A state of the art mobile micro pulse lidar (MPL) is used extensively to study the vertical distribution of aerosols properties. The results obtained using this set-up are presented and discussed in more detail. In addition, results from other observations, using Sun-photometers, Aethelometer, Nephelometer, QCM-Cascade Impactor and Aerosol Size Spectrometer are also discussed and used in the aerosol radiative forcing estimation.

2.1 Aerosol Vertical Profiles

The knowledge of vertical profile of aerosol optical properties is very important for modelling the radiative effect as well as in computations related to the study of boundary layer stability, evaporation rate, sensible heat flux, convection, actinic flux etc. In the present work information on the vertical distribution of aerosol is obtained using the Micro Pulse Lidar (MPL).

2.1.1 Instrumentation

Lidar works on the principle similar to that of RADAR. Measurement of the intensity of the backscattered photons provides information on the properties of the scatterer. The time lapse between the transmitted and received signals is proportional to the distance of scatterer from Lidar. The Micro Pulse Lidar (MPL) used in the present study is a compact co-axial mono-static lidar system. Figure 2.1 shows the photograph of MPL mounted inside a mobile Lidar observatory of the Physical Research Laboratory (PRL) and Table 2.1 summarises important features of MPL. Laser, telescope and diode are housed in single tubular casing. The tubular casing is fixed on two stanchions with pivoting ring in the centre. This allows to change the viewing angle. Also, the arrangement is very useful for estimating the overlap correction (to be discussed later) unlike traditional lidar.
system, which looks only at fixed angle. MPL uses a diode pumped Nd-YLF laser with second harmonic generation facility to give laser light of wavelength 523.5 nm. Advantage of diode pumped laser is that laser unit can be miniaturised and pulse repetition frequency can be increased greatly compared to conventional flash lamp pumped laser. This unit can be operated with pulse repetition rate from 1 to 10000 Hz. While a larger pulse rate is good for improving the signal to noise ratio (SNR) it decreases the power in individual laser pulses, which reduce the SNR. Hence it is necessary to find out the optimal value for the laser repetition rate. In the present case, it is operated with 2500 Hz pulse rate, which is found to be the optimum. Outgoing pulses are expanded by a telescope and sent to the atmosphere. Backscattered photons are received back by the same telescope, which is a 20 cm diameter Schmidt-Cassegrain telescope with field of view (FOV) less than 50 μrad. The small FOV reduces the background noise and complications arising out of multiple scattering to a great extent. A narrow band pass filter further reduces the background noise. MPL uses an interference filter of full width at half maximum (FWHM) equal to 0.1 nm centred at 523.5 nm to screen the incoming photons. Screened photons are detected with silicon avalanche photo diode (Si-APD) operated in counting mode. Si-APD has a quantum efficiency of about 40-50% at wavelength 523.5 nm and a larger dynamic range of detection limit. This enables MPL to work even in day light condition. The electrical pulses from Si-APD are counted by multichannel scaler (MCS) electronics as time gated signal. It is possible to set the binwidth as 200, 500, 1000 or 2000 ns, the time interval during which incoming photons will be summed and recorded in single memory location (bin). For the next same length of period, photons will be counted and recorded in consecutive bin. Binwidth essentially determines the range resolution of system (ΔR) which is equal to BinWidth×c/2, where “c” is the speed of light. The factor two in the denominator is because the photon has to travel distance ΔR twice. In the present study, the bin width is set as 200 ns, which corresponds to a range resolution of 30 m.
Table 2.1. Features of Micro Pulse Lidar system operational at PRL, Ahmedabad

<table>
<thead>
<tr>
<th>Feature</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laser</td>
<td>Diode pumped Nd-YLF</td>
</tr>
<tr>
<td>Wavelength</td>
<td>523.5 nm</td>
</tr>
<tr>
<td>Pulse repetition rate</td>
<td>2500 Hz (Typical)</td>
</tr>
<tr>
<td>Telescope</td>
<td>20 cm dia. Schmidt-Cassegrain</td>
</tr>
<tr>
<td>Field of view</td>
<td>50 μrad</td>
</tr>
<tr>
<td>Filter</td>
<td>Central wavelength: 523.5 nm</td>
</tr>
<tr>
<td></td>
<td>FWHM: 0.1 nm</td>
</tr>
<tr>
<td>Detector</td>
<td>Si avalanche photo diode (Si-APD)</td>
</tr>
<tr>
<td>Data acquisition</td>
<td>Multichannel scaler</td>
</tr>
<tr>
<td>Bin width</td>
<td>200 ns (Typical)</td>
</tr>
</tbody>
</table>

Figure 2.1. Micro Pulse Lidar (MPL) fitted inside the mobile lidar observatory of the Physical Research Laboratory, Ahmedabad.
Pulse repetition frequency (PRF) determines the theoretical limit on the maximum range of a lidar. Time between two pulses is available to photons for travelling to and fro distance. In the case of PRF equal to 2500, this period is equal to 400 μsec. During this period maximum distance a photon can travel is 120 km. However, in actual practice the maximum altitude that is studied for aerosols is up to around 7 km, mainly determined by laser power and background sky radiation intensity but on perfect dark nights backscattered laser light could be detected up to about 20 km using the present system.

![Figure 2.2. Block diagram of the micro pulse lidar system, described in Table 2.1 and shown in Figure 2.1.](image)

2.1.2 Data reduction

Counts obtained using multi-channel scaler (MCS) are saved with one-minute average for 1000 bins. Each record is preceded with a header containing information on laser pulse energy, detector, laser and ambient temperatures, pulse
repetition rate, number of profiles summed, etc. This is called raw data record. Following paragraphs describe various corrections required to apply on the raw data.

MPL raw data counts can be related to extinction coefficient using following relation, known as the lidar equation:

\[
P(r) = \frac{A E \beta(r) T^2(r)}{r^2} + P_b + P_{ap}(r)
\]

Here \( P \) is the actual count measured by the system. \( r \) is the range from lidar to target. \( A \) is an instrument constant, depends upon the size of telescope receiver, photo detector and optics efficiencies. \( E \) is the energy of the outgoing pulse. \( \beta \) is the volume back-scattering coefficient at laser wavelength by all the constituents of atmosphere at range \( r \). \( T \) is the transmission of medium from lidar to range \( r \). Since photons have to travel distance \( r \) twice, square of the transmission is taken. \( P_b \) is the background due to ambient light sources. \( P_{ap} \) is the after-pulse count. In this equation range \( r \) can be calculated from bin number using the following relation:

\[
r = \text{Bin No} \times \text{Bin Width} \times c / 2 - \text{Bin Width} \times c / 4 - \Delta t \times c / 2
\]

where \( c \) is the speed of light. Bin number multiplied by bin-width gives the total time available for travelling the to and fro distance by photon and multiplying it with speed of light and dividing by two gives the distance of that particular bin with respect to Lidar. Since each bin has finite bin-width, distance calculated in this manner corresponds to a range (30 meter in present case). It is customary to designate distance of each bin with central value of range for that bin and the second term in above equation accounts for that. Laser trigger and MCS are synchronised using BNC cable. However there could be offset \( (\Delta t) \) between triggering of laser pulse and MCS. If triggering of laser pulse takes place before MCS starts recording, offset can be found only through laboratory experiments.
In the present case MCS starts data recording before the laser pulse is fired, hence offset can be directly found in the raw data. The bins, which are recording data before the laser is fired, contain only dark current and background noise. The present MPL system has 400 ns (in other words 2 bins) offset between MCS and Laser trigger. Last term in equation 2.2 accounts for this offset $\Delta t$.

![Dead-time correction factor for the MPL detector as a function of photon-counts](image)

Avalanche photo diode has finite time for a single photon avalanche event. During this time interval it will not record another event if it occurs. This time interval is known as dead time of a diode. Overall effect of it will be an underestimation of the actual count rate. Correction of it is particularly important in the situation of high incident rate of photon. Dead-time varies from diode to diode, which can be estimated in laboratory by using standard light source. A lookup table supplied by the manufacturer listing the statistical approximation of dead-time correction factor as a function of photon events per $\mu$s is used in the present study. Figure 2.3 shows the dead-time correction factor used in the present study. The highest
incident rate of photons is immediately after the laser firing. This is because of internal reflection and atmospheric backscattering from the nearest range. Count rate for this case is always high and is typically around 5 counts/μs. Normally this bin is avoided in analysis. It is quite unlikely that atmospheric scattering can exceed this count rate. For the bins excluding third bin count rate normally remains less than 0.4 counts/μs and hence dead-time correction factor is very close to one. \( P(t) \) in the equation 2.1 is dead-time corrected.

Laser pulse before exiting the telescope encounters at least 6 surfaces (Figure 2.2) resulting in significant internal reflection of light that can reach to photodiode. This initial high count rate saturates the Si-APD and the exponentially decaying small leakage current from diode keeps on flowing even after the pulse is over. This current is known as after-pulse current. Since range is estimated from time elapsed after the pulse has been fired, after-pulse \( (P_{\text{ap}}) \) in that way is a function of range (Refer to equation 2.1). After-pulse can be estimated experimentally in dark room by pointing the lidar towards a black target. In this condition signal beyond the blocking point is after-pulse and dark current of detector. However, values of after-pulse also depend on the out-going pulse energy. Linear relationship between after-pulse and laser pulse energy is assumed (Campbell et al., 2002). After-pulse correction factor is normalised with laser energy during the after-pulse calculation experiment.

\[
P_{\text{ap}}(r) = P'_{\text{ap}}(r) \frac{E}{E_0}
\]

In equation 2.3 \( P'_{\text{ap}}(r) \) is the after-pulse factor obtained from the experiment. \( E_0 \) is the energy of laser pulse when \( P'_{\text{ap}}(r) \) was estimated. \( E \) is the energy of laser pulse of the profile to which after-pulse factor is to be applied. In Figure 2.4, the after-pulse profile is shown by dotted line and total raw signal during night time using solid line. Dark current of the detector is also a limiting factor for higher altitude data as after 16 km total raw counts are very close to dark current counts.
Figure 2.4. After-pulse effect in lidar signal. Thin line shows the after pulse and dark current photons and thick line shows the total photon counts obtained during nighttime.

Small field of view of telescope and narrow band-width of filter limits background light to a great extent but they do not completely eliminate it. MPL is operated with viewing angle in vertical direction. It is assumed that count measured for altitude range above 25 km may not contain back-scattered photons but only noise from after-pulse, dark current and sky background. Once the after-pulse and dark current are removed as described in previous section, background counts \( P_b \) (equation 2.1) is calculated by averaging photon counts from the range 25 to 30 km. Sky background will be uniform over this range which is subtracted from lower bins.

In the equation 2.1, instrument dependent constant \( A \) is shown to be independent of range. However, for near range, it is a function of range for two reasons. First, the telescope has small field of view and laser beam is expanded to the size of telescope. In near range telescope will not see the entire image of the beam.
Secondly, rays coming from infinity are focused at focal point where pinhole is placed. However rays coming from near range are focused away from the focal point, and the pinhole blocks some part of ray reaching to the focal plane. If all the optical properties of the system are known overlap correction factor can be calculated theoretically \cite{Dho et al., 1997}, however it is more advisable to obtain overlap correction factor experimentally.

When background and after-pulse corrections are accounted for and backscattered photon counts are normalized for laser energy, equation 2.1 takes the form:

\[
P'(r) = \frac{O(r) A' \beta(r) T^2(r)}{r^2}
\]

Here \( A \) in equation 2.1 is replaced with \( O(r) \) and \( A' \). Where \( O(r) \) is range dependent part of \( A \) and \( A' \) is range independent part. \( P'(r) \) is background and after-pulse corrected signal. Transmission of medium \( T \) is given by

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

Where \( \sigma(r) \) is the extinction coefficient of aerosol and air molecules. When system is pointed horizontally with no obstruction and if atmosphere is homogeneous, \( T(r) \) will be simply \( T(r) = \exp(-\sigma r) \). Substituting this into equation 2.4 for range \( r > n \) (where \( n \) is the distance beyond that overlap correction factor is 1) and taking log on both the sides, we get the following relation:

\[
\ln\left[ P'(r) r^2 \right] = \ln[ A' \beta] - 2\sigma r
\]

which is the equation for straight line with slope equals to \(-2\sigma\) and intercept equals to \( \ln[ A' \beta] \). As shown in Figure 2.5, the coefficients are found by least square fitting for horizontal direction measurements and is used to estimate \( O(r) \) for the range \( r < n \).
**Figure 2.5.** Result from an overlap correction experiment conducted at Mt. Abu, where lidar measurements are made in horizontal direction.

Over an urban region such as Ahmedabad, it is very difficult to get unobstructed horizontal view as well as a horizontally homogeneous atmosphere. So for overlap correction, at every 6 months MPL was taken to Mt. Abu observatory of PRJ., situated on a hilltop at a height of 1.7 km from sea level, where the air is relatively clean. It can be seen in Figure 2.5 that for the range beyond 1.5 km overlap correction becomes one. After removing the instrument related artefact discussed above, data are further screened for presence of cloud and measurement timings. Cloud screening is not straightforward as cloud exhibits scattering features similar to that of aerosol except that cloud backscattered signal is stronger than aerosol. That is why most of the cloud-screening-algorithms rely on the threshold value (Pal et al., 1992, Sassen and Wang, 1999, Venema et al., 2000). One needs to be very careful in selecting threshold value as it is quite possible that a smoke plume may
be screened out as a cloud or a thin cloud could be misunderstood as an aerosol layer. Often data contains large amount of random noise at the altitude where cloud occurs, which makes identification of cloud layer more difficult based on simple algorithm. In this work semi automatic approach is adopted for cloud screening. At first noise from the data is removed using wavelet transform (Namavati and Panigrahi, 2004). Then based on threshold value, which is specific to particular instrument, clouds are screened out using a computer code. Threshold value is deliberately chosen higher so as optically thin clouds are not removed. The profiles are then checked one by one manually along with log-book description and further screening of data is done. Profiles for which cloud occurred above 10 km were retained for analysis. In case of partially cloudy day, if more than 25 % profiles contained the cloud then that day was removed from averaging. However exceptions were made for summer monsoon months i.e. July to September. For these months each profile was manually checked for cloud screening and the data for the day was retained after removing the cloud affected profiles. Since many aerosol properties show diurnal variation, for study of day to day or seasonal variation it is important that measurements done during the same period of the day to be compared. Observations done after sunset and within 3 hours after the sunset are considered for averaging purpose in case of Ahmedabad. There exist observations that are made outside this time limit on special occasions. Timing is explicitly mentioned whenever results from those observations are discussed. Data screened in such manner were used to create 30 minute average profile which were subsequently used to calculate Extinction values as described below.

2.1.3 Theoretical Basis for Extinction Calculation

Corrections discussed above are applied to the measured photon counts. These photon counts are further multiplied by the square of range and range corrected normalized photon counts are obtained. Lidar equation 2.1 for range corrected normalized photon counts will take the form:

\[ \text{Lidar equation 2.1} \]
where $P''(r)$ is the photon counts after all the corrections applied and $A''$ is the calibration constant. There are two approaches to solve equation 2.8 for backscattering or extinction coefficient. One way is to absolutely calibrate the lidar system for $A''$ using simultaneous measurements such as aerosol optical depth (AOD) as described in Voss et al. (2001). However this method has some limitations for example most of the lidar measurements exist during nighttime when sun-photometer measurements does not exist. Indeed many lidar systems are not able to take measurements in day light condition because of very sensitive photo detectors. Another problem comes form the fact that calibration constant $A''$ is not stable and is highly dependent on the conditions of optical parts, detector temperature, laser energy, etc. More popular approach is to calculate extinction or backscattering profile from lidar measurements by using relative calibration. This eliminates the need to evaluate $A''$ but involves certain assumptions. During the last two decades large amount of work has been carried out by several authors to develop algorithm for retrieving aerosol extinction profile and other properties from Lidar measurements, to name a few Klett (1981), Ferguson and Stephens (1983), Fernald (1984), Mulders (1984), Klett (1985), Kaestner (1986), Balin et al. (1987), Gonzalez (1988), Kunz and Leuw (1993). Kovalev (1993), Ray et al. (1993), Bissonette and Hult (1995), Devara et al. (1995), Kunz (1996), Racadenbosch et al. (1999), Voss et al. (2001), Stephens et al. (2001), Sicard et al. (2002), Palm et al. (2002), Kovalev (2003), etc. Algorithms given by Stephens et al. (2001) and Palm et al. (2002) are for lidar measurements from satellite based platform. These algorithms are discussed in Chapter 4 where aircraft based lidar measurements are discussed. Following paragraph describes the two algorithms Klett (1981) and Kaestner (1986) in detail. Klett (1981) algorithm is milestone in the field of lidar based aerosol study and many algorithms named above are originally variants of Klett (1981) algorithm. Kaestner (1986) algorithm is used in present study to retrieve the extinction profiles.
It is not philosophically different from Klett (1981, 1985), however mathematical treatment is quite different.

Equation 2.8 may be expressed in a system-independent form as follows:

\[ S - S_0 = \ln \frac{\beta}{\beta_0} - 2 \int_{r_0}^{r} \sigma dr \]  \hspace{1cm} (2.9)

where \( S = \ln[r^2 P(r)] \), \( S_0 = S(r_0) \), \( \beta_0 = \beta(r_0) \) and \( r_0 \) is given constant reference range. Differentiating equation 2.9 with respect to \( r \) gives

\[ \frac{dS}{dr} = \frac{1}{\beta} \frac{d\beta}{dr} - 2\sigma \]  \hspace{1cm} (2.10)

Above equation can be solved only if we know the relationship between \( \beta \) and \( \sigma \). Traditional way to account for dependency between \( \beta \) and \( \sigma \) is to assume a power law relationship of the form

\[ \beta = B_0 \sigma^k, \]  \hspace{1cm} (2.11)

where \( B_0 \) and \( k \) are constants. Substituting this into equation 2.10 we obtain differential equation of the form:

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

The equation 2.12 is a nonlinear ordinary differential equation but it is of the form of Bernoulli or homogeneous Ricatti equation (Weisstein E. W. at http://mathworld.wolfram.com/RiccatiDifferentialEquation.html) and can be transformed to a first order linear form, solution to which is:

\[ \sigma(r) = \frac{\exp[(s-s_0)/k]}{\sigma_0^{-1} - \frac{2}{k} \int_{r_0}^{r} \exp[(s-s_0)/k] dr} \]  \hspace{1cm} (2.13)
where $\sigma_0 = \sigma(r_0)$. A priori knowledge of $\sigma_0$ is required to evaluate equation 2.13. The major drawbacks of equation 2.13 are that calculations of $\sigma$ involve the ratio of two numbers that normally decrease with range due to attenuation and denominator is a subtraction of two big numbers. This makes solution of the form equation 2.13 highly unstable for small measurement errors. Klett (1981) has shown remarkably simple way out of this problem. One can get equivalent solution of equation 2.12 using reference range $r_m$ so that solution generated for $r \leq r_m$ rather than for $r \geq r_0$ as before. New form of the solution will be

$$
\sigma(r) = \frac{\exp[(s - s_m)/k]}{\sigma_m^{-1} + \frac{1}{k} \int^r_s \exp[(s - s_m)/k]}dr',
$$

where $S_m = S(r_m)$ and $\sigma_m = \sigma(r_m)$. These seemingly small changes contribute significantly to the stability of solution. The solution is in the form of two numbers that becomes progressively larger and larger. In order to get $\sigma$, one has to know $\sigma_m$ a priori, which is usually not available. However, this is not a problem in case of vertical measurements from ground. This is because far end of profile ($r_m$) is in free troposphere or stratosphere where values of $\sigma$ are very small and does not exhibit large day to day variability. One may start with a guess value or climatological value. Calculations for successive lower altitudes depend weakly on the choice of $\sigma_m$ because of the form of the denominator in equation 2.14. In the end of this section it is shown that error due to guess value of $\sigma_m$ is less than 1%. For the full discussion of sensitivity of solution with different guess values one may see Klett (1981). Klett (1985) has shown the improvement for air molecules scattering and variable backscatter-to-extinction ratio (i.e. $B_0$ in the equation $\beta = B_0 \sigma^k$) and presented final equation (equation number 22 in Klett, 1985) in the form of backscattering coefficient. For most practical purposes $k$ is assumed to be equal to one. So extinction profile can be obtained by dividing backscattering profile with $B_0$. However, it is more advisable to get extinction profile straight from solution, as this is the physical quantity required for estimate of aerosol
radiative effect. Kaestner (1986) presents the solution of the lidar equation 2.8 in terms of extinction values. Following equations show the solution given by Kaestner (1986).

\[
\sigma_M(r) = -\frac{\beta_B(r)}{B_M(r)} Z(r) + \frac{Z(r)}{N(r)},
\]

with

\[
Z(r) = \frac{r^2 \cdot P(r)}{B_M(r)} \cdot \exp\left\{ 2 \cdot \int_{r}^{\infty} \left[ \frac{1}{B_p(r')} - \frac{1}{B_s} \right] \beta_B(r') \, dr' \right\},
\]

\[
N(r) = \frac{r^2 \cdot P_m}{\beta_{Rm} + \beta_{Mm} \sigma_{Mm}} + 2 \cdot \int_{r}^{\infty} Z(r') \, dr'.
\]

where \( P \) is equal to \( P' \) in equation 2.8 and hence forth double prime are dropped for sake of brevity. Subscript M denotes the aerosol (Mie scattering) and R denotes the gaseous (Rayleigh scattering) extinction or backscattering as the case may be. Subscript \( m \) denotes the quantity at range \( r_m \) (maximum range). \( \sigma \) and \( \beta \) are volume extinction and backscattering coefficients respectively and \( B \) is backscatter-to-extinction ratio equivalent to \( B_0 \) in equation 2.11. To get the aerosol extinction using equation 2.15 two assumptions are required, one on aerosol extinction at maximum range and another on backscattering-to-extinction ratio, inverse of which is known as lidar-ratio. Henceforth lidar-ratio will be denoted with symbol ‘\( L \)’. Lidar ratio remains to be the largest source of systematic error in retrieving aerosol extinction (Hughes et al., 1985, Kovalev, 1995). Value of the lidar ratio ranges from 20 to 80 sr over time and space as shown by Young et al. (1993), Marenco et al. (1997), Anderson et al. (2000), Welton et al. (2000), Voss et al. (2001), Liu et al. (2002), Welton et al. (2002), Sakai et al. (2003) depending upon aerosol type, size distribution and relative humidity. As far as regional variability is concerned \( L \) value of about 40 sr for arid and semi-arid region, around 30 sr for maritime aerosol and around 60 sr for urban and biomass burning are reported. Ahmedabad is an urban semi-arid region. We have used the constant lidar ratio 40
sr for extinction calculation throughout the period. Lidar ratio was found to be 44 sr during one of the experiment carried out over Mt Abu (a hill-station about 250 km north of Ahmedabad) following Young et al. (1993). One check for correctness of algorithm is how well observations made by one type of instrument matches with other type of instrument. Aerosol optical depth measurements using sun-photometer and aerosol optical depth measurements by integrating extinction profile are measurements of same quantity using two different methods. Correlation between them is checked and slope of the line is found close to one (Figure 2.6).

Figure 2.6. Scatter plot between sun-photometer derived AOD and micro pulse lidar derived AOD. Solid line through the middle of data point is 1:1 line.

In Figure 2.6 scatter of data points arise because of the fact that sun-photometer measurements are mainly during afternoon hours whereas lidar measurements are
mainly after sun-set hours. Also, scatter in data points due to error in lidar-ratio cannot be ruled out completely. In the absence of measurements of lidar-ratio it is justified to use constant lidar-ratio for all seasons because single scattering albedo (SSA) which is like lidar-ratio depends on aerosol type, does not show systematic variation with season over Ahmedabad (discussed in Chapter 3).

![Lidar Ratio](image)

**Figure 2.7. Aerosol extinction profiles obtained by using different values of lidar ratio for the observation made on 15 Feb 2002.**

Errors in the extinction profile are mainly because of various assumptions involved rather than instrument precision and noise. Assumption related to boundary condition i.e. extinction value at far end ($\sigma_{\text{max}}$) is found not to affect the accuracy of extinction profile. We carried out calculations of extinction profiles using different values of $\sigma_{\text{max}}$ for the same set of measurements. When $\sigma_{\text{max}}$ varied from 0 to $10^{-3}$ km$^{-1}$ changes in the extinction coefficient in the boundary layer is found to be less than 1%. The average extinction coefficient in the altitude range 7
to 10 km (i.e. where guess value $\sigma_{\text{max}}$ is required) observed by SAGE-II (http://www-sage2.larc.nasa.gov) satellite is $5 \times 10^{-4}$ km$^{-1}$.

Similar sensitivity analysis was also carried out for lidar ratio. Figure 2.7 shows the calculations of extinction profiles for different values of lidar ratio for the same set of measurements done on 15 February 2002. As described earlier extinction calculations for data analysis were carried out using lidar ratio 40, however lidar ratio for Ahmedabad like stations can be in the range of 30 to 50. Changing lidar ratio in the range of 30 to 50 changes the extinction profile in boundary layer by 15 to 20%. Changes due to change in Lidar ratio are like shifting the whole profile by a constant value. Hence even if error is of the order of 20% it will not affect the conclusions regarding relative distribution of aerosol with height. In the absence of independent measurements to calibrate, error in extinction profile is quoted 25%.

2.2 Total Columnar Aerosol Optical Depth

Aerosol optical depth (AOD) is one of the key aerosol optical property and an important input to climate modelling studies. It critically depends upon the amount, size-distribution and chemical composition of aerosols. It is a measure of the attenuation of radiation while passing from top of the atmosphere to surface by aerosol. The loss of intensity $F$ over the infinitesimal slice $dx$ as a result of light absorption or scattering is (Seinfeld and Pandis, 1998):

$$dF = -\sigma F dx$$ \hspace{1cm} 2.16

where, $\sigma$ is extinction coefficient (m$^{-1}$) of the medium. For a finite path between $x_1$ and $x_2$, integration of equation 2.16 is given

$$F(x_2) = F(x_1) \exp(-\tau)$$ \hspace{1cm} 2.17

where $\tau$ is the optical thickness (dimensionless) between $x_1$ and $x_2$ and is given by
\[ \tau = \int_{x_i}^{x} \sigma(x) dx \]  

2.18

This result is known as the **Beer-Lambert law of extinction**. When \( x \) is measured vertically in the atmosphere the optical thickness is referred to as *optical depth*.

### 2.2.1 Instrumentation

AOD is calculated by integrating aerosol extinction profile obtained using lidar as evident from equation 2.18. Results pertaining to AOD over Ahmedabad, which are discussed in present work, are mainly obtained in this way. However, AOD can also be measured directly by using a Sun-Photometer. Observations of AOD during field campaigns and also over Ahmedabad were also made using Sun-Photometer, which are used in the calculation of radiative forcing discussed in Chapter 5. Present section describes the Sun-Photometers in detail.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Central Wavelength (nm)</th>
<th>Full Width at Half Maximum (FWHM; nm)</th>
<th>Optical Depth ((\tau_{ray} + \tau_{mol}))</th>
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</thead>
<tbody>
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<td>Sun-Photometer (in-house make)</td>
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<td>15</td>
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</tr>
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<td>497</td>
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<td>Microtops-II (columnar ozone and water vapour)</td>
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<tr>
<td></td>
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<td>---</td>
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Table 2.2. Characteristics of the Sun-Photometers used in the present study. Last column shows the total optical depth due to Rayleigh scattering by air-molecules \((\tau_{ray})\) and molecular absorption \((\tau_{mol})\) for standard atmosphere.
There are three different Sun-Photometers used in present study. Table 2.2 summarizes the major characteristics of these Sun-Photometers. A filter Sun-Photometer built in PRL, was used for the measurement of AOD over Maitri-Antarctica (Gadhavi and Jayaraman, 2004), Gujarat, Mt. Abu, Indore (Gupta et al., 2003) and over Ahmedabad until a commercially available Sun-Photometer (MICROTOPS-II) from Solar Light Co. acquired. A Sun-Photometer can also be used for ozone and water vapour measurements by selecting appropriate wavelengths. One such Sun-Photometer is used to measure columnar ozone and water vapour concentrations. Ozone and water vapour measurements are used for correcting AOD measurements as well as radiative forcing calculations.

### 2.2.2 Theoretical Basis and Calibration of the Sun-Photometer

Figure 2.8 shows the block diagram of the Sun-Photometer developed at PRL. A Sun-Photometer essentially measures the sun light intensity at selected wavelengths. Baffle in front of photo detector is used to minimize diffuse radiation by reducing the field of view (FOV) of Sun-Photometer. FOV of PRL built Sun-Photometer is around 4° whereas that of Microtops is 2.5°. Equation 2.17 may be rewritten in following form as discussed in Gupta et al. (2003)

\[
\tau(\lambda) = -\frac{1}{m} \left\{ \ln \left( \frac{J(\lambda)}{J_0(\lambda)} \right) - 2 \ln \left( \frac{\theta}{\lambda} \right) \right\}
\]

2.19
where $\tau$ is total optical depth, $I$ is the instantaneous solar radiation intensity measured at ground level. $I_0$ is a top of the atmosphere (TOA) radiation intensity. $I$ and $I_0$ need not be in absolute unit (W/m$^2$), instead they can be in the form of voltages or current output of the Sun-Photometer as only the ratio of intensities are used. Symbol 'm' denotes relative airmass, a ratio of path length light beam has actually travelled in the atmosphere to that it would have travelled in vertical direction, and depends upon solar zenith angle. Symbols $r$ and $m$ in second term of equation 2.19 are Sun-Earth distances when measurements of $I$ and $I_0$ were made. In plane parallel approximation $m$ can be obtained by taking inverse of cosine of zenith angle. However, this approximation can introduce significant error in airmass calculation at high solar zenith angle because of curvature of the Earth and refractive index of air.

Young (1994) has given one approximate formula to calculate airmass from true solar zenith angle that accounts for the effects of refractive index and curvature of the Earth.

$$m = \frac{1.002432 \cos^2(\theta) + 0.148386 \cos(\theta) + 0.0096467}{\cos^3(\theta) + 0.149864 \cos^2(\theta) + 0.0102963 \cos(\theta) + 0.000303978}$$  \hspace{1cm} 2.20

where $\theta$ is the solar zenith angle. Precision given in the coefficients in equation 2.20 is intended only to prevent round-off errors near horizon; it does not indicate either the true accuracy of the formula or the precision of individual coefficients. Calculations of airmass using equation 2.20 are better than 1% even at horizon and taking into account the changes in meteorological conditions such as temperature, pressure etc. Solar zenith angle can be calculated from latitude, longitude of the place and time of measurements by astronomical formulae. Computer code to calculate solar zenith angle is prepared from the algorithm given by Meeus (1991). Left side of equation 2.19 is total optical depth, which is sum of optical depths by aerosol, scattering by air molecules and absorption by gases at that particular wavelength.
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Figure 2.8. Block diagram of the Sun-Photometer developed at PRL.

$I_0$ in equation 2.19 can be calculated from ground based measurements using Langley method (Liu, 2002). In this technique log of intensity measured at ground level is plotted as a function of airmass. A line is fitted to data points by least square fitting. Intercept of line on Y-axis gives log$_e(I_0)$. Ideally $I_0$ should remain constant over different places and time however because of deterioration in detector or filter characteristics value of $I_0$ can change. Hence, Sun-Photometers are repeatedly calibrated. Figure 2.9 shows a typical Langley plot for data obtained over Maitri-Antarctica on 21st February 2001 for 400 nm.

Inaccuracy in $I_0$ is the largest and most difficult to define source of systematic error (Shaw, 1983). Another sources of error in AOD measurements are radiometric uncertainties due to bias and precision of instrument that is around 5%. Optical depth is calculated by Beer-Lambert law that ignores the contribution of forward scattering to the measured flux. Forward scattering in FOV of a Sun-Photometer tends to decrease the measured AOD. Jayaraman et al. (1998) estimates less than 8% decrease in AOD for 400 nm and less than 4% in case of 1058 nm wavelength for the Sun-Photometer used in the present study. Errors for Microtops II would be certainly less than this as they have smaller FOV. Overall error estimated for hand held Sun-Photometer built in PRL is around 15%.
AOD is obtained by subtracting the Rayleigh optical depth and molecular absorption optical depth from total optical depth. For Rayleigh optical depth, average scattering cross-section for air molecules is calculated using an empirical formula given by Nicodet (1984):

\[ \sigma_{Rs} = 4.02 \times 10^{-28} \frac{1}{\lambda^{4+x}} \text{ cm}^2 \]  

where

\[ x = 0.389 \lambda + 0.09426 / \lambda - 0.3228; \quad 0.2 \mu m < \lambda < 0.55 \mu m \]
\[ x = 0.04; \quad 0.55 \mu m \leq \lambda < 1.0 \mu m \]

and \( \lambda \) is wavelength expressed in \( \mu m \). One can calculate Rayleigh optical depth by multiplying \( \sigma_{Rs} \) to columnar number density of air molecules. It is \( 2.153 \times 10^{25} \) molecules/cm\(^2\) for standard atmosphere at mean sea level. However, it can change due to change in pressure, which is accounted by multiplying Rayleigh optical depth calculated for mean sea level by ratio of measured ambient pressure with
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that of standard atmospheric pressure. Further, to account for finite bandwidth of filters, transmission weighted mean of optical depth is used. Optical depth due to molecular absorption is calculated using radiative transfer code SBDART (further discussed in Chapter 5). Important gases for molecular absorption for the present filters of the Sun-Photometers are Ozone, Water vapour and Oxygen. Ozone and Water vapour are variable gases in the atmosphere. Values of columnar ozone concentration are obtained from MICROTOPS-II measurements and substantiated by TOMS satellite data whenever measurements were not available. Columnar water vapour measurements were done using one of the MICROTOPS-II and substantiated by NCEP reanalysis data whenever measurements of water vapour were not available. Selection of filters for AOD measurements is done in such a manner that highest contribution of molecular absorption optical depth is less than 10%. Last column in Table 2.2 shows the sum of typical optical depth due to Rayleigh ($\tau_{\text{rayleigh}}$) and molecular absorption ($\tau_{\text{mol}}$), which needs to be subtracted from total optical depth in order to get AOD (Equation 2.23). Rayleigh optical depth is calculated for standard atmosphere. Molecular absorption in case of hand held sun-photometer is calculated for average condition over Maitri-Antarctica and that of Microtops-II is for tropical atmosphere.

$$\tau_{\text{aero}} = \tau_{\text{total}} - \tau_{\text{rayleigh}} - \tau_{\text{mol}}$$  \hspace{1cm} 2.23

2.3 Aerosol Scattering and Absorption Coefficients

In addition to extinction profile and AOD, other optical properties of aerosol, which are important for modelling aerosol radiative forcing are single scattering albedo (SSA) and asymmetry parameter ($g$). Single scattering albedo is a measure of the proportion of scattering and absorption in the light extinction by aerosol. It is defined as a ratio of scattering coefficient to extinction coefficient

$$\omega = \frac{\sigma_{\text{sr}}}{\sigma_{\text{ext}}}$$  \hspace{1cm} 2.24
where, $\sigma_{scat}$ is the scattering coefficient (m$^{-1}$) and $\sigma_{tot}$ is the extinction coefficient (m$^{-1}$). Extinction coefficient is a sum of scattering coefficient and absorption coefficient.

2.3.1 Instrumentation

Observations of scattering coefficient are carried out using Nephelometer (Model M903, Radiance Research) at wavelength 530 nm. Nephelometer measures light scattering in an airflow that passes through the scattering chamber of the instrument. Air volume is illuminated by diffuse light source from side. Photomultiplier tube views the dark trap through a scattering conical volume define by baffles with a hole in centre. Baffles are arranged in such a manner that photomultiplier tube does not see any surfaces. This geometry is chosen because light measured by photomultiplier is nearly proportional light scattered in all directions. So by calibrating Nephelometer volume scattering coefficient in the unit of m$^{-1}$ is obtained.

![M903 Optical Design](image)

Figure 2.10. Block diagram of the Nephelometer (model M903 Radiance Research, USA).

Calibration of nephelometer involves two steps known as zero calibration and span calibration. Zero calibration of nephelometer is done by measuring the scattering by particle free air. Measured scattering for particle free air is recorded
for subtraction from sample air. Scattering by air molecules will depend upon pressure, temperature and relative humidity (RH). Nephelometer has in-built pressure, temperature and relative humidity sensors. Since operating condition and calibration condition were same within 3% of pressure, no special correction is applied for the change in pressure. Sample air is pre-heated before measuring the scattering coefficient and hence very small differences are possible in temperature and RH between calibration and actual measurements. Span calibration is done using CO₂ gas whose scattering coefficient is higher by a factor of 2.61 than that of particle-free air. There are two sources of errors in scattering coefficient measurements. One is called wall scattering, which defines all type of stray light entering into Photomultiplier tube. However, this is not a serious problem for nephelometer, which is calibrated regularly, and stray light is expected to be same for particle-free air and aerosol-laden air. Another serious error occurs because of very nature of aerosol scattering i.e. Mie Scattering. Integrating nephelometer cannot measure scattered light at angles close to 0 and 180 degrees. For small particles, this is not a problem as very little light is scattered in forward direction than compared to bigger particles where large amount of light is scattered in forward direction. This error is known as truncation error. Since there is no direct way to correct this error and any indirect way may involve assumptions, which not necessarily improve the results, truncation error in measurements are ignored. Rest of the error in scattering coefficients are in the range 3 to 4 %.

Measurements of absorption coefficients are carried out using Aethalometer (Model AE-42, Magee Scientific) at seven wavelengths i.e. 370, 450, 520, 590, 660, 880 and 950 nm. Aethalometers are primarily used to measure black carbon (BC) or soot concentration in air. Measurements of soot particles are carried out using optical attenuation method. Sample air with constant flow rate for a known period is passed through a quartz filter paper and change in attenuation of light beam is monitored. Soot particles have one of the largest broad-spectrum absorption cross-sections approximately 10 m²/gm at wavelength 550 nm. This absorption is proportional to the wavelength of the incident light. Change in attenuation is
calibrated to mass concentration of BC. However there are other aerosol species in the atmosphere who have absorption in visible wavelength e.g. mineral dust, iron, etc. Hence, measurement of BC may be overestimated or can be corrected by studying the spectral dependence of absorption. However, error in BC mass is not a problem for this work since aethalometer measures the attenuation coefficient that is indeed required to calculate SSA. It is found that error due electronic noise, optical noise, and flow rate inaccuracy is less than 1%. Manufacturer of aethalometer states the accuracy in the range of 2 to 3 % by showing results from simultaneous measurements with other instruments, which uses different mechanism to measure attenuation of BC concentration.

2.4 Aerosol Size Distribution

Asymmetry parameter (g) of aerosol depends on aerosol size distribution and refractive index of particles. It is a ratio of photons scattered in forward direction to total scattered photons. Value of g ranges from -1 to +1. When photons are completely back-scattered, g is -1 and for completely forward scattering g is +1. For symmetrical scattering g will be 0. For ISRO-GBP land campaign (discussed in Chapter 4) calculations of g were carried from size distribution. For rest of the places measurements of size distributions are used as a proxy to constrain the model estimates of g.

2.4.1 Instrumentation

A quartz crystal microbalance cascade impactor (QCM) manufactured by California Measurements Inc, USA is used to measure the mass size-distribution of aerosols (Gadbavi and Jayaraman, 2004). The instrument has 10 stages with each stage sensitive to a specific size range of particles. The radii at which the collection efficiency is maximum are 8.64, 4.26, 2.24, 1.08, 0.55, 0.29, 0.16, 0.07 and 0.03 μm respectively for the stages 2 to 10. The full width at half maximum of efficiency changes from 7 to 0.03 μm for stages 2 to 10. As the name implies QCM works on the principle of cascade impactor but unlike the traditional cascade impactors, which use filter paper, QCM uses quartz crystal to give the aerosol mass size
distribution in near real time (Wood, 1979). In the cascade impactor several stages are cascaded and each subsequent stage has smaller size of nozzle. Quartz crystals are placed opposite to nozzles. Air stream coming out of nozzle takes sharp turn because of the crystal. Depending upon the size of the particle (smaller or bigger) either it will take turn with air stream or it may get impacted on crystal. Crystal surfaces are greased to capture the impacted particles. Quartz crystals have property that their resonance frequency is changed with change in their mass. Hence, by monitoring change in frequency the mass accumulated in given time over crystal is calculated. Since subsequent stages have smaller size of nozzle, speed of air coming out of nozzle will be increased and hence particles which were able to escape with airflow in previous stage, now will be impacted on crystal. There are several sources of error for example re-entrainment of particle from crystal surface, wall loss, change in relative humidity, electronics noise, etc. Error is estimated to be ~15% by running QCM with collocated Anderson impactor (Jayaraman et al., 1998).