INTRODUCTION

1.1 Background

The Earth's climate is controlled by the amount of solar radiation it intercepts and the fraction of that energy absorbed in the atmosphere. Average solar radiation received per square meter of the Earth is about 343 W/m² of which ~103 W/m² is reflected to space. The remaining 240 W/m² is radiated back to space in the form of infrared radiation to maintain equilibrium state. Black body radiation laws predict temperature of the Earth to be about 255 K for this amount of radiation, which is 18 degrees lower than the freezing point of water. Greenhouse gases present in the atmosphere keep the Earth warm at about 288 K and make it liveable (Seinfeld and Pandis, 1998). The climate, defined as the long term mean of weather is not always like the one which we experience currently. It is believed that during cryogenian period (800 to 600 million years ago) permanent sea ice covered almost the entire globe. Paleo-records show that past climatic and environmental changes on the Earth are correlated with atmospheric composition. Records from Vostok (Antarctica) ice core show that the lowest concentration for greenhouse gases such as CO₂ and CH₄ is observed during full glacial conditions. CO₂ and CH₄ concentration are found to be 190 ppmv and 0.35 ppmv respectively during the last glacial maximum, ~20 kyr BP. Increase in their concentration during deglaciation period is about 40% for CO₂ and 200% for CH₄. Ice core data also reveal that during ice age terrestrial and marine contributions to Antarctic aerosol deposition increased significantly. Concentration of calcium and sodium (crustal and marine primary aerosol reference elements, respectively) in Vostok ice were higher by factors of about 30 and 4 respectively, relative to Holocene values (Wuebbles et al., 2003). Atmospheric composition affects climate as well as gets altered by climate. This feedback loop makes climate very sensitive to small perturbations in the atmospheric composition. For example, the warmer Earth
may sustain more water vapour in the atmosphere, which can absorb more infrared radiation making the earth further warm. The causes for natural variability of climate and atmospheric composition are yet to be fully understood. Mean while the present day concentration of greenhouse gases is unprecedented in the last 420 kyr. (Petit et al., 1999) and likewise is the case for aerosols. Determining the influence of such large scale changes in the composition of the atmosphere on climate is the central objective of present day atmospheric science community. Complete understanding of emission rate of all atmospheric species, their transport and transformation, removal rate, seasonal variation, influence in radiation budget, and chemical and physical processes occurring in the atmosphere is key to acquire forecast ability for future changes of climate.

Rapid changes in atmospheric composition observed over the past century have been driven largely by increased usage of fossil fuel, intensive farming, industrial activities and biomass burning. One can deduce the anthropogenic contribution by comparing present day concentration with that of pre-industrial time (~1750 AD). Concentrations of CO₂ and CH₄ during pre-industrial time were ~280 ppmv and ~650 ppbv respectively, while the present day values are ~360 ppmv and ~1700 ppbv respectively. Similarly, sulphate concentration has increased from about 50 mg per tonne to about 200 mg per tonne of ice during the same period in ice core taken from Greenland (IPCC, 2001). Concentrations of CO₂ and CH₄ are global values whereas that of sulphate is more region specific. This is because greenhouse gases such as CO₂ and CH₄ have longer lifetimes and hence they are well mixed in the atmosphere whereas aerosols have shorter lifetime and hence their concentration shows gradient from source to remote region. Apart from large spatial variability, there are other complexities, such as poor understanding of the processes that link aerosol particles, precursor emissions and radiative effect, and lack of comprehensive global data set that can be used to verify models which contribute to large uncertainty in estimating the effect of aerosols on climate (Huebert et al., 2003). Nevertheless, globally averaged anthropogenic aerosol
forcing is estimated, which is found to be equal to the forcing by anthropogenic greenhouse gases but opposite in sign (IPCC, 2001).

Atmospheric aerosols affect Earth's radiation budget in two different pathways, one is by directly scattering and absorbing the electromagnetic radiation and is called direct effect of aerosols. Aerosol particles can also modify the lifetime and microphysical properties of clouds. Increased concentration of aerosols increases the number concentration of cloud droplets but reduce their size. This leads to an increase in cloud reflectance. Also smaller size droplets have lower precipitation efficiency and hence lifetime of clouds increases. This results in increased global cloud cover. These effects of aerosol are known as indirect effects. Influence of parameters such as aerosols which affect the Earth's radiation budget is quantified by a quantity called radiative forcing. Radiative forcing is defined as the change in net irradiance at the tropopause due to an applied perturbation to a particular parameter while holding all other atmospheric variables fixed, and the stratospheric temperature is allowed to adjust to equilibrium value (Haywood and Boucher, 2000). Adjustment of stratospheric temperature is more important for computing radiative forcing by ozone depletion in stratosphere whereas it does not make any measurable difference for aerosols. Also, aerosol radiative forcing study is no more restricted to tropopause but more importance is given to vertical distribution particularly at lower altitudes (Satheesh and Ramanathan, 2000). Concept of radiative forcing was first used to deduce temperature changes for applied perturbations in 1 D models, but was found equally useful for 3 D models. Sign convention for radiative forcing is defined in a manner that positive forcing has warming effect and negative forcing has cooling effect. Until all the feedback effects such as changes in cloud, snow and ice cover, atmospheric water vapour, winds, ocean currents, etc are accounted for, exact effect of applied perturbation to radiation budget cannot be known, nevertheless concept of radiative forcing is useful for first order assessment of the impact of applied perturbation (Ramaswamy et al., 2001).
Importance of aerosols in climate forcing of the Earth is well recognised for a long time. Hansen et al. (1990) estimate the global anthropogenic aerosol radiative forcing between -0.75 to -1.5 W/m². Charlson et al. (1992) estimate global anthropogenic sulphate forcing in the range of -1 to -2 W/m² including both direct and indirect effects. Haywood et al. (1999) have shown by comparing top of the atmosphere irradiance from general circulation model (GCM) and observations from Earth Radiation Budget Experiment (ERBE) satellite that annually and globally averaged tropospheric aerosol radiative forcing (including both natural and anthropogenic) is around -6.74 W/m² over oceans. Haywood and Bowcher (2000) discuss in detail the anthropogenic aerosol radiative forcing and present range of values for sulphate (-0.26 to -0.82 W/m²), mineral dust (+0.09 to -0.46 W/m²), fossil fuel black carbon (+0.16 to +0.42 W/m²) and organic carbon (-0.02 to -0.04 W/m²), black carbon and organic carbon from biomass burning (-0.14 to -0.74 W/m²), nitrate (-0.03 W/m²) and indirect effect of aerosols (-0.3 to -1.8 W/m²). Range of values span more than a factor of two for almost all the aerosol types. Such a large uncertainty is mainly because of insufficient data on inventories, poor understanding of mixing state of aerosols (whether aerosols are internally or externally mixed with other species), different schemes for cloud representation in sub grid level, lack of data on vertical distribution, etc. To reduce the uncertainty and enable better forecast for future climate change, world wide efforts have been made to measure aerosol properties by establishing permanent stations (Subbaraya et al., 2000, Holben et al., 1998) and through major field campaigns such as ISRO-GBP land campaign-1 (Jayaraman et al., 2005), INDOEX (Ramanathan et al., 2001 and 2002), TARFOX (Russell et al., 1999), ACE (Bates et al., 1998), SAFARI (Campbell et al., 2003) etc.

INDOEX study has shown large aerosol radiative forcing existing over the Arabian Sea and the Indian Ocean during winter months when the winds are predominantly NE, blowing from the continents towards the ocean. Large latitudinal gradient is found in aerosol radiative forcing (Figure 1.1). Surface level
aerosol radiative forcing is found to vary from -5 W/m² in southern Indian Ocean to -40 W/m² close to western coast of India (Ramanathan et al., 2001).

![Figure 1.1. Latitudinal variation in aerosol radiative forcing (including both natural and anthropogenic) observed over the Indian ocean, averaged between 40°E to 100°E longitude. (source: Ramanathan et al., 2001)](image)

The modelling studies following INDOEX have identified significant consequences of the aerosol forcing in terms of variation in the spatial distribution of rainfall pattern over India and in modifying the amplitude and frequency of El Niño. Observed decrease in rainfall over India after 1950 is attributed to regional scale aerosol radiative forcing and further emphasized that if current trends in aerosol emission continue, drought frequency will double in coming decades (Chung and Ramanathan, 2004, Ramanathan et al., 2005). Latitudinal gradient in aerosol properties over the Arabian Sea and the Indian Ocean (Jayaraman et al., 2001) suggest that major sources of aerosols lie over land. Venkataraman et al (2005) have shown that more 65% of global black carbon from biofuel is emitted in Asia and about 25% of that is emitted in India. The satellite data show large spatial and temporal variations of aerosol optical depth (AOD)
over land (Figure 1.2). AOD is high over northern and western part of India than peninsular India and the temporal variation is very high over Gujarat, Rajasthan and Gangetic basin of India.

Figure 1.2. Satellite based observation of aerosol optical depth over Indian subcontinent. (source: MODIS; http://modarch.gsfc.nasa.gov/)

Detailed ground based observation for aerosol properties over land remains sparse. Measurements on vertical distribution of aerosol properties are further sparse. Even if radiation fluxes at the top of the atmosphere and surface level are correctly accounted for, the computed heating rate within the atmosphere can be erroneous if the vertical distribution of aerosols is not properly characterised. Depending on whether the aerosols occur above or below the cloud layer, the radiative forcing can be different for the same concentration and type of aerosols, hence accurate prescription of aerosol vertical profile is very important (Haywood and Ramaswamy, 1998, Chung and Ramanathan, 2004). Lifetime of aerosols depends, apart from their size, on the altitude of their occurrence. In the boundary layer, aerosol lifetime is of the order of few days, it is a few weeks in free troposphere.
and few years in the stratosphere. Also satellite remote sensing of various parameters require knowledge on vertical distribution of aerosols (Herman et al., 1997, Torres et al., 1998).

1.2 Scope of the present work

In the context of India, aerosol properties over ocean surrounding peninsular India are extensively studied and their impact on solar radiation fluxes reaching the surface and leaving the atmosphere have been reported (Jayaraman et al., 1998, Satheesh and Ramanathan, 2000, Rajeev and Ramanathan, 2001, Coakley et al., 2002, Satheesh, 2002, Satheesh et al., 2002, Satheesh and Lubin, 2003, Ganguly et al., 2005) but relatively a few measurements on land locations exist (Ramachandran et al., 1994, Devara, 2000, Niranjan et al., 2000, Moorby et al., 2001, Moorby et al., 2002, Gupta et al., 2003, Sagar et al., 2004, Parameswaran et al., 2004). Measurements of vertical profiles of aerosol are further sparse (Jayaraman et al., 1996, Parameswaran et al., 1998, Devara, 2000, Ramachandran and Jayaraman, 2003). The study of stratospheric aerosol have been carried out from Ahmedabad in the past (Jayaraman et al., 1995a, 1995b, and 1996). Ramachandran (1995) has carried out observations of aerosol optical depth from 1991 to 1993 over Ahmedabad but for subsequent period AOD observations are not available over Ahmedabad. Though very important, no information exists on vertical distribution of aerosol in the lower troposphere over Ahmedabad where more than 90 % of aerosols reside during volcanically quiescent period.

Ahmedabad is an urban location in semi-arid region of western part of India (23°3'N, 72°52'E). Being an urban location, studies of aerosol is very important for long-term monitoring of environment degradation and source characterisation. Its proximity to Thaar desert of Rajasthan and the Arabian Sea allows one to study further the effect of changing airmass trajectory on aerosol properties and concentration. Present thesis focuses on the study of vertical distribution of aerosol in the troposphere. Vertical distribution of aerosol extinction coefficient has been studied using micro pulse lidar (MPL) system. More than 20 algorithms
have been surveyed for their feasibility to get extinction profile from lidar data and robustness of the solution for uncertainty in various assumptions. Chapter 2 of the thesis describes the instruments, data retrieval and important algorithms in detail. Seasonal variation in extinction profile, aerosol scale height and aerosol optical depth obtained by integrating extinction profile is presented in Chapter 3. Observations of aerosol absorption and scattering coefficients have been obtained for the first time over Ahmedabad. Their daily mean values for the period of about one year are also presented in Chapter 3.

Chapter 4 of the thesis discusses specific field observations made during special campaigns. Regions covered are highly heterogeneous such as Antarctica, Coastal Area of Gujarat, Rann of Kachchh, Central India, etc. Large difference in the aerosol properties is found from place to place. AOD over Antarctica is an order of magnitude lower than Ahmedabad, while vertical extent of aerosol over central India is found high by about one km in comparison to Ahmedabad.

Information gathered over Ahmedabad and other places has been used to study their impact on Earth’s radiation budget. Results and details of radiative forcing calculations are presented in Chapter 5. Results of radiative forcing are discussed in the context of their seasonal as well as spatial variation and vertical distribution. Significant influence of surface reflectance is found on the aerosol radiative forcing. It is shown that sign of radiative forcing can flip from negative to positive depending on brightness of a surface. Chapter 6 presents the major conclusions and scope for future work.