Abstract

Air quality degradation is emerging to be an issue of major concern in India. Recent investigations have shown that anthropogenic aerosols have significant impact on climate as well as on health. In fourth assessment report of IPCC, it has been mentioned that radiative effects of anthropogenic aerosols constitute one of the major uncertainties in assessing aerosol-induced climate impact. In addition to climate impacts, aerosol causes respiratory and cardiovascular diseases, air quality degradation, acidification of aquatic and terrestrial ecosystems. Characterization of anthropogenic aerosol fraction (defined as the fraction of anthropogenic aerosols to composite aerosols) is an appealing topic of research in current scenario. The first step towards achieving this goal is to separate natural aerosol from composite aerosols, which is a complex task. The main objective of this thesis work is the assessment of air quality and anthropogenic aerosol fraction over India using observations (ground-based as well as satellite-based) and chemistry-transport model. Specifically objectives are (a) assessment of air quality and anthropogenic aerosol fraction over Indian region (b) develop a method to derive natural aerosol properties over land and oceans using multi-satellite data analysis, which is first step towards separating natural aerosol effects from its anthropogenic counter parts and (c) evaluate performance of CHIMERE chemistry-transport model for Indian region and validate its suitability to air quality studies over India. In this thesis, different approaches have been followed such as ground-based observations, multi-satellite data analysis and CHIMERE transport model. We have used multi-year observations of particulate mass (PM) concentration, aerosol black carbon (BC) mass concentration and aerosol optical depth (AOD) from a network of observatories to make an assessment of ambient air quality over India. First, we have developed a method to estimate dust and sea-salt optical depth using multi-satellite data analysis. This enabled the determination of anthropogenic aerosol fraction over land and ocean and we have validated this method by comparing against observations. Surprisingly, even over desert locations in India and Saudi Arabia, the anthropogenic fraction were unexpectedly high (~0.3 to 0.4) and the regionally averaged anthropogenic fraction over India was 0.62±0.06 (for the year 2004). The CHIMERE chemistry-transport model was used to simulate PM, BC and AOD over India and are compared with measurements. Evaluation of CHIMERE output shows that diurnal and seasonal trends are captured reasonably well by the model. It was found that absolute

magnitudes differ substantially during monsoon months. Model simulations are also used to estimate anthropogenic fraction over Indian region and are compared with observations. Implications of the results are discussed.

Mineral dust constitutes the single largest contributor of natural aerosols over continents. The first step towards separating natural aerosol radiative impact from its anthropogenic counterparts over continents is to gather information on dust aerosols. The infrared (IR) radiance (10.5–12.5 mm) acquired from the Kalpana satellite (8-km resolution) was used to retrieve regional characteristics of dust aerosols over the Afro-Asian region during the winter of 2004, coinciding with a national aerosol campaign. Here, we used aerosol-induced IR radiance depression as an index of dust load. The regional distribution of dust over various arid and semi-arid regions of India and adjacent continents has been estimated, and these data in conjunction with regional maps of column aerosol optical depth (AOD) are used to infer anthropogenic aerosol fraction. Surprisingly, even over desert locations in India and Saudi Arabia, the anthropogenic fraction were relatively high (0.3 to 0.4) and the regionally averaged anthropogenic fraction over India was 0.62 ±0.06.

Sea-salt constitutes the single largest contributor of natural aerosols over oceans. We derive sea-salt aerosol distribution using a method utilizing multi-satellite data analysis. This information was used in conjunction with dust aerosols retrieval to calculate anthropogenic fraction over land and ocean. First, we derived a relation between MODIS AOD and NCEP wind speed at the sea-surface. An exponential increase in AOD as a function of wind speed was observed from mid of southern ocean to northern Arabian Sea. Latitudinal variation of wind independent component of optical depth ($\boldsymbol{\tau_0}$) and wind index (**b**) was used to estimate the sea-salt optical depth over Arabian Sea. The value of $\boldsymbol{\tau_0}$ showed an exponential increase as we move towards north from 35°S while **b** showed linear increase. The derived relations for the τ_0 and b have been used to derive the sea-salt AOD distribution over oceanic regions in the domain (Eq-30°N; 30°E-110°E). Then we subtract the natural aerosol contribution from composite AOD data from MISR to obtain anthropogenic aerosol fraction. Over Indian region, high anthropogenic fraction was observed over northern belt specifically Indo-Gangetic Plains (IGP). Annually averaged anthropogenic fraction over Indian domain (4N-29.5N; 67E-88.5E) is ~0.43. Further, we have investigated the impact of sea-surface winds on sea-salt radiative effect in visible and infrared region with the help of SBDART radiative transfer model. The

SBDART simulations have shown that at 15 m s^{-1}, sea-salt induced shortwave cooling at the sea-surface was -86 W m^{-2}.

Derivation of anthropogenic aerosol fraction over whole Indian domain has demonstrated the importance of anthropogenic aerosols. This observation motivated us to examine the air quality over Bangalore, a fast growing city in India. We have analyzed data from ground based measurements of particulate matter, observations from satellites and also model simulations. Comparison with national threshold indicates that more than 50% of observations were above the residential threshold. To represent the air quality of Bangalore we have calculated the air quality index (AQI) for air pollutants. Coarse spatial and temporal resolution of observational data is one major shortcoming in such analysis. Therefore, satellite observations are alternative to quantify the air quality over large area. We have used MODIS AOD and RSPM to develop an empirical relation between these two parameters. A reasonably good agreement was observed between measured RSPM and RSPM derived using satellite data (by applying empirical relation).

The CHIMERE chemistry-transport model was used to simulate PM, BC and AOD over India and are compared with measurements. Evaluation of CHIMERE output shows that diurnal and seasonal trends are captured reasonably well by the model. It was found that absolute magnitudes differ substantially during pre-monsoon and monsoon months. Model simulations are also used to estimate anthropogenic fraction over Indian region and are compared with observations. Implications of the results and future scope are discussed. The validation of model results suggests that CHIMERE model is suitable for simulating air quality over India with reasonable accuracy. This would in turn help us to address the impacts of air pollution on regional climate and help policy makers in order to reduce the air pollution.

In summary, we have developed a new method to infer natural aerosol (sea-salt and dust) properties using multi-satellite data analysis. This technique has been applied to derive anthropogenic aerosol fraction over Indian region. Surprisingly, even over desert locations in India and Saudi Arabia, the anthropogenic fraction were relatively high (0.3 to 0.4) and regionally averaged anthropogenic fraction over India was 0.62±0.06 in 2004. This study indicates that multi-satellite observations can provide a powerful tool in monitoring air quality. We have noticed that anthropogenic fraction was 0.62 in 2004 and reduced to 0.43 in 2008. Major anthropogenic aerosol over India is BC and decreasing trend in BC could be one of the reasons for the decrease in anthropogenic fraction from 2004 to 2008. The CHIMERE chemistry-transport model was used to simulate PM, BC and AOD over India and are compared with measurements. Evaluation of CHIMERE output shows that diurnal and seasonal trends are captured reasonably well by the model. It was found that absolute magnitudes differ substantially during pre-monsoon and monsoon months. Presence of elevated aerosol layers during these seasons could be one of the sources for such discrepancy. Model simulations of anthropogenic fraction over Indian region are compared with observations and found good agreement. Results from this thesis moves us one step forward to reduce the uncertainties involved in anthropogenic aerosol fraction, its spatial and temporal distributions and regional distribution of OC/BC ratio, which are most important parameters in order to assess the climate forcing by anthropogenic aerosols.