AEROSOL RADIATIVE EFFECTS OVER KANPUR REGION, NORTHERN INDIA

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Abstract

This paper presents five-year (2001-2005) aerosol direct radiative forcing over Kanpur region in the Indo-Gangetic Basin (IGB) in the northern India. The anthropogenic components are delineated from the natural components by an optically-equivalent aerosol model, which is developed using the *in-situ* measured and remotely retrieved data of aerosol physical, chemical and optical properties. Absorbing black carbon (BC) and scattering water-soluble (Wsol, e.g. sulfate, nitrate, chloride) components are considered as the main anthropogenic species in the region, while the mineral dust is considered to be the main natural species. The probable mixing state of aerosols is also examined. A new methodology is developed to quantify the absorbing BC columnar concentration, which is the most crucial player in the aerosol DRF. On annual scale, 5.5% BC mass fraction contributes 8.4% to total AOD and 40% to total aerosol surface DRF. The 5 year mean±SD top-of-the-atmosphere (TOA), surface and atmospheric clear-sky aerosol DRF over Kanpur are -4.1 ± 6 , -31.8 ± 10.9 and $+27.7\pm10.4$ W m⁻², respectively. The clouds reduce the aerosol surface DRF, but enhance the aerosol TOA DRF. The aerosols reduce the net surface radiation by 18-20% annually. Finally, the possible implications of the large aerosol DRF to the regional climate are discussed.

1. Introduction

Aerosol direct radiative forcing (DRF) is one of the most uncertain components of the 'global climate change' problem. Aerosols through scattering and absorption of the incoming solar and outgoing terrestrial radiation perturb the Earth's radiation balance. Amongst the major hotspots of aerosol pollution in the world, Indo-Gangetic Basin (IGB) stands close to the top, where the transportation of natural dusts lead to enormous aerosol loading in addition to the anthropogenic activities (*Dey et al.*, 2004); yet the spatio-temporal heterogeneity of aerosol radiative forcing is not understood properly. Very few studies exist in literature regarding the aerosol DRF in the IGB, that too for very short period of time. In this paper, we have investigated the radiative effects due to aerosols in an urban/industrial site in the IGB for a five year (2001-2005) time period.

2. Approach

An optically-equivalent aerosol model is developed considering wsol, BC and dust as the major component. The size distribution parameters of these components are derived from measured size-separated chemical composition during winter (*Dey and Tripathi*, 2007). For rest of the months and for BC, size distribution parameters of *Hess et al.* (1998) are used. BC number concentration is calculated from the direct measurements and is used in the model. The number concentrations of other components are varied iteratively in such a manner that the model derived aerosol optical depth (AOD) spectrum, the spectral dependence of AOD or the Ångstrom exponent, single scattering albedo (SSA) and asymmetry parameter (g) match with the Aerosol Robotic Network (AERONET) retrieved values within their uncertainties. BC concentration is measured by Aethalometer from December 2004-December 2005 period (*Tripathi et al.*, 2005a). For the earlier period, a new methodology is developed to retrieve columnar BC concentration, [BC] from AERONET-retrieved composite refractive index. Maxwell-Garnett and Bruggeman mixing rules are used to derive the BC volume fraction. [BC] is calculated using the model-derived BC volume fraction and AERONET-retrieved volume size distribution (*Dey et al.*, 2006). The aerosol scale height is derived from the vertical profiles of BC and composite aerosol concentration measured using IIT-K aircraft (*Tripathi et al.*, 2005b, 2007).

To infer the probable mixing state, SSA has been calculated from the measured chemical composition in three different seasons, post-monsoon, winter and pre-monsoon, assuming external, internal and core-shell mixing. The model-derived SSA is then compared with the AERONET-retrieved SSA. Four types of core-shell combinations, wsol coating on dust, BC coating on dust, wsol coating on BC and BC coating on wsol, are assumed for the calculations.

The model-derived aerosol optical properties in the shortwave and longwave region are incorporated in SBDART model (*Ricchiazzi et al.*, 1998) to estimate the aerosol DRF at the TOA, surface and atmosphere. Similarly the anthropogenic aerosol DRF (AAF) are computed using the anthropogenic aerosol optical properties. The surface albedo and the cloud parameters are taken from Moderate resolution Imaging Spectroradiometer (MODIS) satellite.

3. Results and Discussion

[BC] in Kanpur exhibits strong seasonal variation. Maximum [BC] has been found during October-January months. Specific absorption cross section decreases non-linearly with increase in [BC] using both Maxwell-Garnett and Bruggeman mixing rules, as increase in BC volume fraction would lead to higher number of BC particles in the eccentric position of the mixture reducing the efficiency in absorption. The external mixing seems to be the most probable mixing state in the winter, whereas the water-soluble coating over dust particles and external mixing emerge as the probable cases in the post-monsoon season. However, very close SSA values for the above cases imply that the external mixing would be reasonable choice to compute aerosol DRF in these seasons. The mixing state is, however, very different in the pre-monsoon season, when the BC coating over dust particles seems to be the most probable case. The dust-BC mixing in the IGB would lead to enhanced absorption.

The mass fractions of various components are shown in Fig. 1a. The relative contribution (RC) of various major components to AOD over Kanpur (Fig. 1b) reveals that RC_{wsol} in January is as high as 67%, which reduces to mere 30% in June and again builds up to reach 69% in December. RC_{BC} is high in winter months (>11%) and low during the summer months (<6%). RC_{dust-f} varies between 1-3% and RC_{dust-c} is highest in May and June (>60%). RC_{wsol} shows strong spectral decrease, whereas RC_{dust-c} shows strong spectral increasing trend. RC_{BC} spectrally decreases with varying trend in different seasons, whereas RC_{dust-f} shows varying spectral trend. The spectral variation of AOD brings out three major information; first, the AOD is high $(AOD_{0.5} > 0.4)$ throughout the year in Kanpur, second, the spectral dependence of AOD shows strong seasonal variability and third, AOD in the shortwave region is much higher than in the longwave region. In general, SSA at the SW region is much higher than in the LW region indicating that the aerosols are more scattering in the SW region. Although the aerosols are highly absorbing (SSA<0.5) in the LW region, due to very low AOD and hence low total extinction, the radiative effects in the LW region are not significant. The fine mode scattering increases drastically when RH exceeds 80% and in the relative scale, the absorption is suppressed.

The 5 year mean±SD (ranges given within brackets) TOA, surface and atmospheric clear-sky aerosol DRF over Kanpur are -4.1 ± 6 (-19.6 to +6.1), -31.8 ± 10.9 (-5.8 to -62.3) and $+27.7\pm10.4$ (+11.6 to +63) W m⁻², respectively. The corresponding cloudy-sky values are +1.4±6.1 (-10.2 to +16.6), -23.3±9.3 (-3.5 to -43.8) and +24.8±9.7 (+5.4 to +51.8) W m⁻², respectively. Aerosol DRF reveals three major observations. First, large negative surface forcing $(> 20 \text{ W m}^{-2})$ is observed in all the months with two peaks, one during the winter and the other during the pre-monsoon when the surface forcing exceeds 30 W m^{-2} . Secondly, TOA forcing was close to zero in January, July and September, negative in other months except August when TOA forcing flips to positive sign. Thirdly, very high atmospheric heating persists throughout the year. The negative surface aerosol DRF in the SW is compensated maximum up to 11% by LW surface heating in the summer months. Inclusion of clouds reduces the aerosol surface DRF, but shifts the TOA aerosol DRF from cooling to warming, and depending on the magnitude of these changes, atmospheric aerosol DRF changes. The monthly variations of RC to total surface forcing of various components (Fig. 1c) are completely different from the variations of their mass fractions or RC to AOD. RC_{BC} varies from a high of ~56% in the winter to a low of ~19% in May. On an annual basis, ~5% f_{BC} contributes ~9% to total AOD_{0.5}, but ~40% to the total aerosol surface forcing. RC_{wsol} contributes to $\sim 20\%$ to the annual surface forcing, and the contribution is almost similar throughout the year except in June ($\sim 10\%$). RC_{dust-f} is lower than 2% for all the months. Annually, RC_{dust-c} is 29% to the total aerosol mass, 34% to AOD_{0.5} and 39% to the total surface forcing. RC_{dust-c} to total surface forcing is more than 55% during April-July and less than 20% during November to February. RC of various components clearly indicates that BC is the most crucial player in the aerosol surface forcing for this region.





AAF is related to total aerosol forcing (Δ F) through: AAF = Δ F * AEF * AF, where AEF is the anthropogenic efficiency factor (i.e. the anthropogenic aerosol forcing efficiency or the AAF per unit AOD, normalized by total forcing efficiency) and AF is the anthropogenic fraction to the composite aerosol mass. AF is estimated from the MODIS-derived aerosol fine mode fraction (AFMF) product. The monthly variations of anthropogenic and natural aerosol surface DRF over Kanpur are shown in Fig. 2. The surface AAF contributes to more than 80% of total forcing from October to February. AAF forcing starts decreasing from March onwards, and from April, natural forcing overtakes the anthropogenic forcing. The annual mean±SD (ranges within brackets) TOA, surface and atmospheric clear-sky AAF over Kanpur are +0.3±2.5 (-2.4 to +4.8), -19.9±9 (-8.5 to -34.3) and +20.2±9.9 (+10.9 to +38.6) W m⁻², respectively.



Fig. 2 Monthly variations of anthropogenic and natural clear-sky aerosol surface DRF over Kanpur during 2001-2005

The mean (\pm SD) annual clear-sky atmospheric heating rate over Kanpur is 0.84 \pm 0.3 K day⁻¹, which is 68% higher than the corresponding wintertime value for the Indian Ocean (*Satheesh et al.*, 2002). The heating rate is highest during December-January and May-June (~ 1 K day⁻¹), due to strong atmospheric absorption. Presence of clouds decrease the surface cooling to some extent, but they themselves induce large cooling, 2-3 times higher than the clear-sky condition. On an average, aerosols reduce the net solar radiation at the surface over the Kanpur region by 17%. The warmer atmosphere close to surface (i.e. high atmospheric heating) and colder surface due to large negative cooling would create an environment where energy flow from Earth's surface to atmosphere would be suppressed. This would increase the low level inversions and strengthen the boundary layer stability, which in turn would increase the aerosol residence time, thus providing a positive feedback.

4. Conclusions

[1] An aerosol model, representative of the Kanpur region has been developed from the collocated aerosol physical, chemical and optical measurements. This opticallyequivalent model provides an in-depth analysis of the seasonal variations of the aerosol characteristics and the radiative effects for Kanpur region.

[2] AOD shows strong seasonal variation with reduced spectral dependence during the pre-monsoon season (March-May) due to enhanced dust loading. Within the five year, no significant increasing or decreasing trend is observed in the AOD_{0.5}. The spectral variation of AOD in the SW and LW wavelengths reveals that the spectrally decreasing trend in the SW diminishes in the LW region. The LW AOD is less than 0.15 in all the months except May and June, when in the thermal IR region, AOD shows slight enhancement. In general, SSA shows decreasing trend with wavelength in the winter season, while in the premonsoon season and sometimes in the monsoon season, the trend gets reversed. In the post-monsoon season, the variation is mostly spectrally insensitive.

[3] The external mixing seems to be the most probable mixing state in the winter, whereas the water-soluble coating over dust particles and external mixing emerge as the probable cases in the post-monsoon season. The mixing state is, however very different in the pre-monsoon season, when the BC coating over dust particles seems to be the most probable case.

[4] On annual scale, 5.5% f_{BC} contributes 8.4% to AOD_{0.5}, 71.3% f_{wsol} contributes 55.1% to AOD_{0.5}, 0.2% f_{dust-f} contributes 2.5% to AOD_{0.5} and 23.1% f_{dust-c} contributes 34% to AOD_{0.5}. Annual mean (±SD) aerosol DRF at TOA, surface and atmosphere over Kanpur for five year period are estimated to be -4.1±6, -31.8±10.9 and +27.7±10.4 W m⁻². The large negative surface forcing in the SW is partially (up to 11%) compensated by LW heating. The relative contribution of natural forcing due to transportation of dusts starts increasing from March reaching maxima of 71% in July. The annual mean (±SD) TOA, surface and atmospheric clear-sky anthropogenic aerosol forcing over Kanpur are +0.3±2.5, -19.9±9 and +20.2±9.9 W m⁻², respectively.

[4] Large negative surface forcing and positive atmospheric forcing in the Kanpur region raise several climatic issues. The atmosphere is getting heated by more than 1 K day ⁻¹ in the winter and pre-monsoon seasons due to highest atmospheric absorption. Anthropogenic aerosols contribute 65.4% to the mean (\pm SD) annual heating rate of 0.84 \pm 0.3 K day⁻¹ Kanpur. Such large aerosol loading in the atmosphere would also alter the cloud microphysics through indirect effect. Hence there is a need to understand the effect of the aerosols on the regional hydrological cycle in much better way, which would require more in-situ measurements to reduce the present-day uncertainty of the radiative forcing.

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