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Key Points:
- Increased aerosol loading in the lower free troposphere during spring season over Indo-Gangetic Plain
- A coincident reduction in near-surface BC concentration during spring, due to convective dispersion
- Enhancement in absorbing aerosols in the free troposphere leads to significant atmospheric warming in spring

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Large-scale enhancement in aerosol absorption in the lower free troposphere over continental India during spring

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Abstract Aerosol absorption in the lower troposphere over continental India was assessed using extensive measurements of the vertical distribution of absorption coefficients aboard an instrumented aircraft. Measurements were made from seven base stations during winter (November–December 2012) and spring (April–May 2013), supplemented by the data from the networks of surface observatories. A definite enhancement in aerosol absorption has been observed in the lower free troposphere over the Indo-Gangetic Plain (IGP) during spring, along with a reduction near the surface. The regional mean aerosol absorption optical depth (AAOD) over IGP, which was derived from aircraft observations (integrated from the ground to 3 km), increased from 0.020 ± 0.009 in winter to 0.048 ± 0.01 in spring. The columnar AAOD depicted weak and distinctly different seasonal variations than that of surface level black carbon mass concentrations. This contrasting difference in the seasonality indicates the presence of elevated layers of absorbing aerosols during spring in association with the long-range transport and vertical convective lofting of aerosols.

1. Introduction

The abundance of absorbing aerosols (mainly black carbon (BC) and dust) over South and Southeast Asia has been the topic of scientific investigation because of the potential implications for the regional climate forcing and hydrological cycle [*Chung and Zhang*, 2004; *Lau et al*., 2006; *Bollasina et al*., 2011; *Nair et al*., 2013; *Vinoj et al*., 2014]. Increasing energy demand for domestic and industrial sectors has resulted in an increase in consumption of fossil fuels, biomasses, and biofuels, which has increased the loading of strongly absorbing BC aerosols with weakly absorbing organics (OC) in the atmosphere over this region [*Kumar et al*., 2014]. The mineral dust, advected from west Asia and the Thar Desert over to the South Asian region, also contributes significantly to atmospheric warming [*Vinoj et al*., 2014]. Due to rapid industrialization and urbanization in recent decades, India, in general, and the IGP, in particular, have witnessed a steady increase in anthropogenic aerosol loading [*Babu et al*., 2013]. Several studies have projected the importance of aerosol absorption on the radiation budget and hydrological cycle over the region using state-of-the-art climate models [*Lau et al*., 2006]; however, most of these models underestimate the quantity of aerosol BC over south Asia [*Nair et al*., 2012]. One of the missing elements in the quantitative assessment of aerosol-induced warming over north/central India [*Vinoj et al*., 2014] or along the southern slopes Himalaya, as proposed by the elevated heat pump hypothesis [*Lau et al*., 2006], is observational evidence for the seasonality of vertical profiles of absorbing aerosols over the region. Considering the inherent limitations of spaceborne or remote sensing techniques in this aspect, in situ measurements using aircrafts and balloons provide more reliable and direct estimates of the altitude distribution of aerosol absorption with high accuracy and vertical resolution [*Babu et al*., 2008; *Babu et al*., 2011] although it is difficult to conduct these measurements periodically.

Even though previous studies conducted in peninsular India have indicated the presence of elevated aerosol layers over the Indian region during spring [*Satheesh et al*., 2008; *Babu et al*., 2011], the characteristics of such layers remained unexplored especially over the Indo-Gangetic Plains (IGP). It is important to analyze these characteristics in order to understand the climate implications of absorbing aerosols over the Indian region. Thus, extensive measurements of the vertical profiles of aerosol absorption coefficients have been carried out using an instrumented aircraft at different locations of Indian landmass as a part of the Regional Aerosol Warming Experiment (RAWEX). The continuous and long-term measurements of BC mass concentrations from the ARFINET (network of aerosol observatories under Aerosol Radiative Forcing over India project of Indian Space Research Organisation (ISRO)) stations are used to supplement the aircraft measurements. The RAWEX was envisaged to characterize the aerosol properties during two contrasting seasons (winter

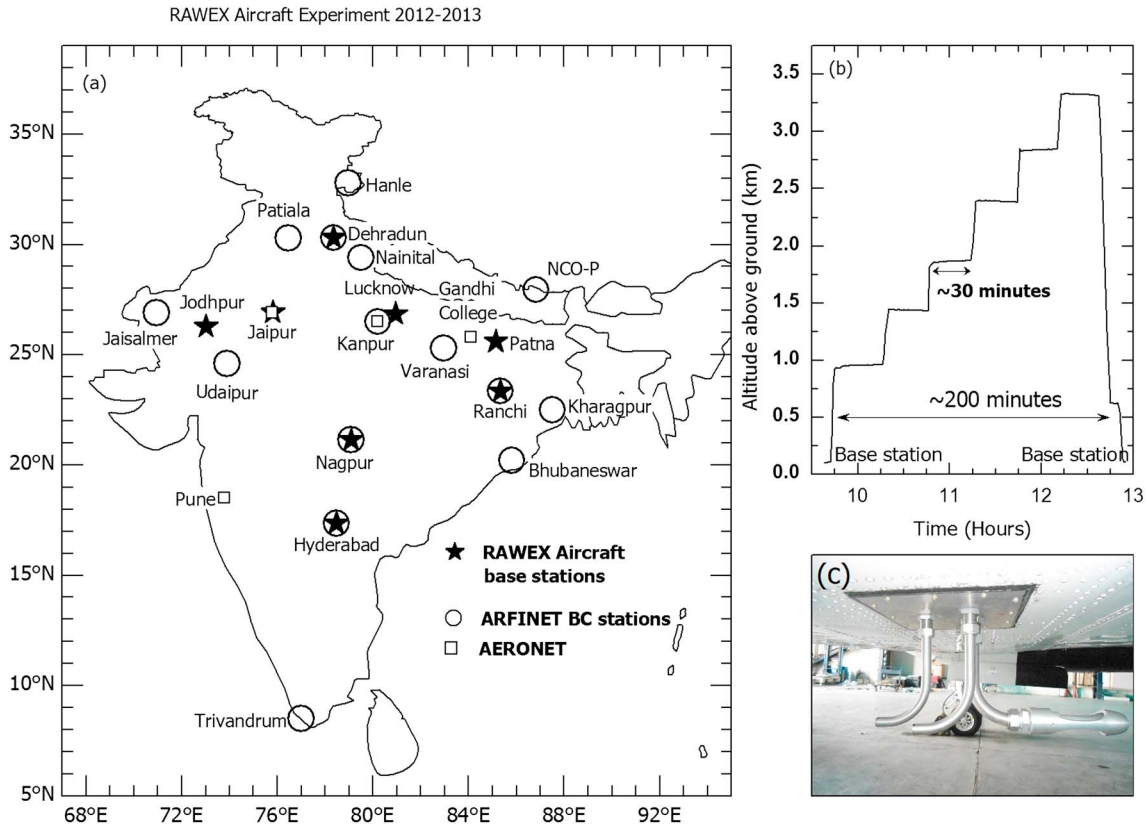

Figure 1. (a) Location map of base stations of aircraft sorties carried out under RAWEX during spring (April–May 2013) and winter (November–December 2012) seasons over central India (Hyderabad, Nagpur), West India (Jaipur, Jodhpur), Indo-Gangetic Plain (Lucknow, Patna/Ranchi), and Himalayan foothills (Dehradun). Corresponding measurement dates are Hyderabad (18 November to 1 December and 25 to 27 April), Nagpur (3 to 5 December and 30 April to 2 May), Lucknow (8 to 10 December and 5 to 7 May), Ranchi (27 to 29 December), Patna (9 to 12 May), Jaipur (22 to 24 December and 22 to 24 May), Jodhpur (17 to 19 December and 18 to 20 May), and Dehradun (12 to 15 December and 14 to 16 May). Simultaneous BC mass concentration measurements at or near to the RAWEX base stations were taken from the ARFI network (ARFINET, open circle). AERONET radiometer locations (Kanpur, Gandhi College, Pune, and Jaipur) are also shown (open circle). (b) A pattern of the typical staircase profiling carried out at Lucknow on 7 May 2013. (c) Photograph showing the air sampling inlet installed under the belly of the aircraft.

and spring) with distinctly different mesoscale and synoptic meteorology that would influence the local aerosol features as well as the long-range transport. The details of the measurements and the results are presented and discussed in this paper.

2. Campaign Details and Data

The campaign mode measurements of the vertical distribution of the atmospheric aerosols were carried out in two phases using the instrumented aircraft (Beechcraft B200) of National Remote Sensing Centre (NRSC), Hyderabad [*Babu et al*., 2016]: (a) November–December 2012 (winter phase) and (b) April–May 2013 (spring phase). Seven base stations were selected to represent distinct regions of India: Central India (Hyderabad and Nagpur), Western India (Jaipur and Jodhpur), Indo-Gangetic Plain (Lucknow and Ranchi/Patna), and Himalayan foothills (Dehradun) (Figure 1a). Due to aviation safety considerations and the highly undulating terrain near the Himalayan foothill station, the sorties at Dehradun could not be conducted at altitudes below 3000 m (AGL). In general, we have followed a six level typical staircase pattern (Figure 1b) for vertical profiling as described in *Moorthy et al*. [2004], with a minimum duration of 30 min at each level. All the sorties were conducted around noon (representing a fully evolved convective boundary layer so that the vertical distribution of aerosols is better homogenized at least within the atmospheric boundary layer) and away from the city area. The instruments were mounted in the cabin, which was unpressurized, and as such, the sorties were limited to a ceiling altitude of 4 km above the mean sea level. The data for the first 5 min after attaining the desired altitude at each vertical level is discarded in the analysis to ensure that instruments were stabilized to the pressure variations and operated under near isokinetic condition. The data collected between the two

levels were also discarded. For statistical significance, three profiling were carried out on consecutive days from the same station, and the average of these was used as the representative profile over that location.

A shrouded solid diffuser inlet (University of Hawaii) was installed under the belly of the aircraft (fuselage) for a near isokinetic ambient air sampling inside the aircraft, which is shown in Figure 1c. The air flow through the inlet was maintained at 70 L/min, using a pump, for an aircraft speed of 300 to 350 km h^{-1}, and all the instruments aspirated air from the manifold common air sampling inlet (more details are provided in *Babu et al*. [2016]). A high-resolution GPS system on board provided accurate information on the latitude, longitude, and altitude of the aircraft, and all the instruments were synchronized with the GPS time prior to the experiment. More details of the experimental design and sampling protocols are available in *Babu et al*. [2016].

In situ measurements of aerosol absorption at 781 nm were made using a photoacoustic soot spectrometer (PASS, Droplet measurements Technologies, USA) that was installed in the aircraft. Using PASS, the aerosol absorption is estimated by measuring the acoustic waves emanating from the aerosol sample while interacting with a laser beam using a high sensitive microphone [*Arnott et al*., 1999]. The instrument was operated at a flow rate of 1 L/min, and measurements were made at every 1 s time interval. The calibration of the instrument was carried out before and after the campaigns following the standard protocol described in *Arnott et al*. [2000]. The uncertainties associated with the calibration are given in *Arnott et al*. [2000] and are not repeated here. Due to technical/operational constraints within the airfield, we could not measure the absorption coefficient at the surface level for a sufficient amount of time prior to each aircraft sortie. Because of this measurement limitation, the surface values were taken from the collocated or nearby ground-based optical attenuation measurements carried out at ARFINET stations (as shown in Figure 1a) using multiwavelength aethalometers. Aerosol absorption coefficients at its seven wavelengths are estimated from the measured attenuation values following *Arnott et al.* [2005] [e.g., *Nair et al*., 2008; *Babu et al*., 2016]. The following supplementary data were used: the aerosol absorption optical depth (AAOD) derived from the ozone monitoring instrument (OMI) on board the EOS-Aura satellite and the optical properties of aerosols (aerosol optical depth, single scattering albedo, and asymmetry parameter) that were measured using Sun-sky spectral photometers installed at Aerosol Robotic Network (AERONET)stations (Pune, Jaipur, Gandhi College, and Kanpur) over the Indian region. These stations are also marked in Figure 1a.

3. Results and Discussion

3.1. Surface Level BC and Its Seasonal Changes

The mean values of the BC mass concentration at the surface level, estimated from long-term measurements over the distinct ARFINET stations during December (representing winter) and May (representing spring), are shown in Figure 2. The BC measurements were made using multiwavelength aethalometers. Out of the 34 stations having BC measurements over India, 14 stations are selected, which (i) represent distinct environments such as the peninsular, central and west India, Indo-Gangetic Plain, and Himalayas (ii) that are close to the RAWEX aircraft base stations (iii) with more than 2 years of data.

The near-surface BC mass concentrations depicted large spatial heterogeneity and seasonality: there were very high values observed during winter that decreased continuously through spring to an annual minimum during the summer monsoon season. This seasonal pattern is observed over most of the Indian mainland stations [*Nair et al*., 2007, 2012; *Kumar et al*., 2015] except at the high-altitude Himalayan stations, where BC peaks during the spring in association with large-scale convection in the central India and Ganges valley [*Nair et al*., 2013]. The high aerosol loading during winter has significant implications on the daily life of the people living in this valley due to low air quality and visibility. It has been extensively documented that the meteorological conditions play a crucial role in the buildup of high aerosol loading during winter through confinement of aerosols within the shallow boundary layer [e.g., *Nair et al*., 2007]. As the season advances, the BC mass loading decreases, with consequent improvement in the air quality through spring to the summer monsoon that gradually returns to the annual peak by next winter. This seasonal cycle in near-surface BC raises two questions: (i) whether BC sources are not active during spring/summer and whether this leads to the seasonal reduction and (ii) whether the aerosols system is lofted to the free troposphere due to increase in convection during spring and summer followed by extensive washout by the monsoon rains. This finding calls for vertical profiling of BC.

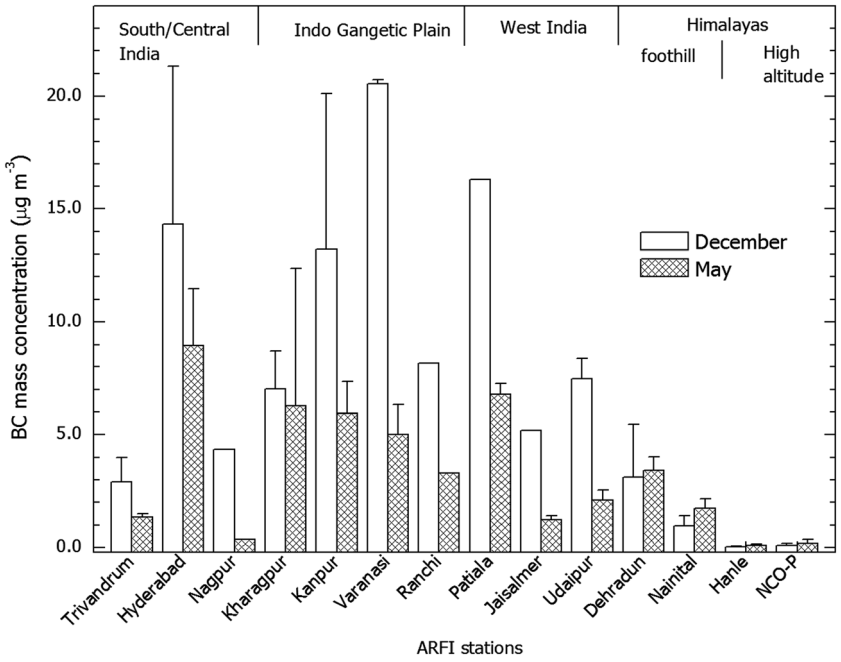

Figure 2. Climatological variation of BC mass concentrations during December (represent winter) and May (represent spring) at various locations over the Indian region.

3.2. Vertical Distribution of Aerosol Absorption

The mean vertical distribution of aerosol absorption coefficients obtained from the aircraft measurements for Central India, Indo-Gangetic Plain, Western India, and Himalayan foothills during the winter and spring seasons are shown in Figure 3. Large seasonality is observed over the Indo-Gangetic Plain and Western India, whereas seasonal variations were nonsignificant for the Central India and Himalayan foothills. The absorbing aerosols are mostly confined to the lower altitudes ($<$1 km) during winter with very low values (close to or below the detection limit of the instrument) at altitudes above 1 km over most of the regions. In contrast, the aerosol concentration increases within lower free tropospheric altitudes by threefold to fivefold during the spring season over the Indo-Gangetic Plain and Western India. It is striking to note that the aerosol absorption over the Himalayan foothill regions is comparable in magnitude with the lower free troposphere and at the surface levels in both spring and winter seasons. Consistently, high aerosol absorption was observed above 3 km over the Himalayas during winter and spring. The source attribution and dynamical aspects of this high aerosol loading (over Himalayas) within the lower free troposphere, especially during winter, are yet to be explained. In comparison with the Indo-Gangetic Plain and Central India, the magnitude of aerosol absorption is smaller over Western India, where there are far less industrial and biomass burning activities compared to the other parts of the country, during winter and spring. However, it should be noted that the biomass burning activities associated with agriculture crop residue burning is significant over this region during the autumn (October–November) season [*Rastogi et al*., 2016].

The vertical distribution of aerosol absorption over the Indian landmass has been mostly derived from the vertical profiling of the black carbon mass concentration onboard aircraft [*Moorthy et al*., 2004; *Babu et al*., 2008, 2011], and several studies have indicated the presence of elevated layers of enhanced black carbon concentration above the boundary layer at several locations. However, seasonal and spatial heterogeneities in the vertical distribution of direct measurements of absorption coefficient were nonexistent over the region prior to the RAWEX.

3.3. Absorption Optical Depth and Vertical Heterogeneity

Aerosol absorption optical depth (AAOD) is estimated by integrating the vertical profiles of aircraft-derived absorption coefficient up to 4 km. The AAOD at 781 nm wavelength has been extrapolated to the midvisible wavelength (550 nm) using the power law function with a wavelength exponent (Angstrom exponent) equal

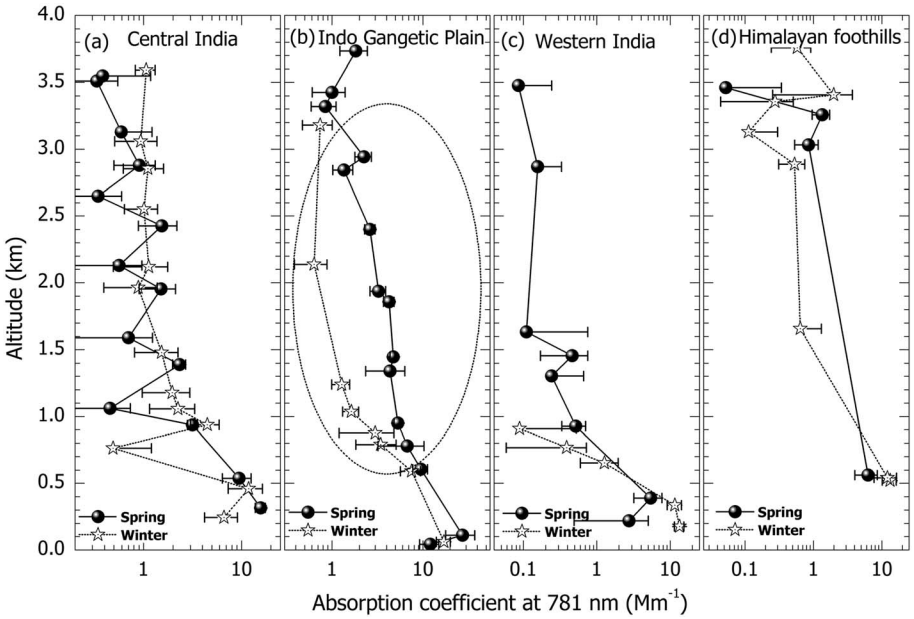

Figure 3. Vertical profiles of aerosol absorption coefficient measured over (a) Central India (Hyderabad, Nagpur), (b) Indo-Gangetic Plain (Lucknow, Patna/Ranchi), (c) Western India (Jaipur, Jodhpur), and (d) Himalayan foothill (Dehradun) during winter and spring seasons.

to unity, assuming that the aerosol absorption is solely attributed to fossil fuel burning [*Kirchstetter et al*., 2004]. We have considered this spectral extrapolation as the lower bound of the estimation since biomass burning and dust aerosols have much steeper spectral dependence (wavelength exponent > 2) than that from fossil fuel burning [*Kirchstetter et al*., 2004]. Disentangling the contribution of black carbon, brown carbon, and dust to the aerosol absorption using spectral measurements of absorption coefficients is a potential tool but that is highly challenging too. Several studies have quantified the contribution of various absorbing species to the total absorption over the south Asian region [*Chung et al*., 2012; *Babu et al*., 2016]. Figure 4 depicts the AAOD estimated at 550 nm over different base stations during the winter and spring seasons and the AAOD retrieved using OMI/Aura. In general, (i) the AAOD values are high over the Indo-Gangetic Plain during winter and spring. (ii) The AAOD values remained comparable during both the seasons over the Indo-Gangetic Plain, despite the large seasonality in the vertical profiles of absorption coefficient. (iii) An increasing gradient toward the Indo-Gangetic Plain from south to north (latitudinal) and west to east (longitudinal) is also observed.

The variations in the regional mean values of black carbon mass concentrations measured at different ARFINET stations and AAOD at 675 nm estimated from the AERONET observations at the Indo-Gangetic Plain, Western India, and Central India are shown in Figure 5. The mean AAODs over Lucknow derived from aircraft measurements were 0.029 and 0.04, respectively, for winter and spring seasons. These values are lower than the seasonal mean values of AAOD (0.055 for winter and 0.045 for spring) retrieved using AERONET at Kanpur. This difference and perhaps the opposite seasonality are attributed mainly to (i) the difference in retrieval techniques, (ii) the uncertainties and large variabilities in the near-surface aerosol properties between the two location, (iii) the aircraft measurements being limited to ~3 km altitude while the AERONET retrieval represents the columnar values, and (iv) to the possible contribution from different aerosols types (dust) residing above 4 km altitude, especially during spring. The surface level BC mass concentration depicts an approximate twofold to threefold decrease from winter to spring, whereas columnar AAOD does not show a significant decrease toward spring. The AAOD values increased (from 0.020 ± 0.009 in winter to 0.048 ± 0.01 in spring over the Indo-Gangetic Plain) or remained constant (over west India) except for Central India. This finding clearly indicates the vertical heterogeneity and presence of elevated layers (possibly above 4 km) of absorbing aerosols during spring over the northern parts of India. The decrease in BC mass concentration at the surface from the winter to the spring season does not mean that the BC aerosols are removed from the atmosphere; rather, the black carbon is vertically redistributed and accumulated in the

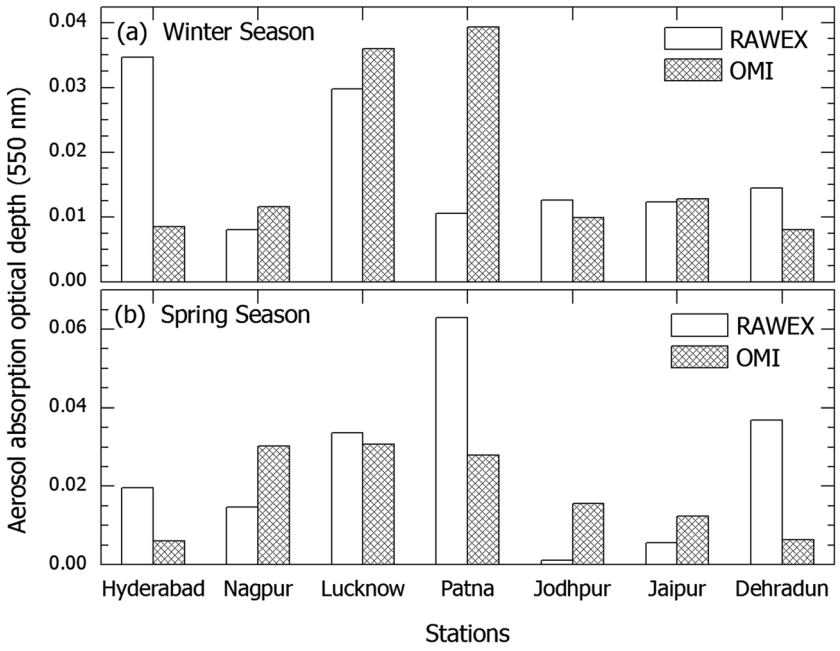

Figure 4. The aerosol absorption optical depth (AAOD) at 550 nm estimated from the vertical profiles of aerosol absorption coefficient measured over Hyderabad, Nagpur, Lucknow, Jodhpur, Jaipur, and Dehradun during spring and winter seasons. Simultaneous AAODs retrieved from OMI is also shown in the figure.

lower free troposphere. Advection of West Asian dust within the lower free troposphere also contributes to the observed high AAOD values during the spring season. In general, Figures 3 and 5 indicate that the absorbing aerosol concentration increases above the boundary layer during the spring season. In contrast to the seasonal variations of surface and columnar aerosol absorption over the Indo-Gangetic Plain and West India, the BC mass concentration and columnar AAOD over Central India decreased from winter to spring, which is in line with the seasonal variation of the vertical profiles of absorption coefficient measured during the RAWEX aircraft experiment.

3.4. Radiative Effects and Climate Implications

The spring time enhancement in elevated aerosol absorption over the Indo-Gangetic Plain has the potential to increase the atmospheric absorption of radiation at free tropospheric altitudes during spring. This enhancement leads to an increase in atmospheric direct radiative forcing ($DRF_{atmosphere}$) over the region. The seasonal variation of clear-sky $DRF_{atmosphere}$ due to aerosols (the difference in solar flux reaching the top of the atmosphere and surface with and without the presence of aerosols) over the Indo-Gangetic Plain and Central and West India is computed using the Santa Barbara DISORT model [*Ricchiazzi et al*., 1998] by feeding the AERONET measurements of aerosol optical depth (AOD), single scattering albedo and asymmetry parameters (in the wavelength range from 0.44 to 1.02 micrometer), and the collocated satellite measurements of spectral surface albedo, columnar ozone, and water vapor. Here we relied on AERONET-derived aerosol parameters as the input to the radiative transfer model since we (i) could not make simultaneous columnar AOD measurements and (ii) could not sample the near surface aerosol loading accurately during the aircraft campaign. It is found that aerosols absorb +24.9 W m^{-2} over the Indo-Gangetic Plain during winter and +34.0 W m^{-2} during spring. A similar pattern is observed over Western India, where atmospheric forcing was + 14.7 W m^{-2} during winter and +19.8 W m^{-2} during spring. Although the $DRF_{atmosphere}$ values at Central India (+24.6 W m^{-2}) are comparable to that over the Indo-Gangetic Plain during winter, the Indo-Gangetic Plain experiences ~8 W m^{-2} and ~14 W m^{-2} more atmospheric warming during spring compared respectively to Central India and Western India. Atmospheric forcing over Central India also depicts increase from winter to spring season. This increase is in line with the inferences drawn from Figure 5. However, the availability of the solar radiation increases significantly during the spring season, which enhances aerosol absorption in the atmosphere by ~9 W m^{-2} from winter to spring even though the optical

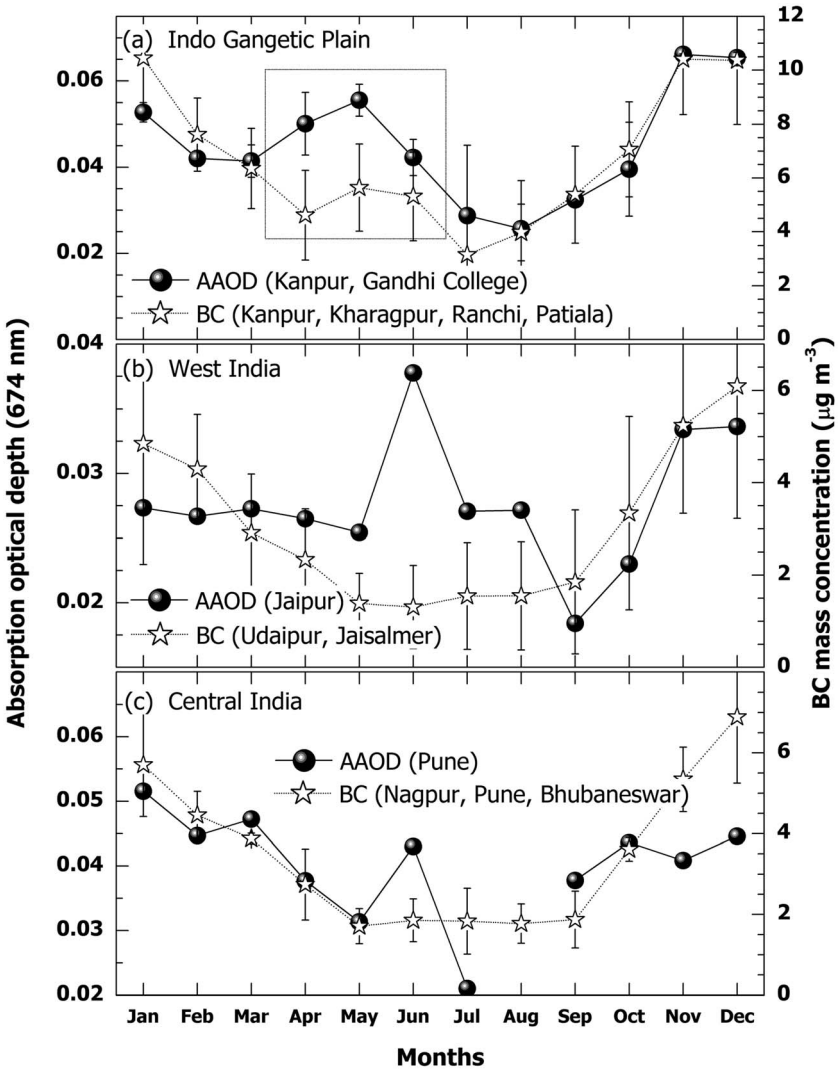

Figure 5. Mean variation of aerosol absorption optical depth at 674 nm estimated from AERONET measurements and BC mass concentration measured at ARFINET stations located at (a) Indo-Gangetic Plain, (b) west India, and (c) central India. The name of the station data used for estimating the regional mean is shown in the bracket.

depth (AOD = 0.016) did not vary significantly. Additionally, the west Asian dust transport to North India will also contribute significantly to the spring time enhancement in atmospheric forcing. However, the relative contribution of BC and dust to the aerosol absorption is yet to be quantified. Based on GOCART model simulations, we have estimated that the BC aerosols over the Indo-Gangetic Plain contribute 56% to the atmospheric forcing during winter, which further decreases to 30 % during spring. It seems that a partial decrease in columnar BC loading might have compensated by the spring time dust transport to the region.

Allen and Landuyt [2014] have demonstrated the significant role of vertical distribution of aerosol absorption in climate forcing estimations. Similarly, *Chung and Zhang* [2004] have shown that diabatic heating due to aerosols above the planetary boundary layer could lead to a change in the monsoon pattern over the Indian region. The vertical heterogeneity in aerosol absorption, especially due to the presence of elevated layers of strongly absorbing aerosols [e.g., *Babu et al.*, 2011], highly limits the extension of surface level BC measurements to the entire column. The RAWEX measurements indicate the enhanced aerosol absorption in the lower free troposphere, which increases diabatic heating, especially over north and west India. The present study shows that the lower free tropospheric values of aerosol absorption over the Indo-Gangetic Plain were very high during the spring season compared to that during winter. This finding is, in general, consistent with the elevated heat pump hypothesis by *Lau et al.* [2006]. However, the magnitude of diabatic heating

required for influencing the circulation pattern and monsoon dynamics are highly uncertain and debatable [*Nigam and Bollasina*, 2010]. Although several studies addressed the direct effect of spring aerosols on Indian monsoon, it is rather unclear on the implication of the indirect and semidirect effect due to these aerosol layers. Hence, it is essential to conduct field experiments to resolve the uncertainties in the aerosol-monsoon interaction due to absorbing aerosols over the Indian region during the spring and summer monsoon seasons.

4. Conclusions

Exhaustive measurements of the vertical profiles of the aerosol absorption coefficients have been carried out under the RAWEX aircraft campaign during the winter and spring seasons over the Indian region. The main highlights of the RAWEX aircraft campaign are the presence of elevated aerosol absorption at the lower free tropospheric altitudes during the spring season, especially over the Indo-Gangetic plain and western India, which is coincident with a significant decrease at near the surface. The monthly mean variation of aerosol absorption optical depth and surface level BC mass concentration showed a distinctly different pattern implying vertical heterogeneity (elevated aerosol layer) in aerosol absorption over the Indo-Gangetic plain and west India during the spring season. Although the surface level black carbon mass concentration decreased significantly from winter to spring by twofold to fivefold, AAOD did not show significant seasonal variations (rather a small increase), indicating the role of vertical redistribution of aerosols and long-range transport of dust aerosols in the lower free troposphere over north India. The heterogeneity in the column and surface values further affirms the importance of conducting vertical profiling of BC aerosols for the quantitative assessments of the impact of absorbing aerosols on regional climate.

Acknowledgments

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