Abstract

According to the latest IPCC report, eastern and south Asia, particularly India and China, continue to significantly contribute to the global aerosol burden. India is the home to the second-largest population in the world. Therefore, climate change and air pollution have significant socio-economic consequences, especially in an emerging economy like India. Short-lived climate pollutants (SLCPs) are a class of aerosols that act as both climate forcers and air pollutants. SLCPs exist in the atmosphere from a few hours to a few days or months, and carbonaceous aerosols (CAs) are an essential category of SLCPs. CAs are mainly produced either directly (as primary particles) or indirectly (as secondary particles due to chemical reactions in the atmosphere) due to incomplete combustion of fossil fuels and open-air biomass burning. CA constitutes black carbon (BC) and organic carbon (OC). BC exerts a positive forcing while OC exerts a negative forcing at the top of the atmosphere. Therefore, mitigating emissions of CAs are expected to result in crucial climate-health co-benefits.

Despite their major role in climate forcing, considerable knowledge gaps exist in this region regarding their life cycles and radiative feedback. These gaps arise due to complex topography, heterogeneous aerosol distribution, measurements at limited locations, and disagreement of the inter-model estimates. Knowledge gained from field studies is limited to point locations, and the meteorological response to the CA radiative forcing cannot be investigated using observations. The magnitude of dynamic response can be estimated by climate models. Accurate representation of CAs in the climate models is critical for reducing uncertainty in estimated climate forcing. Model uncertainties can result from the model’s inability to accurately represent aerosol processes, composition and emission fluxes.

This thesis investigates the direct radiative effects of CAs on the Indian climate using a regional climate model - RegCM4.6. To achieve the thesis objective, at first two of the above-

mentioned model uncertainties – (i) ageing (aerosol process) of the carbonaceous aerosols in a regional climate model RegCM4.6 and (ii) emission inventory have been addressed. The model is simulated over the South-Asian CORDEX domain for 2010 only for the two-step augmentation. Then the augmented RegCM4.6 has been simulated with total and source-apportioned anthropogenic aerosols to study the CA direct radiative effects for the period 2006-2015. All the numerical experiments conducted in this thesis considered anthropogenic aerosols only.

First, a dynamic ageing parameterization scheme has been implemented in RegCM4.6. Freshly emitted CAs are hydrophobic in nature. Conversion of hydrophobic to hydrophilic carbonaceous aerosol is called 'ageing' and, in general, is influenced by physical processes like condensation and coagulation. In RegCM4.6, an e-folding fixed ageing time of 1.15 days (~27.6 hours) is considered, while in reality, the ageing is dynamic and depends on coagulation and condensation. After implementing the dynamic ageing scheme, it is found that the conversion of hydrophobic to hydrophilic aerosols took place in <10 hours over the polluted regions, especially over the Indo-Gangetic Basin (IGB), with almost 6-7% increment in anthropogenic AOD (AAOD). The results demonstrate the importance of improving aerosol representation in the climate models for a more realistic climate impact assessment. Furthermore, this work is the first-ever study on aerosol 'ageing' conducted in the Indian context.

The dynamic ageing scheme represented a more realistic atmosphere, but the underestimation in CA mass concentrations persisted. In the next step, a region-specific emission inventory has been incorporated in the model. Simulations have been carried out to understand the relative importance of aerosol processes and emission fluxes and their combined impact in improving the model performance. Results demonstrated that the combined effect of the new ageing scheme and regional emissions reduced the model-to-observation mismatch to a greater extent than the improvement due to only either of them. Therefore, the effect of each of the model uncertainty is non-linear, and the mean normalised bias for BC concentration reduced from -69% to -51% w.r.t the in-situ data, with the highest improvement observed in the polluted IGB where the regional background aerosol loading is high. The customization enhanced AAOD from 6-7% (improvement due to only ageing) to ~19% (due to ageing and regional emissions together). The results suggest that the combined effect of emission fluxes and more realistic aerosol processes are crucial to improving the aerosol representation in the climate models.

After customizing RegCM4.6, direct radiative effects of anthropogenic aerosols have been investigated. Considering only CA aerosols are far from the realistic atmosphere, total anthropogenic direct radiative effects have been considered. However, only CA simulations have been performed to separate out the sulphate contribution. The results show that the augmented model is able to capture the seasonal CA distribution even on a longer timescale. Additionally, the anthropogenic feedback mechanism on the Indian climate is not uniform from 2006-2015 and varies seasonally. Tracer concentrations are highest during winter and lowest during the monsoon. AAOD showed higher values over more polluted IGB, resulting in a strong surface dimming. This, in turn, lowered the monsoon rainfall over the IGB. Variations in wind convergence, transported anthropogenic aerosols and higher(lower) residence time due to lower(higher) removal of the tracers resulted in positive (negative) ΔAAOD. The current study is the first of its kind where the CA direct radiative forcing has been analysed simultaneously for all four seasons.

The CA direct radiative forcing is source-specific over Asia. Therefore, in the final working chapter of the thesis, anthropogenic direct radiative effects have been investigated for five major anthropogenic sources. For this, the ‘subtraction’ method is followed, where in each successive simulation, emissions from one particular sector have been switched off, and the changes in radiative forcing and meteorological response relative to the simulations in the previous chapter (control for this case) are interpreted as the contribution of that particular sector. The domestic sector emerged as the dominant contributor to BC and OC over the IGB, while the industries and the energy sector are the dominant contributors to sulphate (Sadavarte & Venkataraman, 2014). In fact, domestic sector emerged to be the most significant contributor to atmospheric heating (almost 40 – 50%) throughout the year and over the entire country. Other sectors contributed an annual average of, within, 30% to the atmospheric heating with varying seasonal magnitudes.

The current thesis work demonstrates the importance of addressing model uncertainties for an improved model-to-observation ratio. Also, long-term, seasonal and source-apportioned analysis is crucial for investigating aerosol direct radiative forcing under changing climate.