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Can the State of Mixing of Black Caron Aerosols Explain the Mystery of 'Excess' Atmospheric Absorption?

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Aerosols are capable of modifying incoming solar and outgoing infrared radiation. The consequent change in radiative fluxes caused by aerosols is termed 'radiative forcing'. In order to understand the response of the Earth-Atmosphere system to anthropogenic aerosol forcing, it is essential to know the relative impact of various aerosol species on the radiative budget. Most of the models used for estimating the direct radiative forcing have assumed that various aerosol species are mixed externally or internally. Recent investigations have suggested that more atmospheric absorption (two to four times larger than present estimates) was needed to yield the best agreement between observed and simulated surface radiative fluxes and hypothesized the presence of an 'unidentified' absorber. However, it is possible that one aerosol species may be coated over another species to form core-shell structure and resulting radiative impact may be significantly different than those of internally mixed or externally mixed aerosols. It is common practice to incorporate measured aerosol properties in radiative transfer models to estimate radiative forcing. This often involves assumptions about how various aerosol species are mixed (called 'mixing state'). Most of the estimates of aerosol direct forcing due to absorbing aerosols have assumed that aerosols are either externally-mixed (various aerosol species exist independently) or internally mixed (where optical properties are volume averaged). Here, we show using observed data on size segregated aerosol chemical composition along with observations from ground based radiometers over tropical Indian Ocean that past estimates of climate forcing due to anthropogenic black carbon aerosols represent the lower bound and the actual values may be quite larger than that estimated by Intergovernmental Panel for Climate Change (IPCC). Our study suggests that changes in the state of mixing of black carbon aerosols may be one of the possible causes for 'excess' atmospheric absorption reported earlier by many investigators.