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Short communication

The vertical profile of atmospheric heating rate of black carbon aerosols at Kanpur in northern India

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Abstract

Altitude profiles of the mass concentrations of aerosol black carbon (BC) and composite aerosols were obtained from the collocated measurements of these quantities onboard an aircraft, over the urban area of Kanpur, in the Ganga basin of northern India during summer, for the first time in India. The enhancement in the mean BC concentration was observed at ~1200 m in the summer, but the vertical gradient of BC concentration is less than the standard deviation at that altitude. The difference in the BC altitude profile and columnar concentration in the winter and summer is attributed to the enhanced turbulent mixing within the boundary layer in summer. This effect is more conspicuous with BC than the composite aerosols, resulting in an increase in the BC mass fraction (F_{BC}) at higher levels in summer. This high BC fraction results in an increase in the lower atmospheric heating rate in both the forenoon, FN and afternoon, AN, but with contrasting altitude profile. The FN profile shows fluctuating trend with highest value (2.1 K day^{-1}) at 300 m and a secondary peak at 1200 m altitudes, whereas the AN profile shows increasing trend with highest value (1.82 K day^{-1}) at 1200 m altitude.

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1. Introduction

Black carbon (BC) aerosols perturb the Earth's radiation budget by absorbing the solar radiation; consequently heating the atmosphere and cooling the surface (IPCC, 2001; Jacobson, 2001). The vertical profile of BC, more essentially the vertical

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distribution of BC mass fraction, F_{BC} (i.e. fraction of BC to the composite aerosol mass concentration), is the most important factor in the estimation of BC radiative forcing (Haywood and Ramaswamy, 1998; Babu et al., 2004).

Very limited information worldwide, and even fewer in India, is available on the BC altitude profile despite its importance. Only two such attempts have been made in India, one in Hyderabad, southern India (Moorthy et al., 2004) and the other in Kanpur, northern India (Tripathi et al., 2005a).

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These two BC altitude profile measurements in the winter season were found to be contrastingly different, particularly in the region below ~ 1.5 km. An elevated BC layer near the boundary layer (at around 900 m) was observed in Kanpur during the winter season, which was not observed over Hyderabad. Moreover, both these attempts were limited to the measurement of the altitude distribution of BC concentration only, and there were no idea on how $F_{\rm BC}$ varied with altitude.

Aircraft measurements were carried out in Kanpur during the summer season with three major objectives: (1) whether the wintertime enhanced BC layer persists in the summer, (2) to collect information about $F_{\rm BC}$ altitude variation and (3) to study the heating rate altitude profiles. In this paper, we present the salient differences of BC altitude profiles over Kanpur between the winter and summer season. Further we discuss the altitude variation of $F_{\rm BC}$ in the summer. These data are used to estimate, for the first time, the altitude variation of lower atmospheric heating rates due to atmospheric BC aerosols in Kanpur.

2. Experimental details

2.1. Site location and observations

The aircraft borne experiments were conducted over Kanpur (80°20'E, 26°26'N, 142 m altitude from mean sea level) during January (winter season, Tripathi et al., 2005a) and June (summer season), 2005. The winter sorties (forenoon, FN and afternoon, AN) contained only BC measurements (Tripathi et al., 2005a) while the other experiments measured BC and composite aerosol concentrations simultaneously, respectively, using an Aethalometer and an Optical Particle Counter (OPC). The instrument details are given in the next section. Altogether two sorties were made in the month of June, one during evening of June 18 (hereafter referred as experiment 1) and the second during morning of June 19 (hereafter referred as experiment 2). During the experiment 1, the evening flight took off at around 17:15 local time (LT) and the aircraft flew up to an altitude $\sim 2 \text{ km}$ and continuous measurements were made during the flight until it landed at about 18:30 LT. Each sortie comprised of flights at 4-5 levels (below the upper limit of 2 km above local ground level, AGL), with the aircraft flying for $\sim 5 \min$ at each levels, separated by \sim 300 m altitude. Continuous measurements of both BC and composite aerosol concentration, presented in this paper, are the average of whole measurements carried out at the particular altitude levels. Similar path has been followed during experiment 2, when aircraft flew from $\sim 07:35$ to 09:00 LT up to ~ 1.5 km altitude.

2.2. Instrumentation and data analysis

The instruments were mounted in a Piper Super Cub PA-18 aircraft (for details see Tripathi et al., 2005a). Even though this aircraft can fly up to an altitude of 3 km without oxygen, during the measurements the altitude was restricted up to 1.5 km only due to safety reasons. The instruments were powered with two batteries and a UPS with power back up for about one and half hour. Measurements have been taken continuously from 10 min before the take off of the aircraft and continued up to 10 min after its landing. Temperature data were measured simultaneously with aerosol measurements and the coordinate of each measurement has been measured with a hand-held global positioning system (GPS) receiver during the experiments.

The Aethalometer (AE-21-ER, Magee Scientific, USA) that used a size selective inlet BGI Sharp cut cyclone (Model SCC-1.828, which allows particles below 10 μ m) was operated at a mass flow rate of 6.5 standard 1min⁻¹ and at a time base of 1 min. Details of the operation of Aethalometer, data deduction, and error budget are discussed elsewhere (e.g., Babu et al., 2004; Tripathi et al., 2005b). As the pump speed varied with altitude to maintain the set mass flow rate, each measurement of BC concentration (M_1) was converted to actual BC concentration (M_2) using Eq. (1) following Moorthy et al. (2004):

$$M_2 = M_1 \left[\frac{P_1 T_2}{P_2 T_1} \right]^{-1}, \tag{1}$$

where P_1 , P_2 , T_1 and T_2 are standard atmospheric pressure (1017 hPa), the atmospheric pressure at the measurement altitude, standard temperature (293 K) and temperature at the measurement altitude, respectively.

The OPC (Model 1.108 of Grimm Aerosol Technik GmbH, Germany) is a small portable unit, used for the continuous measurements of particles in the air. The measurements were carried out in mass mode i.e. particle mass per unit volume

ranging from 0.1 to $10^5 \,\mu g \,m^{-3}$ with an operational flow rate of $1.21 \,min^{-1}$ with 1 min sampling time. This instrument uses the light (from a diode laser source) scattering by single particles to count the particles. The counts are used to get the number density i.e. distributions in 15 channels in the range from 0.3 to 20 μm . The numbers are converted to mass assuming spherical particles, and a mean particle density of $1.66 \,g \,cm^{-3}$ (more details are available in www.grimm-aerosol.com). The data of Aethalometer and OPC averaged for each altitude level are presented here with the corresponding standard deviations.

The Aethalometer measures the particles of size $<10\,\mu$ m, whereas the OPC measures the size distribution in the range of 0.3-20 µm. The difference in the size distribution range in the two instruments would lead to overestimation of $F_{\rm BC}$ in the size range $<0.3\,\mu\text{m}$ and underestimation of $F_{\rm BC}$ in the size range > 10 µm. As the BC particles mostly lie in the fine mode (Hess et al., 1998) and rarely in the coarse mode only when it gets mixed with coarse dust (Clarke et al., 2004), the resulting $F_{\rm BC}$ would be underestimated. Few recent studies have indicated that BC mixing with dust in the summer is a possibility for this region (Dey et al., 2007; Satheesh et al., 2006), which indicates that BC also exists in the coarse mode leading to reduction in the overestimation of $F_{\rm BC}$. However, as there are no measurements of BC size distribution, this effect cannot be quantified.

3. Results and discussion

3.1. Altitude profiles of BC and composite aerosol

In Fig. 1a, we have shown the altitude profiles of BC mass concentration obtained during the summer experiments along with the altitude profiles obtained in the earlier experiments in winter by Tripathi et al. (2005a) for completeness as well as for comparison. The enhancement in mean BC concentration (shown with broken arrow mark) was observed at ~900 m in the winter. In summer season, the mean BC concentration shows an increase at $\sim 1200 \,\mathrm{m}$ (shown by solid arrow); however, the vertical gradient is less than the corresponding standard deviations. The mean FN profile shows a steadily decreasing trend up to 900 m, whereas the mean AN profile shows a fluctuating trend about a mean value of $7.5 \,\mu g \, m^{-3}$. Due to some technical problem in the Aethalometer,

it was not possible to measure BC concentration above 1.2 km during experiment 2. However, the trend in the mean AN profile suggests the increment in BC concentration above 900 m, although the enhancement is not statistically significant. In the lower altitude region ($< 500 \,\mathrm{m}$) the FN concentrations are higher than the AN values for winter. while the reverse is seen in summer, which is due to the shallow boundary layer and reduced convection in the winter compared to summer. The FN BC gradients during the summer (0.5 and $0.32 \,\mu g \,m^{-3}$ per 100 m, respectively) are less than that during the winter season $(1.04 \,\mu g \,m^{-3}$ per 100 m, Tripathi et al., 2005a) indicating stronger convective activities during summer dispersing the pollutants from the surface.

As compared with BC profiles, the total particle concentration (Fig. 1c) in summer, interestingly shows a different pattern with a steady decrease in concentration with altitude throughout in both FN and AN. Fine mode aerosols are larger ($\sim 73\%$) compared to coarse mode aerosols ($\sim 27\%$) throughout the altitude. The temperature profiles for two sorties (Fig. 1b) reveal no significant inversion, higher boundary layer height and more mixing as compared to the winter profiles (Tripathi et al., 2005a). The contrasting natures of BC profiles and total particle concentration profiles give rise to fluctuating $F_{\rm BC}$ profiles. $F_{\rm BC}$ was lower (5%) at the surface as compared to the wintertime vales ($\sim 10\%$, Tripathi et al., 2005a), but increases with the altitude. In the AN, $F_{\rm BC}$ increases up to 12% at 1200 m, where the enhanced layer exists. In the FN, the value is even higher (17%), but the sharper decrease of total particle concentration from surface to 500 m with respect to BC concentration leads to very high $F_{\rm BC}$ (30%) at 500 m altitude.

BC is lighter compared to the composite aerosol with typical density of $\sim 1 \text{ g cm}^{-3}$ (Hess et al., 1998). During summer, dust, which is heaviest among other species (2.6 g cm⁻³), is present in abundance, which means the density of the composite aerosol is even higher. Crop wastes were being burnt in the nearby rural areas in the AN emitting BC. Freshly emitted BC particle concentration of $\sim 10^5 \text{ cm}^{-3}$ needs only 7 min to mix with existing particles, assuming a coagulation coefficient of $10^{-7} \text{ cm}^3 \text{ s}^{-1}$ (Seinfeld and Pandis, 1998, p. 662). There seems to be sufficient time available for internal mixing of the BC emitted from the open crop-waste burning. The lighter BC is transported more easily by convective wind to mix more with the existing particles at



Fig. 1. (a) Vertical profiles of aerosol black carbon for the months of January (winter) and June (summer); (b) same as (a) but for temperature; (c) same as (a) but for total aerosol mass; (d) same as (a) but for black carbon mass fraction (F_{BC}). The enhanced BC layer in the winter profiles is shown by dotted arrow and in the summer profile by solid arrow.

higher altitude, thus increasing the mass fraction with altitude.

3.2. Columnar BC and composite aerosol concentration

We have calculated columnar BC concentration using Simpson's rule for each 500 m column height interval for summer profiles along with the winter profiles from Tripathi et al. (2005a) for better comparison (Fig. 2). In both profiles, maximum BC concentration was confined up to first 500 m altitude. The absolute columnar BC concentration within this level is higher in FN than AN in the winter season, but shows reverse trend in the summer season due to differences in boundary layer



Fig. 2. Columnar BC concentration in the FN and AN of summer and winter over Kanpur.

mixing height variation. During summer months, absolute value of BC concentration is higher in each column height levels during AN hour compared to FN hour mainly due to biomass-burning event in a wide region observed during the sortie. During both the sorties, although maximum BC is more confined up to first 500 m height level, it is almost uniformly distributed in each column height due to more vertical mixing in the AN hours.

During winter months also maximum BC concentration was confined up to first 500 m, which is more pronounced in the FN hour (~68% of the total columnar value) compared to AN hour (~47%) due to shallow boundary layer. Absolute value of columnar BC at each altitude level is higher during both FN and AN hours of summer month compared to winter month; however, the percentage contribution of columnar BC at lower height i.e. up to 500 m, is about 26% higher during FN hour and about 13% higher during AN hour of winter month compared to summer month.

Columnar total aerosol concentration (not shown) has similar pattern as columnar BC during summer month, which is mostly confined up to 500 m during FN and AN hour. Aerosols are almost uniformly distributed in upper column heights as BC in AN. Aerosol mass size distributions, measured by OPC during FN and AN, were fitted with bimodal log normal distribution to calculate the mass mode radius in the fine $(R_{m,f})$ and coarse mode $(R_{m,c})$ fractions. The mean $R_{m,f}$ and $R_{m,c}$ for the FN profile are 0.258 and 1.44 µm and the corresponding AN values are 0.28 and 1.96 µm, respectively. The altitude variations of $R_{m,f}$ and $R_{m,c}$ (Fig. 3) are contrastingly different in the FN and AN profiles in summer. Both $R_{m,f}$ and $R_{m,c}$ are larger for the AN profile than FN implicating upward transport of more number of larger aerosols during AN hours due to stronger convective activity.

3.3. Vertical distribution of heating rate due to BC absorption

Vertical distribution of composite aerosol mass, BC mass and F_{BC} to composite aerosol mass were used as anchoring points to optical properties of aerosols and clouds (OPAC) continental polluted aerosol model (Hess et al., 1998). The size distribution parameters of Hess et al. (1998) are used in the calculations of the optical properties. The extinction coefficient, single scattering albedo and asymmetry parameter at each atmospheric layer were estimated



by OPAC using the measured values of BC mass, total aerosol mass concentration and F_{BC} at various altitude levels. The procedure for estimating aerosol optical properties using measurements of composite and BC aerosol mass is well documented in literature (Babu et al., 2004; Satheesh, 2002).

The aerosol optical properties thus deduced from measured altitude profiles are then incorporated in a Discrete Ordinate Radiative Transfer model developed by University of Santa Barbara (SBDART) (Ricchiazzi et al., 1998) to estimate the net radiative fluxes (downward minus upward) at the top and bottom of each atmospheric layer. The aerosolinduced flux change at each atmospheric layer is derived from the net radiative flux estimations with and without aerosols. The net flux change due to aerosols between top and bottom boundary of each layer is the flux absorbed in the layer. The atmospheric heating rate due to aerosol absorption at each layer (ΔF) is given by

$$\frac{\partial T}{\partial t} = \frac{g}{C_{\rm p}} \frac{\Delta F}{\Delta P},\tag{2}$$

where $\partial T/\partial t$ is the heating rate (K day⁻¹), g is the acceleration due to gravity, C_p is the specific heat capacity of air at constant pressure (~1006 J kg⁻¹ K⁻¹) and ΔP is the atmospheric pressure difference between top and bottom boundary





Fig. 4. Vertical profiles of heating rate due to aerosol black carbon calculated from $F_{\rm BC}$ profiles.

of each layer, respectively (Satheesh and Ramanathan, 2000). The vertical profiles of atmospheric heating rate due to aerosol absorption thus calculated are shown in Fig. 4 (in units of $K day^{-1}$).

The heating rate depends on the abundance as well as BC mass fraction. While abundance determines the magnitude of heating, BC mass fraction depends on the efficiency of heating. In both FN and AN, composite aerosol mass indicates sharp decrease as a function of altitude. The heating rates in FN and AN are similar ($\sim 0.9 \,\mathrm{K} \,\mathrm{day}^{-1}$) at surface, but show distinct variations in their profiles. FN profile shows fluctuating trend with maximum value $(2.1 \,\mathrm{K} \,\mathrm{day}^{-1})$ being observed at 300 m altitude, where $F_{\rm BC}$ is ~31%. A second peak $(1.75 \text{ K day}^{-1})$ appears at around 1200 m, where the $F_{\rm BC}$ is ~17%. Higher heating rate at this altitude than the heating rate at 600 m altitude despite having slightly lower $F_{\rm BC}$ is due to much lower ΔP value at this level as compared with that at lower level. On the other hand, AN profile shows steadily increasing trend, reaching maxima $(1.82 \,\mathrm{K} \,\mathrm{day}^{-1})$ at 1200 m. The presence of the high F_{BC} results in enhancement of heating rate with altitude within the boundary layer. Such high BC concentrations aloft increases the radiative forcing as compared to steadily decreasing BC profile (Tripathi et al., 2005a). As the high BC concentration was found to persist in the dry season (winter-summer) in the region, such high atmospheric heating would result in strengthening of boundary layer inversion leading to poor dispersal of aerosols, thus providing a positive feedback. Further in situ measurements are needed to investigate the spatial extent of the high $F_{\rm BC}$ at higher altitude in the Ganga basin to understand its role in modifying the regional climate.

4. Conclusions

We have presented the heating rate profiles in the lower atmosphere due to aerosols for Kanpur region for the first time. The major conclusions of our study are as follows:

- 1. Similar to the winter profile of BC mass concentration in Kanpur, higher BC concentration as well as F_{BC} is found in summer at higher altitude (~1200 m), due to increased convective activities. However, the vertical gradient in BC concentration is lower than the variations of BC concentrations at that altitude. The seasonal changes in boundary layer characteristics affect the BC altitude variation much as compared to the total aerosol, thus increasing F_{BC} with altitude in summer.
- 2. $R_{m,f}$ and $R_{m,c}$ profiles are contrastingly different in the FN and AN in summer. $R_{m,f}$ and $R_{m,c}$ are both larger in the AN than in the FN indicating upward transport of more number of coarse aerosols during AN hours due to stronger convective activity.
- 3. The heating rate profile of FN follows zigzag pattern with highest value (2.1 K day^{-1}) at 300 m altitude, whereas a second peak appears at 1200 m altitude. On the contrary, the AN profile shows steadily increasing heating rate reaching maxima $(1.82 \text{ K day}^{-1})$ at the same altitude. Persistence of the high BC concentration over Kanpur region in the winter as well as summer season would have significant impact on the regional climate forcing.

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