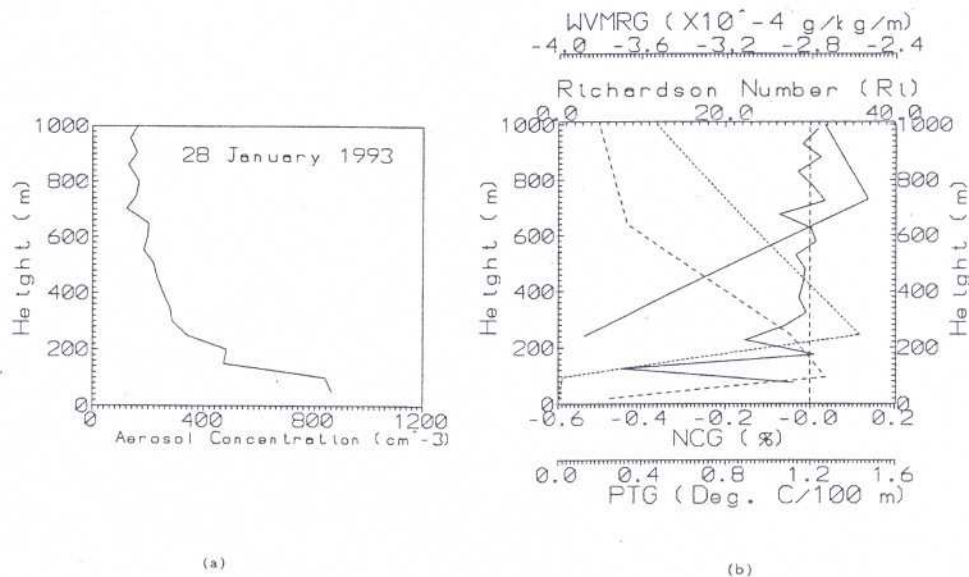


Fig2. Vertical distributions of [a] aerosol number concentration and [b] relationship among the height gradients of aerosol number concentration (NCG), potential temperature (PTG) and water vapour mixing ratio (WVMRG), and Richardson number (Ri) observed on 28 January 1993.

The lidar experiments were conducted on three days with clear night sky conditions on 23-24 December 1992 and 14 & 28-29 January 1993. On each day, the observations have been obtained at close height intervals of 50 m (total height steps of 20) in the altitude range of 50-1000 m in the early night period on 23 December 1992 and 14 & 28-29 January 1993 coinciding with the radiometersonde (an audio-modulated radiosonde with an additional radiometer attached) and pibal experiments of the India Meteorological Department (IMD), Pune which is situated at an aerial distance of about 3 km east of the lidar site. As the lidar observations on the above three days were obtained during the early night hours around 1900 LST, the wind, temperature and relative humidity datasets collected during the evening ascents are considered here for comparison. Furthermore, as the pilot balloon and radiometersonde data are available at height intervals varying from 40 to 500 m, respectively, values at intermediate heights were obtained by interpolation methods.

A series of vertical profiles of aerosol concentration was also obtained at 15 min and 1 h intervals, respectively, on the nights intervening 23-24 December 1992 and 28-29 January 1993. The lidar observations obtained in the early night period on 14 and 28 January 1993 were used to study the vertical distribution, structure and stratification of aerosols and their relationship with atmospheric stability parameters. Such a study could not be undertaken with the lidar observations obtained on 23 December 1992 due to non-availability of radiometersonde data on that day. The aerosol vertical profile series obtained from early night through early morning hours on 23-24 December 1992 and 28-29 January 1993 were utilized to investigate the height-time variability of aerosol layer structure and stratification. The temporal variation of aerosol concentration close to the ground (50 m) which reflects, by and large, the local effects due to surrounding human activities and the experimental terrain, is studied in conjunction with the surface-level meteorological parameters.

In order to investigate the height variation of aerosol layer structure and its relation with the stable layer characteristics, normalized concentration gradient (NCG) profiles have been derived from the aerosol profiles obtained on 14 & 28 January 1993 by following the method described by DEVARA and RAJ (1990). The normalization procedure followed in this method eliminates the absolute calibration of the lidar system and also the local variations in the aerosol concentration. The largest negative gradient nearest to the surface in the lidar backscatter (or the aerosol concentration) is associated with the sharpest decline from high to low particulate concentrations and thus marks the mixing depth (ENDLICH et al., 1979). Positive gradients often occur above the mixing depth and they denote the aerosol layer structures associated with the stable conditions. The atmospheric stability parameters such as Richardson Number (Ri), which includes the directional wind-shear, potential temperature gradient (PTG) and water vapour mixing ratio gradient (WVMRG) were computed from the temperature, relative humidity and wind profiles collected. Simultaneous wind speed, temperature and relative humidity observations at 5 m level were also collected in the vicinity of the lidar setup during the throughout night lidar experiments performed on 23-24 December 1992 and 28-29 January 1993.

DISCUSSION OF RESULTS

The vertical distributions of aerosol concentration retrieved from the lidar observations carried out on 14 and 28 January 1993 along with the vertical profiles of NCG, Ri, PTG and WVMRG are shown in Figures 1 (a&b) and 2(a&b). The aerosol concentration showed variation from 93 to 2478 cm^{-3} and from 160 to 861 cm^{-3} between 50 m and 1000 m altitudes, respectively on 14 and 28 January 1993 [Figs. 1 (a) and 2 (a)]. Furthermore, the profiles indicate high aerosol concentration in the surface layer, decreasing rapidly with altitude (steep negative gradient) up to about 150 m and thereafter varies smoothly with altitude. The higher aerosol concentration and its steep decrease in the lowest layers are the regular features seen in almost all the profiles of aerosol concentration at this experimental site because of the combined effects of terrain-induced circulation processes and local pollution sources (DEVARA and RAJ, 1991).

Stable stratifications are normally present at the altitudes where the air temperature profiles exhibit inversions. During the periods of clear night sky, mixing processes are mainly controlled by advection and clear air radiative cooling (RUSSELL et al., 1974; MAHRT et al., 1979). The influence of wind-shear-caused turbulent layers on the shape of the aerosol profiles and association between the height regions having larger values of Ri (very stable) and stratified aerosol layer structures can be clearly seen from the Figures 1 (b) and 2 (b). This is consistent with the stratified turbulence due to wind-shears in the NBL as observed by MAHRT (1985). It may be noted here that during night, the depth of significant cooling increases with time while the depth of the turbulence region remains constant or decreases with time which leads to the stratification of aerosol layer. The stratified turbulence in the NBL arises mainly due to wind-shears (MAHRT et al., 1979; MAHRT, 1985). Horizontal mixing of aerosols also takes place in the turbulent layers and the regions in between such turbulent layers favour the formation of aerosol layers.

Besides the stratified aerosol layer structures, the multiple stably stratified aerosol layers at around 310 m and 725 m may also be noted respectively on 14 and 28 January 1993 from the above figures. As suggested by SASANO et al (1982), these layers can be explained on the basis of height variation of potential temperature gradient. Smaller negative ratio values of water vapour mixing ratio gradient seem to have favoured the formation of significant layer structures. According to NILSSON (1979) the increase in aerosol size with relative humidity more than 80% results in an enhancement in scattered intensity. The relative humidity in the region of observations was less and it showed variation from 21 to 33% and 30 to 43%, respectively, on 14 and 28 January 1993.

The depth of mixed-layer was determined from the NCG profile by marking the height corresponding to the maximum negative gradient of aerosol concentration closest to the ground (DEVARA and RAJ, 1990). This method of marking the mixing depth has been widely used to study the evolution of the daytime mixed-layer. The nighttime mixed-layer height extending up to 1500 m during the post-sunset period in fine weather has also been reported by some researchers using such method (SASANO et al., 1983). The mixing depth thus deduced was found to be 149.5 m on

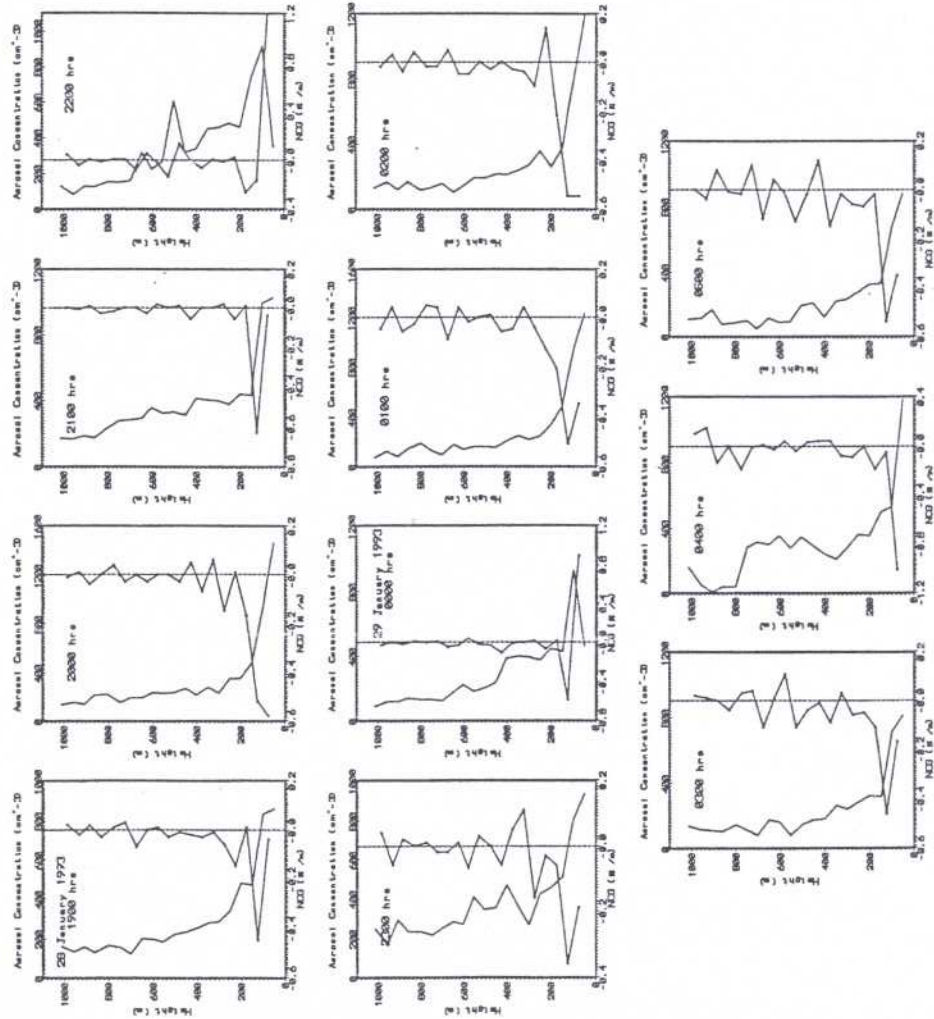


Fig 3. Time-height variations of aerosol number concentration and corresponding NCG (shown with centred symbols) observed on the night of 28-29 January 1993.

14 January and it was 220 m on 28 January 1993. The mixing depth limits the vertical transport of pollutants and the rate at which these pollutants are transported can be determined from ventilation coefficients. The higher the ventilation coefficient, the more the atmosphere is able to dispose the pollutants and hence these coefficients provide an index of air pollution potential. Thus these coefficients have been estimated by coupling the mixing depths with the average wind velocity in the mixed-layer, and they were found to be $657.80 \text{ m}^2\text{s}^{-1}$ and $880 \text{ m}^2\text{s}^{-1}$, respectively on 14 and 28 January 1993. According to the US Air Pollution Forecast Programme (GROSS, 1970), ventilation coefficients $< 6000 \text{ m}^2\text{s}^{-1}$ during afternoon and less than $2000 \text{ m}^2\text{s}^{-1}$ during morning hours indicate high air pollution potential. Thus the experimental station appears to have higher pollution potential during the late evening hours of the above observational days. This is consistent with the earlier results of lidar observations which had indicated that the winter late evenings at Pune tend to have higher pollution potential (DEVARA et al., 1994).

Figure 3 displays a series of vertical profiles of aerosol concentration and the corresponding NCG obtained at 1 h interval on the night of 28-29 January 1993. It is clear that the aerosol concentration, on an average, was higher during the post-sunset period as compared to during the pre-sunrise period. The NCG profiles revealed the presence of significant aerosol layers between the altitudes 300 m and 700 m, and they appear to redistribute vertically with time. Formation of these layers is mainly due to the lack of thorough mixing in those height regions. The variations in the aerosol layer depth in both space and time are considered to be due to the changes in atmospheric stability.

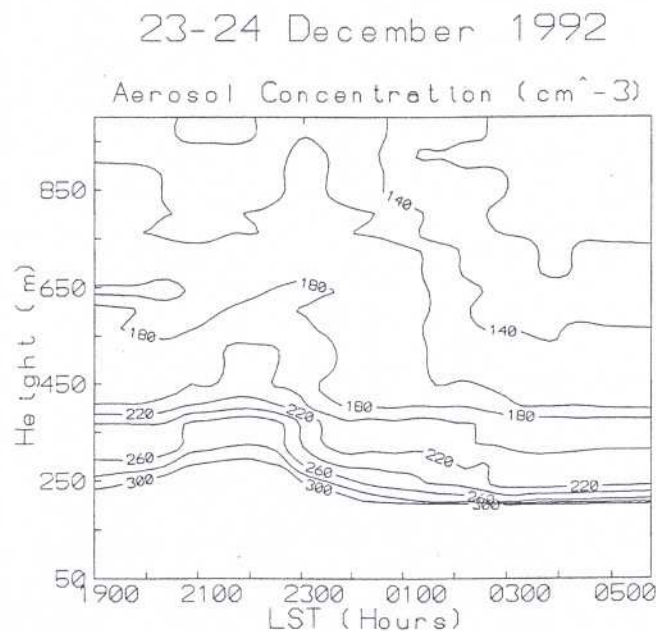


Fig 4. Contour plot showing time-height cross-section of aerosol number concentration observed on the night of 23-24 December 1992. Contour values above 300 cm^{-3} up to 150 m altitude are not in the figure for the reasons cited in the text.

Figure 4 depicts the three-dimensional graphic representation of a series of vertical profiles of aerosol concentration archived at 15 min interval on the night of 23-24 December 1992. This clearly shows significant space-time variations of aerosol concentration. The aerosol concentrations observed above 300 cm^{-3} were not shown in the figure in order to project clearly the fine-scale variations in concentrations in each data set. A significant temporal variation in aerosol concentration is pronounced at lower altitudes which is considered to be due to the variations in meteorological parameters at different altitudes and in local anthropogenic sources which are believed to be contributing to the aerosol observations at the experimental site.

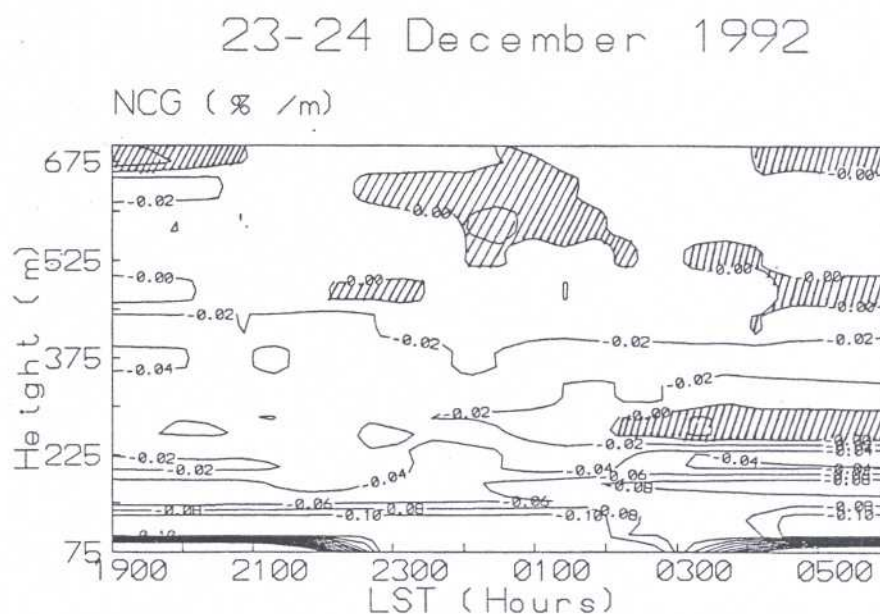


Fig 5. Temporal variation of NCG indicating the time evolution of nocturnal mixing depth and stable layer on the night intervening 23-24 December 1992. Hatched contours show the regions of stability.

A contour plot of the NCG distributions corresponding to the fine-scale variations of aerosol concentrations shown in Figure 4 is depicted in Figure 5. The time variation of both mixing and stable regions, derived from the procedure as explained above, can be seen from the figure. Besides the large negative NCG observed at around 125 m in almost all the profiles which is considered to be due to the effect of experimental terrain, the average mixing depth was found to be 300.5 m (175-426 m) while the most probable height of stable layer was 525 m (325-725 m) during the observational period. The observed mixed and stable layer heights and their temporal variations in the present study were found to be consistent with our earlier-reported results (RAJ and DEVARA, 1993).

As explained above, the mixing depth and its time variation, from the series of NCG profiles, obtained on the nights of 23-24 December 1992 and 28-29 January 1993 can be important input parameters in the assessment of air quality at the place

of observations. Generally, the nocturnal mixed-layer height is known to vary strongly with time - growing, collapsing, and then growing again, several times over the course of the night. Thus the drifts in the mixed-layer height may have direct bearing on the changes in the dispersal of pollutants near the ground at the observing station.

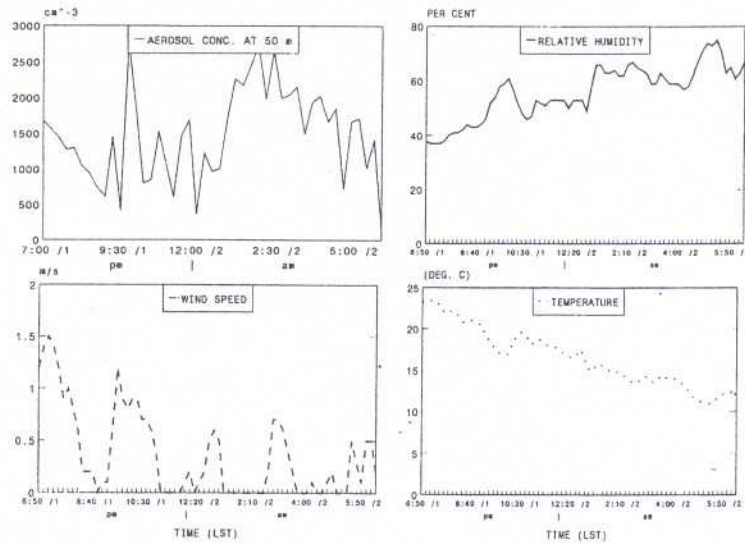


Fig 6. Comparison between the temporal variation of the aerosol number concentration at 50m and the concurrent surface-level meteorological parameters observed on 23-24 December 1992.

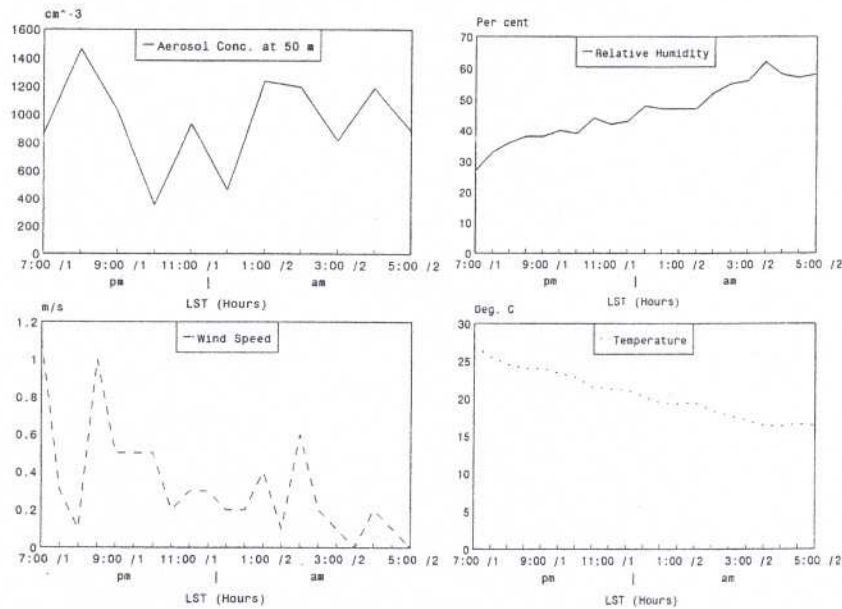


Fig 7. Comparison between the temporal variation of the aerosol number concentration at 50m and the concurrent surface-level meteorological parameters observed on 29 January 1993.

Since the surface-level meteorological parameters are known to influence more the aerosol concentration near ground, the aerosol concentration at the lowermost altitude (50 m) was picked-up from the series of profiles collected on 23-24 December 1992 and 28-29 January 1993. Wind speed, relative humidity and dry-bulb temperature at 5.0 m level at the lidar site were also collected simultaneously. The temporal variations in the aerosol concentration at 50 m altitude and concurrent meteorological parameters are plotted in Figures 6 and 7. It is evident from the figures that there exists a close correspondence between the variations in aerosol concentration and meteorological parameters in such a way that the aerosol concentration increases when there is increase in wind speed and temperature. In the case of relative humidity the relationship was found to be relatively complex. Generally, as the relative humidity increases, water vapour condenses onto the particulates suspended in the atmosphere and thereby one can expect increase in scattering intensity. Such correlations between the nocturnal atmospheric structure derived from the lidar aerosol profile measurements and aerometric observations have been reported in the literature (GOROCH et al., 1980; RENAUT and CAPTINI, 1984; SASANO, 1985; COOPER and EICHINGER, 1994). But as reported by NILSSON (1979), this is true only when the relative humidity values are high (more than 80%). The relative humidity showed variation from 37% to 75% on 23-24 December 1992 and from 27% to 62% on 28-29 January 1993; but in opposite direction i.e. from high value to low on 23-24 December and vice versa on 28-29 January 1993. This could be the reason as to why the variations in aerosol number density and relative humidity on these nights did not show any significant relationship. In addition, the low concentrations of aerosols during these months over this station may be due to the weak generation mechanisms. It also appears from the figures that the aerosol number density is lagging behind the wind velocity. This may happen because the wind measurements were made at about 5 m level whereas aerosol measurements correspond to 50 m height. Furthermore, the aerosol particles which are lifted during daytime due to convective activity would be suspended for a considerable amount of time in the lowest layers of the atmosphere during nighttime because of low fall velocities of the particles for sizes considered in the present study. This could be the reason for the observed association between aerosol concentration and wind even when the magnitude and variation in the latter is low enough to carry the particles from 5 m to 50 m altitudes.

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