# Possible climatic implications of high altitude emissions of black carbon 

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## Authors' reply to comments from the second reviewer

First of all, we would like to thank the anonymous reviewer for appreciating our work, giving constructive suggestions and overall positive recommendations.

## General comments from the reviewer:

1. This study investigates the reasons behind measured high-altitude BC layers over India for 3 separate days using a chemistry transport model. The topic is relevant and within the scope of ACP. The introduction is clear and well-written. When it comes to the general presentation of the results and the figures, however, I think this manuscript needs further work before I can recommend it for publication. First, the aircraft emissions in this study were scaled, but this is only mentioned in one sentence and never mentioned again in the abstract or conclusion.
-Authors' reply:
We accept the aforementioned suggestion from the reviewer. The manuscript will be modified and the corresponding changes will be made.
2. The authors correctly states that the uncertainties in emissions are large, but it is not clear to me why the emissions were scaled, by how much, and how this could be justified.
-Authors' reply:
This point raised by the reviewer is similar to the point raised by the first reviewer in the third specific comment. The aircraft emissions of BC from MACCity inventory have been modified in this study by using published data regarding fuel efficiency of an aircraft, recent estimates of the emission factor of aircraft fuel for BC and the published data of air traffic over Hyderabad. The explanation to the modification/scaling of the emissions can be found in part 'b' of the
authors' reply to the third specific comment from the first reviewer.
3. Second, the manuscript is written like a story ('then we did this and then we did that'), and it needs to be rewritten to a more common form with 'Methods' and 'Results' clearly separated. I also miss a 'Discussion' section where the authors discuss some of the uncertainties with their study, e.g. the limitation of data (only 3 days), the scaling of the emissions etc. Third, the figures need to be plotted on the same format, and some of them can be combined or removed. Finally, I think there is a tendency for the interpretation of the results to be slightly overstated. The study itself is interesting, and it would read much better with a plain description of the results, without any 'convincing'. There are also use of words like 'significance difference' without any significance testing mentioned.
-Authors' reply:
We partly agree with the observation of the reviewer on the writing style and in the revised version we have changed it to a large extent taking cue from the comments. A discussion section has also been added in the revised version of the manuscript. To strengthen our results, we have carried out 1 month long model simulations (with prescription of aircraft BC emissions) for each of the 4 seasons. A brief discussion about the results of those simulations has been presented in the reply to the first general comment from the first reviewer. Those results will be discussed in greater details in the modified version of the manuscript. A few figures have also been combined into one figure for better understanding. Additionally, as suggested, wherever necessary, the statistical parameters like correlation coefficient, significance level have been computed.

## Specific comments from the reviewer:

## 1. Abstract

Reviewer's comment:
Please specify the 3 dates for the measurements, and that you scaled the aircraft emissions.
-Authors' reply:
The corresponding modifications will be made in the abstract.

## 2. Fig. 1

Reviewer's comment:
What does the error bars illustrate? Do you know why the error bars are much larger in a) and b) compared to c)?
-Authors' reply:
The error bars represent ensemble standard deviation. The balloon data is not equi-spaced along the vertical. The raw aethelometer data are averaged over regular altitude intervals combining the data obtained during the ascending and descending phases; and the bars represent the standard deviations representing the spread of the values in each ensemble.


Figure 1. Observed vertical profile of BC over Hyderabad obtained from balloon measurements (a). during 17 March 2010 (b). during 8 January 2011 (c). during 25 April 2011. The high altitude BC peaks in each profile are identified by letters P1 to P5.

Larger amplitudes of these bars represent larger fluctuation in the data. The bars are shorter in fig. 1c, due to averaging over a larger vertical extent (a little reduced vertical resolution), because of the larger variation in the ascent speed of the balloon in that flight.
3. Chapter 2:

Reviewer's comment:
Can you add a small paragraph on how BC were measured using an Aethalometer, e.g. filter-based absorption. What are the uncertainties on separating out other absorbing components, like dust? Can you also report on the MAC that was used? I suggest specifying that 'elemental carbon' is measured (eBC), as suggested in literature (Petzold et al. (2013) and references within). I suggest to drop Fig. 3 of the manuscript.
-Authors' reply:
We have replied to the reviewer's comment one by one.
-Reviewer's comment:
Can you add a small paragraph on how BC were measured using an Aethalometer, e.g. filter-based absorption. Can you also report on the MAC that was used?
-Authors' reply:
The mass concentration of 'Equivalent Black Carbon' (EBC) was measured using an Aethelometer (Model AE-42, of

Magee Scientific, USA). The aethelometer estimates the mass concentration of EBC by measuring the change in the transmittance of a quartz filter to 880 nm light upon the deposition of aerosol. The value of effective Mass Absorption Cross-section (MAC) used in these measurements is $16 \mathrm{~m}^{2} \mathrm{~g}^{-1}$ (Hansen, 1996, 2005). The effective MAC includes the amplification of absorption due to multiple scattering on the filter fiber matrix and the decrease due to shadowing. The aethalometer was configured for volumetric flow with an external pump, providing a flow rate of 14 liters per minute (LPM) at ground, and operated at a time base of 5 s . The data from the aethelometer was telemetered down along with the GPS co-ordinates. A few studies have reported uncertainties in the aethalometer estimated EBC (for e.g., Weingartner et al. (2003); Arnott et al. (2005); Sheridan et al. (2005); Corrigan et al. (2006)). Following the suggestions from the reports, we have used an amplification factor of 1.9 and an ' $R$ ' factor (shadowing effect) of 0.88 in this study. Nevertheless, when the aerosols are aged or mixed with others, as they would be away from the direct emissions, the shadowing effect would be negligible (Weingartner et al., 2003).
The modified manuscript will have the paragraph written above. The figure 3 has been removed from the manuscript. -Reviewer's comment:

- What are the uncertainties on separating out other absorbing components, like dust?
-Authors' reply:
- The Mass Absorption Cross-section (MAC) for dust is two to three orders of magnitude lesser than the for BC (Hansen, 2005). Hence if the mass of dust is substantially higher than EBC, only then same optical absorption will be produced. Thus under normal conditions, the effects of dust on measured EBC mass concentrations would be negligible.
-Reviewer's comment:
- I suggest specifying that 'elemental carbon' is measured (eBC), as suggested in literature (Petzold et al 2013 and references within).
-Authors' reply:
- As suggested by the reviewer, we will follow the terminology specified by Petzold et al. (2013). The mass concentration measurements made using aethelometer will be reported as the 'Equivalent Black Carbon' (EBC) instead of 'Black Carbon' (BC).


## 4. Reviewer's comment:

There are many figures and I think it would be easier to read if some of the figures could be combined. For instance, may be Fig. 1 and Fig. 2 and Fig. 5/6 from the manuscript can be merged into one bigger figure (with 9 equally sized panels) so it is easier to compare the peaks? In that case; can you plot the panels on the same y axis, some goes up to 10 km vs. 9 km vs 12 km . I find it confusing that the axis and panels are plotted differently for each figure, e.g. comparing Fig. 5 and Fig. 6 from the manuscript, where the black line in Fig. 5 are the same as blue line in Fig. 6 (?). I suggest dropping Fig. 5 from the manuscript as the same info is in Fig.6. Fig 9 and Fig 10 from the manuscript can also be combined (with 9 panels).
-Authors' reply:

We agree with the modifications suggested by the reviewer. The corresponding suggestions are incorporated in the modified version of the manuscript. The modified figures can be found below as fig. 2 and fig.3.


Figure 2. Observed vertical profile of BC over Hyderabad obtained from balloon measurements (a). during 17 March 2010 (b). during 8 January 2011 (c). during 25 April 2011. The high altitude BC peaks in each profile are identified by letters P1 to P5. Observed vertical profile of $\mathrm{dT} / \mathrm{dz}\left(\mathrm{K} \mathrm{km}^{-1}\right)$ over Hyderabad region obtained from balloon measurements (d). during 17 March 2010 (e). during 8 January 2011 (f). during 25 April 2011. The locations corresponding to high altitude BC peaks are identified by letters P1 to P5. Model simulated vertical profile of BC over the area in the vicinity of the balloon flight region for (blue line) NoACEM/Ctrl (control run) and (red line) ACEM (runs with prescription of aircraft BC emissions) during (g). 17 March 2010, (h). 8 January 2011 and (i). 25 April 2011.

## 5. P6, L16:

## Original sentence from the manuscript:

While showing in general reduction in temperature with height similar to the observations (Red line, fig.4d), the model (Blue line, fig.4d) shows discrepancies in actual magnitudes vis-a-vis the observations, as in case of the other meteorological variables.
Reviewer's comment:


Figure 3. Vertical profiles of atmospheric heating rates for the observed BC profiles during (a).March 2010, (b). January 2011 and (c). April 2011. Vertical profiles of atmospheric heating rates corresponding to the observed BC profiles during (a).March 2010, (b). January 2011 and (c). April 2011. Vertical profiles of atmospheric heating rates corresponding to the model simulated BC profiles during (d and g). 17 March 2010 (e and h). January 2011 (f and i). 25 April 2011.

Can you report numbers for the differences in magnitudes you are referring to? It is hard to read on the figure.
-Authors' reply:
The model simulated vertical profile of temperature (Blue line, fig.4d) in-general shows similarity with the observed profile of temperature (Red line, fig.4d) retrieved from the thermometer attached as a pay-load to the balloon. However, the two profiles differ in the exact magnitudes of the temperature. The differences in the observed and the simulated temperature magnitudes lie within -1.7 to +3.4 K .

## 6. P6, L25:

## Original sentence from the manuscript:

The magnitudes of the simulated BC significantly match the observations only over the lower most altitudes i.e. below 3.5 km .

Reviewer's comment:
What do you mean by 'the simulated BC significantly match the observation'? Did you do any significance testing on


Figure 4. Comparison between the model simulated meteorological variables in NoACEM/Ctrl (control) run and the corresponding observations over the balloon flight region on 17, March 2010 for (a) wind speed (b) wind direction (c) Potential temperature (d) Temperature
this? Again, it is difficult to compare the model and the observations since the plots are so different (axis).
-Authors' reply:
During the comparison of the observed vertical profile of BC obtained from the balloon measurements with the model simulated vertical profile of BC in the control run (i.e. without the prescription of BC emissions from aircrafts), upon the visual inspection of the two profiles we notice that, the profiles roughly match each other within the lower atmosphere but differ drastically from each other beyond 3.5 km . The model does not depict the sharp elevated layers of BC (as seen in the observed BC profiles) in the simulations without the prescription of aircraft emissions of BC. So, in this comparison, we just wanted to highlight that, the rough agreements in the profiles are seen below 3.5 km and the disagreements in the profiles beyond 4 km altitude. Relatively better agreements between the model simulated vertical profile of BC and the observed BC profile are seen within 0.5 km to 2 km altitude band. Due to coarse resolution of model within this band (hence lesser number data points) vis-a-vis the observations, we could not find out the correlation coefficient between the two profiles and hence the significance of the correlation coefficient of this match could not be found out. Nevertheless, we have computed the ratio of the observed BC and the modeled BC all throughout the vertical for these profiles and same are shown in fig. 5 . It could be clearly seen that the ratio shows relatively lower values (ratio $\sim 3$ ) upto 4 km altitude, specifically between the altitude band of 0.5 to 2 km (ratio ranges between 0.6 to 2 ). On the other hand, the ratio shows values greater than 10 beyond 4 km . Thus, we say that the model simulated BC shows better comparison with the observations below 3.5 km altitude. We will accordingly modify the sentence in the manuscript. The modified
figure (Fig.2) also has the modeled and observed vertical profiles of BC on the same vertical axis.


Figure 5. Ratio of the observed BC to the model simulated BC (in the control run)

## 7. P7, L5:

## Original sentence from the manuscript:

The meteorology (observed as well as simulated) being benign for all the cases, and possibility of any long-range transport of BC from other location being an unlikely cause for such high concentrations

Reviewer's comment:
What do you mean by the meteorology being benign? And that the long-range transport of BC is unlikely?
-Authors' reply:
In this study, we first noticed that, the model WRF-Chem, simulated the meteorological parameters with satisfactory agreements with the corresponding observations obtained from the balloon measurements. In spite of such satisfactory agreements between the modeled and the observed meteorological parameters, the model fails to capture the high altitude sharp peaks in BC as observed. Thus we noticed that the local meteorology may not have played a major role in causing the observed sharp and confined high altitude BC peaks. Hence, we say that the local meteorology may not explain or be responsible for the observed sharp high altitude BC peaks.
To assess the role played by long range transport of BC, Babu et al. (2011) used the Lagrangian back trajectory model, Hybrid Single Particle Lagrangian Integrated Trajectory HYSPLIT. The authors plotted the isentropic 7 day back trajectories of air parcel arriving over the balloon flight location at different vertical levels. It was found that, the air parcels beyond 5 km have origin in the African region or beyond that. It was also found that, BC showed very low concentrations in those altitudes up to around 8 km ; before increasing again. Interestingly, between $8-9 \mathrm{~km}, \mathrm{BC}$ showed one more
peak in concentration (Fig.2a), but the trajectories did not show any conspicuous shift. Thus, possibility of long-range transport in causing the peaks in BC was ruled out.
Thus, we rule out the local meteorology and the long-range transport as possible causes for the existence of such high altitude BC peaks. A citation will be provided to Babu et al. (2011) in this regards.

## 8. Reviewer's comment:

Your manuscript is written more like a story, and how you came about with the different hypothesis, but I suggest rewriting the manuscript with the more common separation of 'Methods' and 'Results', which means e.g. that 4.3 and 4.6 (until L15 on P10) should be moved to 'Methods' along with everything else in Results that explains the methods you have used. Parts of 4.7 explaining background for convective lifting can be moved to introduction (and the rest to methods). Same goes for Conclusions.
-Authors' reply:
We appreciate the aforementioned modifications suggested the reviewer and the corresponding changes in the layout of the manuscript will be made.

## 9. P7 L32:

## Original sentence in the manuscript:

Thus, considering such underestimations of $\operatorname{EI}(\mathrm{BC})$ in the current emissions inventory, we modified the emissions and forced our model with such emissions at 23 levels with appropriate mapping to model levels.
Reviewer's comment:
This is the first time and only place you say that you have scaled the aircraft emissions you have used. How did you modify the emissions? You also need to mention this in the abstract/introduction, throughout the text and in the conclusions.
-Authors' reply:
This point raised by the reviewer is similar to the point raised by the first reviewer in the third specific comment. The aircraft emissions of BC from MACCity inventory have been modified in this study by using published data regarding fuel efficiency of an aircraft, recent estimates of the emission factor of aircraft fuel for BC and the published data of air traffic over Hyderabad. The explanation to the modification/scaling of the emissions can be found in part 'b' of the authors' reply to the third specific comment from the first reviewer.
As suggested by the reviewer, the modifications in the emissions will be mentioned in the manuscript.
10. P9 L29:

## Original sentence from the manuscript:

This aerosol mixture has water soluble species ( $28000 \mathrm{\#} \mathrm{~cm}^{-3}$ ), insoluble species ( $1.5 \mathrm{\#} \mathrm{~cm}^{-3}$ ) and soot ( 130000 \# $\mathrm{cm}^{-3}$ ).

Reviewer's comment:
How is 'soot' defined in this context?
-Authors' reply:

The soot component considered in the Optical Properties of Aerosols and Clouds (OPAC) model is used to represent absorbing Black Carbon. The BC particles are not soluble in water and hence are assumed not to grow with relative humidity. The density of soot is taken as $1 \mathrm{~g} \mathrm{~cm}^{-3}$. The optical properties are calculated assuming the size distribution with many very small particles (which would have the density $2.3 \mathrm{~g} \mathrm{~cm}^{-3}$ ). More details about the soot component defined in OPAC can be found in Hess et al. (1998).

## 11. P8, L7

## -Original sentence from the manuscript:

the nature of the layers (sharp and confined) looks very similar to those seen in the measurements
-Reviewer's comment:
'the nature of the layers (sharp and confined) looks very similar'. Please rewrite. Also, the peaks are much smaller in magnitude and are not located at the same altitudes as the observations?
-Authors' reply:
We acknowledge that the model simulated high altitude BC peaks are smaller in magnitudes vis-a-vis the observed high altitude BC peaks. Additionally, the model simulated peaks occur at different altitude as compared to the observation. Nevertheless, the overall pattern or character of the model simulated high altitude sharp and confined BC peaks resembles the observed high altitude BC peaks. Moreover, the correlation coefficients between the model simulated peaks and the observed peaks within the altitude band of the respective peaks comes out to be 0.8 and 0.97 for March and January profiles. The correlation coefficients are $95 \%$ and $99.9 \%$ significant respectively.
-Modified sentence:
the sharpness of the modeled BC layers make them look similar to the observed BC layers.

## 12. P8, L11:

## Original sentence from the manuscript:

This clearly highlights the role played by aircraft emissions of BC in creation of the high altitude BC peaks
-Reviewer's comment:
'This clearly highlights the role played by aircraft emissions' However, you did scale the emissions? You need to emphasis this more. Do you see the peaks when you run with unscaled emissions?
-Authors' reply:
In this study, the aircraft emissions of BC from MACCity inventory have been modified by using published data regarding fuel efficiency of an aircraft, recent estimates of the emission factor of aircraft fuel for BC and the published data of air traffic over Hyderabad. The explanation to the modification/scaling of the emissions can be found in part ' b ' of the authors' reply to the third specific comment from the first reviewer. The model does not capture the high altitude BC
peaks with unscaled emissions.

## 13. P8, L34:

Original sentence from the manuscript:

Beyond 4 km , the profiles are identical, implying that the elevated BC layers are insensitive to surface BC emissions. -Reviewer's comment:
'Beyond 4 km , the profiles are identical'. It is hard to say this without any significance testing.
-Authors' reply:
As suggested by the reviewer, we have further analysed the model simulated vertical profiles of BC in the two cases i.e. with and without the surface emissions of BC. Specifically, we have compared the the two profiles beyond 4 km altitude. The correlation coefficient between the two BC profiles beyond 4 km comes out to be 0.97 which is $99.99 \%$ significant. Moreover, the the magnitudes of BC in the two profiles show good agreement with a difference limited to only $0.1 \mu \mathrm{~g}$ $\mathrm{m}^{-3}$. This analysis suggests that the two profiles significantly match each other.

Modified sentence:
Beyond 4 km , the profiles are similar
14. P9, L23:

## Original sentence from the manuscript:

During the winter flight (January 2011, fig.3b), the observed BC profile causes more heating near the surface than that at higher heights.
-Reviewer's comment:
: '..the observed BC profiles causes more heating near the surface'. Did you show that the observed BC profiles cause this heating?
-Authors' reply:
We in this study, compute the atmospheric heating rates caused by aerosol in general. In computation of the heating rates we use vertical profiles of the extinction coefficient obtained from CALIOP LIDAR on-board the CALIPSO satellite, over the Hyderabad region and the observed vertical profile of BC, using the methodology discussed in section 4.6.1 of the manuscript. Hence the heating rate profiles are not caused by BC alone but by the entire aerosol mixture. The corresponding changes in the manuscript will be made in this regards.

## Modified sentence:

the observed BC profiles along with extinction coefficient profiles from CALIPSO, cause more heating near the surface
15. P10, L29:

## Original sentence from the manuscript:

The identical nature of the heating rates profiles during March 2010 and April 2011, brings out the average features of heating rate profile during summer months over the region of study.

## Reviewer's comment:

: 'The identical nature of (..), brings out the average pattern'. I do not understand this sentence.
-Authors' reply:

The vertical profiles of atmospheric heating rates due to observed BC profiles and the extinction coefficient profiles from CALIPSO during March 2010 (Fig.3a) and April 2011 (Fig.3c) show largely similar patterns. The correlation coefficient between the 2 profiles comes out to be 0.96 which is $99.99 \%$. Such large similarities in the 2 profiles of heating rates over Hyderabad region, computed for 2 different years, highlight the consistent broad-features of the heating rate profiles over Hyderabad region during the pre-monsoonal period. Thus, we say that the identical pattern of the heating rate profiles during pre-monsoonal month (Fig.3a and Fig.3c), bring out the broad features of the the pre-monsoonal heating rate profile.

## Modified sentence:

The largely similar nature of the heating rates profiles during March 2010 and April 2011, brings out the average features of heating rate profile during summer months over the region of study.

## 16. P11, L21:

## Original sentence from the manuscript:

We entitle 'starting point of the trail' as that area in the aircraft emitted BC trail, where emission magnitudes are substantially higher than that over the remaining part of the trail

Reviewer's comment:
I am not sure if I understand your explanation of 'normal' and 'extreme' profiles in terms of the starting point. Area in the model?

Authors' reply:
The high altitude emissions of BC from aircrafts are in the form of a narrow trail. The area where two or more such trails cross each other (mainly in the vicinity of an airport) show higher magnitudes of emissions than the remaining part of the trail. So, we mainly refer to these areas when we use the phrase 'starting point of the trail'. Nevertheless, to avoid such confusion, the use of the phrase 'starting point of the trail' has been avoided. Instead, we simply specify such area as the one which shows higher magnitudes of aircraft emissions relative to the remaining part of the trail.

## 17. Fig. 13 from the manuscript:

Reviewer's comment:
Fig. 6 is very difficult to read.
-Authors' reply:
We have made modifications in the caption of the figure, for better understanding. The modified caption can be found with the figure 6 and is written in this reply as well.


Figure 6. The time-series of the vertical profile of extinction coefficient at stratospheric altitude, stratospheric AOD and the vertical profile of particle depolarization ratio at stratospheric altitude are plotted for each of the 5 regional boxes shown in fig. 12 (previous figure from the manuscript). The time-series are plotted from January 2010 to December 2012. The time-series of the aforementioned parameters for each region are joined to each other to form a single time-series for each parameter. Such appended time-series is shown for a) vertical profile of extinction coefficient at stratospheric altitude b) stratospheric AOD and c) the vertical profile of particle depolarization ratio at stratospheric altitude. The letter ' $M$ ' signifies the monsoon season (June-July-August-September) embedded within the time-series for each region. In 'b', the background stratospheric AOD for the entire tropical belt (Kremser et al., 2016) is shown by the dotted red line.

## Modified caption:

The time-series of the vertical profile of extinction coefficient at stratospheric altitude, stratospheric AOD and the vertical profile of particle depolarization ratio at stratospheric altitude are plotted for each of the 5 regional boxes shown in fig. 12 (previous figure from the manuscript). The time-series are plotted from January 2010 to December 2012. The time- series of the aforementioned parameters for each region are joined to each other to form a single time-series for each parameter. Such appended time-series is shown for a) vertical profile of extinction coefficient at stratospheric altitude b) stratospheric AOD and c) the vertical profile of particle depolarization ratio at stratospheric altitude . The letter ' M ' signifies the monsoon season (June-July-August-September) embedded within the time-series for each region. In 'b', the background stratospheric AOD for the entire tropical belt (Kremser et al., 2016) is shown by the dotted red line.

## Technical corrections:

1. P2, L1

## Original sentence from the manuscript:

Several earlier studies have shown that such an atmospheric heating by a layer of aerosol with high BC abundance
-Reviewer's comment:

What is 'such an' refer to here? I suggest to remove.
-Authors' reply:
We follow the suggestion from the reviewer. The words 'such an' have been removed from the text.

## 2. P2, L12:

## Original sentence from the manuscript:

Additionally, the atmospheric heating at higher altitude can give rise to local stable scenario below, which can affect convection and consequently impact precipitation.
-Reviewer's comment:
Can you provide references to precip/convection?
-Authors' reply:
The surface cooling and atmospheric warming due to black carbon can alter the vertical profiles of temperature, induce more stability and reduce convection and the resulting rainfall. Fan et al. (2008) discusses these effects of BC on convection and rainfall in greater details. The study will be cited in the modified manuscript.
3. P6, L3:

Original sentence from the manuscript:
up to $1^{s t} 2 \mathrm{~km}$ of the lower troposphere
-Reviewer's comment:
$1^{\text {st }}$-first
-Authors' reply:
The corresponding changes will be made in the manuscript.

## 4. P6, L8:

## Original sentence from the manuscript:

barring a few discrepancies such as the extent of stable and well mixed layer in the lower atmosphere, magnitude of $\theta$ in the vicinity of the primary BC maxima in observations (around $4-4.5 \mathrm{~km}$, fig. 2 a ) etc.
-Reviewer's comment:
either remove 'etc..' or replace by explaining the other discrepancies.
-Authors' reply:

The word 'etc' will be removed from the text.

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