Spectral Aerosol Optical Depths and Radiative Forcing : Seasonal and Spatial Variations

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Abstract

Atmospheric aerosols modulate the Earth - atmosphere radiation balance by scattering and absorbing the incoming solar and outgoing terrestrial radiation, and the strength of this perturbation is quantified as the "*aerosol radiative forcing*". The thesis work concerns with the regional and seasonal variations of aerosol optical and radiative properties, and the uncertainties in aerosol radiative forcing associated with mixing state of aerosols. The mixing state of aerosol can also have regional and temporal variations depending upon the availability of different aerosol species and favorable meteorological conditions. The objective of the work is to study the spatio-temporal variabilities in aerosol optical properties and their effect on aerosol radiative forcing over, urban, polluted and other different environments spread across the globe. The probable mixing state of aerosol is determined using measured and modeled spectral aerosol optical properties, and the seasonal variations of aerosol mixing state over environmentally distinct locations (such as urban, polluted, dust dominant, and biomass burning influenced regions) over the globe including the Indo-Gangetic plain (IGP), and the implications of aerosol mixing state on aerosol radiative forcing are studied.

Aerosol radiative forcing at the Earth's surface is estimated by collocated measurements of broad-band global fluxes and aerosol optical depths (AODs) over an urban location, Ahmedabad in western India, during 2008. AOD at 0.5 μm shows large seasonal variability with higher values (0.52) during monsoon. The enhancement in AOD during monsoon is mainly due to increase in relative humidity which overwhelms the effects of wet removal of aerosols and addition of sea salt. The influence of different single scattering albedo (SSA) values on aerosol radiative forcing is determined using the SSA obtained from ground-based measurements and columnar SSA derived from remote sensing instrument and model. Forcing efficiency for monsoon season is lower when compared to other seasons due to higher SSA in monsoon. Model estimated surface forcing using surface based SSA is about two times higher than observed forcing for different seasons except for monsoon. However, model estimated forcing using columnar SSA agrees well with observations except in monsoon.

The influence of aerosol mixing state on aerosol optical properties and radiative forcing is investigated over Ahmedabad. Aerosol forcing at the surface calculated from measurements matches well with forcing estimated for probable mixing states determined for SSA derived from Ozone Monitoring Instrument (OMI) except in monsoon. Probable mixing states of aerosols and variable mass fractions of aerosol species (black carbon (BC), dust, sea salt, water soluble (WS) and insoluble) involved in core-shell mixing vary as a function of season and with SSA. Atmospheric forcing for probable mixing states derived for in situ SSA is $\sim$ 3 to 7 times higher when compared to those obtained for OMI SSA. The study reveals that aerosol mixing state is important in assessing the impact of aerosols on regional and global climate.

Seasonal variations in mixing states of aerosols over an urban and a rural location in the Indo-Gangetic plain (IGP) are determined. More than one probable mixing state is identified during a season over the urban (Kanpur) and rural (Gandhi College) locations of IGP. The degree of mixing i.e. percentage mass fraction of aerosols involved in core-shell mixing is found to exhibit seasonal variations. Different fractions of BC and WS aerosols in core-shell mixing emerges as probable mixing state during winter, monsoon and post-monsoon over Kanpur. Differences exist between measured and model derived asymmetry parameter (g) owing to non-sphericity of aerosols. However, aerosol radiative forcing is found to be weakly sensitive to the variation in g.

Aerosol mixing states are determined over environmentally distinct locations *viz.*, Maryland, Mexico City, Tamanrasset, Djougou, Mongu, Abu Dhabi, Karachi, Singapore, Gwangju and Osaka spread across the globe and their influence on aerosol radiative forcing is examined. Over locations influenced by biomass burning aerosols BC (core)-water soluble (WS, shell) is a preferred mixing state, while dust gets coated with anthropogenic aerosols (BC, WS) over urban regions influenced by dust. In Abu Dhabi, the change in aerosol forcing is maximum during pre-monsoon and monsoon owing to significant differences in SSA between external mixture (low SSA) and core-shell (high SSA). Karachi, a dust dominated urban location, exhibits similar features as that of Abu Dhabi. The spatiotemporal variations in mixing state of aerosols and their radiative radiative impact will be useful in regional and global climate assessment.

Key words: *Atmospheric aerosols, Aerosol optical depth, Single scattering albedo, Radiative forcing, Observations, Model estimates, Mixing state, Urban, polluted and rural locations, Environmentally distinct regions, Regional-Global scale influence.*