Atmospheric aerosols are particles in solid/liquid/mixed phase dispersed in the atmosphere. Their sizes span over nearly 5 orders of magnitude from $10^{-3}$ to $10^2$ μm and number concentrations (by over 10 orders of magnitude and more) in the size range and depending on location. Even though they constitute only parts per billion by volume to the atmosphere, they have the potential to implicate in many areas ranging from climate change to human health, due to the complexity of their properties. They are generated from a wide variety of natural and anthropogenic sources like sea-spray, wind blown soil dust, combustion products, volcanic eruptions, meteoric dust, vegetation, etc. On a global scale, natural aerosols are probably 4 to 5 times larger than anthropogenic component. But the regional variations in anthropogenic activities and air pollution can change this ratio significantly in areas with large-scale anthropogenic/industrial activities. Aerosols are removed from the atmosphere mainly by dry and wet deposition processes. The complex chemical composition, microphysical properties, state of mixing (external, internal or core-shell structure), size transformation processes such as coagulation and condensational growth, removal mechanisms, long-range transport to locations far away from source regions by atmospheric circulations/waves, its vertical distributions make the species highly heterogeneous-spatially, temporally and spectrally, and this heterogeneity is the primary feature which makes the species distinct from other atmospheric constituents. This large heterogeneity would result in greater uncertainties in climate impact assessment of the species.

Nevertheless, the various atmospheric manifestations (such as such as the twilight glow, the noctilucent clouds, and the occasional blue sun and moon) and its environmental and health effects have been both fascinating and worrying the mankind over decades. The mist, fog, smog and visibility have environmental concern in one hand, while inhalation of ultra-fine and toxic aerosols cause health hazard on the other. While the investigations of the impacts of aerosols on visibility and human health have a long history, the concerns on regional and global climate implications are of more recent origin. However, sustained investigations over the last several years have considerably improved our understanding and awareness on the importance of aerosols in regional and global climate. The Intergovernmental Panel for Climate Change (IPCC) did not consider aerosols as potential...
source of observed climate changes in the 20th century prior to 1995, mainly because of the lack of information of various aerosol properties, and their interaction with the Earth-Atmosphere system on one hand and the difficulties in assimilating aerosol properties in climate models with adequate sensitivity in the other. Extensive experimental investigations using different measurement techniques over multiple platforms and model simulations have been carried out during last two decades to understand the characteristics and impacts of atmospheric aerosol system. These studies revealed that, climate implications of atmospheric aerosols mainly depend on its direct interaction with the solar radiation via scattering and absorption and indirectly by modifying the albedo and lifetime of the clouds. Both these mechanisms lead to the perturbations in the radiative balance of the Earth-Atmosphere system. The large uncertainties in the aerosol radiative impacts raise questions on the quantitative assessments of the role of aerosols on the observed climate changes and future climate projections. Reduction in these uncertainties calls for coordinated and campaign mode observations well-focusing on specific themes, research strategies that will successfully integrate data from multiple platforms (e.g., ground-based networks, satellite, ship, and aircraft) and techniques (e.g., in-situ measurement, remote sensing, numerical modeling, and data assimilation). In view of this, flurry of exhaustive campaigns were carried out to characterize the aerosol properties and accurate estimation of its radiative effects under the distinct aerosol regimes over the land and ocean during the last two decades. These efforts contributed significantly to increase the level of scientific understanding from very low to medium as reported by IPCC 2007.

Notwithstanding its high density of population, increasing industrialization and urbanization, diverse living habits, geographical features and contrasting meteorological processes, studies focusing on the spatio-temporal heterogeneity of aerosol properties are rather sparse over the Indian sub continent. Thus, in backdrop of the limitations of earlier campaigns, Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB), has been formulated as the most exhaustive, multi-platform, multi-instrumented, field campaign ever conducted over the South Asian region to characterize the aerosol properties using a synergy of experimental observations from ground based network observatories, cruise measurements over the entire oceanic regions around India with high spatial resolution, concurrently with aircraft measurements of aerosol composition. Besides, being an active participant of this campaign, the author has done the analysis of the extensive
database generated over the network of observatories to evolve a space-time synthesis of aerosol characteristics over the Indian region for the first time. The observations have also been compared with the simulations using globally accepted emission inventory models.

The natural variabilities modulating the aerosol characteristics by large scale atmospheric motions or atmospheric waves is one of the least understood components in the aerosol characteristics. The modulations of aerosol properties at intra-seasonal and inter annual scales by large scale circulations and atmospheric waves of various time scales, ranging from mesoscale to planetary scale have been quantified for the first time over the subcontinent in this thesis. The associated changes in aerosol microphysics due to the natural variabilities and subsequent changes in source strengths have been examined. Also the implications of these in radiative forcing estimations have been speculated. This thesis deals with the above, organized in seven chapters.

Chapter 1 gives the brief introduction of the atmospheric aerosol system, its sources, sinks, physical, optical and chemical properties. The three dimensional distribution of aerosols and its different types of variabilities are also described subsequently. The role of aerosols in perturbing the radiation balance, hydrological cycle and the health impacts of aerosols are also discussed. A summary of earlier attempts to characterize the aerosol properties over land and oceans is discussed with special emphasis to those over Indian subcontinent. The chapter also gives description of the present campaign, ICARB and the scope of this study. The extensive database of aerosol properties from the network observatories generated during the land segment of ICARB field campaign and the period following the campaign, have been examined over the subcontinent for the first time.

Chapter 2 deals with the instrumentation and measurement techniques used for this study. These include (i) spectral aerosol optical depth (AOD) measurements using Microtops Sun-photometer, Multi-wavelength solar radiometer and CIMEL Sun-photometer (ii) BC mass concentration using multi wavelength Aethalometer (iii) total and size segregated composite aerosol mass concentrations using Quartz Crystal Microbalance.

Chapter 3 deals with the space-time synthesis of aerosol characteristics over Indian landmass and the islands with a view to examining spatial distribution of aerosol properties and its seasonal transformations. The roles of short and long-range transport are examined by identifying different advection pathways. By performing a cluster analysis, the contributions from different source regions are delineated and quantified using the
Concentration Weighted Trajectory (CWT) analysis. The study revealed that during the ICARB period, the west Asia and north-west India remained the potential source regions, which contributed significantly to the enhancement of AOD and turbidity parameter, $\beta$, and the decrease of the Angstrom wavelength exponent, $\alpha$ over the entire mainland; if it is the peninsular regions that are more impacted in April, the north Indian region including the Indo Gangetic Plain get affected the most during May. For the other seasons, varying amounts of advection from the adjacent oceanic regions, West Asia, and East Asian locations make the aerosol characteristics extremely heterogeneous both spatially and temporally.

Chapter 4 deals with the space-time synthesis of the surface mass concentrations of aerosol Black Carbon (BC), a chemically inert fine species, produced mainly from anthropogenic activities, and contributes significantly to the absorption of solar radiation and thus has strong implications to radiative forcing. The results showed vary large spatial variation in BC mass concentrations with values ranging from values as high as 25 $\mu$g m$^{-3}$ over industrial/urban locations to as low as 0.065 $\mu$g m$^{-3}$ over Arabian sea. While the diurnal patterns of these are found to vary widely depending upon the topography of the measurement locations, and the human activities in the adjoining areas, the long-range transport produce significant modulations at meso/synoptic time scales. Measured BC mass concentrations ($M_B$) from the network stations are compared with the corresponding estimates made using GOCART model simulations, which revealed that the model values are highly unrealistic over the Indian region. Detailed analysis of the time series data strongly indicated that the intra-seasonal oscillations are poorly represented in the model.

Meso-scale, synoptic scale and planetary scale atmospheric oscillations produce significant modulations to columnar AOD and surface mass concentrations through horizontal advection, convective coupling and transport. While the dispersing and recycling of species produced by a mountain grass-land fire, at an apparently upwind station through the circulation cell demonstrated the potential of meso-scale processes, the eastward propagating circulation anomalies such as Madden Julian Oscillations (periodicities of 30-60 days) and the westward propagating 13-22 days (quasi 16 day) Rossby waves were found to strongly modulate the aerosol (columnar) properties at intra-seasonal time scales upto ~ 45% of the seasonal mean values and in its spectral dependencies (change in aerosol abundance and type) over the tropics. Furthermore, the analysis of long-term time series of
monthly mean AOD delineated and quantified the modulations of columnar AOD by stratospheric Quasi Biennial Oscillations (QBO) through deep convection, troposphere stratosphere exchange and the cirrus clouds. The details are given in Chapter 5 and implications are discussed.

Changes in the synoptic airmass types (either seasonal and periodic or ergodic in nature) could produce corresponding signatures on AODs and its spectral dependencies, which result in significant deviation of the wavelength variation of AOD from the linear relationship with wavelength in a logarithmic scale, and introduce curvatures in both wavelength and optical depth domains. This causes the wavelength dependence to deviate from the simple monotonic relation expressed using the Angstrom relation. This aspect has been examined using the long term database from two stations, representing (a) coastal (TVM) and (b) oceanic (PBR) environments, in Chapter 6. Considerable variations are observed in the relationship between wavelength and AOD in log-log scale, which changed distinctively with seasons; being convex spectral curvature during winter monsoon season and concave spectral curvature during summer monsoon season as the synoptic airmass changes from continental to marine leading to drastic changes in the columnar size distributions of aerosols over Trivandrum; whereas over Port Blair, the fluctuations are more rapid irrespective of the seasons, in which the spectral curvature switches between the two types within a season/month itself. Combining these with back trajectory analysis, the role of distinct source regions in producing these are delineated and the implications of these in estimation of aerosol radiative forcing are discussed.

The major results emerged from these studies (explained in Chapter 3 to Chapter 6) are summarized in Chapter 7, along with the future scope of the studies.