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Implications of multiple scattering on the assessment of black carbon aerosol radiative forcing

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ABSTRACT

The effects of radiative coupling between scattering and absorbing aerosols, in an external mixture, on the aerosol radiative forcing (*ARF*) due to black carbon (BC), its sensitivity to the composite aerosol loading and composition, and surface reflectance are investigated using radiative transfer model simulations. The *ARF* due to BC is found to depend significantly on the optical properties of the 'neighboring' (non-BC) aerosol species. The scattering due to these species significantly increases the top of the atmospheric warming due to black carbon aerosols, and significant changes in the radiative forcing efficiency of BC. This is especially significant over dark surfaces (such as oceans), despite the *ARF* due to BC being higher over snow and land-surfaces. The spatial heterogeneity of this effect (coupling or multiple scattering by neighboring aerosol species) imposes large uncertainty in the estimation *ARF* due to BC aerosols, especially over the oceans.

1. Introduction

Despite extensive studies in the recent years to reduce the uncertainties in the magnitude of the aerosol direct radiative forcing (*ARF*) [4,11,12], the accurate estimation of *ARF*, especially the speciated *ARF* (*ARF* due to individual aerosol component of the composite aerosol system), still remains a challenge [21]. In order to delineate the climate implications of the anthropogenic aerosols and to propose the mitigation strategies, it is essential to understand (i) the fractional contribution of different aerosol species to the total aerosol load, (ii) the three dimensional distribution of speciated aerosol properties, (iii) contribution of sectorial and regional emission sources and (iv) the species specific *ARF*. Even though, the magnitude of the global mean aerosol optical depth simulated using the climate models are comparable to those observed by satellites, significant biases are observed in simulating the speciated aerosol properties; for example black carbon (BC) over South Asia [9,14]. However, it is a common practice to report the speciated aerosol properties and radiative forcing values, regardless of limited validation experiments. Even the IPCC reports have considered individual *ARF*s for sulfate, BC, organic carbon (OC) and dust aerosols based on model simulations [4,12].

During the recent years, speciated *ARF*s have been extensively studied using global/regional climate models, capable of simulating the three dimensional distribution of aerosol species in time [18,11,21]. Historically, species-specific *ARF* estimations have been carried out for a single component, hypothetical aerosol system; mainly for sulfate and BC [2]. During the past few decades, simple and single component aerosol system has been evolved to multi-component, externally or internally mixed aerosol system having complex interactions with cloud, cryosphere, and biosphere in addition to aerosol–radiation

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interactions. In this context, it is highly relevant to understand the effect of neighboring aerosols on speciated *ARF*, since there are several studies using the conventional single component approach for calculating the speciated *ARF*, without considering the effect of neighboring aerosols. In contrast to the externally mixed aerosol system, several studies were carried out to understand the contribution of neighboring aerosols to the speciated aerosol properties of an internal mixture or core–shell structured aerosol system, where the amplification of optical properties of aerosols is quite obvious [5].

The current state-of-the-art models simulate multi-component aerosol mixtures (external and internal mixtures) and a few studies investigated the non-linear effects of optical properties and radiative forcing due to aerosol mixing [5,7]. There exists a large uncertainty in the state of mixing and it is difficult to quantify the relative contribution of external and internal mixing in the global aerosol system. In general, the level and percentage of mixing is yet to be quantified accurately. The gas-to-particle reactions, coagulation, hygroscopic growth, cloud processing and ageing of aerosols; all support the transformation of externally mixed aerosol system to internally mixed aerosol system [7]. In general, representation of the mixing state of aerosols is equivocal. For example, while climate models represent the aerosol system as fully internally mixed or externally mixed [19], satellite retrievals and in-situ measurements generally assume the aerosol system to be externally mixed. Most of the climate models that participated in the IPCC, [4] report, considered externally mixed aerosol system for radiative impact assessments. The actual state of mixing may be a combination of internal and external mixing, with spatio-temporal variations in the percentage of mixing rather than a complete internal or external mixture. However, it is reasonable to expect that some of the hydrophobic aerosols (like BC and dust) may exist as externally mixed especially over source and outflow regions. So several studies considered externally mixed aerosol system for calculating the aerosol radiative forcing [15].

In the case of an external mixture, the estimation of speciated *ARF* by considering only one particular aerosol species in the atmosphere could lead to a systematic error due to not accounting for the contributions of scattering-coupling due to 'neighboring' aerosols (i.e., other aerosol species present in the composite mixture); nevertheless it has not been seriously attempted. Investigating this in detail, in this paper, we quantify this effect in the estimation of speciated *ARF* in the presence of externally mixed aerosol species and its sensitivity to composite aerosol loading, composition and surface reflectance.

2. Experimental data, model and radiative transfer simulations

To examine the effect of neighboring aerosols on the speciated *ARF* and its sensitivity to different aerosol/surface conditions, numerous simulations were carried out using a radiative transfer model, and aerosol chemical models derived from the measurements and chemical transport model simulations. The aerosol data used for the sensitivity analysis in this study have been taken from the Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB) [10] experiment, conducted over Bay of Bengal and Arabian Sea during March–May, 2006. The rationale for this has been the availability of extensive data on the physical, chemical and optical properties of aerosols from collocated measurements. Several earlier publications have already discussed the instruments, methodology, data collection, uncertainties and cruise details of ICARB, hence these are not repeated here (references are there in [10]). Nevertheless, these results are applicable, in general, for any externally mixed composite aerosols. Based on the measured properties of composite aerosols, an aerosol model has been developed using the Optical Properties of Aerosols and Cloud (OPAC) model [3] by constraining spectral aerosol optical depth (AOD), spectral single scattering albedo and BC mass fraction as described in Moorthy et al. [10]. This aerosol model has major contribution from water soluble components (69%) to the total AOD (0.3 at 500 nm) followed by sea salt (15%), BC (12%), dust (3%) and insoluble (1%) aerosol species [13]. In this study, we consider only externally mixed aerosol system in which optical depth $\tau = \sum_i \tau_i$, single scattering albedo $\omega = (\sum_i \tau_i \omega_i / \sum_i \tau_i)$ and asymmetry parameter $g = (\sum_i \tau_i \omega_i g_i / \sum_i \tau_i \omega_i)$, where τ_i, ω_i and g_i are the aerosol optical depth, single scattering albedo and asymmetry parameter of the i^{th} aerosol species [3]. Using the ICARB aerosol model, speciated AOD, single scattering albedo and phase function have been estimated following the Hess et al. [3] model [10] by assuming an externally mixed aerosols system. Clear-sky radiative fluxes reaching at the surface and top of the atmosphere are simulated for aerosol (F_{aero}) and no aerosol ($F_{no\text{-}aero}$) conditions using Santa Barbara DISORT (SBDART) model [16] by inputting the measured aerosol properties and meteorological parameters. More details on the estimation of *ARF* are available in Moorthy et al. [10]. Unless mentioned specifically, we consider the diurnally averaged *ARF* (integrated over entire day at a time step of half an hour) at the top of the atmosphere in the following sections:

$$ARF = \frac{1}{24}\int_0^{24} (F_{no-aero} - F_{aero})dt \quad (1)$$

3. Results and discussions

3.1. Speciated aerosol radiative forcings

Speciated aerosol radiative forcing ($ARF_{eff\ x}$) of particular aerosol species, '*x*' (black carbon/sulfate/organic carbon/dust/sea salt), in presence of other aerosol species in a heterogeneous aerosol system is given as

$$ARF_{eff\ x} = ARF_x + ARF_{coupling} \quad (2)$$

where ARF_x is the radiative forcing due to the aerosol species '*x*' for the hypothetical atmosphere having only the single aerosol species (x), while $ARF_{coupling}$ accounts for the interactions between the aerosol species '*x*' and the neighboring aerosol species of the mixture. This coupling