

# Possible climatic implications of high altitude emissions of black carbon

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## Authors' reply to comments from the first reviewer

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

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### General comments from the reviewer:

The authors first showed vertical distributions of BC simulated by WRF-Chem around Hyderabad/India for three different days (17 March 2010, 08 January 2011, and 25 April 2011). The previous study showed that the observed BC has prominent spikes in 4 km or higher altitudes. The authors identified that the source of the spikes of BC is the aircraft emission. Second, the authors confirmed that the BC particles from the aircraft emission reach in lower stratosphere by using both the simulation and observation of CALIPSO. The manuscript and logic are straightforward, but I would like to ask the authors to add more explanation for more deeply understanding.

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Especially, how much are the results representative in each season? Although the measurements are very limited, i.e., one day per one season, the simulation seems to be easily conducted in at least one month to generalize and strengthen the conclusions. In overall, the manuscript would be acceptable for publication if these comments can be satisfactorily addressed.

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### Authors' reply:

We appreciate this suggestion from the reviewer regarding seasonal simulations. As suggested by the reviewer, we have carried out 1-month long model simulations for each season i.e. winter, pre-monsoon, monsoon and post-monsoon, of the year 2010. The months January, March, July and October are taken as representatives of the respective seasons. The analysis of the monthly mean vertical profiles of BC from the model simulations brings out following main results:

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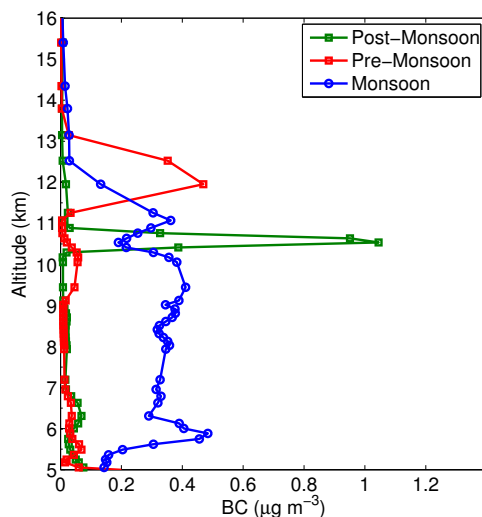
1. During the winter, pre-monsoon and the post-monsoon seasons, a large number of locations within the model domain show more than 50% probability of occurrence of a high altitude (9-11 km) BC peak (a few locations show more than 60% probability).

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2. The upper levels winds and the aircraft BC emissions themselves play a major role in deciding the horizontal location of the high altitude (9-11 km) BC peaks.

3. During the monsoon season, such probability values and the locations with high-altitude (9-11 km) BC peak reduce. The main reason for such reduced occurrence of high altitude BC peaks during the monsoon season appears to be the monsoonal

5 convective lifting of BC which uniformly distributes BC at the upper levels (blue line, fig.1), which increases the total BC mass concentration at the upper levels, while getting rid of the sharp BC peaks.



**Figure 1.** The model simulated vertical profile of BC mass concentration ( $\mu\text{g m}^{-3}$ ) in the vicinity of balloon flight domain during different seasons, Pre-Monsoon (red line), Monsoon (blue line) and Post-Monsoon (green line), of the year 2010.

The detailed analysis of the seasonal simulations will be included in the modified manuscript.

10 **Specific comments from the reviewer:**

1. **P.2, L27:**

- **Original sentence from the manuscript:**

Synthesizing multi-platform measurements Satheesh et al. (2008) have revealed the existence of elevated aerosol layers in the middle troposphere.

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- **Reviewer's comment:**

Could the authors clarify actual heights of "the middle troposphere" here?

- **Authors' reply:**

Here "the middle troposphere" refers to a height of 4 to 5 km.

2. **P.5, L2**

- **Original sentence from the manuscript:**

5 The model takes into account the following aerosols species: BC1 (Hydrophobic), BC2 (Hydrophilic)

-**Reviewer's comment:**

How do the authors classify the BC1 (hydrophobic) and BC2 (hydrophilic)? Do the authors consider atmospheric aging processes of BC?

- **Authors' reply:**

10 The regional chemistry transport model WRF-Chem, used for these simulations considers black carbon aerosol to be present in two different modes: hydrophobic (BC1) and hydrophilic (BC2). The characteristic conversion e-folding lifetime from hydrophobic to hydrophilic i.e. BC1 to BC2 is considered to be 2.5 days. The primary emissions of BC are assumed to occur in the hydrophobic mode (BC1). The BC is assumed to be removed from atmosphere by dry deposition (for both the hydrophobic and hydrophilic modes) and wet deposition (for the hydrophilic mode). More details about the  
15 treatment of BC in WRF-Chem can be found Kumar et al. (2015).

3. **P.7, L32-33:**

- **Original sentence from the manuscript:**

Thus, considering such underestimations of EI(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.

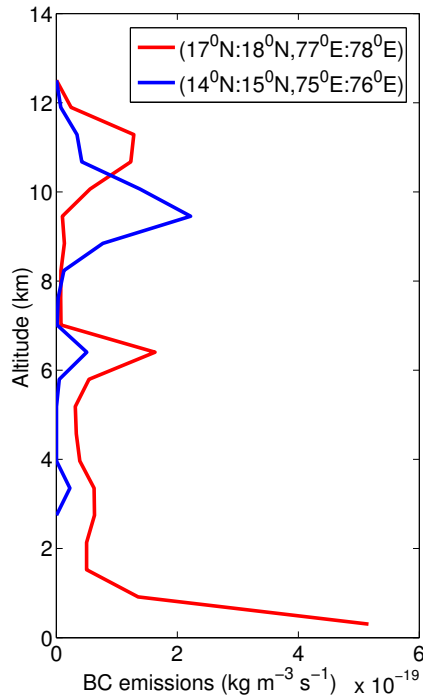
20 -**Reviewer's comment:**

The modification is not clear. Especially, the readers cannot understand the vertical profile of the BC emission. Please clarify it.

-**Authors' reply:**

**a). Vertical profile of the BC emissions from the aircrafts within the MACCity inventory:**

25 The vertical profile of BC emissions from aircrafts, averaged over a  $1^0 \times 1^0$  grid box centering Hyderabad (fig.2, red line) and that over another grid box (fig.2, blue line) of the same dimension just  $2^0$  south-west of the Hyderabad grid box, are shown in fig.2. It could be clearly noted that the vertical profile of BC emissions from aircrafts shows multiple-peaks over the Hyderabad region (fig.2, red line), with highest emissions near the surface followed by a peak at 6 km and at 11 km. On the other hand, over the other grid box which is relatively farther from the airport location, the vertical profile of  
30 BC emissions from aircrafts (fig.2, blue line) shows more dominant peak at higher levels (10 km) with relatively lesser BC emissions at 6 km and 4 km altitudes. The near-surface peak in BC over Hyderabad (fig.2, red line) could be due to the Landing and Take-Off (LTO) activities within the airport. The peaks at an altitude of 10 km (fig.2, red line) could be due to the aircrafts which are flying at the cruising altitudes and not landing at or taking-off from Hyderabad airport.



**Figure 2.** The vertical profile of BC emissions ( $\text{kg m}^{-3} \text{s}^{-1}$ ) from aircrafts in the MACCity inventory over  $1^\circ \times 1^\circ$  grid boxes a) centered over Hyderabad (red line) b) located  $2^\circ$  south-west of Hyderabad (blue line) during the month of March, 2010.

### b). Modification of aircraft BC emissions:

The MACCity emission inventory database (Lamarque et al., 2010), used in our study for the emissions of BC from aircrafts, is prepared using AERO2k database of global aircraft movement (Eyers et al., 2004), fuel consumption from PIANO aircraft emission tool (Simos, 2004) and the BC emission factors reported by Eyers et al. (2004). To assess this inventory over Hyderabad (our study region), we estimated BC emissions from aircrafts over this region using previously reported values for fuel efficiency of different airline carriers (Kwan and Rutherford, 2015), seating capacity of the carriers (websites of different airlines), density of aviation fuel used (Cookson and Smith, 1990; Arkoudeas et al., 2003; Outcalt et al., 2009; Blakey et al., 2011) in Indian region, BC emission index for the aviation fuel (Stettler et al., 2013), and actual data on air traffic over Hyderabad obtained from Air Traffic Controller, Hyderabad (Babu et al., 2011), following the method given below.

In view of the reliable information of the fuel efficiency of different airline carriers in India, we used the fuel efficiency of Trans-Atlantic aircrafts, reported by Kwan and Rutherford (2015), which gives values in the range 27 passenger-km/L for British Airways to 40 passenger-km/L for Norwegian airlines (Passenger – km per litre = No. of passengers in an aircraft  $\times$  km/L for 1 passenger). We also estimated the average seating capacity of different aircrafts (regional, short-

haul, medium haul, long-haul etc) of the various airlines in India from their respective websites, and these are given in Table.1.

**Table 1.** Calculations regarding BC emitted by aircrafts in our single grid volume.

Type of air journey (haul)	Average number of seats	Average number of passengers	Fuel consumed per km by an aircraft (L/km)	Fuel consumed per km by an aircraft (kg/km)	BC emitted ( $\mu\text{g m}^{-3}$ ) by an aircraft in our model grid $\times 10^{-3}$	BC emitted ( $\mu\text{g m}^{-3} \text{ s}^{-1}$ ) by aircrafts in our single grid volume $\times 10^{-6}$
Regional	186	140	4.376	3.413	1.5	3.338
Short	181	135	4.234	3.3	1.452	3.22
Medium	288	216	6.774	5.28	2.325	5.167
Long	357	268	8.39	6.545	2.88	6.398

We used an average load factor of 75% for obtaining the values in the third column, following Bhullar (2017), which reports the average load factor for the year 2011-12 for the Indian domestic flights. Using this average passenger load, we estimated the fuel consumed in L/km in each case and these are given in the fourth column of the table 1. The mean density of aircraft fuel is taken as  $780 \text{ kg m}^{-3}$  (Cookson and Smith, 1990; Arkoudeas et al., 2003; Outcalt et al., 2009; Blakey et al., 2011) and the fuel consumed in kg/km for the different aircrafts are calculated and given in fifth column of table 1. These values are further used to estimate the quantity of BC emitted. However the BC emission index, (the amount of BC emitted per kg of aviation fuel) has a large uncertainty and several studies (Herndon et al., 2008; Onasch et al., 2009; Timko et al., 2010) have shown that, this parameter spans 4 orders of magnitude. Stettler et al. (2011) suggests that the currently used emissions inventories could underestimate EI(BC) by an order of magnitude or more. In this study, we have used an EI(BC) value of  $0.088 \text{ g(BC)/kg}$  of aircraft fuel burnt as suggested in a recent study by Stettler et al. (2013). Using the aforementioned EI, the emission intensity within a single grid volume of our model (i.e.  $2000\text{m} \times 2000\text{m} \times 100\text{m}$ ), if an aircraft crosses the grid volume, is estimated for different air journey types and is given in the seventh column of table 1. Using the above information, and the information obtained on the number of aircrafts that used the Hyderabad flight corridor (as per the information obtained from the ATC reported in Babu et al. (2011)), we made an estimate of the average BC emission rate for each aircraft type as given in column 7, table 1. Using the mean of seventh column, table 1 and assuming  $100 \text{ km}^2$  as the influence zone of Hyderabad ATC, we estimate the total emissions of BC over the Hyderabad region within the flight corridor of 8-10 km to be  $2.2536 \times 10^{-4} \mu\text{g m}^{-3} \text{ s}^{-1}$ . The corresponding total emissions within the flight corridor over Hyderabad region from MACCity inventory comes out to be  $3.606 \times 10^{-7} \mu\text{g m}^{-3} \text{ s}^{-1}$ , which is far lesser than our estimates. Thus one scaling factor comes from this comparison. Additionally, acknowledging the coarse resolution of the MACCity inventory, the ‘line-source’ nature of the freshly emitted aircraft trail and the finer resolution of our model simulations, we confine the aircraft BC emissions into a width of 2 km and a height of 100 m. The mass conservation of the emitted BC due to such confinement leads

to an additional scaling factor. The total scaling factor becomes the product of these two scaling factors. The modified emissions are formulated by multiplying the original emissions by the aforementioned scaling factors.

4. **P.13, L20-22:**

- **Original sentence from the manuscript:**

5 It can be seen that, the stratospheric AODs from our analysis over the 5 regional boxes are roughly 4-6 times higher than the background tropical stratospheric AOD.

-**Reviewer's comment:**

How do the authors determine the background tropical stratospheric AOD? Please add some references or evidence to the manuscript.

10 -**Authors' reply:**

In our study, the background tropical stratospheric aerosol conditions are taken from a recent paper by Kremser et al. (2016), which reviews the current knowledge regarding stratospheric aerosols. The paper reports mean stratospheric (tropopause to 40 km) AOD over the tropics (20<sup>0</sup>S-20<sup>0</sup>N) for the years 1985 to 2012 (Figure 4 (bottom), Kremser et al. (2016)). The corresponding values are taken from SAGE II (The Stratospheric Aerosol and Gas Experiment II) instrument for the years 1985-2005, GOMOS (Global Ozone Monitoring by Occultation of Stars) satellite for 2002 to 15 2010, and CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite for 2006-2012. In our study, we examine the mean tropical stratospheric AOD from Kremser et al. (2016) for the years 2008 to 2012. The maximum value of the mean tropical stratospheric AOD during that period is considered to be the background tropical stratospheric AOD in our study.

20 5. **P.14, L10-11:**

-**Original sentence from the manuscript:**

One of the major non-spherical aerosol species over this region is mineral dust. But dust is relatively too heavy and coarse to be lifted up to such altitudes.

-**Reviewer's comment:**

25 The reason to eliminate the existence of dust is not unclear for me, because there is a possibility to existence of dust particles in the fine mode. The fine particles of dust perhaps exist in the 21-22 km layers. Could the authors add more evidence or include the possibility of mineral dust to strengthen the reason of the elimination of dust in your analysis?

-**Authors' reply:**

30 The source for mineral dust aerosol species in stratosphere could be largely related to a) volcanic eruptions b) meteoritic debris and c) convective transport of tropospheric dust. As mentioned in the manuscript our study has been carried out for a period which is relatively volcanically quiescent. Thus, volcanically erupted dust may not have contributed much to the stratospheric aerosol burden during our study period. The meteoritic debris form a minor part (5-10%) of the stratospheric aerosol composition, especially at altitude less than 30 km (Turco et al., 1981), making them difficult to be captured by a LIDAR. Moreover, the meteoritic debris are reported to be coarse in size (i.e. having radius above 1  $\mu$ m)

**Table 2.** Year to year change in global air traffic reported by Airports Council International (ACI).

Year	% change in passenger traffic	% change in air cargo traffic	% change in flight movements
2013	4.6	0.9	0.6
2014	5.7	4.7	1.3
2015	6.4	2.6	2

(Turco et al., 1981; Mackinnon et al., 1982) (and hence would have large deposition rates) and are largely spherical in shape (Mackinnon et al., 1982) with lower values of particle depolarisation ratio (PDR less than 0.1 (Klekociuk et al., 2005)). The extinction coefficient associated with plume of meteoritic debris are 3 orders of magnitude lesser (Gorkavyi et al., 2013) than the values we notice over our study domain. These points together rule out the possibility of associating the observed values of extinction coefficient and PDR to the meteoritic dust. The convectively lifted dust and other air pollutants while can get into the higher altitude regime (Andreae et al., 2001; Randel et al., 2010; Vernier et al., 2011; Fadnavis et al., 2013; Corr et al., 2016), their transport over the Indian region is limited to around 20 km (Fadnavis et al., 2013), which is mainly governed by the height of convective towers (Meenu et al., 2010). Moreover, dust aerosol is 2.6 times heavier than BC (Hess et al., 1998) and also less solar absorbent; thus it is unlikely to get vertically lifted up beyond 20 km by large-scale or self lifting mechanisms (de Laat et al., 2012). Hence we eliminate the possibility of existence of tropospheric dust as well beyond 22 km altitude over our domain.

## 6. P.15, L10:

### **Original sentence from the manuscript:**

The continued negative trend in stratospheric ozone over 40<sup>0</sup>S-40<sup>0</sup>N from 1984 to 2014 (Bourassa et al., 2014) could possibly be related to the ever increasing aircraft traffic.

### **-Reviewer's comment:**

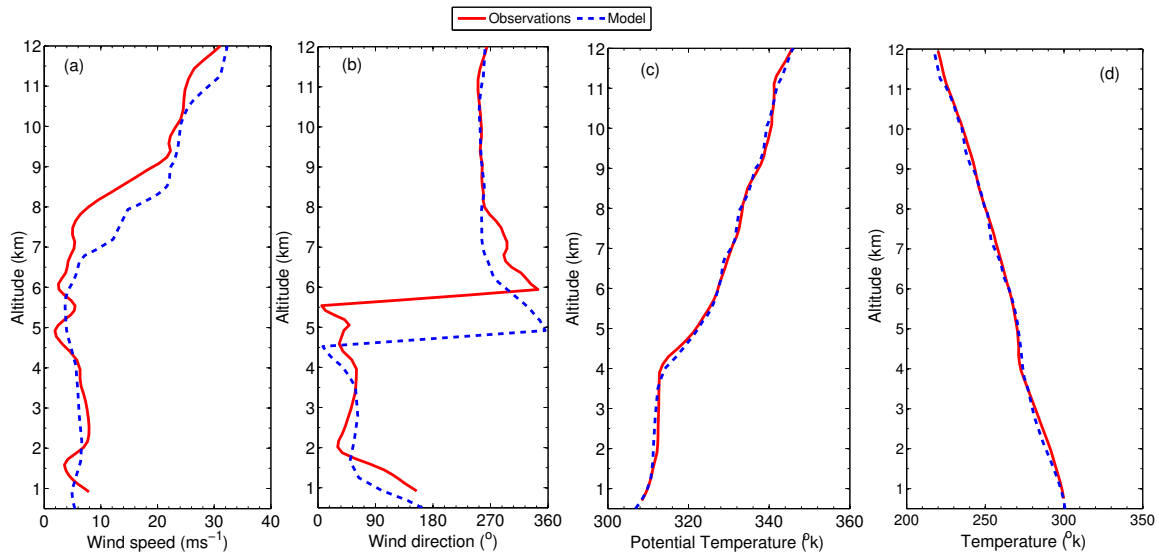
Do the authors have any evidence of the increasing aircraft traffic?

### **-Authors' reply:**

Airports Council International (ACI) (an international organization which is a representative of the world's airport authorities) reports following increasing trends (table 2) in global passenger traffic, cargo traffic and aircraft movements for the years 2013, 2014 and 2015 in their annual reports (Paraschis and Gittens, 2014; Piccolo and Gittens, 2015, 2016). Moreover, a continuous increasing trend in global passenger traffic has also been reported from the year 2003 (Paraschis and Gittens, 2014). These together confirm an ever increasing trend in global air-traffic for more than a decade.

7. Fig 4:

Figure from the manuscript:



**Figure 3.** 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

-Reviewer's comment:

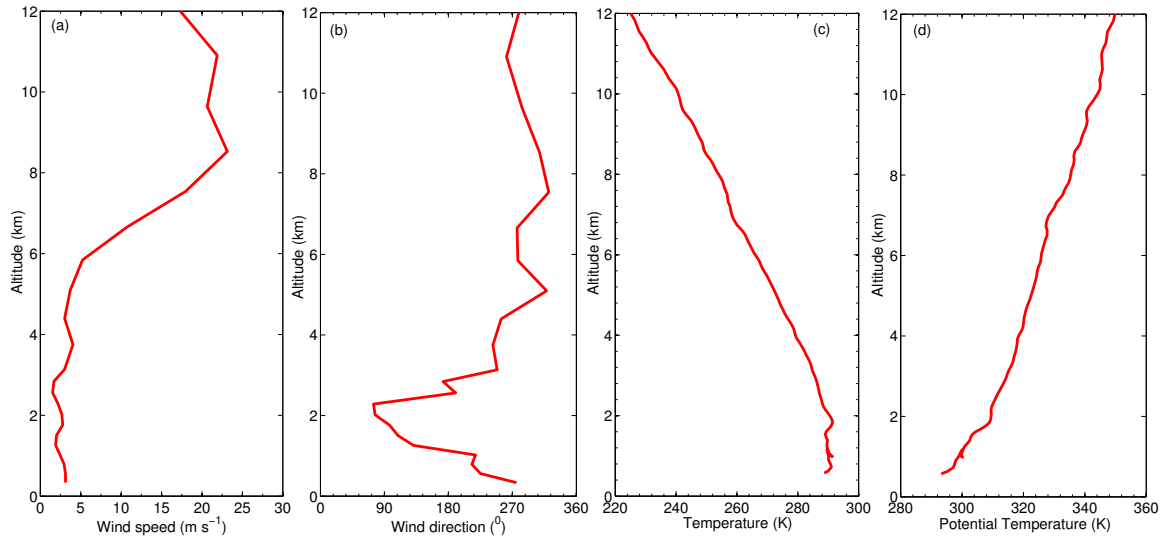
5 Is this weather a typical around Hyderabad. Also, how about the other days (8 January 2011 and 25 April 2011)?

-Authors' reply:

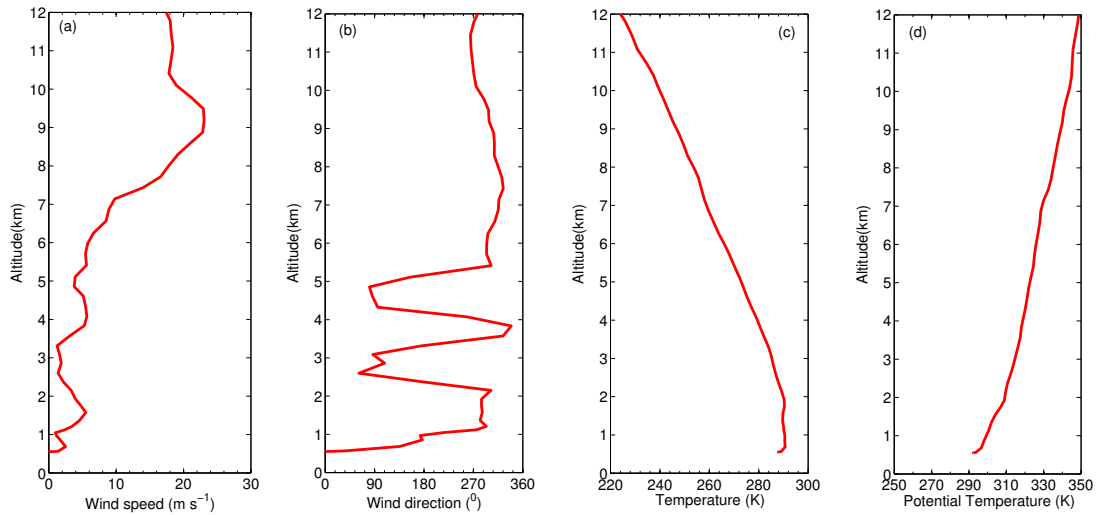
To examine this further we have plotted the meteorological fields over Hyderabad for the other days i.e. 8 January 2011 and 25 April 2011. The vertical profiles of wind speed, wind direction, temperature and potential temperature can be found in fig.4 for 8 January 2011 and fig.5 for 25 April 2011. It could well be noticed that the from fig.4a of the manuscript (fig.3a of this document), fig.4a and fig.5a that, the lower values of wind speed from surface to upto around 5-6km and gradual increase in wind speed beyond that, are seen as a consistent feature on all the 3 days over Hyderabad. Thus, such a feature looks to be a permanent character of winds over Hyderabad. Also, the prevalence of westerly to north westerly winds beyond 6-7 km also appears to be a common feature on all the 3 days (fig.3b, fig.4b and fig.5b).

15 The temperature and potential temperature on 17th March (fig.3d and fig.3c) show a different behavior vis-a-vis that during 8 January 2011 (fig.4c and fig.4d) and 25 April 2011 (fig.5c and fig.5d). While the temperature during March shows a decreasing trend from surface to around 4 km and an isothermal layer from 4 to 5 km (fig.3), the isothermal





**Figure 4.** Vertical profiles of meteorological variables obtained from balloon measurements during 8th January 2011, (a) wind speed (b) wind direction (c) Temperature (d) Potential Temperature



**Figure 5.** Vertical profiles of meteorological variables obtained from balloon measurements during 25th April 2011, (a) wind speed (b) wind direction (c) Temperature (d) Potential Temperature

layer of temperature is seen from surface to first 2km during January (fig.4c) and April (fig.5c) flights. The potential temperature in January (fig.4d) and April flights (fig.5d) also shows an increasing tendency throughout the vertical with a small layer around 2 km altitude with near-steady potential temperature, unlike the well mixed layer upto 4km during March (fig.3c). Thus, the winds fields show a consistent behavior during all the days, while temperature fields show some differences.

#### 8. Figs 5 and 6:

-Reviewer's comment:

What is the difference in NoACEM/Ctrl between Fig 5 and Fig 6? I am confused.

-Authors' reply:

The figures 5 and 6 have now been combined with fig.1 and fig.2 of the manuscript. The modified figure can be found attached below (fig.6). In the modified figure NoACEM simulations means the model simulations without emissions of BC from aircrafts.

#### 9. Fig 7:

-Reviewer's comment:

How much did the meteorological fields, i.e., air temperature, change by implementing the BC radiative impacts?

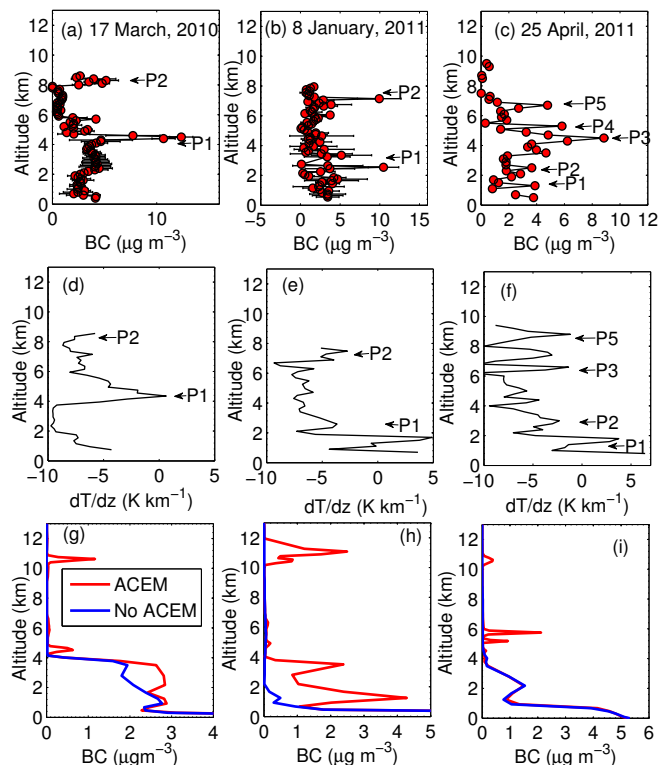
-Authors' reply:

The vertical profile of the changes in air temperature ( $\Delta T = T_{ACEM} - T_{NoACEM}$ ) due to the prescription of BC emissions from aircrafts are plotted in fig.7. In addition to  $\Delta T$ , the vertical profiles of BC after the prescription of emissions from aircrafts are also plotted. The analysis is done for all the 3 balloon flight days. It could be well noticed that, the prescription of BC emissions from aircrafts increases the temperature in the vicinity of the elevated sharp BC layers (around 4 to 6 km and 10 to 12 km during March, around 10 to 12 km during January and around 4 to 6 km during April). Additionally, such a prescription also reduces temperature just below the sharp BC layer (just below 4 km during March, near the surface during January and near the surface and just above 4 km during April). Such increments and reduction in the temperatures could be due to the absorption of shortwave and long wave radiation by aircraft emitted BC. The magnitude of such increment and reduction in temperature appears to be around  $\pm 0.2$  to  $0.3$  K. However, it may be noted that, the model simulated BC magnitudes within the elevated sharp layers are on an average 5-6 times lower than that within the observed elevated sharp layers (fig.6), thus the atmospheric heating and the corresponding changes in temperature within the model simulations would also be proportionally lower vis-a-vis the reality.

### 30 Technical Corrections

#### 1. P4, L23:

-Original sentence from the manuscript:



**Figure 6.** 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  $dT/dz$  ( $K km^{-1}$ ) 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.

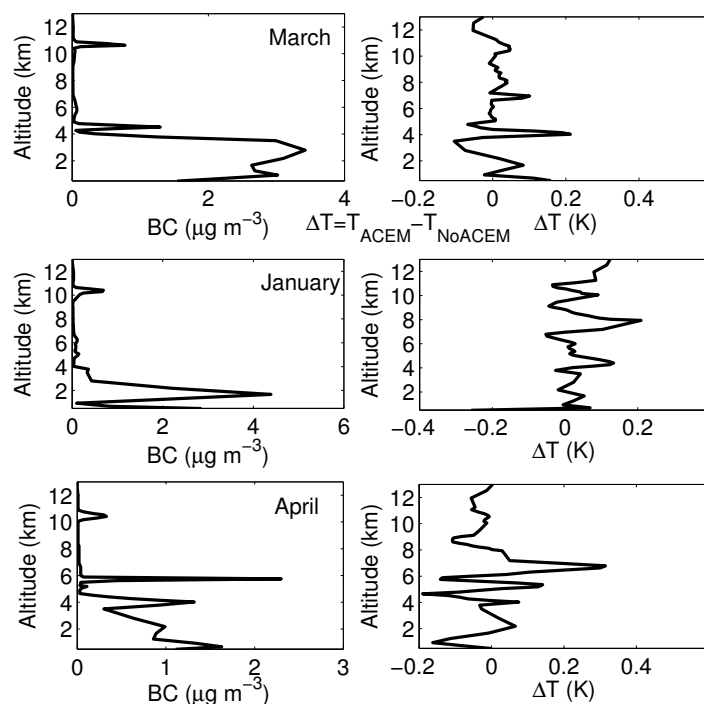
The WRF-Chem model used in this study employed horizontal grid spacing of 2 km with 70 vertical levels

-Reviewer's comment:

How do the authors define the 70-vertical layer? Please clarify each level.

-Authors' reply:

- 5 The model simulations have been carried out by specifying 70 vertical levels. The levels are specified as listed in the table 3 and are discussed below. Roughly, first 10 levels resolve the atmospheric boundary layer (i.e. upto 2km). Next 4 levels cover an altitude of 2km, with vertical spacing of 500m. Beyond 4km, next 16 levels are spaced with 100m vertical spacing and they reach upto a height of 6 km. Such a fine resolution has been set to resolve the sharp and confined observed layers BC within this altitude band. These layers are followed again by the layers with 500m vertical spacing within the



**Figure 7.** Change in temperature due to the prescription of BC emissions from aircraft within the model simulations. The left column shows the simulated vertical profile of BC, while the right column shows the changes in temperature due to the prescription of BC emissions from aircrafts. The upper panels show the scenario fro March 2010, the middle panel shows the scenario for January 2011 while the bottom panel is for April 2011.

altitude band of 6-8 km. These coarsely spaced layers are then followed by 2 zones (8-9 km and 10-11 km) of 1 km vertical depth with 100 m vertical resolution accompanied by 2 coarsely spaced layers (covering 9-10 km altitude band) in-between. Such a fine vertical resolution has been kept to resolve the sharp and confined layers of BC if simulated by the model. Beyond 11 km, 10 more layers are specified to cover the altitude upto 20 km.

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## 2. P8, L21:

### -Original sentence from the manuscript:

Additionally, we carried out one more model simulation, in which we prescribed the emissions of BC from biomass burning activities using FINN-version 1.5 inventory (Wiedinmyer et al., 2011) biomass burning data. This Fire INven-

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### -Reviewer's Comment:

The acronym "FINN" should be defined in L20 not L21.

**Table 3.** Vertical levels prescribed in WRF-Chem simulations

Level Index	Altitude (km)
01-10	0-2
11-14	2-4
15-31	4-6
32-37	6-8
38-49	8-9
50-51	9-10
52-59	10-11
60-70	11-20

- **Authors' reply:**

The manuscript will be modified with the corresponding correction.

3. **Fig 3:**

5 -**Reviewer's Comment:**

The map is too simple to know the information about city name and topography. Most of readers are not familiar to this area.

- **Authors' reply:**

Accepting the suggestions from both the reviewers, this figure will be removed from the manuscript.

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