Abstract

Increasing aerosol loading over the south Asian region, holds a potential of, perturbing the south Asian monsoon system and the Himalayan glacial coverage, and thus affecting lives of $(1/4)^{th}$ of the world's population. Though space-based or ground-based measurements assists us in keeping an account of the composite aerosol loading over the region, they have their own limitations. Numerical models on the other hand help us, regionalize our understanding of aerosol-climate interactions and moreover, predict future scenarios, which makes them a favorite tool of policy makers. Nonetheless, realizing the consequences of such policies on the human-life (especially over the densely populated south Asian region), such numerical models are first needed to be examined for 'present-scenario' before utilizing them for future predictions.

Motivated by the need of evaluation of such chemistry transport models over the Indian region, we in the first part of this thesis, have examined the performance of 3 such models over the region. The performance of the models in simulating columnar Aerosol Optical Depth (AOD) and mass concentration of the aerosol species- Black Carbon (BC) are evaluated by comparing against satellite based retrievals of AOD, and station as well as aircraft based measurements of BC carried out over multiple locations across the country. It is found that the online regional chemistry transport model WRF-Chem underestimates the columnar AOD over the Indian region vis-a-vis the satellite retrievals by factors ranging upto 2. Additionally, the model is seen to underestimate the near-surface BC mass concentrations as compared to the station measurements by a spatio-temporally varying factor ranging between 2-5. The overestimation related to the simulations of meteorological parameters namely the planetary boundary layer height and the near-surface wind speeds, and the unrealistic emissions of BC within the model appear to be the primary causes for such BC related underestimation within the model.

The unrealistic emissions also appear to be responsible for the underestimated AOD with the model simulations. Continuing this evaluation exercise, we next have inter-compared the performance of WRF-Chem with a global chemistry transport model SPRINTARS in simulating aerosol loading over the Indian region. The models differ in many aspects including horizontal resolution, domain (regional vs global) and meteorological formulation (free meteorology vs nudged meteorology). In-spite of such a large differences in their set-up, both the models look to underestimate near-surface BC mass concentration vis-a-vis the station measurements by similar margins. Thus, a model like SPRINTARS with 'close-to-realistic' meteorological formulation also underestimates near-surface BC over the Indian region, and thus underlines the limited role played by meteorological forcing and the relatively major role played by emissions in such BC related underestimations. Utilizing, the realistic formulation of meteorology within SPRINTARS, we further quantify the underestimation in 5 widely used emission inventories for BC over the Indian region, which is found to range within 1.5 to 2.9. Unlike BC, the model simulated AODs considerably differ from each other, due to the differences associated with-1) simulated dust mass concentrations, dust emission formulations and near-surface winds especially over the dust source region, 2) optical properties of BC within the model simulations. The limited role played by meteorology in causing BC related underestimations within the model simulations, is further explicitly verified in evaluation of the global chemistry transport model HadGEM-3 over the Indian region. The model has been run in 4 different combinations including different chemistry mechanisms (CLASSIC and GLOMAP-mode+UKCA) and meteorological formulations (free meteorology and nudged meteorology). It is found that the model underestimates the near-surface BC mass concentration vis-a-vis the station measurements, by a large margin, in all the 4 different combinations. Surprisingly, the HadGEM-3 model overestimates AOD over the Indian region, especially over the Indo-Gangetic-Plain (IGP), vis-a-vis the satellite and ground-based measurements. The overestimated values of boundary layer relative humidity, which are further related to cold-bias in surface temperature and overestimated specific humidity, appear to be partly responsible for such a behavior of the model. Additionally, the Mass-Extinction-Cross-section (MEC) and its variation with RH, for Sulphate aerosol within the HadGEM model appear to be mainly responsible for such an overestimated AOD

over the IGP region. Re-visiting, the major role played by emissions in causing BC related underestimations in model simulations over the Indian region, we next have modified emission of BC within WRF-Chem by introducing a diurnal cycle and a spatially uniform multiplication factor of 3. The model's performance in simulating BC over the Indian region gets substantially improved upon such modifications. The modified version of the model still underestimates the AOD over the Indian region vis-a-vis the satellite retrievals. As a result, it is found that the model underestimates the aerosol radiative forcing at surface, at Top-of-the Atmosphere and within atmosphere vis-a-vis previous observational estimates over different stations across India.

Acknowledging, unrealistic AOD simulation as one of the outstanding issues for chemistry transport models over the Indian region, we in the second part of this thesis, have examined the effects of assumptions regarding state of mixing of aerosol species on AOD simulations over the Indian region. Thus, to first examine the ambient state of mixing of aerosol species over the Indian region, aerosol samples are collected using TISCH High Volume Sampler in the metropolitan city of Bengaluru, in southern part of India. Upon the subsequent Scanning Electron Microscope (SEM) analysis coupled with Electron Dispersive X-rays (EDX) technique to identify the morphology of mixing and the corresponding chemical composition, it is found that the prevailing state of mixing of aerosol is 'shell-core' with SiO_2 (dust-origin) as the core and carbon-SiO_2-others (Calcium, Aluminum, Magnesium) (i.e. BC+Dust) combination as the shell. On an average, 63% area of the core is found to be coated by the shell. Carbon amounts to 28% of the entire composition which is equivalent to 45% of the shell area while the remaining portion of the shell is seen to be composed of SiO_2 and other dust-origin elements. The AOD and other related parameters are computed for the ambient state of aerosol mixing by making use of Dr. Warren Wiscombe's mie-scattering code for layered spheres, and are further compared with that for the purely external mixture scenario (common assumption in chemistry transport models). It is found that the observed state of mixing induces only +15% changes in the AOD vis-a-vis purely external mixture scenario. The computed SSA also shows limited reductions (upto 5% near-surface) vis-a-vis the external mixture case. Moreover, the changes in AOD due to shell-core state of mixing are found to be robust and lesser sensitive to the available

mass of dust and BC, and the assumed size distribution related properties of the dust species. The corresponding relative radiative perturbations due to the prescription of the shell-core state of mixing of BC aerosol vis-a-vis the purely external mixture scenario are found to be 2-5 times lower than the previous estimates. Thus, this exercise suggests that the prescription of the ambient state of mixing of aerosol species over the Indian region would induce lesser changes in AOD and related radiative forcing vis-a-vis the external-mixture assumption, than previously thought and thus the 'external-mixture' assumption for aerosol species may not induce large errors in AOD computations over the Indian region

Simulated vertical profiles of aerosol species play a critical role in AOD computations within a model. Realizing its significance, in the last part of this thesis, we have examined the ability of the modified version of regional chemistry transport model WRF-chem in simulating the elevated sharp and confined layers of BC, observed in the recent in-situ measurements using high-altitude balloons, over the Indian region. The causes behind formation such confined layers of BC at high altitudes are investigated. While the model realistically simulates the meteorology over the balloon-flight region, it does not capture the high-altitude confined layers of BC, without prescription high altitude emissions of BC. With the help of additional simulations, our study demonstrates that, high-flying aircrafts are the most likely cause for those elevated BC layers. Furthermore, we show that such aircraft-emitted BC can get transported to even upper tropospheric/ lower stratospheric heights ($\sim$ 17 km) aided by the strong monsoonal convection occurring over the region, which is known to overshoot the tropical tropopause leading to injection of tropospheric air mass (along with its constituent aerosols) into the stratosphere. We show observational evidence for such an intrusion of tropospheric BC into the stratosphere over Indian region, using extinction coefficient and particle depolarization ratio data from CALIOP LIDAR on-board the CALIPSO satellite. We hypothesize that such intrusions of BC to lower stratosphere and its consequent longer residence time in the stratosphere would have significant implications for stratospheric ozone, considering the already reported ozone depleting potential of BC.

Thus, our study has highlighted the causes behind unrealistic simulations of aerosols over the Indian region. It has also shown that, the possible effects of realistic state of mixing of

BC on AOD and related radiative forcing over the Indian region, could be lesser than previously thought. These could serve as guidelines for further developments in chemistry transport models for the Indian region. Our study has also demonstrated the significance of high-altitude emissions of BC, especially over the strong convective regions of the world.