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Modulation of aerosol radiative forcing due to mixing state in clear and cloudy-sky: A case study from Delhi National Capital Region, India

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Aerosol properties change with the change in mixing state of aerosols and therefore it is a source of uncertainty in estimated aerosol radiative forcing (ARF) from observations or by models assuming a specific mixing state. The problem is important in the Indo-Gangetic Basin, Northern India, where various aerosol types mix and show strong seasonal variations. Quantifying the modulation of ARF by mixing state is hindered by lack of knowledge about proper aerosol composition. Hence, first a detailed chemical composition analysis of aerosols for Delhi National capital region (NCR) is carried out. Aerosol composition is arranged quantitatively into five major aerosol types - accumulation dust, coarse dust, water soluble (WS), water insoluble (WINS), and black carbon (BC) (directly measured by Athelometer). Eight different mixing cases - external mixing, internal mixing, and six combinations of core- shell mixing (BC over dust, WS over dust, WS over BC, BC over WS, WS over WINS, and BC over WINS; each of the combinations externally mixed with other species) have been considered. The spectral aerosol optical properties - extinction coefficient, single scattering albedo (SSA) and asymmetry parameter (g) for each of the mixing cases are calculated and finally 'clear-sky' and 'cloudy-sky' ARF at the top-of-the-atmosphere (TOA) and surface are estimated using a radiative transfer model.

Comparison of surface-reaching flux for each of the cases with MERRA downward shortwave surface flux reveals the most likely mixing state. 'BC-WINS+WS+Dust' show least deviation relative to MERRA during the pre-monsoon (MAMJ) and monsoon (JAS) seasons and hence is the most probable mixing states. During the winter season (DJF), 'BC-Dust+WS+WINS' case shows the closest match with MERRA, while external mixing is the most probable mixing state in the post-monsoon season (ON). Lowest values for both TOA and surface 'clear-sky' ARF is observed for 'BC-WINS+WS+ Dust' mixing case. TOA ARF is 0.28±2.4, 2.2±1.1, -1.4±1.4, -0.15±0.13, while, surface ARF is -16.4±3.1, -7.6±1.7, -31.5±4.7, -17.1±8.4, respectively for the MAMJ, JAS, ON and DJF seasons. Post-monsoon and winter season shows negative values of TOA ARF, hence suggest 'cooling'. The associated heating rate profiles show higher values for 'WS-BC+Dust+WINS' case as compared to other cases, with relatively large values during the winter and post-monsoon seasons, while lower value was observed for 'BC-WINS+WS+Dust'.

We examined the modulation of clear sky ARF by 'water-cloud' and 'ice-cloud' separately. The seasonal mean ARF for both water and ice clouds show nearly similar characteristics as observed for clear-sky case, with relatively large ARF at TOA and surface in water cloud case as compared to ice cloud during all the seasons. As a result, the associated heating rate is also relatively higher in water cloud case as compared to ice cloud. Such large modulation of ARF due to mixing state calls for a coordinated effort to create a mixing state database for this region to reduce the uncertainty in climate forcing.