

Thesis Statement

The aerosol optical and physico-chemical properties at a given location mainly depend on the local and surrounding source region of aerosol formation and are modified by local and regional meteorology besides mixing with other particles of non-local origin. The atmospheric physical processes including coagulation, growth by aging and humidification, gas-to-particle conversion, long-range transport and deposition of the aerosol particles in the atmosphere modify the aerosol characteristics, their size distribution and vertical and columnar properties. The understanding of these underlying processes which modify the distribution of aerosols in the atmosphere is of great importance for climatic studies.

Since the surface aerosol properties can be markedly different from columnar ones, due to the presence of distinct aerosol layers aloft, the surface measurements cannot represent the columnar characteristics. Similarly, the passive remote sensing instruments can only provide the columnar aerosol properties without distinguishing the different aerosol layers in the atmosphere which may be of different origin. In addition, the altitude distribution of aerosols has also important bearing in the aerosol radiative forcing, particularly when different aerosol types co-exist in the vertical.

Spatial distribution of aerosols and their heterogeneity is strongly controlled by the long-range atmospheric transport, vertical mixing, altitude distribution of aerosols, aerosol abundance in the source regions and the prevailing aerosol removal mechanisms. Aerosol loading and their properties show very strong spatio-temporal variations over the oceanic regions around the Indian subcontinent and the characterization of marine aerosols is necessary to develop a reasonable assessment of their transport impact and role of aerosols in the climate. Studying pristine marine environments can also provide a background against which the anthropogenically-influenced regions may be compared.

Abstract

This Thesis addresses the surface, vertical and columnar measurements of aerosol characteristics carried out over Hyderabad using a suite of various instruments in addition to the dedicated field campaign (W-ICARB) over Bay of Bengal (BoB) as well as satellite observations over south Asia viz. Arabian Sea (AS), BoB, Northern Indian Ocean (NIO) and continental India, in order to investigate the long-range transport of aerosols and their climatic implications.

In general, high AOD₅₀₀ with abundance of coarse-mode aerosol is seen over Hyderabad during the pre-monsoon (March to May) and monsoon (June to September) seasons associated with low Angstrom exponent ($\alpha_{400-1025}$), while during post-monsoon (October-November) and winter (December – February) seasons fine-mode aerosols dominance and steep AOD spectra are the basic features. Aerosol radiative forcing (ARF) shows larger negative ARF values at surface (-65 to -80 Wm⁻²) and top of the atmosphere (TOA) (~-17 Wm⁻²) during pre-monsoon and early monsoon, while the atmospheric heating is higher (~50-70 Wm⁻²) during the period of January to April resulting in heating rates of ~1.6-2.0 Kday⁻¹. The measurements of Black Carbon (BC) aerosol component show that BC mass fraction (F_{BC}) varies from 5-15%, while the mean values during winter, pre-monsoon, monsoon and post-monsoon are 13±1.9%, 8.19±2.16%, 7.3±1.8% and 11.8±0.18% respectively. The annual average BC mass fraction was found to be 10±3 % that contributes to the atmospheric radiative forcing by 55 ± 10 % of the composite aerosol. The BC ARF at TOA is positive for all months, thus suggesting an overall heating of the regional climate. The seasonal variations in the nature of aerosols were inferred using the graphical visualization scheme as well as columnar aerosol size distribution analysis. The results reveal a well mixed aerosol type via coagulation and condensation of particles during winter and post-monsoon while during pre-monsoon freshly emitted fine particles via secondary aerosol formation through gas-to-particle conversion and hydration process are observed. The data suggest that humidification process lead to increased hygroscopic growth of water soluble particles during monsoon season.

The lidar measurements further revealed considerable variations in the vertical distribution and concentration of aerosols within the boundary layer and in the free troposphere over Hyderabad. The majority of aerosols were found in the boundary layer below 2 km and in some cases they reached higher altitudes (~4 km) during pre-monsoon. In the boundary layer the aerosols

originated from local sources, with a larger contribution to the columnar AOD during winter, while, during pre-monsoon season these mainly originate from distinct sources from long range transport. The contribution of atmospheric boundary layer AOD (ABL-AOD) to the total AOD was found to be $93.6\pm 4.4\%$, $92.5\pm 6.9\%$, $68.5\pm 13.4\%$, $88.2\pm 8.1\%$ during winter, pre-monsoon, monsoon and post-monsoon season respectively. This seasonal variation is similar to the one that was found for the BC mass fraction indicating that the BC aerosols play an important role in the determination of columnar aerosol loading in the urban environment. The aerosol vertical profiles also revealed multiple thin aerosol layers on certain days during monsoon season.

Field measurements during the W-ICARB cruise campaign over BoB showed that the aerosol number size distribution to be bi-modal in the 72% of the cases and the highest total aerosol number concentration N_T in the range $350 - 550 \text{ cm}^{-3}$, with $\text{AOD}_{500} > 0.7$ and Angstrom exponent $\alpha_{380-870}$ values of 1.2-1.25 were observed in western and northern BoB with lower values in the southern and parts of central BoB. The eastern part of BoB, which was investigated for the first time during this campaign showed high values of N_T (200 and 300 cm^{-3}), AOD_{500} (0.39 ± 0.07) and $\alpha_{380-870}$ (1.27 ± 0.09). The altitude variations of aerosol number density measured using in-situ observations with a high flying tethered balloon, were found to be nearly steady at all locations within the convective boundary layer (up to $\sim 400 \text{ m}$), while above this height the aerosol concentration decreased except for far east BoB. A collocated examination of the air-mass back trajectories and the aerosol size distribution indicates that the aerosols advected from continental India have a pronounced natural (coarse mode) component, while those originating from east Asia are in general accumulation mode. Similarly, the satellite (Terra-MODIS) observations of AOD over the whole south Asia during the last decade (2000-2009) show an overall increasing trend of 10.17% in AOD which exhibits large spatio-temporal variation. The decreasing AOD_{550} trend over the densely-populated Indo-Gangetic Plain (IGP) during the period of April-September is an impressive finding and is found to be associated with decrease in dust activity. Since the long range transport plays a key role in aerosol load and its optical properties of a place, this study clearly showed the significant contributions of aerosols arriving over Hyderabad from the surrounding oceanic region as well as from the continental emission.