The Asian region, comprising of China, India and other East-Asian countries is a rapidly developing region with increasing levels of pollution caused by industries, vehicles and other anthropogenic activities [Menon et al., 2002; Akimoto, 2003]. Our present study is largely motivated by the rising concern of the global scientific community during post-INDOEX (Indian Ocean Experiment) period to know more about aerosols over the South Asian region in greater detail, as some of the earlier studies have shown that aerosols over this region can even slow down the hydrological cycle thereby resulting in severe consequences not only for this region but can produce global impacts [Ramanathan et al., 2001a, 2001b]. This chapter presents results on various physical and optical parameters of aerosols continuously measured over Ahmedabad, an urban location in western India, from early 2002 till the end of 2005 and discusses their seasonal and inter-annual variabilities which has implications for aerosol radiative forcing over the region. Important aerosol parameters that have been studied over this location include: aerosol optical depth (AOD) spectra, aerosol mass concentration, size distribution, BC mass concentration, wavelength dependency in aerosol absorption, aerosol scattering coefficient, single scattering albedo and aerosol vertical distribution. All measured parameters have been classified in terms of four major seasons prevailing over the region, viz. Dry, Pre-Monsoon, Monsoon and Post-Monsoon season. For the first time continuous measurements of such large number of aerosol parameters are being presented from Ahmedabad and the results presented are of
special importance for all researchers interested in investigating the role of aerosols prevalent over this region on regional scale climate forcing and their possible global impacts.

2.1 Site Description and Meteorology

The measurement site is at the campus of the Physical Research Laboratory [23°03'N, 72°55'E, 50 m amsl], located in the western edge of the Ahmedabad city. The city has a population of about 5.5 million and has several small and large scale industries, mostly located on its eastern and northern outskirts. There is also a rapid increase in the number of vehicles (about 10% per year) in the city which includes buses, cars, two-wheelers (motorbikes, scooters) and three-wheelers (Indian auto-rickshaws) all of which contribute significantly to the production of various kinds of pollutants including aerosols. Moreover, since the city is located in the south-east direction of Thar desert and the entire region being semi-arid, the anthropogenically produced sub-micron aerosols get mixed with the naturally produced coarser mineral dust aerosols over the measurement location.

In the present work, the data are grouped in terms of four major seasons viz. Dry (December to March), Pre-Monsoon (April-May), Monsoon (June-September) and Post-Monsoon (October-November), primarily based on different meteorological conditions prevailing over this site during different months of the year. Figure 2.1 shows the seasonal variation of major meteorological parameters over Ahmedabad during last four years (2002 – 2005). Vertical lines on top of each bar in all the four panels represent ±1σ variation about mean value of the corresponding parameter in that season. The inter-annual variability in most of the meteorological parameters has been much less over the period of our study except the rainfall amount which is different for different years. Also we find that within a year, the pattern of seasonal variation in different meteorological parameters has remained similar in all four years. Average daily mean temperature is minimum, in the range of 22 – 24°C, during Dry season and maximum of about 32°C during Pre-Monsoon season. In general, relative humidity is found to be less than 30% during Dry season while it ranges between 30 and 40% during both Pre-Monsoon and Post-Monsoon seasons. The season which markedly differs from all other seasons in any year is the Monsoon season, spread over four months from June to September, when average relative humidity over this site is more than 70% and surpasses 90% mark quite a few times within this season.
2.1. Site Description and Meteorology

Figure 2.1: Seasonal variations of four important surface meteorological parameters measured over Ahmedabad during the four year period from 2002 to 2005

The Indian summer Monsoon, active during this season is part of a large scale circulation pattern which develops in response to the thermal gradients between the warm Asian continent in the north and cooler Indian Ocean in the south. A strong south westerly flow in the lower troposphere brings a substantial supply of moisture into India which is released as precipitation almost across the entire country. Ahmedabad receives most of the rainfall only during this season (Figure 2.1). In the year 2002, total annual rainfall over Ahmedabad was much lower than the climatological mean, which is about 700 mm, whereas during 2003 and 2004 though the total rainfall was within the climatological mean, its day to day variability was quite large during the Monsoon months. On the other hand, in the
Seasonal and inter-annual variations in aerosol characteristics over Ahmedabad

year 2005, not only the total annual rainfall over Ahmedabad was more than the climatological mean but also it was distributed over the entire Monsoon period from June to September. For semi-arid region like Ahmedabad, amount of rainfall and aerosol loading in the atmosphere are very intricately related to each other. This is because on the one hand, amount and type of aerosols which act as cloud condensation nuclei (CCN) together with the available moisture in the atmosphere decides the amount of rainfall that occurs over a region, while on the other, more frequent rainfall leads to moist soil and more vegetation, which curtails the amount of soil derived dust aerosol loading in the atmosphere and possibly can have impact on subsequent weather pattern. Although, establishing an exact relation between aerosol loading in the atmosphere and rainfall occurring in the region is beyond the scope of the present work, nevertheless, since aerosols are key to the formation of CCN required for rainfall, seasonal and inter-annual variabilities of major aerosol parameters measured over this site will be very useful for future studies on the role of aerosols in monsoon rainfall over the region.

Near surface wind speeds over Ahmedabad are the highest during Pre-Monsoon season (~ 4.8 m/s) which is closely followed by during Monsoon season (~ 4.3 m/s), whereas winds are usually calm during Post-Monsoon season (~ 2.3 m/s) and increases slightly by the end of Dry season (~ 3.3 m/s). Seasonal variation in wind speed remained the same in all four years. However, we noticed a slightly decreasing trend in the maximum wind speed during Pre-Monsoon and Monsoon seasons over the period of our study from 2002 to 2005 (Figure 1). Apart from speed, wind direction can also play a role in determining the type of aerosols present over a location as they help in bringing aerosols from different neighboring regions to the measurement site. For example, figure 2.2 shows the wind speed and direction prevailed over Ahmedabad during all the four seasons for the year 2003. The seasonal wind directions can be considered representative for all the four years. During Dry season, surface winds over Ahmedabad are either north easterly or north westerly, making the lower level air masses reaching our measurement site mostly of continental origin and hence they are rich in land derived dust particles apart from other sub-micron aerosols produced due to anthropogenic activities. Surface wind vectors slowly change their direction to become north westerly or south westerly during Pre-Monsoon and Monsoon seasons. Therefore during these seasons, aerosols over Ahmedabad will have a possible influence of the marine air masses reaching this region from
2.1. Site Description and Meteorology

Arabian Sea in the south. However, even during these seasons, when the surface winds are dominated by south westerly flow from the Arabian sea, air back-trajectory analysis using NOAA HYSPLIT-4 (Hybrid Single-particle Lagrangian Integrated Trajectory) model shows that air parcels reaching over our measurement site at higher heights (> 2 km), could be completely of land origin in the west. Thus, there are high chances of coarser mineral dust aerosols getting transported along these trajectories while they cross through large areas of arid and semiarid regions in northern Africa, west Asia and north western India [Figure 2.6(a)]. During Post-Monsoon season, winds are not only calm but wind directions are also found to be random. Primarily, due to lower wind speed prevailing
Seasonal and inter-annual variations in aerosol characteristics over Ahmedabad during this season, amount of land derived dust aerosols are expected to be less in the atmosphere compared to other seasons of the year.

2.2 Instrumentation and Measurements

Physical and optical parameters of aerosols have been studied in detail by observational means using a variety of techniques. The instruments used in the present study along with a brief description of their measurement techniques and working principles are presented below. As the same instruments have been used for the measurement of aerosol parameters over other study locations, discussions on methodology and working principle of these instruments are not repeated in subsequent chapters.

2.2.1 Sun-Photometer

Measurements of aerosol optical depth (AOD) have been carried out using a hand held multichannel sun-photometer named Microtops-II (Solar Light Co. U.S.A.), capable of measuring AOD at five wavelength bands centered around 0.380, 0.440, 0.500, 0.675 and 0.870 \( \mu \text{m} \) [Morys et al., 2001]. Field of view of each collimator is about 2.5\(^{\circ}\) and band-width of each filter is about 0.01 \( \mu \text{m} \). A second Microtops is also used to measure AOD at 1.02 \( \mu \text{m} \) along with total columnar concentrations of ozone and water vapor in the atmosphere. AOD values are derived from the direct measurement of solar radiance at each channel and using a calibration constant corresponding to that channel. Derivation of AOD is based on Beer-Lambert-Bouguer law as follows:

\[
\tau_{\lambda} = \frac{-1}{m} \left[ \ln \left( \frac{I_{\lambda}}{I_{0\lambda}} \right) - 2 \ln \left( \frac{r_0}{r} \right) \right].
\]  

(2.1)

where \( \tau_{\lambda} \) is the total columnar optical depth of the atmosphere at wavelength \( \lambda \), \( I_{\lambda} \) is the instantaneous solar radiation intensity recorded by the Microtops and \( I_{0\lambda} \) is the solar radiation intensity reaching the top of the atmosphere derived using Langley plot technique, \( m \) is the relative air mass, \( r \) is the instantaneous sun-earth distance and \( r_0 \) is the sun-earth distance when \( I_{0\lambda} \) is evaluated. Young, [1994] gave an empirical relation for the relative air mass as follows:

\[
m = \frac{1.002432 \cos^2 z + 0.148386 \cos z + 0.0096467}{\cos^3 z + 0.149864 \cos^2 z + 0.0102963 \cos z + 0.000303978}
\]  

(2.2)
where $z$ is the Solar zenith angle at the time of measurement. This expression takes care of atmospheric refraction and Earth's curvature effect to provide better accuracy at all zenith angles. However, for smaller zenith angles (< 60°), equation 2.2 simplifies into $m = \sec z$.

From the total optical depth, aerosol optical depth $\tau_a$ is obtained by subtracting the contribution due to Rayleigh scattering ($\tau_{RA}$) and molecular absorption ($\tau_{MA}$).

$$\tau_\lambda = \tau_a + \tau_{RA} + \tau_{MA}$$  

(2.3)

Air column density of $2.16 \times 10^{25}$ molecules $cm^{-2}$ applicable for tropical atmosphere has been used for the estimation of $\tau_{RA}$. Molecular absorption has been taken into account for the air molecules $O_3$, $H_2O$ as well as $O_2$ and $N_2$ based on their concentrations valid for the tropical atmosphere. Regular calibration of this instrument is important [Ichoku et al., 2002], which otherwise could lead to erroneous values of AOD computed by this user friendly instrument. In the present study, both Microtops have been regularly calibrated at least twice a year and fresh up-to-date values of calibration constants obtained from Langley plot analyses carried out from Mount Abu (26.6°N, 72.7°E), a relatively pristine site at a height of about 1.7 km above the mean sea level, are used for every subsequent period of about six months. Figure 2.3 shows the Langley plot analysis for the 0.38 and 0.87 $\mu m$ channels carried out from Mount Abu on 14 March 2004. Radiance data measured during forenoon (1st session) and afternoon (2nd session) hours of the day are separately fitted with different straight lines to obtain the value of $I_0\alpha$.

**Figure 2.3:** Langley plots for the (a) 0.38 and (b) 0.87 $\mu m$ channels of Microtops carried out from Mount Abu on 14 March 2004. Radiance data measured during forenoon (1st session) and afternoon (2nd session) hours of the day are separately fitted with different straight lines to obtain the value of $I_0\alpha$. 
forenoon and afternoon hours of the day are separately fitted with different straight lines to obtain the value of $I_{0\lambda}$ (equation 2.1) for all the channels of Microtops. Table 2.1 lists the calibration constants ($I_{0\lambda}$) for the 6 AOD channels of Microtops obtained from separate Langley plot analysis carried out during the period 2002 – 2005. Also, most of the time, the measurements are repeated for three times and the one with lowest AOD is considered for further analysis. The absolute uncertainty in the AOD values are less than 0.03 at all wavelengths.

### 2.2.2 QCM cascade impactor

Aerosol mass concentrations are measured using a Quartz Crystal Microbalance (QCM) cascade impactor (model PC-2, California Measurements Inc. U.S.A.) [Ganguly et al., 2005a]. It draws the ambient air at a constant flow rate of $0.24 \text{ l min}^{-1}$ and segregates aerosols present according to their aerodynamic diameters into ten different size ranges viz. 25, 12.5, 6.4, 3.2, 1.6, 0.8, 0.4, 0.2, 0.1, 0.05 $\mu m$ from stages 1 to 10 respectively. Typical sampling duration is kept as 5 minutes. In this instrument, aerosol concentration is determined from the relative change in frequency between a pair of sampling and reference quartz crystal sensors employed for each stage. Filtered air is flushed through the instrument prior to actual measurement to facilitate the crystals to attain stability. During actual measurements, relative frequency between the sampling and reference crystal are recorded, thereby ensuring that the changes in air temperature and relative humidity does not affect our measurement. Jayaraman et al. [1998] and Ramachandran and Jayaraman [2002] have estimated a maximum overall uncertainty of about 25% in the mass measurement.
for all stages of QCM. Higher chances of error in the mass measurements by QCM occurs under high relative humidity conditions during Monsoon season. This is because, during the measurement time of about 5 to 10 minutes, there could be an evaporation loss of the adsorbed water from the water soluble particles which are being collected under low pressure condition inside the impactor stages [Jayaraman et al., 1998]. Aerosol mass concentrations obtained at different size ranges are then used to retrieve aerosol number size distributions using appropriate mass density applicable for an urban environment [Hess et al., 1998].

2.2.3 Aethalometer

Black carbon mass concentration and absorption coefficient of aerosols ($\beta_{abs}$) are obtained using a seven channel Aethalometer (model AE-42, Magee Scientific, USA). This instrument measures attenuation of light beam at seven different wavelengths viz. 0.37, 0.47, 0.52, 0.59, 0.66, 0.88 and 0.95 $\mu m$, transmitted through the aerosols deposited continuously on a quartz fiber filter [Hansen et al., 1982, 1984] that acts as a perfect diffuse scattering matrix with light absorbing particles embedded in it. The difference in light transmission through the particle-laden sample spot and a particle free reference spot of the filter is attributed to the absorption caused by the aerosol. The attenuation data are converted into BC mass loading using a set of wavelength dependent calibration factors [see http://mageesci.com/Aethalometer_book22009.pdf]. The instrument was operated at a flow rate of 3 $l\ min^{-1}$ and with a data averaging time set to 5 minutes. 0.88 $\mu m$ channel is considered as the standard channel for BC measurement because at this wavelength, BC is the principle absorber of light and other aerosol components have negligible absorption. Data obtained from other channels can be considered as equivalent BC mass that will produce the same absorption. If the absorbing component of the aerosols being sampled consists only of black carbon then the BC values obtained from all the channels of Aethalometer would be identical.

Absorption coefficient of the aerosols at all the seven channels of Aethalometer are calculated using the following relation [Bodhaine, 1995, Weingartner et al., 2003; Ganguly et al., 2005b]

$$\beta_{abs}(\lambda) = -\frac{1}{C.R} \frac{A.ln(I_2/I_1)}{Q.A \Delta t}$$  \hspace{1cm} (2.4)

where $I_1$ and $I_2$ are the ratios of the intensities recorded by the detector for the sensing
beam to the reference beam before and after each sampling time interval $\Delta t$. $Q$ is the volumetric flow rate of air sampled through the filter during interval $\Delta t$ and $A$ is the area of the spot where aerosols are collected. $C$ is a correction factor applied to account for any change in absorption occurring in the filter embedded aerosols over that of the airborne particles while $R$ is an empirical correction factor describing the change in instrumental response with increased particle loading on the filter. Finally Aethalometer calculates the aerosol black carbon concentration [Gundel et al., 1984, Bodhaine, 1995, Weingartner et al., 2003] using the following relation

$$M_{BC}(\lambda) = \frac{\beta_{abs}(\lambda)}{\sigma_{abs}(\lambda)} = \frac{\beta_{abs}(\lambda).C}{\sigma_{atn}(\lambda)}$$

(2.5)

where $\sigma_{abs}(\lambda)$ and $\sigma_{atn}(\lambda) = \sigma_{abs}(\lambda).C$ are the mass specific absorption and attenuation cross-sections respectively. The values of $\sigma_{atn}(\lambda)$ used are 14625/\(\lambda\) nm m$^2$ g$^{-1}$ [Weingartner et al., 2003]. Knowledge about the correction factors $C$ and $R$ is very important for the determination of the absorption coefficient [Weingartner et al., 2003 and Kirchstetter et al., 2004]. In the present study we have used the wavelength dependent values $C(\lambda)$ obtained from the work of Bodhaine, [1995] and assumed $R$ to be equal to unity. Our calculation shows that the error in the estimation of absorption coefficient due to various instrumental artifacts such as flow rate, filter spot area, detector response is about 1%. However, maximum error can come due to empirical factors $C$ and $R$. Previously Moosmuller et al. [1998] found the difference in absorption coefficient and BC mass from Aethalometer with those from Photoacoustic instrument, PSAP, Integrating Plate etc. to be in the range of 2 - 6%.

2.2.4 Nephelometer

Scattering coefficient of aerosols ($\beta_{sea}$) are measured using an Integrating Nephelometer (Model M903, Radiance Research, California, USA) [Ganguly et al., 2005c]. This instrument draws the ambient air through a temperature controlled inlet, which is then illuminated by a flash lamp and the scattered light intensity is measured at 530 nm by a photomultiplier tube. Pressure and temperature measurements are made in the scattering chamber which are further used to calculate the scattering by air molecules and is subtracted from the total scattering to get the scattering by aerosols. Figure 2.4 shows the block diagram of the Nephelometer, model M903 of Radiance Research, USA, used in the present study.
Figure 2.4: Block diagram of the Nephelometer (model M903 Radiance Research, USA) used in the present study for the measurement of aerosol scattering coefficient.

The instrument is regularly calibrated in laboratory according to the procedure described by Charlson et al. [1968], in which the Nephelometer is adjusted to read zero by passing particle free air and span calibration is done by passing the gases of high scattering coefficient such as CO₂. The instrument has been operated in a continuous mode with data averaging time of 5 minutes.

2.2.5 Aerosol Size Spectrometer

Number size distribution of near surface aerosols is measured using an aerosol size spectrometer (model 1.108, Grimm aerosol technik, Germany) giving particle count for all aerosols in the size range of 0.3 to 20 μm by grouping them into 14 different size bins [see http://www.grimm-aerosol.com/index.htm]. This instrument constantly draws the ambient air using a volumetric pump at a flow rate of 1.2 l min⁻¹, passes them through a laser beam and records the scattered signal from particles at an angle of approximately 90°. Strength of each recorded signal carries information on particle size while number of such signals above a set threshold is proportional to the number of particles in that size range. Continuous data are recorded by this instrument at 5 minute interval.
2.2.6 Micro Pulse Lidar

We have used a co-axial mono-static Micro Pulse Lidar (MPL) system (SES Inc., USA) to get the vertical distribution of aerosols in the atmosphere and retrieve their extinction profiles. Lidar is based on active remote sensing principle in which backscattering of laser radiation is used to retrieve information on the structure and composition of the atmosphere. Usually a Lidar employs a pulsed laser as light source and the time delay between the transmitted pulse and the backscattered signal is used to derive information about the range of the scatterer from the lidar system.

Micro Pulse Lidar used in the present study uses an AlGaAs diode laser pumped Nd:YLF laser to get a frequency doubled output at 523.5 nm. This Lidar can be operated with a laser pulse repetition frequency varying anywhere from 1 Hz to 10000 Hz and pulse duration of 10 nS. However, in order to optimize the power associated with individual laser pulses and the signal to noise ratio, we have operated the MPL system with a pulse repetition frequency of 2500 Hz. High pulse repetition frequency is advantageous for averaging large number of low energy pulses in shorter time as it helps in achieving high signal to noise ratio [Welton et al., 2000]. Pulse repetition frequency decides the maximum range detection limit for the Lidar system according to the relation

\[ R_{\text{max}} = \frac{c}{2 \times PRF} \]  

(2.6)

where \( c \) is the velocity of light and \( PRF \) is the pulse repetition frequency of the laser. The output energy for each pulse is in the range of 2 – 4 \( \mu \)J. The MPL uses a common beam expansion configuration with a Schmidt-Cassegrain telescope serving as both transmitter of the outgoing laser beam as well as receiver of the backscattered photons. Diameter of the telescope is 20 cm and it has a field of view of about 50 \( \mu \)rad. In order to reduce the background noise, photons received by the optical transceiver are passed through an interference filter whose peak transmission is centered around 523.5 nm and a FWHM (Full Width at Half Maxima) of 0.1 nm. The MPL system uses a Si avalanche diode (Si-APD) based photon counting detector. The photon counting detector generates electrical pulses according to the photon numbers reaching the active element of the detector with a quantum efficiency of about 45% at 523.5 nm. The electrical pulses are counted by a Multi Channel Scaler (MCS) as time gated signal. Synchronization between multi channel scaler and the outgoing laser pulse is achieved by triggering the MCS with the laser pulse itself.
2.2. Instrumentation and Measurements

Currently allowed bin widths for our MPL system are 200, 500, 1000 and 2000 nS which correspond to the spatial resolution of 30, 75, 150 and 300 m respectively. In the present study MPL was operated with a bin width of 200 ns, corresponding to a range resolution of 30 m. Bin width of Lidar corresponds to the time for which backscattered signals are counted and recorded in a buffer of the multi channel scaler unit. In the present configuration of MPL, the first bin records the electrical pulses generated by photons scattered from the first 30 m of the atmosphere. Second bin holds the number of pulses generated by the scattered photons from the atmosphere between 30 and 60 m, third bin holds the number of pulses for 60 to 90 m and so on. For all practical purposes, each bin is designated by the central value of the corresponding range. Lidar signals are averaged and stored at 1 minute interval during data collection, which are averaged for longer periods of 5 minutes to one hour at the time of data analysis. Figure 2.5 shows the Micro Pulse Lidar system fitted inside the Mobile Lidar Observatory of Physical Research Laboratory and also a block diagram of MPL system. For the above geometry of the MPL, raw signal intensity can be

\[ P(r) = \frac{CO(r)E\beta(r)T^2(r)}{r^2} + N_b + A(r) \] (2.7)

In the above equation \( P(r) \) represents the instantaneous signal intensity recorded by the
system, arriving from an altitude \( r \). \( C \) is the MPL system constant which depends on the transmitter and receiver efficiencies and is a function of the MPL optics, \( O(r) \) is the overlap correction factor (discussed later) and \( E \) is proportional to the output energy of the laser pulse. \( \beta(r) \) is the volume backscattering coefficient of the atmosphere at laser wavelength and at range \( r \). This gives the fractional amount of the incident energy scattered per steradian in the backward direction per unit atmospheric path length and has a unit of \( m^{-1}Sr^{-1} \). \( N_b \) is the background noise signal, primarily due to sunlight during daytime, at laser wavelength (523.5 nm in the present case). \( A(r) \) is the after-pulse correction signal (discussed later). \( T(r) \) is the transmission of atmosphere from Lidar to range \( r \) and is given by

\[
T(r) = \exp\left\{ - \int_0^r \sigma(r')dr' \right\}
\]

where \( \sigma(r') \) is the extinction coefficient of the atmosphere and has a unit of \( m^{-1} \). Replacing \( T(r) \) of equation 2.7 by the above expression, equation 2.7 takes the following form

\[
P(r) = \frac{CO(r)E\beta(r)\exp\left\{ - 2 \int_0^r \sigma(r')dr' \right\}}{r^2} + N_b + A(r)
\]

In this equation, the terms \( \beta \) and \( \sigma \) contain the required information of the atmospheric properties in which we are interested. However, \( \beta \) and \( \sigma \) values depend on a number of interacting physical processes as both these terms represent the sum of contribution from air molecules and aerosols. Also, it can be seen from equation 2.9 that the raw signals obtained from the Lidar need to be subjected through several correction procedures such as dead time correction, after pulse correction, overlap correction, background correction etc before proceeding any further towards the evaluation of extinction coefficient of aerosols [Campbell et al., 2002; Welton et al., 2002]. Some of the correction factors applied to the raw signal from MPL are described below, however, the actual procedures followed to make the signals free from any instrumental artifact are described in Gadhavi, [2005].

**Detector dead-time correction**: This correction is needed because the silicon avalanche photodiode detectors do not respond perfectly linearly under high incident rate of photons. Incident rate of photons being high for the first few bins, detector underestimates the actual count rate of photons for these bins. Usually the manufacturer supplies a correction table relating the actual incident count rate and the detected count rate, from which the dead-time correction factor can be calculated for all the bins of MPL.
2.2. Instrumentation and Measurements

**After-pulse correction:** This correction is needed because the internal reflection of the laser pulse within the MPL system, saturates the Si-APD detector at the beginning of the sampling period and an exponentially decaying leakage signal is recorded along with the actual signal. As the signal from different altitudes takes different times to reach the detector after the laser pulse is fired, after-pulse noise is a function of range. Correction for this after-pulse noise is determined by measuring the normalized signal when the returning photons are completely blocked beyond the first range bin. In this situation, the signal does not contain any return from the atmosphere and the measured signal is completely due to the after-pulse effect. In the after-pulse correction procedure, this beam-blocked after-pulse profile is subtracted from all actual atmospheric profiles at the time of post-processing the raw data. After-pulse noise being dependent on the energy of the laser pulse, the corresponding correction factor is normalized with the laser energy.

**Background subtraction:** When the MPL is operated in daytime, sky background is much stronger than the actual signal. Since the backscattered signal (both Rayleigh and Mie scattering) returning from high altitudes (more than 25 km) are negligible, counts recorded in the bins corresponding to such long distances are mainly due to sky background or to a small extent due to after-pulse effect. Based on this assumption, photon counts measured in the last 5 km (between 25 to 30 km) has been averaged and subtracted from the photon counts measured at all bins.

**Overlap correction:** Overlap correction is needed because due to a small difference existing between the field of view of the transmitter and receiver (detector) in MPL, there is an overspilling effect of the focused image of backscattered photons from the near vicinity of the detector which could be up to 5 km. At near-range, closer than about 3-5 km, the spot size of the outgoing laser pulse yields an image larger than the detector field of view, which results in an under-representation of near-range signal. In order to correct for this near-field instrumental artifact, MPL is operated with the beam oriented horizontally. Under appropriate well-mixed atmospheric conditions, the Lidar profile is expected to display an exponential dependence on range and show a linear relationship with it when plotted on a semi-log plot. The measured profile shows a straight line behaviour at far range, but due to imperfect overlap, it shows a kind of roll-off tending towards zero signal at near range. The overlap correction factor is evaluated for each bin in the near range by taking the ratio of expected signal strength obtained by extrapolating the straight line behaviour from far
range to near range bins and the measured signal for the corresponding bins.

After incorporating all the correction factors listed above, Lidar equation 2.9 takes the following form:

\[ r^2 \hat{P}(r) = \hat{C} \beta(r) \exp \left\{ -2 \int_0^r \sigma(r') \, dr' \right\} \]  \hspace{1cm} (2.10)

where \( \hat{P} \) is the Lidar count at range \( r \) and \( \hat{C} \) is called as the calibration constant of the Lidar. The above equation is made system independent by taking a relative calibration approach and the same equation 2.10 takes the following form:

\[ S - S_0 = \ln \frac{\beta}{\beta_0} - 2 \int_0^r \sigma(r') \, dr' \]  \hspace{1cm} (2.11)

where \( S = \ln[r^2 \hat{P}(r)] \), \( S_0 = S(r_0) \), and \( \beta_0 = \beta(r_0) \) and \( r_0 \) is a reference distance. Above equation can be written in the form of a differential equation of the following form:

\[ \frac{dS}{dr} = \frac{1}{\beta} \frac{d \beta}{dr} - 2 \sigma \]  \hspace{1cm} (2.12)

There are two unknowns in the above equation viz, \( \beta \) and \( \sigma \) and a solution of the above equation is possible only if we have a relation between these two parameters. For all practical purposes, a linear relationship of the following form is assumed between these two quantities

\[ \sigma = L \beta \]  \hspace{1cm} (2.13)

where \( L \) is known as Lidar ratio. Klett, 1985 gave a solution of equation 2.12 for \( r < r_m \) of the following form

\[ \sigma(r) = \frac{\exp[S - S_m]}{\left\{ \sigma_m^{-1} + 2 \int_r^{r_m} \exp[S - S_m] \, dr' \right\}} \]  \hspace{1cm} (2.14)

Extinction values obtained using the above relation contains contribution due to both air molecules and aerosols \( \sigma(r) = \sigma_M(r) + \sigma_R(r) \), where suffix \( M \) and \( R \) stand for contribution due to aerosols and air molecules respectively. However, extinction due to aerosols can be calculated by subtracting the extinction values due to air molecules derived using tables of McClatchey et al. [1972] for standard tropical atmosphere from \( \sigma(r) \) of equation 2.14.

In the present study, extinction coefficient due to aerosols is determined using a solution of equation 2.10 for the range \( r < r_m \) of the following form

\[ \sigma_M(r) = -L_M(r) \beta_R(r) + \frac{Z(r)}{N(r)} \]  \hspace{1cm} (2.15)
2.3. Results and Discussion

with

\[
Z(r) = L_M(r) r^2 P(r) \exp\left\{2 \int_r^{r_m} \left[ L_M(r') - L_R \right] \beta_R(r') \, dr' \right\}
\]

(2.16)

\[
N(r) = \frac{L_M r_m^2 P_m}{L_M r_m \beta_R + \sigma_M} + 2 \int_r^{r_m} Z(r') \, dr'
\]

(2.17)

where index \( m \) refers to the value of the corresponding quantity at the maximum range \( r_m \). Also, Lidar ratio for the air molecules denoted as \( L_R \) is a constant and is equal to \( 8\pi/3 \). Usually Lidar ratio for aerosols lie in the range from 20 to 80 Sr over different environments [Young et al., 1993; Voss et al., 2001; Welton et al., 2002]. For our study, we have assumed Lidar ratio to be independent of altitude of the aerosol layer.

Meteorological data discussed in the present study are obtained from the meteorological center of Indian Meteorological Department (IMD) at Ahmedabad.

2.3 Results and Discussion

In order to study the characteristics of aerosols present over the urban location Ahmedabad, understand their behavior and estimate their effect on the radiative balance of the Earth-Atmosphere system on a regional scale, continuous measurements of various physical and optical parameters of aerosols are being made since early 2002. In this chapter, we present results on seasonal and inter-annual variabilities of these aerosol parameters studied over Ahmedabad during the period 2002 to 2005. Implications of the observed variabilities in various aerosol parameters are discussed separately in Chapter-6.

2.3.1 Aerosol optical depth

The time series of seasonal variation in AOD at two representative wavelengths viz. 0.38 and 1.02 \( \mu m \), measured over the entire period of our study from 2002 to 2005 are shown in figure 2.6(b). Vertical lines on top of each bar represents \( \pm 1\sigma \) variation about the mean value measured during that season. AOD values corresponding to both these wavelengths are higher for the year 2002 than during 2005, indicating higher total columnar loading of aerosols during 2002 than compared to 2005. Annual patterns of seasonal variability in AOD shows more consistency at longer wavelengths than at smaller wavelengths (0.38 \( \mu m \)). In all four years, AOD at 1.02 \( \mu m \) has been lower and comparable (within \( \pm 1\sigma \) variation) during Dry and Post-Monsoon seasons, while higher values are obtained during
Pre-Monsoon and Monsoon seasons of the same year. Also, from Dry to Pre-Monsoon season, we notice an increase in AOD at all wavelength channels. This happens because of two reasons: first, due to increased wind speed during Pre-Monsoon season, large quantities of soil derived dust aerosol gets lifted in the atmosphere from the dry lands of semi-arid region all around our measurement location. Secondly, boundary layer height or the mixed layer thickness increases from Dry to Pre-Monsoon season, which provides a larger room for all kinds of natural and anthropogenic aerosols to get accommodated into it. Also, AOD values at higher wavelengths are more affected by naturally produced coarser aerosols, while the sub-micron sized aerosols produced mostly due to various anthropogenic activities contribute maximum to AODs at smaller wavelengths. While there is no significant variation occurring in the local production of aerosols from anthropogenic activities such as fossil fuel combustion or industrial emissions, there is always an increase of dust aerosols in the atmosphere from Dry to Pre-Monsoon season. Thus both these factors cause AOD values at all wavelength channels to increase over the first half of the year, the trend being much stronger and consistent at higher wavelengths. Further, AODs at almost all wavelength channels continue to maintain higher values during Monsoon season because not only winds, which are capable of lifting dust aerosols remain high during
this season, but also the higher relative humidity facilitates hygroscopic growth of several water soluble aerosols in the atmosphere. Although Monsoon season spans from June to September and Ahmedabad gets almost all of its total annual rainfall during this period, it is expected to see a decrease in AOD values due to wet removal of aerosols during rain events. However, this decrease does not seem to be always taking place primarily because even during Monsoon, rainfall in Ahmedabad is not very uniformly distributed over the entire season but occurs in certain spells with large intermittent gaps. Moreover as shown in figure 2.6(a), although the surface level wind flow is predominantly south-westerly, transport of dust aerosols from distant regions of West Asia and Northern Africa to this region continue to occur at higher altitudes during this season. Also, since the rainfall amount as well as its distribution over the Monsoon season has been quite different in different years, we observe different trends in AOD at different wavelengths and in different years. In the year 2002, when total annual rainfall itself was very scarce, AODs at 0.38 and 1.02 \( \mu m \) both show a continuously increasing trend from Dry to Monsoon season. In 2003, when Ahmedabad received more rainfall during Monsoon, AOD at higher wavelengths remained similar to their Pre-Monsoon values, but AOD at 0.38 \( \mu m \) increased further to reach still higher values. This trend is reversed in the year 2004, when AOD at 1.02 \( \mu m \) decreases from Pre-Monsoon to Monsoon season while AOD at 0.38 \( \mu m \) increased during the same period. This observed decrease in AOD at higher wavelengths from Pre-Monsoon to Monsoon period occurred primarily due to decline of dust loading during Monsoon season. As a result of good rainfall occurring this year, surface remained damp and offered sufficient resistance to lifting of soil dust by wind. However, increasing trend in AOD at lower wavelengths suggest that the effect due to increase in boundary layer height dominated over any loss due to wet removal processes in case of smaller aerosols of sub-micron size. But unlike all previous years, not only greater amount of rainfall occurred in the year 2005 but also it was more uniformly distributed over the entire Monsoon period. Due to such frequent spells of rainfall continuing throughout the Monsoon time, level of every aerosol type decreased in the atmosphere, which is evident from lower values of AOD at all wavelength channels measured during intermittent cloud free skies available during Monsoon season of 2005. Total aerosol loading in the atmosphere over any location depends on the differences between production of aerosols from all possible sources (either locally produced or transported to that location by wind) and their sinks (either gravita-
tional settling, rain wash or transported to other locations by wind). This means that even if there is no difference in source strength of aerosols, any weakening of the sink mechanisms can result in pile up of aerosols in the atmosphere which may manifest itself in terms of higher aerosol optical depth. This is what happens during Post-Monsoon season of all the years studied. Due to much weaker wind speeds during Post-Monsoon, dispersal of aerosol becomes much slower and those produced within city are not properly ventilated or get transported to other downwind locations. This causes AODs to attain much higher values during Post-Monsoon compared to following Dry season.

Although a time series of AOD at any single wavelength helps to study changes in total columnar aerosol loading, comparison of spectral AOD measured during different seasons can provide further insight on relative changes in columnar aerosol size distribution occurring at different times of a year. Figure 2.7(a) shows a comparison of mean AOD spectrum for different seasons, averaged over four years. Vertical lines in the figure represent ±1 σ about the mean AOD value for a particular season and corresponding to a particular wavelength channel. Another important parameter estimated from multi-spectral measurement of aerosol optical depths is Angstrom wavelength exponent α, which is the slope of a plot between logarithm of AOD versus logarithm of wavelength in micron units. Angstrom parameter α is useful to compare and characterize the wavelength dependence of AOD and columnar aerosol size distribution [Eck et al., 1999; Cachorro et al., 2001]. For example, a relative increase in the number of larger sized particles with respect to the smaller ones result in a decrease in the value of α and vice-versa. However, α estimated from sun-photometer measured AOD depends on the wavelength pair used for the computation. In a separate study, Redd et al. [1999] have shown that α computed using shorter wavelength pair is more sensitive to changes in the amount of nucleation and accumulation mode sized particles than when estimated using longer wavelength pairs. In order to know the dominating factor which causes α to change from one season to another, we have computed this parameter for different wavelength intervals. Figure 2.7(b) shows the seasonal variation of α computed for three different wavelength intervals, each one of which is averaged over four years of AOD data from 2002 to 2005. Vertical lines on top of each bar represents ±1 σ variation about the mean value for that season during the entire period of study. First, Angstrom parameter α is calculated using AOD values for all the channels (from 0.38 to 1.02 μm) and is also computed for two different wavelength pairs viz. 0.38 – 0.5 and
2.3. Results and Discussion

0.5 – 1.02 µm. In the present study, we find that α value computed using the complete spectrum of measured AOD are almost same when computed for the longer wavelength pair of 0.5 – 1.02 µm in all seasons, except during Dry and Post-Monsoon seasons, when α value derived for the shorter wavelength pair is found to be smaller than its corresponding value derived for larger wavelengths. Nevertheless, the pattern of seasonal variation of α computed using all the three wavelength pairs are similar.

![Graph showing comparison of mean AOD spectrum for different seasons and seasonal variation of Angstrom parameter α computed for three different wavelength intervals.](image)

**Figure 2.7:** (a) Comparison of mean AOD spectrum for different seasons, averaged over four years of data available from 2002 to 2005. (b) Seasonal variation of Angstrom parameter α computed for three different wavelength intervals, each one of which is averaged over four years from 2002 to 2005.

Lower AODs for the entire spectrum obtained during Dry season suggest that total aerosol loading in the atmosphere is usually low during this season compared to all other seasons. When Dry season changes to Pre-Monsoon, we notice an increase in AOD values at all wavelength channels. However, importantly, this increase is found to be spectrally non-uniform as percentage increase in AOD at 1.02 µm during this seasonal transition is more than 100% while the corresponding increase in the case of AOD at 0.38 µm is merely 17%. On the other hand, we find a large drop in α values over the same period. As mentioned earlier, decrease in α can occur either due to a relative decrease in number of smaller sized particles with respect to larger ones or due to an increase in coarser particles with respect to smaller ones. It can be seen from figure 2.7(b) that α values computed using the longer wavelength pair, which is more sensitive to changes in the amount of coarse particles, exhibit maximum decrease while changing from Dry to Pre-Monsoon season.
Also, since AODs at higher wavelengths are more sensitive to changes in coarser particles while those at smaller wavelengths are susceptible to changes in smaller sized particles, this suggests that an increase in the amount of coarser particles (mostly super-micron) relative to other smaller (mostly sub-micron) aerosols is the dominating factor responsible for the observed non-uniform shift in AOD spectrum during this change of season from Dry to Pre-Monsoon. No significant change in the average AOD spectrum is observed from Pre-Monsoon to Monsoon season except some marginal increase in the AOD values at wavelengths shorter than 0.625 \( \mu m \), with a marginal increase in the \( \alpha \) value estimated using all the wavelength pairs. These observations suggest that there is an increase in the total amount of sub-micron aerosols in the atmosphere from Pre-Monsoon to Monsoon season. It will be shown later using lidar data that the observed increase actually occurs due to increased volume being available to these aerosols to distribute themselves vertically in the atmosphere. During Post-Monsoon season, AOD spectrum takes different shape with further increase in AOD values at wavelengths shorter than 0.5 \( \mu m \) while a large drop is seen in the AOD values at longer wavelengths, with a simultaneous increase in \( \alpha \) values computed using all the three wavelength pairs [figure 2.7(b)]. Once again we find that the percentage increase in \( \alpha \) value derived using the longer wavelength pair is maximum, suggesting that as the season changes from Monsoon to Post-Monsoon period, amount of smaller particles increase and coarser particles decrease in the total atmosphere. The decrease in the amount of coarser particles play a dominating role in shaping the AOD spectrum during Post-Monsoon season. This decrease in the amount of coarser particles is expected as the wind speeds are minimum during this season and also after the monsoon rainfall, the growth of vegetation and grass offers resistance to the production mechanism of wind derived dust aerosols.

2.3.2 Aerosol mass concentration

Aerosol mass concentrations measured separately in 10 different size bins have been further classified into three categories viz. coarse mode particles with size ranging between 1 and 10 \( \mu m \) (total mass collected in stages 2, 3 and 4 of the QCM), accumulation mode particles in the size range of 0.1 to 1 \( \mu m \) (total mass collected in stages 5, 6, 7 and 8) and the nucleation mode particles with size less than 0.1 \( \mu m \) (total mass collected in stages 9 and 10). Aerosol mass obtained from stage 1 of the QCM device is not included in this
classification, as it integrates all particles of size greater than 12.5 \( \mu m \) and no meaningful mean radius could be assigned to this stage.

![Graph showing seasonal variation of near surface aerosol mass concentration](image)

**Figure 2.8**: Seasonal variation of near surface aerosol mass concentration classified into three categories viz. coarse mode, accumulation mode and nucleation mode particles, averaged for three years of data available from 2002 to 2004.

Figure 2.8 shows the seasonal variation of near surface aerosol mass concentrations for all the three size modes and averaged over a period of three years from 2002 to 2004. Measurements of aerosol mass concentration could not be continued during 2005 due to technical problem with the QCM device. Though the problem could be rectified during late 2005 and regular measurements were resumed the data are not considered in the present study. Over the period for which data is available, PM10 (particulate matter of size less than 10 \( \mu m \)) concentration mostly varied from low values close to 40 \( \mu g/m^3 \) to high of about 106 \( \mu g/m^3 \) with an average value of around 66 \( \mu g/m^3 \). Lower values for PM10 mass concentration are observed during Monsoon season. This is contrary to the trend seen in the case of columnar AODs which exhibit higher values during Monsoon season (except for the year 2005). This condition could arise only if changes occur in the vertical distribution of aerosols in such a way that a significant contribution of extinction to the column AOD comes from aerosols present at higher altitudes. In an earlier study con-
Seasonal and inter-annual variations in aerosol characteristics over Ahmedabad

ducted over another tropical Indian station, Gadanki (13.5°N, 79.2°E), Krishnan and Kunhikrishnan, [2004] observed the atmospheric boundary layer height to become maximum during Pre-Monsoon period and minimum during Dry months. Over Ahmedabad we find a summertime increase in atmospheric boundary layer, as a result of which, a larger volume becomes available for the aerosols to get distributed in the atmosphere during this period. Because of this increased boundary layer height, a larger mixing of aerosols occurs within this layer, which in turn dilutes the aerosol concentration near surface level. It will be shown later in the chapter that using a simultaneously operated Micro Pulse Lidar system we have observed such changes occurring in boundary layer height and aerosols are detected up to much higher heights in summer, contributing significantly to AOD.

In general, nucleation mode aerosols contribute least to the total mass, except during Post-Monsoon season when the contribution by all the modes are found to become almost equal. Also, since most of the nucleation mode aerosols (soot, sulfate etc.) are associated with anthropogenic sources (fossil fuel or biomass/biofuel burning), their concentration is found to increase when the surface level wind flow is observed from the north, north-east or north-west. During Pre-Monsoon and Monsoon seasons, surface level wind flow is from the Arabian sea in south and hence the air masses reaching Ahmedabad at lower levels are relatively less polluted compared to air masses which are of purely continental origin during other seasons. During Dry and Post-Monsoon seasons, aerosols which are either locally produced or transported from other regions are constrained within a shallow boundary layer having smaller ventilation coefficient and this causes their surface level concentration to rise. High values of mass concentration for the nucleation mode aerosols are obtained during Post-Monsoon and Dry season. One of the reasons is that during the months from November to January (winter months) we observe a lot of waste burning activities in the open, such as burning dry leaves and shrubs in various parts of the city. In addition, another important source which contributes significantly to the production of aerosols and precursor gases is burning of wood, paper or other solid wastes in the open by population dwelling in the city slums, to keep themselves warm during cold winter nights. It will be shown later in the chapter that signatures of such anthropogenic activities are very well captured in the data from other two surface based aerosol instruments viz. Aethalometer and Nephelometer.

The accumulation mode aerosols are produced by the condensation growth and co-
agulation of nucleation mode aerosols. In general, we have found accumulation mode aerosols contributing maximum to the total $PM_{10}$ mass. On an average, their mass varied from a low concentration of about 22 $\mu g/m^3$ recorded during Monsoon or Post-Monsoon seasons to a high concentration of about 33 $\mu g/m^3$ during Dry season. Over the period of our study, accumulation mode aerosols have shown minimum variability in their mass concentration during all seasons and over the entire period of measurement (a maximum of 34% departure from the mean). Except for the nucleation mode, we have seen large inter-annual differences in the seasonal variation of aerosol mass concentration as specific patterns are obtained in different years. The intricacies behind such inter-annual differences in the pattern of seasonal variation of surface level aerosol mass concentration is difficult to explain and we restrict our discussion only to the average pattern of seasonal variation over the entire period of study.

Coarse mode aerosols over land are mainly soil derived dust particles produced by wind blowing over land surfaces, particularly when it is dry and has less vegetation cover. Most of these particles are mineral aerosols made up of materials derived from the Earth's crust and are therefore rich in iron and aluminium oxides and calcium carbonate [Rastogi and Sarin, 2005]. The North Indian Ocean is surrounded by several arid and semi-arid areas which are the major sources of these particles. The largest one being the Rub Al Khali desert in central Saudi Arabia but there are several other sources located in Iran, Pakistan, Afghanistan and north-west India [Leon and Legrand, 2003].

During the period of our study, average value of coarse mode aerosol mass measured over Ahmedabad is around 25 $\mu g/m^3$. This amount varied anywhere from low values close to 12 $\mu g/m^3$ measured during Monsoon season to high values around 40 $\mu g/m^3$ measured during Dry or Pre-Monsoon seasons. Higher values of coarse mode aerosol mass are recorded during Dry season because during this time surface level wind flow over Ahmedabad is mainly from north-west direction (figure 2.2) and there are high possibilities of dust particles being carried to the city from vast areas of arid and semi-arid regions in north-west India including Thar Desert. This transport of dust aerosols from arid and semi-arid regions in the west of Ahmedabad continues even during other seasons of the year but those occurring at higher levels [figure 2.6(a)] are not detected in surface measurements. Near surface values of coarse mode aerosol mass decreases during Monsoon season because during this time surface level wind flow is from the Arabian Sea in south
and south-west of Ahmedabad and therefore it is depleted in dust content but could bring in some amount of sea salt aerosols. Also, during Monsoon season, occasional spells of rain keeps the surface damp and availability of loose soil to be lifted by the wind becomes less. Most of the dust aerosols measured during Monsoon season are locally produced and higher values of coarse mode aerosol mass are measured in the years when rainfall amount is less and soil remains dry. Lower values of coarse mode aerosol mass are also measured during Post-Monsoon season because of two reasons, firstly lower wind speed during this season is not very efficient in lifting dust from the surface and secondly rainfall occurring during previous Monsoon increases grass and other vegetation cover over the surface, which offers resistance to lifting of soil dust by wind.

2.3.3 Aerosol number size distribution

QCM observations have been further used to obtain the number size distribution for the near surface aerosols. Mass concentrations measured in the 10 size ranges are used to derive the aerosol number distribution using appropriate mass density valid for the urban atmosphere and prevailing relative humidity conditions [d'Almeida, 1991; Hess et al., 1998]. We have observed maximum seasonal variation in the number concentration data corresponding to nucleation mode aerosols, with nearly 60% spread in its value about the mean for the entire study period. This is followed by accumulation and coarse mode data showing about 26 and 17% spread in their values respectively. The size distribution of aerosols at any location is mainly governed by the relative strengths of different production and removal mechanisms [Jaenicke, 1993]. Several authors [Porter and Clarke, 1997; Bates et al., 1998; Koponen et al., 2003; Ramachandran and Jayaraman, 2002; Ganguly et al., 2005a] have shown that the size distribution of tropospheric aerosols is a combination of many log-normal distributions, where different modes represent different production sources. Figure 2.9 shows the aerosol number size distribution for two typical seasons of the study period with vertical bars in the figure representing ±1 σ variation about the average number concentration for different size bins. All size distribution plots obtained during the study period exhibited presence of three distinct modes and hence they could be fitted using three log-normal modes of the form:

\[
\frac{dn(r)}{d\log r} = \frac{N}{\sqrt{2\pi}\log\sigma} \exp \left[ -\frac{\log^2 \left( \frac{r}{r_m} \right)}{2(\log\sigma)^2} \right]
\]

(2.18)
2.3. Results and Discussion

Figure 2.9: Aerosol number size distribution for two typical seasons of the study period with vertical bars in the figure representing ±1σ variation about the average number concentration for different size bins.

where N is the number concentration (cm$^{-3}$), σ is the width of the distribution and $r_m$ is the mode radius for a particular mode. We find that the shape of the aerosol size distribution remained same over the study period while number concentration changed in all the three modes during different seasons of different years. This indicates that the amount of aerosols as well as their in situ production strengths changed while various physical processes responsible for their distribution in the atmosphere remained unaltered. Table 2.2 gives the average values of various modal parameters obtained by log-normal curve fitting carried out on the measured distribution. Mode radii of the distribution corresponding to mode—1 lie in the range of 0.009 — 0.018 μm and the number concentration values for this mode are found to be highest during Pre-Monsoon season while low values are obtained in Monsoon period. Similarly, mode radii of the distribution corresponding to mode—2 and mode—3 lie in the range of 0.08 — 0.18 μm and 2.2 — 3.8 μm respectively. Comparing the modal parameters of Table 1 with those of various aerosol types classified by Hess et al. [1998], we find that water soluble aerosols and soot particles constitute mode—1 aerosols while the $r_m$ value of mode—2 matches with those of sulfate aerosols or some of the accumulation mode mineral aerosols. These sulfate aerosols are mostly produced by chemical reactions in the atmosphere by gaseous precursors, mainly sulfur dioxide (SO$_2$) emitted from various anthropogenic sources [Charlson et al., 1992]. Finally, possible aerosol types
Seasonal and inter-annual variations in aerosol characteristics over Ahmedabad

constituting mode—3 of the measured size distribution are either insoluble particles such as soil dusts getting transported from arid and semi-arid regions of surrounding areas or coarser mineral aerosols of desert origin. It can be seen from table 2.2 that there is an increase in the number concentration of particles in mode—3 during Post-Monsoon season, which is contrary to the trend seen in the case of coarse mode aerosols [figure 2.8]. However, the seasonal variation of mode radii corresponding to mode—3 shows a maximum during Dry and Pre-Monsoon seasons and minimum during Monsoon and Post-Monsoon periods. Although number concentration of mode—3 aerosols is less during Dry and Pre-Monsoon compared to Monsoon and Post-Monsoon seasons, it contributes maximum towards the mass concentration of coarse mode aerosols due to their larger mode radii during Dry and Pre-Monsoon seasons. One possible reason for getting larger mode radii for mode—3 aerosols during Dry and Pre-Monsoon seasons is that during these seasons, surface level winds coming from north-west direction of Ahmedabad can bring in coarser desert dust particles to the measurement location (figure 2.2). On the other hand particles constituting mode—3 aerosols during Monsoon and Pre-Monsoon seasons being locally produced soil dust, their mode radii are found to be less compared to other desert dust mineral aerosols prevalent during other seasons.

Table 2.2: Average values of size distribution parameters obtained by fitting log-normal curves to the measured aerosol number size distribution over Ahmedabad

<table>
<thead>
<tr>
<th>Season</th>
<th>Mode-1</th>
<th>Mode-2</th>
<th>Mode-3</th>
</tr>
</thead>
<tbody>
<tr>
<td>N (cm⁻³)</td>
<td>σ (μm)</td>
<td>r_m (μm)</td>
<td>N</td>
</tr>
<tr>
<td>Dry</td>
<td>410000</td>
<td>1.95</td>
<td>0.009</td>
</tr>
<tr>
<td>PrM</td>
<td>230000</td>
<td>2.03</td>
<td>0.009</td>
</tr>
<tr>
<td>Mon</td>
<td>60000</td>
<td>1.81</td>
<td>0.017</td>
</tr>
<tr>
<td>PoM</td>
<td>380000</td>
<td>1.89</td>
<td>0.012</td>
</tr>
</tbody>
</table>

2.3.4 Black Carbon mass concentration

In the present day global scenario, one of the crucial climate forcing agents is black carbon (BC) aerosols, mostly produced from the incomplete combustion of fossil fuels such as coal, diesel, petrol etc. as well as due to usage of biofuel and biomass burning across various parts of the world [Novakov et al., 2000; Jacobson, 2001; Venkataraman et al., 2005]. Although BC aerosols have been recognized as crucial climate forcing agents contributing
2.3. Results and Discussion

Figure 2.10: Seasonal variation of (a) BC mass concentration, (b) aerosol scattering coefficient at 0.53 $\mu$m and (c) single scattering albedo at 0.525 $\mu$m, averaged over all data available from 2003 to 2005. Vertical lines on top of each bar represent $\pm 1 \sigma$ variation about the mean value for the corresponding parameter in that season.

significantly to the present day global warming [Jacobson, 2002], they occupy special importance over populous regions of the world like India and China because of their large local production and possible impact on regional climate [Ramanathan et al., 2001; Menon et al., 2002]. Jacobson [2002] has shown that large positive radiative forcing produced by BC aerosols makes it second most important agent for global warming after $CO_2$ in terms of direct radiative forcing estimates. Total BC emissions from India including all sources such as fossil fuel, open burning and biofuel combustion is around $610 \pm 200 \ Gg \ year^{-1}$ [Venkataraman et al., 2005]. This constitutes a large fraction of total global emission and
therefore demands regular monitoring of BC aerosols across various parts of India to know their radiative impacts not only on a regional scale but also to study their global climatic effects.

BC measurements over Ahmedabad started during Monsoon 2003 and since then almost continuous data are available over this location till date. Figure 2.10(a) shows the seasonal variation of BC mass concentration averaged over 2003 to 2005. Vertical lines on top of each bar represent ±1 σ variation about the mean mass concentration available for that season. Observed seasonal variation in BC mass is similar to that observed in the case of nucleation mode aerosols [figures 2.10(a) and 2.8]. This shows that BC aerosols constitute a major fraction of nucleation mode aerosols over Ahmedabad. Highest value of BC mass is obtained during Post-Monsoon season (mean value 7.3 ± 3.7 μg/m³). This amount decreases slightly during Dry season (5.5 ± 2.8 μg/m³) and comparatively much lower values of BC mass are measured during Pre-Monsoon (2.2 ± 1.0 μg/m³) and Monsoon seasons (1.5 ± 0.8 μg/m³). It is also found that not only the absolute values of BC mass concentration are high in Post-Monsoon and Dry seasons, mass fraction of BC in the total aerosol concentration also remains high during these seasons. On an average, we find BC mass fraction over Ahmedabad is around 10% during Post-Monsoon season while this percentage changed to 7, 3 and 3 during Dry, Pre-Monsoon and Monsoon seasons respectively. Previously Babu et al. [2002] have reported the BC concentrations over Bangalore (13°N, 77°E) for the month of November to be in the range of 0.4 – 10.2 μg/m³. Also, from a month long campaign in December 2004, Tripathy et al. [2005] have reported BC concentrations over Kanpur city (23.43°N, 80.33°E) to be in the range of 6 – 20 μg/m³ and BC mass fraction to vary from 7 to 15%. Ganguly et al. [2005b] found the daytime (10 – 16 hrs) average BC mass over another urban location, Hyderabad (17 47° N, 78 45° E) to be around 4.8 μg/m³. Venkataraman et al. [2002] have reported the BC concentrations for Mumbai (19.38° N, 72.83° E) during INDOEX-IFP (Jan-Mar, 1999) as 12.5 ± 5 μg/m³. At another coastal urban station, Trivandrum (8.55°N; 77°E), Babu and Moorthy, [2002] found the mean BC concentration to vary from 5 to 1.5 μg/m³ from Dry to Monsoon seasons. Also, during an intensive field campaign in December 2004, our group found the average BC mass over Delhi to be 29 ± 14 μg/m³ (highest among all above mentioned values) [Ganguly et al., 2006]. Comparing results from the present study and BC measurements conducted over other urban locations in India, we find that BC mass over Ahmedabad is
lower than the two urban cities viz. Delhi and Kanpur located in the Indo-Gangetic belt while it is higher than coastal locations like Trivandrum and comparable with two other urban cities viz. Bangalore and Hyderabad both located in peninsular India.

Figure 2.11: Average patterns in diurnal variation of BC aerosol mass concentration for different seasons of a year, with vertical lines representing ±1σ variation about the mean value of BC at a particular time on different days of a particular season.

High values of BC mass and its mass fraction in the total aerosol concentration measured over Ahmedabad during Post-Monsoon and Dry seasons have serious implications on regional aerosol radiative forcing as these soot particles are strong absorber of both incoming solar radiation as well as outgoing terrestrial infrared radiation, and alter the radiation budget of the Earth by trapping heat within the atmosphere [Jacobson, 2001]. Figure 2.11 shows the average patterns in diurnal variation of BC aerosol for different sea-
sons of the year. These patterns are actually combined outcome of various factors such as production source of aerosols, their source strengths, their removal mechanisms and also the surface meteorology. Vertical lines in all the panels represent \( \pm 1 \sigma \) variation about the mean BC value measured at a particular time on different days of a particular season. Most common feature in the diurnal variation of BC mass during all seasons is the presence of two maxima and two minima in all these plots. During Dry and Post-Monsoon seasons, first maxima of the day is observed in the morning hours, sometime between 08 – 09 hrs, while this peak shifts to earlier period between 07 – 08 hrs during Pre-Monsoon and Monsoon seasons. Second maxima in the diurnal variation of BC mass concentration is recorded in the late evening or nighttime between 20 – 21 hrs. First minima in BC mass concentration is observed in the early morning between 03 – 04 hrs, while second minima corresponding to the lowest concentration of a day occurs in the afternoon, sometime between 15 – 17 hrs depending on the season. Identical patterns appearing in all seasons suggest that the diurnal variation of BC is mainly governed by the diurnal evolution of atmospheric boundary layer which remains low during morning hours then gradually increases and reaches a higher value at noontime and starts decreasing in the evening [Krishnan and Kunhikrishnan, 2004]. This cycle is repeated on all clear sky days throughout any year. However, seasonal variations in absolute magnitude of BC largely occurs due to differences in the extent of contraction and expansion of the atmospheric boundary layer primarily due to differential Solar heating of the Earth’s surface as well as differences in production sources and their source strengths in different seasons. Increased production of BC aerosols and gradual formation of a surface based inversion opposing vertical mixing in the atmosphere cause BC concentrations to escalate from around 16 hrs and reach maxima in the evening after 20 hrs. One important reason for the increased production of BC aerosols in the evening time is related to road traffic and open burning of solid wastes such as dry leaves and other garbage materials, particularly during Dry and Post-Monsoon seasons. In addition to waste burning, wood and shrubs are also burnt at night by several people to keep themselves warm during cold winter months. Maxima in BC concentrations between 07 – 09 hrs is primarily due to morning peak in traffic level and several small scale industrial activities spread around the city. After around 09 hrs, BC concentrations start decreasing as some of the sources are not very active during this time while boundary layer height of the atmosphere start rising with increased insolation of
2.3. Results and Discussion

The Earth’s surface which kicks off convective mixing of air capable of lifting aerosols to higher levels in the atmosphere and causing a dilution of their concentration at the surface level [Krishnan and Kunhikrishnan, 2004]. BC values reach lowest concentrations between 15 – 17 hrs due to minima in traffic levels in the afternoon hours and higher boundary layer height. After around 20 hrs, the city traffic decreases by a large extent, making one of the main production sources of BC over this urban location weaker while removal of particles from the atmosphere by gravitational settling process continues, which give rise to a minima in BC concentration during early morning between 03 – 04 hrs.

2.3.5 Aerosol absorption coefficient

Although measurements of BC concentration itself is important over populous parts of the world to assess the air quality, but the parameter required to calculate the impact of light absorbing particles on the regional and global climate forcing is the absorption coefficient $(\beta_{abs})$ of these aerosols. In the present study, absorption coefficient of aerosols are calculated using the raw absorbance data recorded at all the seven wavelength channels of Aethalometer viz. 0.37, 0.47, 0.52, 0.59, 0.66, 0.88 and 0.95 $\mu$m. Measurements on spectral dependence of aerosol absorption is very useful because it contains some characteristic features of the sources producing these absorbing species [Kirchstetter et al., 2004]. This fact is particularly important over the Indian region where contribution of BC from biomass/biofuel burning is as important as fossil fuel combustion and the same amount of BC from biofuel can exhibit stronger absorption characteristics [Venkataraman et al., 2005; Ganguly et al., 2005b]. Wavelength dependence of absorption by aerosols have been investigated using a power law relationship of the form $\beta_{abs}(\lambda) = K \lambda^{-\alpha}$ where $K$ and $\alpha$ are the absorption Angstrom coefficients and $\alpha$ is a measure of spectral dependence of aerosol absorption [Kirchstetter et al., 2004; Ganguly et al., 2005b]. Figure 2.12 shows the spectra of aerosol absorption coefficient for different seasons, averaged over all available data from 2003 to 2005. Vertical lines in this figure represent $\pm 1 \sigma$ about the mean value $\beta_{abs}$ for a particular season and corresponding to a particular wavelength channel of Aethalometer. As reported earlier, BC mass concentration over Ahmedabad during Post-Monsoon and Dry seasons are higher than those obtained during Pre-Monsoon and Monsoon seasons. Figure 2.12 clearly shows that not only mass concentration of these soot particles but also the absorbing capability of the aerosols in general increases enormously, particularly at
shorter wavelengths, during Post-Monsoon and Dry seasons. In order to quantify these relative changes in absorption characteristics of aerosols, spectra of absorption coefficients for each season are fitted using the power law relation mentioned earlier and \( \alpha \) values have been estimated. We find the average values of absorption Angstrom parameter \( \alpha \) to be 2.2, 2.0, 1.9 and 2.1 for Dry, Pre-Monsoon, Monsoon and Post Monsoon seasons respectively. Although, differences in absolute values of absorption coefficient are not very large in different seasons, large differences occur in the values of \( \beta_{\text{abs}} \) at shorter wavelengths due to exponential relation of \( \beta_{\text{abs}} \) with \( \lambda \). \textit{Kirchstetter et al.} [2004] reported stronger spectral dependence in absorption (\( \sim \lambda^{-2.5} \)) shown by biomass/firewood aerosols from the Southern African Regional Science Initiative (SAFARI) experiment while a much weaker spectral dependence (\( \sim \lambda^{-1} \)) exhibited by roadway samples from Berkeley consisting of motor vehicle aerosols produced due to fossil fuel burning. \textit{Kirchstetter et al.} [2004] also observed that extraction of biomass burning aerosols with acetone, removed significant amount of organic carbon (OC) from the sample, reduced spectral dependency in absorption and brought down the \( \alpha \) value close to 1.2. Other studies [eg. \textit{Jacobson et al.}, 2000; \textit{Bond}, 2001; \textit{Bergstrom et al.}, 2002] have also shown that mixture of aerosols in which absorption is mainly due to BC, exhibit a weak spectral dependence (\( \lambda^{-1} \)).
of aerosol absorption obtained over Ahmedabad shows signatures for the presence of absorbing aerosols produced from sources other than burning of fossil fuel. Comparison of our finding with the results published by other researchers shows that there are significant amounts of absorbing aerosols in the atmosphere produced from biomass/biofuel burning occurring in all seasons. Higher values of absorption Angstrom parameter $\alpha$ during Dry and Post-Monsoon season shows that activities related to biomass/biofuel burning increase during these seasons which produces larger amounts of such absorbing species in the atmosphere. These activities mainly include open burning of dry leaves or other waste materials which serves as a rather easy way of getting rid of such garbage materials. Other source of carbonaceous aerosols is the usage of biofuel such as wood, animal-dung cakes etc. for cooking by a large population living in slums of the city [Reddy and Venkataraman, 2002]. Also a detailed work by Venkataraman et al. [2005] have shown that in India about 44% of the total BC emission comes from biofuel combustion. Most of the sources of carbonaceous aerosols discussed so far involve low temperature and incomplete combustion processes which release sufficient amount of organic carbon (OC) apart from soot or BC. Kirchstetter et al. [2004] have shown that presence of OC in addition to BC in biomass/biofuel smoke aerosols affects spectral dependence in absorption by aerosols, contributing significantly to the measured light absorption in visible and ultraviolet regions. However, it is a subject for investigation whether OC from biomass burning, which is considered to be the scattering component of carbonaceous aerosols [Novakov et al., 2005], itself contributes to absorption in addition to BC or it forms some kind of coated shell around BC aerosols that increases the absorption efficiency of these complex mixtures [Jacobson, 2000; Jacobson, 2001]. Further laboratory experiments under controlled conditions with known aerosol types as well as modelling studies are required to confirm the exact reason which causes the observed increase in spectral dependence of absorption by aerosols at places where biomass/biofuel burning are major sources of carbonaceous aerosols in the atmosphere. Nevertheless, high values of BC mass with increased absorption efficiency measured over Ahmedabad is expected to produce higher values of aerosol radiative forcing.
2.3.6 Aerosol scattering coefficient

Measurements of aerosol scattering coefficient ($\beta_{\text{sc}}$) started during Monsoon 2003, along with the BC measurements. Figure 2.10(b) shows the seasonal variation of aerosol scattering coefficient at 0.53 $\mu\text{m}$, averaged over all data available from the year 2003 to 2005. Vertical lines on top of each bar represent ±1 $\sigma$ variation about the mean of all data on $\beta_{\text{sc}}$ available for that season. Aerosol types which contribute maximum to the scattering coefficient include water-soluble inorganic species such as sulfates, nitrates etc., arising from emissions of $SO_2$ and $NO_x$ associated mainly with fossil fuel combustion, ammonium from fertilizers and biological sources and some organic aerosols arising from biomass combustion [Charlson et al., 1992; Penner et al., 1994]. In addition to these, nitrates and organic aerosols from industrial emissions also contribute significantly to the scattering characteristics of aerosols. Comparing the results presented in figure 2.10(a) and 2.10(b), we find that the seasonal variation in aerosol scattering coefficient is very much similar to that of BC mass concentration. This shows that majority of absorbing and scattering type aerosols prevalent over Ahmedabad have common production sources. Also, similarity of both these plots with the seasonal variation of nucleation mode aerosols (figure 2.8) shows that most of these scattering and absorbing type aerosols fall in the nucleation mode size range, which are mostly produced due to various anthropogenic activities. Highest values of $\beta_{\text{sc}}$ are measured during Post-Monsoon season and this is followed by values obtained during Dry, Pre-Monsoon and Monsoon seasons respectively in the decreasing order of their magnitudes. Higher values of $\beta_{\text{sc}}$ measured during Post-Monsoon and Dry seasons is an outcome of the combined effect of several favorable factors. During these seasons, there is an increase in waste burning activities, such as burning dry leaves, shrubs etc. in various parts of the city and all these contribute significantly to the emission of both scattering and absorbing type species in the atmosphere. Unlike Pre-Monsoon and Monsoon seasons, when the surface level wind flow is from the Arabian sea in south or south-west, relatively pristine air masses with less continental influence reach our measurement location, while air masses reaching Ahmedabad during Post-Monsoon and Dry seasons are mostly of continental origin contributing more aerosols from surrounding locations to the already high level of aerosols produced within the city. Moreover, during Dry and Post-Monsoon seasons, aerosols which are either locally produced or transported from other regions are constrained within a shallow boundary layer having smaller ventilation coefficient and
2.3. Results and Discussion

this causes their concentration to rise near the surface level. Also, as will be shown later while discussing the vertical profiles of aerosols, during Pre-Monsoon and Monsoon seasons, atmospheric boundary layer height increases, making more volume available for the aerosols to get distributed and this results in a dilution of their concentration near surface level.

Figure 2.13: Average patterns in diurnal variation of aerosol scattering coefficient ($\beta_{sca}$) at 0.53 $\mu$m for different seasons of a year, with vertical lines representing $\pm 1\sigma$ variation about the mean value of $\beta_{sca}$ at a particular time on different days of a particular season.

Figure 2.13 shows the average patterns in diurnal variation of $\beta_{sca}$ for different seasons of the year. Vertical lines represent $\pm 1\sigma$ variation about the mean value of $\beta_{sca}$ measured at a particular time on different days of a particular season. Most striking similarity among the plots on diurnal variation of $\beta_{sca}$ and BC mass concentration is the occurrence
of twin maxima and minima within a day in all seasons, except the first maxima and sec-
ond minima are not resolvable as both appear at a same level during Monsoon season
(Figures 2.11 and 2.13). Highs and lows in the diurnal variation of $\beta_{sca}$ are governed by
several factors such as production source of these scattering type aerosols, variations in
their source strength, removal mechanisms, boundary layer height variations and also the
surface meteorology. First maxima in $\beta_{sca}$ is observed between 08 — 09 hrs during Dry
and Post-Monsoon seasons while this occurs little early between 07 — 08 hrs during Pre-
Monsoon season. Evening peak in scattering coefficient is reached around 21 hrs during Dry and Post-Monsoon season while it shifts little early, between 20 — 21 hrs during Pre-
Monsoon and Monsoon seasons. This peak is a result of increased production of aerosols
in the evening hours due to various anthropogenic activities such as peak in traffic level
after 17 hrs, burning of waste materials (dry leaves, shrubs, papers etc.), usage of biofuel in
the slums of the city etc. All these result in piling up of aerosols produced during evening
hours which continue up to late night, in the surface layer of the atmosphere. In addition to
increased production of aerosols, formation of inversion layer in the evening hours inhibits
convective mixing of air and therefore aerosols present in it. However, after around 21 hrs,
production of aerosol particles from most of these sources start declining but their removal
from the atmosphere, mostly by gravitational settling process, remains still active. This re-
moval process continues for the whole night and $\beta_{sca}$ reaches a minima between 04 — 05
hrs in the early morning. However, various human activities start from the morning itself
and release many precursor gases for the production of these scattering aerosols. These
activities start when the boundary layer height continues to be low and therefore aerosols
produced during this time get very less volume in the atmosphere to distribute themselves
causing $\beta_{sca}$ to rise and reach a maxima between 07 — 09 hrs depending on the season. Af-
ter 09 hrs, due to increased insolation of the Earth’s surface, atmospheric boundary layer
height starts rising along with an entrainment at the top of the inversion layer. This kicks
off convective mixing of air capable of lifting aerosols to higher levels in the atmosphere
and causing a dilution of their concentration at the surface level. This mixing of air contin-
ues upto afternoon hours when we see a minima in the traffic level. As a result of this, $\beta_{sca}$
measured for near surface aerosols reaches a minima between 16 — 17 hrs. Under favorable
conditions such as increasing relative humidity, collapsing boundary layer and increased
production of aerosols in the evening hours, scattering coefficient of aerosols starts rising
2.3. Results and Discussion

after 17 hrs to reach the evening maxima. This cycle in the diurnal variation of $\beta_{sca}$ is repeated on all clear sky days throughout the year, except with some abnormalities during Monsoon season. During Monsoon season, boundary layer height mostly remains high and shows a very weak diurnal variation. Also, the formation of surface based inversion layer and nocturnal boundary layer are not very strong and distinct during this season as a result of which we do not get a well defined peak of $\beta_{sca}$ in the morning hours and even the second maxima during evening hours is not very prominent as observed in other seasons.

2.3.7 Single scattering albedo

As scattering and absorbing type aerosols are present together in the atmosphere, their effect in terms of cooling or warming of the atmosphere depends on the single scattering albedo ($\omega$) of the mixture of aerosols, which is the ratio of scattering to extinction coefficient of aerosols. The magnitude of $\omega$ is considered as an index for the relative dominance of scattering with respect to absorbing type of aerosols, which can range from 0 (purely absorbing) to 1 (purely scattering). Exact knowledge of single scattering albedo is very crucial as small error in its magnitude can produce large difference in the estimated values of aerosol radiative forcing [Takemura et al., 2002]. Ganguly et al. [2005a] have shown that for the same aerosol optical depth (AOD) and mass loading over Bay of Bengal, atmospheric forcing by aerosols is very sensitive to $\omega$. Over land areas, knowledge of $\omega$ is even more critical and any small change its value can have larger impact resulting from errors in the estimated flux changes within and below the aerosol layer such as differential heating rates, changes in atmospheric stability and cloud formation [Ackerman et al., 2000; Russell et al., 2002]. Even satellite retrieval of aerosol optical depth (AOD) is also intricately dependent on the assumption of $\omega$ in the aerosol model for any study region [Ignatov et al., 1995; Stowe et al., 1997]. Although there are some reports on measurements of BC mass concentration over the Indian mainland [Babu et al., 2002; Babu and Moorthy, 2002; Venkataraman et al., 2002; Tripathy et al., 2005; Latha and Badrinath, 2005], reports on measurement of single scattering albedo over Indian mainland is almost nil, except during a field study over central India in February 2004 by Ganguly et al. [2005c] and during another campaign at New Delhi in December 2004 by our group [Ganguly et al., 2006]. Moreover, most of these studies [Babu et al., 2002; Venkataraman et al., 2002; Ganguly et al., 2005c; Tripathy et al., 2005]
are part of intensive campaigns organized for short periods. What is needed for proper assessment of possible impacts produced by these aerosols prevalent over the Indian subcontinent is to make long term measurements of these important aerosol parameters at various stations spread all across the country.

In this particular study, we have estimated the single scattering albedo of aerosols from the ratio of scattering coefficient at 0.53 \( \mu m \) using Nephelometer and its sum with the absorption coefficient at 0.52 \( \mu m \) measured using Aethalometer. Measured values of scattering coefficient are associated with some angular truncation loss which is an inherent and unavoidable problem for all Nephelometers [Heintzenberg and Charlson, 1996]. Detailed Mie theory calculation using the simultaneously measured aerosol size distribution shows that, an angular truncation of 8\(^\circ\) each in the forward and backward direction could result in an underestimation of scattering coefficient by about 15\%. However, this causes a maximum error of about 3 – 4 percent (in the extreme case) in the estimated values of \( \omega \) for the range of absorption and scattering coefficient values measured over Ahmedabad. Similarly, a maximum error of 10\% in the estimation of absorption coefficient using Aethalometer causes about 2% error in the estimated values of \( \omega \) in the extreme case. Taking into account all possible sources of error, overall uncertainty in the estimated value of \( \omega \) during the present study is around 6\%. Nevertheless, for estimation of aerosol radiative forcing, presented in Chapter-6, diurnally averaged values of \( \omega \) are used which have a spread of about 13\%.

Figure 2.10(c) shows the seasonal variation of single scattering albedo at 0.525 \( \mu m \), averaged over all data available from the year 2003 to 2005. Vertical lines on top of each bar represents \( \pm 1 \sigma \) variation about the mean value of \( \omega \) for that season. Over the period of our study, single scattering albedo at 0.525 \( \mu m \) are found to be 0.73 ± 0.1, 0.84 ± 0.04, 0.81 ± 0.03 and 0.73 ± 0.08 during Dry, Pre-Monsoon, Monsoon and Post-Monsoon seasons respectively. Although patterns of seasonal variation in \( \beta_{abs} \) and \( \beta_{sca} \) appear almost identical [figures 2.10(a) and 2.10(b)], small differences in the percentage variation of individual parameters \( \beta_{abs} \) and \( \beta_{sca} \) from one season to other cause the observed pattern in seasonal variation of \( \omega \). Relatively lower values of \( \omega \) obtained during Dry and Post-Monsoon season shows that although both BC mass and \( \beta_{sca} \) exhibit higher values during these seasons, effect of increase in BC mass dominates in terms of contribution to the total extinction caused by all aerosols present near the surface level. This happens primarily
because during these seasons, there is an increase in the activities related to burning of waste materials such as dry leaves, shrubs, papers etc. which are mainly collected as part of cleanliness drives in various parts of the city and putting them on fire serves as an easy way for getting rid of these garbage materials. As discussed earlier and also seen in some of the earlier studies, aerosols emitted due to biomass/biofuel burning exhibit stronger absorption characteristics than those produced due to fossil fuel burning (Kirchstetter et al., 2004; Ganguly et al., 2005b, 2006). Therefore it is a matter of concern from the climate point of view as such practices are not only loading large quantities of aerosols of various kinds in the atmosphere but they are also producing soot particles with increased absorption efficiency, bringing down the single scattering albedo to lower values, thereby trapping heat within the atmosphere which can lead to climate forcing (Ramanathan et al., 2001; Menon et al., 2002).

Comparing results from Ahmedabad with previously published values of single scattering albedo for other regions (Anderson et al., 1999; Jayaraman et al., 2001; Russel et al., 2002, Bergstrom et al., 2003; Han et al., 2003; Qiu et al., 2004), we find that \( \omega \) values estimated over Ahmedabad are much lower indicating the dominance of absorbing aerosols. Babu et al. (2002) estimated \( \omega \) at 0.5 \( \mu m \) over Bangalore for the month of November, based on measurements of BC mass fraction and using the OPAC model developed by Hess et al. (1998), to be around 0.73. Following the same method, Singh et al. (2005) and Tripathy et al., (2005) reported \( \omega \) (0.5 \( \mu m \)) value for Delhi and Kanpur to be 0.67 and 0.76 during Pre-Monsoon period and December month respectively. Also, by inverting sun/sky radiometer data, Pandithurai et al. (2004) estimated \( \omega \) (0.5 \( \mu m \)) value for Pune (18.53°N, 73.85°E) to be around 0.81. Unlike previous examples, Ramana et al. (2004) estimated \( \omega \) (0.5 \( \mu m \)) value for Kathmandu (27.67°N, 85.31°E) to be in the range of 0.7 – 0.9, combining the actual measurements of scattering and absorption coefficient using a Nephelometer and a Particle Soot Absorption Photometer (PSAP). Similar to present study, Ganguly et al. (2005c) reported the single scattering albedo (0.525 \( \mu m \)) values estimated using simultaneously operated Nephelometer and Aethalometer measuring scattering and absorption coefficient of aerosols respectively for different locations (mostly rural) over the central Indian region to be in the range of 0.75 – 0.90. Also, during an intensive campaign at New Delhi in December 2004, our group estimated single scattering albedo at 0.525 \( \mu m \) following the same method as the present study and found \( \omega \) to vary between 0.6 and 0.8 with an average
value close to 0.68 for the entire period of campaign [Ganguly et al., 2006].

Figure 2.14: Average patterns in diurnal variation of single scattering albedo ($\omega$) at 0.525 $\mu m$ for different seasons of a year, with vertical lines representing $\pm 1\sigma$ variation about the mean value of $\omega$ at a particular time on different days of a particular season.

Figure 2.14 shows the average patterns in diurnal variation of $\omega$ (0.525 $\mu m$) for different seasons of the year. Vertical lines represent $\pm 1\sigma$ variation about the mean value of $\omega$ estimated for a particular time on different days of a particular season. Among the most common features, notable in all four panels of figure 2.14 include occurrence of two dips and an afternoon maxima in the diurnal cycle of $\omega$ observed in different seasons. Since the single scattering albedo is a combined effect of scattering and absorption by aerosols, diurnal pattern in the variation of $\omega$ is determined by the contemporary behavior of scattering and absorption coefficient of aerosols. We find that $\omega$ values attain a minima sometime
2.3. Results and Discussion

between 07 – 09 hrs, almost coinciding with the peak in BC mass concentration (figures 2.11 and refAhmddiurnalssa). After this dip, \( \omega \) starts rising very slowly up to a time when BC concentration reaches a minima in the afternoon time between 15 – 17 hrs. Main reason for getting the observed trend in \( \omega \) is the presence of a very small phase lag between the diurnal variation of BC mass and aerosol scattering coefficient (figures 2.11 and 2.13). This phase lag exists because while soot particles from vehicular exhausts or biomass/biofuel burning are directly released as particles in the atmosphere, most of the scattering type aerosols viz. sulfates and nitrates are secondary aerosols formed by gas-to-particle conversion processes. Therefore, although most of the precursor gases are released at the same time together with absorbing soot particles, it takes slightly longer time for the precursor gases to get converted into sub-micron particles. Also, as long as concentration of these precursor gases are sufficient and conditions in the atmosphere remain conducive, their transformation into particles continues. After about 15 hrs when traffic level starts rising along with increase in other types of burning activities, BC mass and absorption coefficient values start increasing in response to increased production of aerosols from these sources. Although scattering coefficient increases after 15 hrs, a faster increase shown by BC aerosols causes \( \omega \) value to decrease up to a time when both scattering and absorption coefficient values reach a more or less steady state after around 20 hrs.

2.3.8 Aerosol vertical profiles

A Micro Pulse Lidar (MPL) system (SES Inc., USA) has been used to get the vertical distribution of aerosols in the atmosphere and retrieve their extinction profiles. Earlier, several researchers emphasized the importance of measuring vertical distribution of aerosols as they have pointed out that one major contributor to the uncertainty in atmospheric forcing comes from uncertainty in the vertical distribution of aerosols and their single scattering albedo [Haywood and Ramaswamy, 1998; Chung et al., 2005]. MPL observations at Ahmedabad started in February 2002 and continuous data on vertical distribution of aerosols over our measurement site are available up to February 2005. Due to technical problem with the MPL system, vertical profile measurements could not be continued beyond March 2005. Throughout our study period, a significant day to day variability in the vertical profiles of aerosol extinction coefficient is observed. For the sake of simplicity and for the purpose of the present work, we have grouped the aerosol extinction profiles in terms of four
Seasonal and inter-annual variations in aerosol characteristics over Ahmedabad

Figure 2.15: Clear sky aerosol extinction profiles for different seasons, averaged over all data available for the years 2002 to 2005. Horizontal bars represent ±1σ variation about the mean value of extinction coefficient retrieved for a particular altitude on different days of a particular season.

different seasons (as discussed earlier) and for all years. We have further averaged the data on aerosol extinction profiles available for same season but in different years. Figure 2.15 shows the seasonal variations in clear sky aerosol extinction profiles, averaged over all data available from the year 2002 to 2005. Horizontal bars represent ±1σ variation about the mean value of aerosol extinction coefficient retrieved for a particular altitude on different days of a particular season. In the present work, we restrict our discussions to the four seasonally averaged profiles shown in figure 2.15. Seasonal variabilities in the shape of these extinction profiles explain reasons for observing relatively opposing trends in seasonal variation of surface aerosol parameters [Figure 2.10] and the columnar aerosol
optical depths [Figure 2.6(b)]. These profiles clearly show the kind of evolution of the atmospheric boundary layer height occurring during different seasons of any year. Area under all these curves represent the average aerosol optical depth for that season. We find the extinction profiles for Dry and Post-Monsoon seasons to be much similar to each other while the profiles obtained during Pre-Monsoon seasons appear intermediary between the two extreme type of profiles obtained during Dry and Monsoon seasons. Aerosol vertical profiles for Dry and Post-Monsoon seasons are characterized by very high values of extinction coefficient within first few hundred meters (~ 200 m) from the surface where we find a sharp decrease in the extinction values with increasing height. The reason for observing such high values of aerosol extinction coefficient at lower heights is the formation of a surface based inversion layer during Dry and Post-Monsoon seasons. This layer being very stable, opposes vertical mixing of various constituents in the atmosphere which results in piling up of aerosols within the layer and pushes extinction values towards higher side [Krishnan and Kunhikrishnan, 2004]. During Dry and Post-Monsoon seasons, atmospheric boundary layer height remains low, primarily due to less insolation at the Earth’s surface. It can be seen from figure 2.15 that extinction values from the top of this surface layer (~ 200m) decrease rather slowly and approach zero level, almost asymptotically, at an altitude of about 4 km. On the other hand, no such surface layer is observed during Pre-Monsoon and Monsoon seasons but in these seasons, height of the atmospheric boundary layer goes up due to increased insolation at the Earth’s surface on cloud free days. This provides a larger room for the aerosols to distribute themselves in the atmosphere, assisted by stronger convective circulations in lower parts of the atmosphere. Vertical distribution of aerosols in Pre-Monsoon season appears to follow a combination of two exponentially decreasing profiles connected through a small inversion layer between them (base of the inversion layer occurs at ~ 0.6 km). During Monsoon season, we find an exponentially decreasing type of distribution in the first ~ 0.5 km, above which rests an inversion layer with its peak close to 1.0 km. Most striking feature in the vertical distribution of aerosols during Monsoon season is the presence of a thick and stable aerosol layer between 0.5 and 2.0 km. This layer contributes significantly to the columnar aerosol optical depth values but its effect is not felt in any of the surface level measurements. This is the main reason why higher AODs are observed on clear sky days during Monsoon season while surface measurements of BC mass and aerosol scattering coefficient shows low value in the same
season. Vertical profiles of aerosol distribution shows finite contribution of extinction coefficients almost up to 5 km from the surface during Pre-Monsoon and this reaches close to 6 km during Monsoon season.

Large seasonal variabilities are observed in all aerosol parameters measured over Ahmedabad during the period from 2002 to 2005. These are bound to produce large differences in radiative forcing due to aerosols in different seasons of the year. All results on different aerosol parameters measured over Ahmedabad are used as inputs in a radiative transfer model and implications of the observed variabilities have been studied in terms of regional scale aerosol radiative forcing. Several interesting results on model estimates of aerosol radiative forcing and its sensitivity to the observed variabilities in aerosol parameters are discussed separately in Chapter-6.

2.4 Summary

Results from continuous measurements of physical and optical parameters of aerosols made over Ahmedabad, an urban location in western India, from early 2002 till the end of 2005 are presented. Important aerosol parameters studied over this location include: column AOD spectra, aerosol mass concentration, number size distribution, BC mass concentration, wavelength dependency in absorption, scattering coefficient, single scattering albedo and vertical distribution of aerosols in the atmosphere. All parameters showed large variability in their values during different seasons and in different years. All available data are classified in terms of four major seasons observed over Ahmedabad viz. Dry (December to March), Pre-Monsoon (April-May), Monsoon (June-September) and Post-Monsoon (October-November), primarily based on different meteorological conditions prevailing during different months of the year. Seasonal and inter-annual variabilities in meteorological parameters played an important role in shaping the observed patterns of seasonal variations for different aerosol parameters. AOD at all wavelength channels show an increasing trend over first half of the year, which is found to be stronger and consistent at higher wavelengths. Angstrom parameter $\alpha$, estimated for three different wavelength pairs showed almost same pattern of seasonal variation in all years with higher values obtained during Dry and Post-Monsoon seasons and lower values during Pre-Monsoon and Monsoon seasons. We find a dominance of smaller size particles during Dry and
Post-Monsoon seasons while an increase in coarse mode particle concentration during Pre-Monsoon and Monsoon seasons. Over the period of our study, PM10 mass concentration varied between low values close to $40 \mu g/m^3$ up to high of about $106 \mu g/m^3$ with an average value of around $66 \mu g/m^3$. Lower values of PM10 mass concentrations are usually measured during Monsoon season. QCM observations have been further used to obtain the number size distribution of near surface aerosols. We have seen maximum seasonal variation in the number concentration of nucleation mode aerosols, with nearly 60% spread in their value about the mean concentration for entire period. This is followed by accumulation and coarse mode data showing about 26 and 17% spread in their values respectively. Aerosol size distribution for all seasons exhibit presence of three distinct modes each one of which could be fitted using a log-normal curve. The shape of the aerosol size distribution remained more or less same over the study period while number concentration changed in all three modes during different seasons, indicating that the amount of aerosols as well as their in situ production strengths changed while various physical processes responsible for their distribution in the atmosphere remained unaltered. High values of BC mass are obtained during Post-Monsoon season (7.3 ± 3.7 $\mu g/m^3$). This decreased slightly during Dry season (5.5 ± 2.8 $\mu g/m^3$) and comparatively much lower values of BC mass are measured in Pre-Monsoon (2.2 ± 1.0 $\mu g/m^3$) and Monsoon season (1.5 ± 0.8 $\mu g/m^3$). Diurnal variation of BC mass shows the presence of two maxima and two minima in all seasons. Wavelength dependency of aerosol absorption shows signatures of presence of significant amount of absorbing aerosols produced from biofuel/biomass burning. Average values of absorption Angstrom parameter $\alpha$ are found to be to be 2.2, 2.0, 1.9 and 2.1 for Dry, Pre-Monsoon, Monsoon and Post Monsoon seasons respectively. Highest values of aerosol scattering coefficient are measured during Post-Monsoon which is followed by values measured during Dry, Pre-Monsoon and Monsoon seasons respectively in the decreasing order of their magnitude. Over the period of our study, single scattering albedo at 0.525 $\mu m$ is found to be 0.73 ± 0.1, 0.84 ± 0.04, 0.81 ± 0.03 and 0.73 ± 0.08 during Dry, Pre-Monsoon, Monsoon and Post-Monsoon seasons respectively. A Micro Pulse Lidar (MPL) has been used to get the vertical distribution of aerosols in the atmosphere and retrieve their extinction profiles. MPL observations at Ahmedabad started in February 2002 and continuous data on vertical distribution of aerosols over our measurement site are available up to February 2005. Aerosol vertical profiles for Dry and Post-Monsoon seasons are...
characterized by very high values of extinction coefficient within first few hundred meters (~ 200m) from the surface where we find a sharp decrease in the extinction values with increase in height. MPL observations during Pre-Monsoon and Monsoon season shows the presence of aerosols upto higher levels in the atmosphere, contributing significantly to the columnar aerosol optical depth values.