

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 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.

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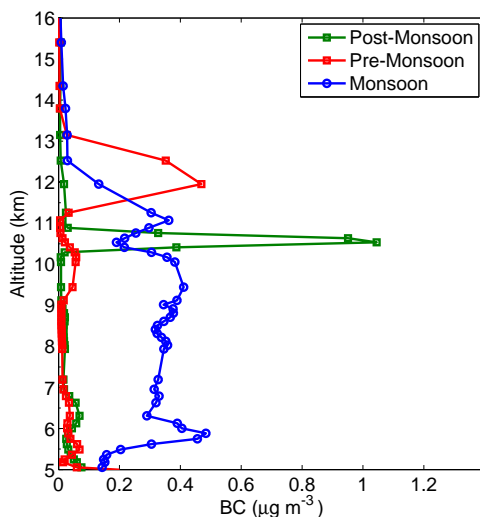


Figure 1. The model simulated vertical profile of BC mass concentration ($\mu\text{g m}^{-3}$) in the vicinity of balloon flight domain during different seasons, Pre-Monsoon (red line), Monsoon (blue line) and Post-Monsoon (green line), of the year 2010.

The detailed analysis of the seasonal simulations can be found in section 3.6 of the track-changed and the modified version of the manuscript.

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Specific comments from the reviewer:

1. P.2, L27:

- Original sentence from the manuscript:

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

- 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.

- **Modifications in the manuscript:**

The corresponding modifications can be found at P3, L5 of the track-changed version of the manuscript and P3, L3 of the revised version of the manuscript.

2. **P.5, L2**

- **Original sentence from the manuscript:**

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:**

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 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.

-**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:

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 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.

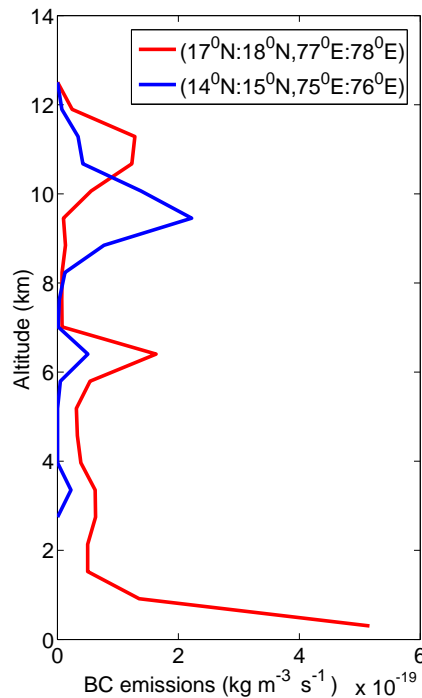


Figure 2. The vertical profile of BC emissions ($\text{kg m}^{-3} \text{s}^{-1}$) from aircrafts in the MACCity inventory over $1^{\circ} \times 1^{\circ}$ grid boxes a) centered over Hyderabad (red line) b) located 2° south-west of Hyderabad (blue line) during the month of March, 2010.

b). Modification of aircraft BC emissions:

5 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., 10 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.

15 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

5 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 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 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 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.

- **Modifications in the manuscript:**

5 This modification has been discussed in the revised version of the manuscript in section 2.3.

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

- **Original sentence from the manuscript:**

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.

10 -**Reviewer's comment:**

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

-**Authors' reply:**

15 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 (20⁰S-20⁰N) 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 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.

- **Modifications in the manuscript:**

This has been clarified at P19, L23 in the track-changed version and at P16, L8 in the revised version of the manuscript.

25 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:**

30 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:**

The source for mineral dust aerosol species in stratosphere could be largely related to a) volcanic eruptions b) me-

teoritic 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 μm) (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.

- **Modifications in the manuscript:**

This has been clarified at P20, L15 in the track-changed version and at P16, L34 in the revised version of the manuscript.

6. **P.15, L10:**

Original sentence from the manuscript:

The continued negative trend in stratospheric ozone over 40⁰S-40⁰N 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.

- **Modifications in the manuscript:**

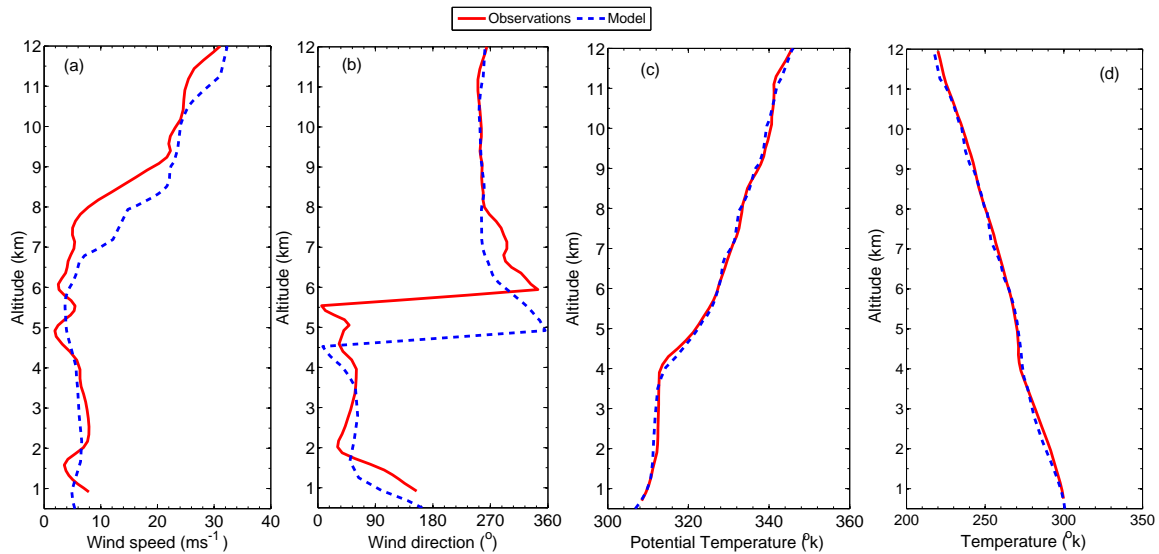
The references have been included at P21, L27 in the track-changed version and at P18, L13 in the revised version of the manuscript.

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

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

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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).

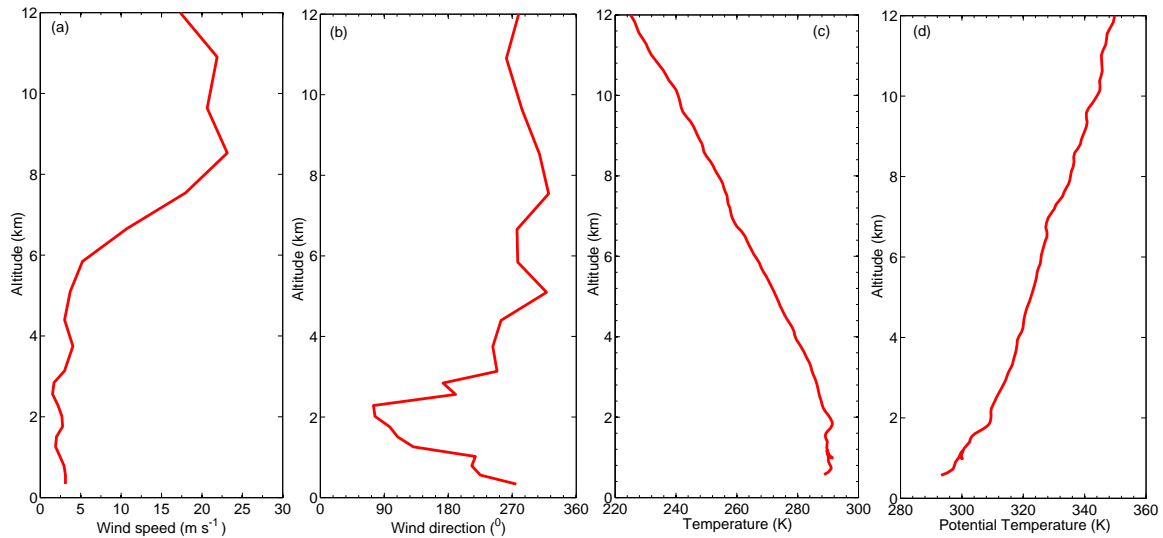


Figure 4. Vertical profiles of meteorological variables obtained from balloon measurements during 8th January 2011, (a) wind speed (b) wind direction (c) Temperature (d) Potential Temperature

5 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 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.

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8. Figs 5 and 6:

-Reviewer's comment:

15 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

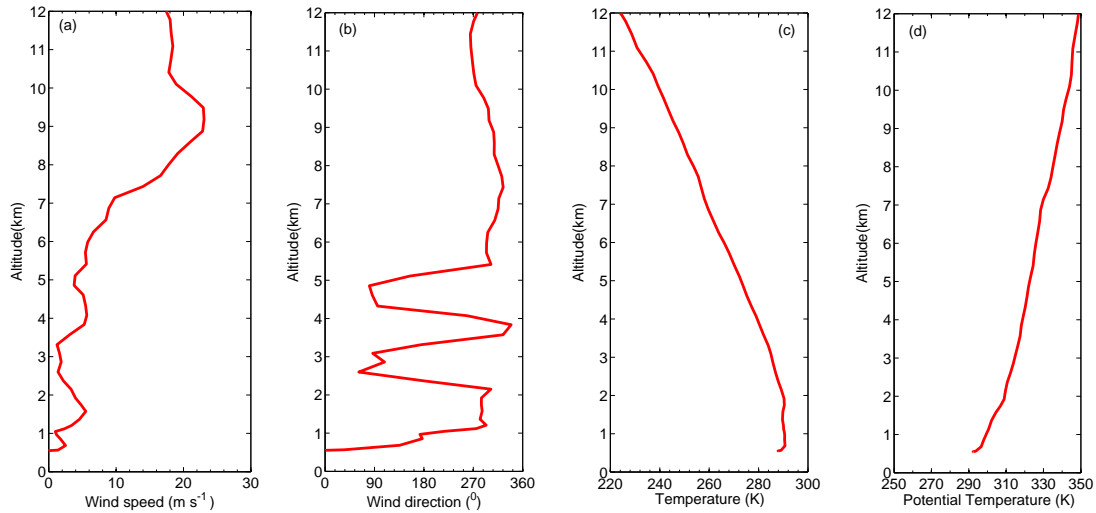


Figure 5. Vertical profiles of meteorological variables obtained from balloon measurements during 25th April 2011, (a) wind speed (b) wind direction (c) Temperature (d) Potential Temperature

attached below (fig.6). In the modified figure NoACEM simulations means the model simulations without emissions of BC from aircrafts.

9. Fig 7:

5 -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 ΔT , 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 ± 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

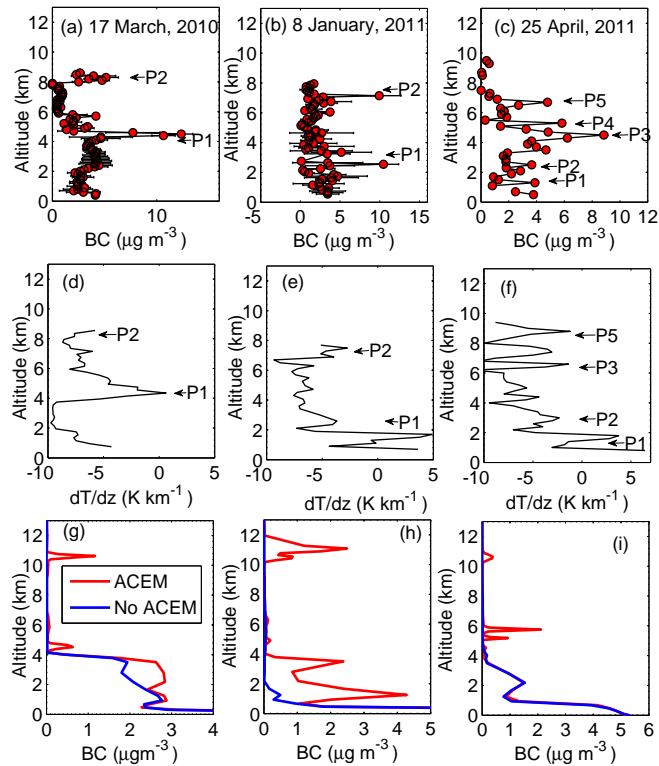


Figure 6. Observed vertical profile of BC over Hyderabad obtained from balloon measurements (a). during 17 March 2010 (b). during 8 January 2011 (c). during 25 April 2011. The high altitude BC peaks in each profile are identified by letters P1 to P5. Observed vertical profile of dT/dz ($K km^{-1}$) over 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.

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.

Technical Corrections

5 1. P4, L23:

-Original sentence from the manuscript:

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.

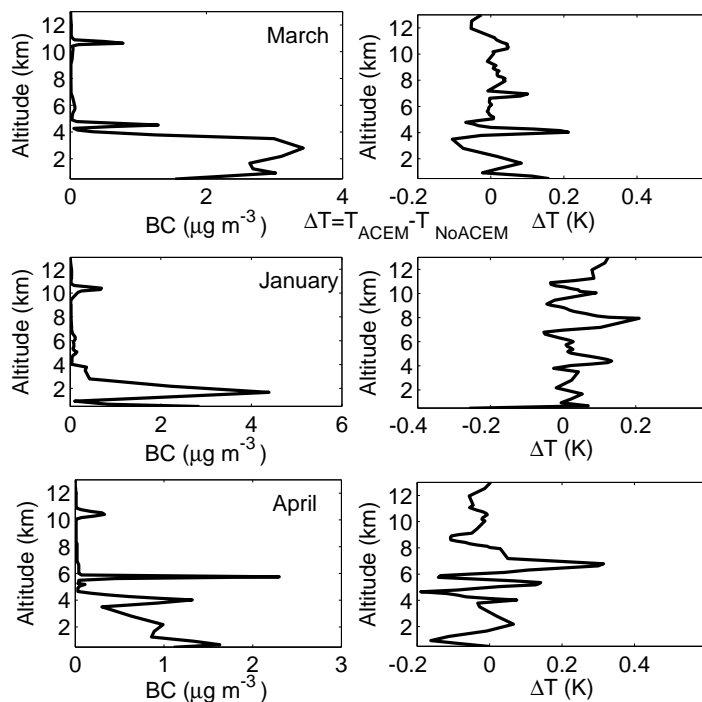


Figure 7. Change in temperature due to the prescription of BC emissions from aircraft within the model simulations. The left column shows the simulated vertical profile of BC, while the right column shows the changes in temperature due to the prescription of BC emissions from aircrafts. The upper panels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

-Authors' reply:

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 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.

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

- Modifications in the manuscript:

This has been clarified at P5, L23 in the track-changed version and at P4, L32 in the revised version of the manuscript.

2. P8, L21:

-Original sentence from the manuscript:

5 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 INventory from NCAR (FINN) provides high resolution, global emission estimates from such open burning activities.

-Reviewer's Comment:

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

10 **- Authors' reply:**

The manuscript has been modified with the corresponding correction.

- Modifications in the manuscript:

The corresponding correction can be found at P12, L32 in the track-changed version and at P11, L17 in the revised version of the manuscript.

15 **3. Fig 3:**

-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:

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

References

- Andreae, M. O., Artaxo, P., Fischer, H., Freitas, S. R., Grégoire, J.-M., Hansel, A., Hoor, P., Kormann, R., Krejci, R., Lange, L., Lelieveld, J., Lindinger, W., Longo, K., Peters, W., de Reus, M., Scheeren, B., Silva Dias, M. A. F., Ström, J., van Velthoven, P. F. J., and Williams, J.: Transport of biomass burning smoke to the upper troposphere by deep convection in the equatorial region, *Geophysical Research Letters*, 28, 951–954, doi:10.1029/2000GL012391, <http://dx.doi.org/10.1029/2000GL012391>, 2001.
- 5 Arkoudeas, P., Kalligeros, S., Zannikos, F., Anastopoulos, G., Karonis, D., Korres, D., and Lois, E.: Study of using JP-8 aviation fuel and biodiesel in {CI} engines, *Energy Conversion and Management*, 44, 1013 – 1025, doi:[https://doi.org/10.1016/S0196-8904\(02\)00112-7](https://doi.org/10.1016/S0196-8904(02)00112-7), <http://www.sciencedirect.com/science/article/pii/S0196890402001127>, 2003.
- Babu, S. S., Moorthy, K. K., Manchanda, R. K., Sinha, P. R., Satheesh, S. K., Vajja, D. P., Srinivasan, S., and Kumar, V. H. A.: Free tropospheric black carbon aerosol measurements using high altitude balloon: Do BC layers build “their own homes” up in the atmosphere?, *Geophysical Research Letters*, 38, doi:10.1029/2011GL046654, <http://dx.doi.org/10.1029/2011GL046654>, 108803, 2011.
- 10 Bhullar, B.: Hand Book On Civil Aviation Statistics 2015-16, Director General Of Civil Aviation, http://dgca.nic.in/pub/Handbook_2015-16.pdf, 2017.
- Blakey, S., Rye, L., and Wilson, C. W.: Aviation gas turbine alternative fuels: A review, *Proceedings of the Combustion Institute*, 33, 2863 – 2885, doi:<https://doi.org/10.1016/j.proci.2010.09.011>, <http://www.sciencedirect.com/science/article/pii/S1540748910003950>, 2011.
- 15 Bourassa, A. E., Degenstein, D. A., Randel, W. J., Zawodny, J. M., Kyrölä, E., McLinden, C. A., Sioris, C. E., and Roth, C. Z.: Trends in stratospheric ozone derived from merged SAGE II and Odin-OSIRIS satellite observations, *Atmospheric Chemistry and Physics*, 14, 6983–6994, doi:10.5194/acp-14-6983-2014, <http://www.atmos-chem-phys.net/14/6983/2014/>, 2014.
- Cookson, D. J. and Smith, B. E.: Calculation of jet and diesel fuel properties using carbon-13 NMR spectroscopy, *Energy & Fuels*, 4, 152–156, doi:10.1021/ef00020a004, <http://dx.doi.org/10.1021/ef00020a004>, 1990.
- 20 Corr, C. A., Ziemba, L. D., Scheuer, E., Anderson, B. E., Beyersdorf, A. J., Chen, G., Crosbie, E., Moore, R. H., Shook, M., Thornhill, K. L., Winstead, E., Lawson, R. P., Barth, M. C., Schroeder, J. R., Blake, D. R., and Dibb, J. E.: Observational evidence for the convective transport of dust over the Central United States, *Journal of Geophysical Research: Atmospheres*, 121, 1306–1319, doi:10.1002/2015JD023789, <http://dx.doi.org/10.1002/2015JD023789>, 2015JD023789, 2016.
- 25 de Laat, A. T. J., Stein Zweers, D. C., Boers, R., and Tuinder, O. N. E.: A solar escalator: Observational evidence of the self-lifting of smoke and aerosols by absorption of solar radiation in the February 2009 Australian Black Saturday plume, *Journal of Geophysical Research: Atmospheres*, 117, doi:10.1029/2011JD017016, <http://dx.doi.org/10.1029/2011JD017016>, d04204, 2012.
- Eyers, C., Norman, P., Middel, J., Plohr, M., Michot, S., Atkinson, K., and Christou, R.: AERO2K global aviation emissions inventories for 2002 and 2025, *QinetiQ*, 2004.
- 30 Fadnavis, S., Semeniuk, K., Pozzoli, L., Schultz, M. G., Ghude, S. D., Das, S., and Kakatkar, R.: Transport of aerosols into the UTLS and their impact on the Asian monsoon region as seen in a global model simulation, *Atmospheric Chemistry and Physics*, 13, 8771–8786, doi:10.5194/acp-13-8771-2013, <http://www.atmos-chem-phys.net/13/8771/2013/>, 2013.
- Gorkavyi, N., Rault, D. F., Newman, P. A., da Silva, A. M., and Dudorov, A. E.: New stratospheric dust belt due to the Chelyabinsk bolide, *Geophysical Research Letters*, 40, 4728–4733, doi:10.1002/grl.50788, <http://dx.doi.org/10.1002/grl.50788>, 2013.
- 35 Herndon, S. C., Jayne, J. T., Lobo, P., Onasch, T. B., Fleming, G., Hagen, D. E., Whitefield, P. D., and Miake-Lye, R. C.: Commercial Aircraft Engine Emissions Characterization of in-Use Aircraft at Hartsfield-Jackson Atlanta International Airport, *Environmental Science & Technology*, 42, 1877–1883, doi:10.1021/es072029+, <http://dx.doi.org/10.1021/es072029+>, pMID: 18409607, 2008.

- Hess, M., Koepke, P., and Schult, I.: Optical Properties of Aerosols and Clouds: The Software Package OPAC, *Bulletin of the American Meteorological Society*, 79, 831–844, doi:10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2, [http://dx.doi.org/10.1175/1520-0477\(1998\)079<0831:OPOAAC>2.0.CO;2](http://dx.doi.org/10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2), 1998.
- 5 Klekociuk, A. R., Brown, P. G., Pack, D. W., ReVelle, D. O., Edwards, W. N., Spalding, R. E., Tagliaferri, E., Yoo, B. B., and Zagari, J.: Meteoritic dust from the atmospheric disintegration of a large meteoroid, *Nature*, 436, 1132–1135, doi:<http://dx.doi.org/10.1038/nature03881>, http://www.nature.com/nature/journal/v436/n7054/supinfo/nature03881_S1.html, 10.1038/nature03881, 2005.
- Kremser, S., Thomason, L. W., von Hobe, M., Hermann, M., Deshler, T., Timmreck, C., Toohey, M., Stenke, A., Schwarz, J. P., Weigel, R., Fueglistaler, S., Prata, F. J., Vernier, J.-P., Schlager, H., Barnes, J. E., Antuña-Marrero, J.-C., Fairlie, D., Palm, M., Mahieu, E., Notholt, J., Rex, M., Bingen, C., Vanhellemont, F., Bourassa, A., Plane, J. M. C., Klocke, D., Carn, S. A., Clarisse, L., Trickl, T., Neely, R., James, A. D., Rieger, L., Wilson, J. C., and Meland, B.: Stratospheric aerosol—Observations, processes, and impact on climate, *Reviews of Geophysics*, 54, 278–335, doi:10.1002/2015RG000511, <http://dx.doi.org/10.1002/2015RG000511>, 2015RG000511, 2016.
- 10 Kumar, R., Barth, M. C., Pfister, G. G., Nair, V. S., Ghude, S. D., and Ojha, N.: What controls the seasonal cycle of black carbon aerosols in India?, *Journal of Geophysical Research: Atmospheres*, 120, 7788–7812, doi:10.1002/2015JD023298, <http://dx.doi.org/10.1002/2015JD023298>, 2015JD023298, 2015.
- 15 Kwan, I. and Rutherford, D.: Transatlantic airline fuel efficiency ranking, 2014, The International Council on Clean Transportation, <http://www.theicct.org/transatlantic-airline-efficiency-2014>, 2015.
- Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, *Atmospheric Chemistry and Physics*, 10, 7017–7039, doi:10.5194/acp-10-7017-2010, <http://www.atmos-chem-phys.net/10/7017/2010/>, 2010.
- 20 Mackinnon, I. D. R., McKay, D. S., Nace, G., and Isaacs, A. M.: Classification of the Johnson Space Center stratospheric dust collection, *Journal of Geophysical Research: Solid Earth*, 87, A413–A421, doi:10.1029/JB087iS01p0A413, <http://dx.doi.org/10.1029/JB087iS01p0A413>, 1982.
- 25 Meenu, S., Rajeev, K., Parameswaran, K., and Nair, A. K. M.: Regional distribution of deep clouds and cloud top altitudes over the Indian subcontinent and the surrounding oceans, *Journal of Geophysical Research: Atmospheres*, 115, doi:10.1029/2009JD011802, <http://dx.doi.org/10.1029/2009JD011802>, d05205, 2010.
- Onasch, T. B., Jayne, J. T., Herndon, S., Worsnop, D. R., Miake-Lye, R. C., Mortimer, I. P., and Anderson, B. E.: Chemical Properties of Aircraft Engine Particulate Exhaust Emissions, *Journal of Propulsion and Power*, 25, 1121–1137, doi:10.2514/1.36371, <http://dx.doi.org/10.2514/1.36371>, 2009.
- 30 Outcalt, S., Laesecke, A., and Freund, M. B.: Density and Speed of Sound Measurements of Jet A and S-8 Aviation Turbine Fuels, *Energy & Fuels*, 23, 1626–1633, doi:10.1021/ef800888q, <http://dx.doi.org/10.1021/ef800888q>, 2009.
- Paraschis, Y. and Gittens, A.: Annual Report 2013, The Voice of the World’s Airports,, Airports Council International, <http://www.aci.aero/Publications/Full-Publications-Listing>, 2014.
- 35 Piccolo, F. and Gittens, A.: Annual Report 2014, The Voice of the World’s Airports,, Airports Council International, <http://www.aci.aero/Publications/Full-Publications-Listing>, 2015.
- Piccolo, F. and Gittens, A.: Annual Report 2015, The Voice of the World’s Airports,, Airports Council International, <http://www.aci.aero/Publications/Full-Publications-Listing>, 2016.

- Randel, W. J., Park, M., Emmons, L., Kinnison, D., Bernath, P., Walker, K. A., Boone, C., and Pumphrey, H.: Asian Monsoon Transport of Pollution to the Stratosphere, *Science*, 328, 611–613, doi:10.1126/science.1182274, <http://science.sciencemag.org/content/328/5978/611>, 2010.
- Satheesh, S. K., Moorthy, K. K., Babu, S. S., Vinoj, V., and Dutt, C. B. S.: Climate implications of large warming by elevated aerosol over India, *Geophysical Research Letters*, 35, n/a–n/a, doi:10.1029/2008GL034944, <http://dx.doi.org/10.1029/2008GL034944>, 119809, 2008.
- Simos, D.: PIANO: PIANO User’s Guide Version 4.0, Lissys Limited, UK www.piano.aero (last access: 16 February 2010), 2004.
- Stettler, M., Eastham, S., and Barrett, S.: Air quality and public health impacts of {UK} airports. Part I: Emissions, *Atmospheric Environment*, 45, 5415 – 5424, doi:<http://dx.doi.org/10.1016/j.atmosenv.2011.07.012>, <http://www.sciencedirect.com/science/article/pii/S135223101100728X>, 2011.
- 10 Stettler, M. E. J., Boies, A. M., Petzold, A., and Barrett, S. R. H.: Global Civil Aviation Black Carbon Emissions, *Environmental Science & Technology*, 47, 10 397–10 404, doi:10.1021/es401356v, <http://dx.doi.org/10.1021/es401356v>, PMID: 23844612, 2013.
- Timko, M. T., Onasch, T. B., Northway, M. J., Jayne, J. T., Canagaratna, M. R., Herndon, S. C., Wood, E. C., Miake-Lye, R. C., and Knighton, W. B.: Gas Turbine Engine Emissions—Part II: Chemical Properties of Particulate Matter, *Journal of Engineering for Gas Turbines and Power*, 132, 061 505–061 505–15, doi:10.1115/1.4000132, <http://dx.doi.org/10.1115/1.4000132>, 2010.
- 15 Turco, R. P., Toon, O. B., Hamill, P., and Whitten, R. C.: Effects of meteoric debris on stratospheric aerosols and gases, *Journal of Geophysical Research: Oceans*, 86, 1113–1128, doi:10.1029/JC086iC02p01113, <http://dx.doi.org/10.1029/JC086iC02p01113>, 1981.
- Vernier, J.-P., Thomason, L. W., and Kar, J.: CALIPSO detection of an Asian tropopause aerosol layer, *Geophysical Research Letters*, 38, doi:10.1029/2010GL046614, <http://dx.doi.org/10.1029/2010GL046614>, 107804, 2011.
- Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning, *Geoscientific Model Development*, 4, 625–641, doi:10.5194/gmd-4-625-2011, <http://www.geosci-model-dev.net/4/625/2011/>, 2011.
- 20

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.

5

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.

10

-Authors' reply:

We accept the aforementioned suggestion from the reviewer. The manuscript has been modified and the corresponding changes have been made.

15

- Modifications in the manuscript:

The corresponding changes in the abstract can be found at P1, L7 in the track-changed version and in the revised version of the manuscript. The conclusion section has also been modified with aforementioned correction. The corresponding changes in the conclusion section can be found at P22, L27 in the track-changed version and at P19, L14 in the revised version of the manuscript.

20

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.

5

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.

10

-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 are discussed in greater details in section 3.6 of the track-changed and the revised 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.

15

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- Modifications in the manuscript:

The ‘Methods and ‘Results’ sections can be found in section 2 and section 3 of the track-changed and the revised version of the manuscript. Relevant discussion regarding uncertainties related to the study can be found at P21, L30 in the track-changed version and at P18, L17 in the revised version of the manuscript.

25

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.

30

-Authors’ reply:

The corresponding modifications are made in the abstract.

- Modifications in the manuscript:

The corresponding modifications can be found at P1, L6 in the track-changed version and in the revised version of the manuscript.

2. Fig.1

Reviewer's comment:

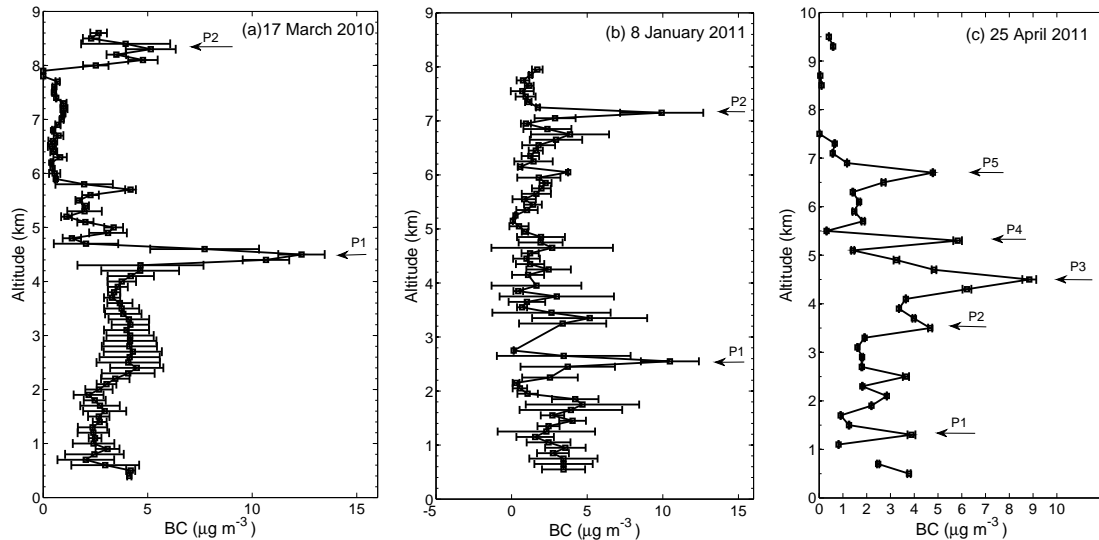


Figure 1. Observed vertical profile of BC over Hyderabad obtained from balloon measurements (a). during 17 March 2010 (b). during 8 January 2011 (c). during 25 April 2011. The high altitude BC peaks in each profile are identified by letters P1 to P5.

5 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. 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.

10

3. Chapter 2:

15

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.

5 **-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:

10 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 $16 \text{ m}^2 \text{ g}^{-1}$ (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
15 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).
20

-Reviewer’s comment:

- What are the uncertainties on separating out other absorbing components, like dust?

-Authors’ reply:

25 - 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).

30 **-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).

- Modifications in the manuscript:

35 This discussion has been included in the revised version of the manuscript. It can be found at P4, L3 in the track-changed

and at P3, L32 in the revised version of the manuscript. The figure 3 has been removed from the manuscript.

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.

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:

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.

- Modifications in the manuscript:

This has been included at P10, L23 in the track-changed and at P9, L33 in the revised version of the manuscript.

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

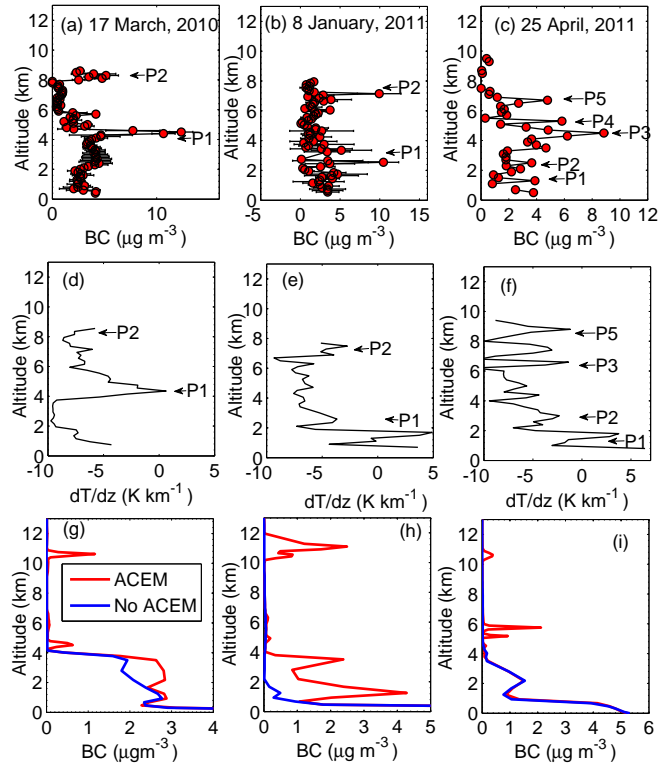


Figure 2. Observed vertical profile of BC over Hyderabad obtained from balloon measurements (a). during 17 March 2010 (b). during 8 January 2011 (c). during 25 April 2011. The high altitude BC peaks in each profile are identified by letters P1 to P5. Observed vertical profile of dT/dz ($K km^{-1}$) over 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.

this? Again, it is difficult to compare the model and the observations since the plots are so different (axis).

-Authors' reply:

5 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

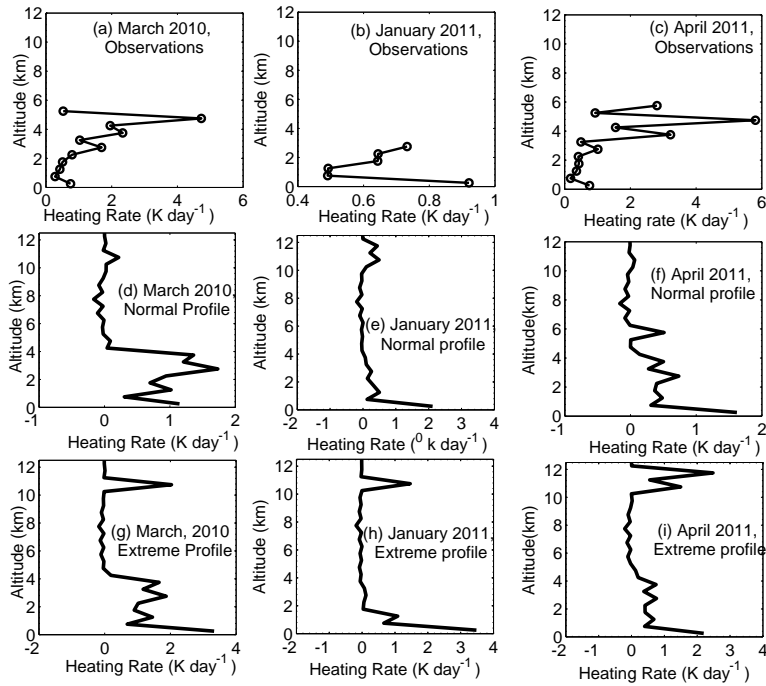


Figure 3. Vertical profiles of atmospheric heating rates for the observed BC profiles during (a).March 2010, (b). January 2011 and (c). April 2011. Vertical profiles of atmospheric heating rates corresponding to the observed BC profiles during (a).March 2010, (b). January 2011 and (c). April 2011. Vertical 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.

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~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.

- **Modifications in the manuscript:**

The corresponding changes can be found at P10, L33 in the track-changed version and at P10, L8 in the revised version of the manuscript.

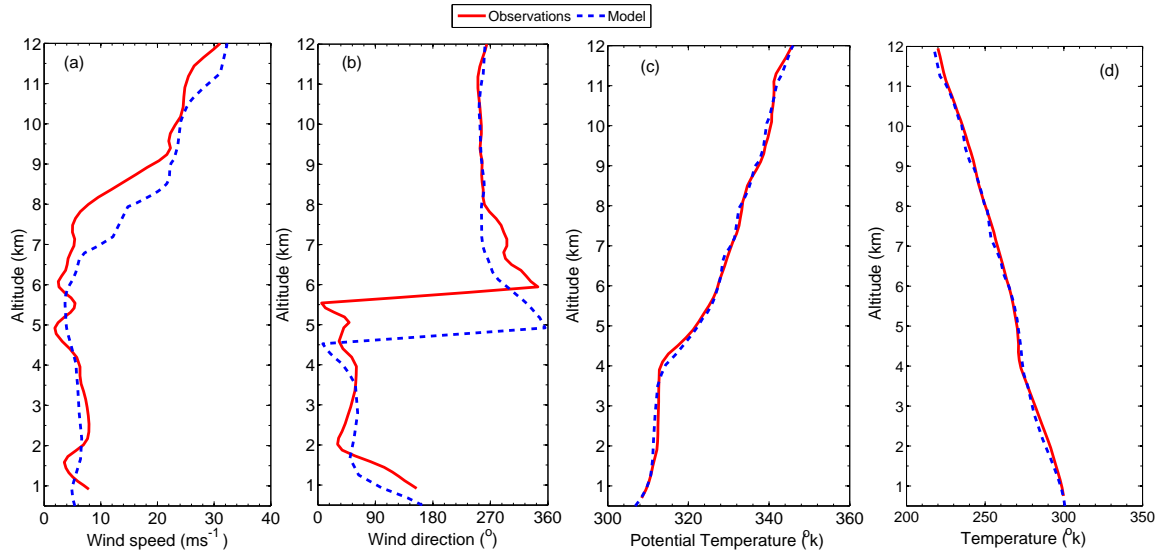


Figure 4. Comparison between the model simulated meteorological variables in NoACEM/Ctrl (control) run and the corresponding observations over the balloon flight region on 17, March 2010 for (a) wind speed (b) wind direction (c) Potential temperature (d) Temperature

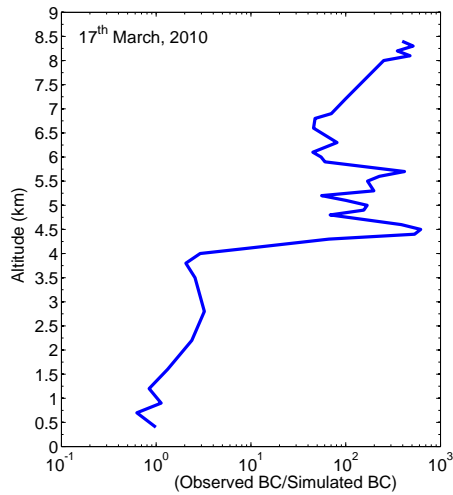


Figure 5. Ratio of the observed BC to the model simulated BC (in the control run)

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 trans-

port 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:

5 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
10 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 km, BC showed one more
15 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 has been provided to Babu et al. (2011) in this regards.

20 **- Modifications in the manuscript:**

The corresponding modification can be found at P11, L17 in the track-changed version and at P10, L23 in the revised version of the manuscript.

8. **Reviewer's comment:**

Your manuscript is written more like a story, and how you came about with the different hypothesis, but I suggest
25 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:

30 We appreciate the aforementioned modifications suggested the reviewer and the corresponding changes in the layout of the manuscript will be made.

- Modifications in the manuscript:

The corresponding modifications regarding layout of the manuscript have been made. The modified 'Methods' section

can be found in section 2 of the track-changed and the revised version of the manuscript, while the modified ‘Results’ section can be found in section 3 of the track-changed and the revised version of the manuscript.

9. **P7 L32:**

Original sentence in the manuscript:

5 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.

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.

10 **-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.

10. **P9 L29:**

Original sentence from the manuscript:

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

Reviewer’s comment:

How is ‘soot’ defined in this context?

-Authors’ reply:

25 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 g cm^{-3} . The optical properties are calculated assuming the size distribution with many very small particles (which would have the density 2.3 g cm^{-3}). More details about the soot component defined in OPAC can be found in Hess et al. (1998).

11. **P8, L7**

30 **-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.

- Modifications in the manuscript:

The corresponding modifications can be found at P12, L18 in the track-changed version and at P11, L4 in the revised version of the manuscript.

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 MACCCity 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 $0.1\mu\text{g m}^{-3}$. This analysis suggests that the two profiles significantly match each other.

Modified sentence:

Beyond 4 km, the profiles are largely similar

5 - **Modifications in the manuscript:**

The corresponding modifications can be found at P13, L11 in the track-changed version and at P11, L30 in the revised version of the manuscript.

14. **P9, L23:**

Original sentence from the manuscript:

10 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?

15 **-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

- Modifications in the manuscript:

25 The corresponding modifications can be found at P16, L24 in the track-changed version and at P14, L1 in the revised version of the manuscript.

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.

30 **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.

- Modifications in the manuscript:

The corresponding modifications can be found at P16, L30 in the track-changed version and at P14, L6 in the revised version of the manuscript.

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.

- Modifications in the manuscript:

The corresponding modifications can be found at P17, L24 in the track-changed version and at P14, L25 in the revised version of the manuscript.

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.

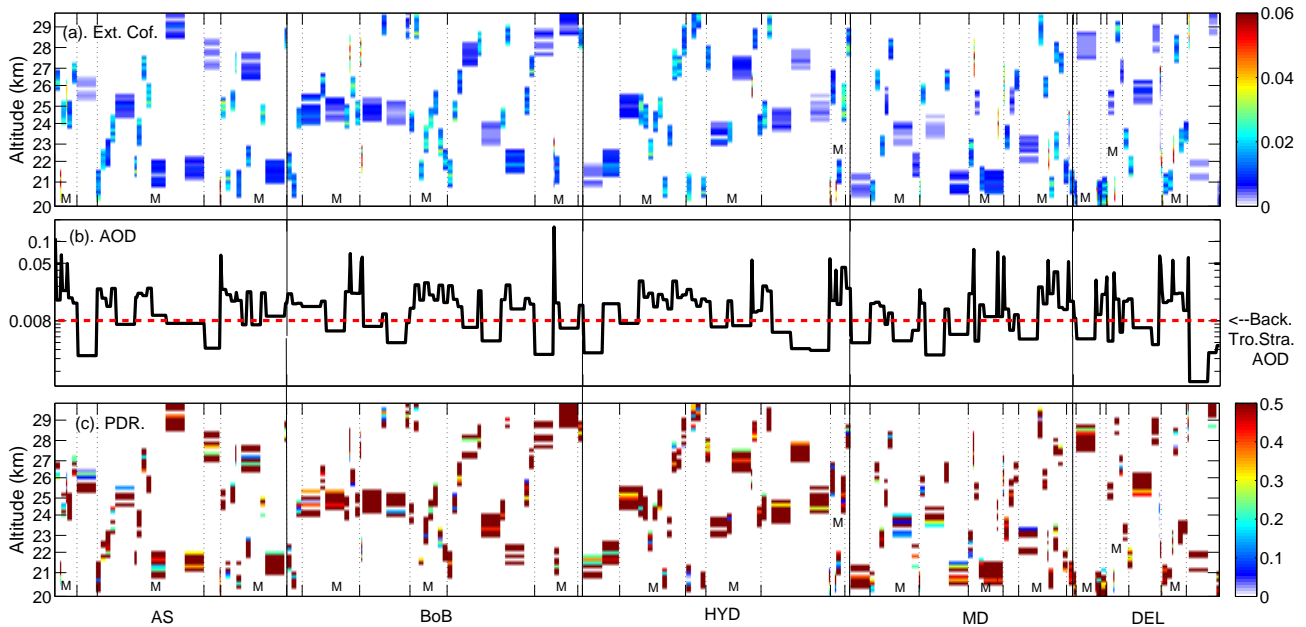


Figure 6. The time-series of the vertical profile of extinction coefficient at stratospheric altitude, stratospheric AOD and the vertical profile of particle depolarization ratio at stratospheric altitude are plotted for each of the 5 regional boxes shown in fig.12 (previous figure from the manuscript). 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.

- **Modifications in the manuscript:**

The corresponding modifications can be found in P35 of the track-changed version and P32 of the revised version of the manuscript.

Technical corrections:

5

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:**

10

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.

- **Modifications in the manuscript:**

The corresponding modifications can be found at P2, L2 in the track-changed version of the manuscript.

15

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:**

20

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 has been cited in the modified manuscript.

25

- **Modifications in the manuscript:**

The corresponding modifications can be found at P2, L14 in the track-changed version and at P2, L12 in the revised version of the manuscript.

3. **P6, L3:**

Original sentence from the manuscript:

30

up to 1st 2 km of the lower troposphere

-**Reviewer's comment:**

1st -first

-**Authors' reply:**

The corresponding changes have been made in the manuscript.

- **Modifications in the manuscript:**

The corresponding modifications can be found at P10, L10 in the track-changed version and at P9, L20 in the revised version of the manuscript.

5 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 θ in the vicinity of the primary BC maxima in observations (around 4-4.5 km, fig.2a) etc.

-**Reviewer's comment:**

10 either remove 'etc..' or replace by explaining the other discrepancies.

-**Authors' reply:**

The word 'etc' has been removed from the text.

- **Modifications in the manuscript**

The corresponding modifications can be found at P10, L15 in the track-changed version of the manuscript.

References

- Arnott, W. P., Hamasha, K., Moosmüller, H., Sheridan, P. J., and Ogren, J. A.: Towards Aerosol Light-Absorption Measurements with a 7-Wavelength Aethalometer: Evaluation with a Photoacoustic Instrument and 3-Wavelength Nephelometer, *Aerosol Science and Technology*, 39, 17–29, doi:10.1080/027868290901972, <http://dx.doi.org/10.1080/027868290901972>, 2005.
- 5 Babu, S. S., Moorthy, K. K., Manchanda, R. K., Sinha, P. R., Satheesh, S. K., Vajja, D. P., Srinivasan, S., and Kumar, V. H. A.: Free tropospheric black carbon aerosol measurements using high altitude balloon: Do BC layers build “their own homes” up in the atmosphere?, *Geophysical Research Letters*, 38, doi:10.1029/2011GL046654, <http://dx.doi.org/10.1029/2011GL046654>, 108803, 2011.
- Corrigan, C. E., Ramanathan, V., and Schauer, J. J.: Impact of monsoon transitions on the physical and optical properties of aerosols, *Journal of Geophysical Research: Atmospheres*, 111, n/a–n/a, doi:10.1029/2005JD006370, <http://dx.doi.org/10.1029/2005JD006370>, d18208, 10 2006.
- Fan, J., Zhang, R., Tao, W.-K., and Mohr, K. I.: Effects of aerosol optical properties on deep convective clouds and radiative forcing, *Journal of Geophysical Research: Atmospheres*, 113, n/a–n/a, doi:10.1029/2007JD009257, <http://dx.doi.org/10.1029/2007JD009257>, d08209, 2008.
- Hansen, A.: Magee Scientific Aethalometer User’s Guide, Magee Sci., Berkeley, California, 56pp, 1996.
- Hansen, A.: The Aethalometer, Magee Sci., Berkeley, California, 2005.
- 15 Hess, M., Koepke, P., and Schult, I.: Optical Properties of Aerosols and Clouds: The Software Package OPAC, *Bulletin of the American Meteorological Society*, 79, 831–844, doi:10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2, [http://dx.doi.org/10.1175/1520-0477\(1998\)079<0831:OPOAAC>2.0.CO;2](http://dx.doi.org/10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2), 1998.
- Kremser, S., Thomason, L. W., von Hobe, M., Hermann, M., Deshler, T., Timmreck, C., Toohey, M., Stenke, A., Schwarz, J. P., Weigel, R., Fueglistaler, S., Prata, F. J., Vernier, J.-P., Schlager, H., Barnes, J. E., Antuña-Marrero, J.-C., Fairlie, D., Palm, M., Mahieu, E., Notholt, 20 J., Rex, M., Bingen, C., Vanhellefont, F., Bourassa, A., Plane, J. M. C., Klocke, D., Carn, S. A., Clarisse, L., Trickl, T., Neely, R., James, A. D., Rieger, L., Wilson, J. C., and Meland, B.: Stratospheric aerosol—Observations, processes, and impact on climate, *Reviews of Geophysics*, 54, 278–335, doi:10.1002/2015RG000511, <http://dx.doi.org/10.1002/2015RG000511>, 2015RG000511, 2016.
- Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S.-M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, 25 C., Wiedensohler, A., and Zhang, X.-Y.: Recommendations for reporting “black carbon” measurements, *Atmospheric Chemistry and Physics*, 13, 8365–8379, doi:10.5194/acp-13-8365-2013, <http://www.atmos-chem-phys.net/13/8365/2013/>, 2013.
- Sheridan, P. J., Arnott, W. P., Ogren, J. A., Andrews, E., Atkinson, D. B., Covert, D. S., Moosmüller, H., Petzold, A., Schmid, B., Strawa, A. W., Varma, R., and Virkkula, A.: The Reno Aerosol Optics Study: An Evaluation of Aerosol Absorption Measurement Methods, *Aerosol Science and Technology*, 39, 1–16, doi:10.1080/027868290901891, <http://dx.doi.org/10.1080/027868290901891>, 2005.
- Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B., and Baltensperger, U.: Absorption of light by soot particles: determination of the absorption coefficient by means of aethalometers, *Journal of Aerosol Science*, 34, 1445 – 1463, doi:[https://doi.org/10.1016/S0021-8502\(03\)00359-8](https://doi.org/10.1016/S0021-8502(03)00359-8), <http://www.sciencedirect.com/science/article/pii/S0021850203003598>, intercomparison of Soot Measurement Techniques, 2003. 30

Possible climatic implications of high altitude emissions of black carbon

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Abstract. On account of its strong absorption of solar and terrestrial radiations, Black Carbon (BC) aerosol is known to impact large scale systems such as the Asian monsoon, Himalayan glaciers etc., besides affecting the thermal structure of lower atmosphere. While most studies focus on the near-surface abundance and impacts of BC, our study, using online regional chemistry transport model (WRF-Chem) simulations, examines the implications of sharp and confined layers of high BC concentration (called ‘elevated BC layers’) at altitudes ~~of about 4.5 km and 8~~ more than 4 km over the Indian region, as revealed in the recent in-situ measurements using high-altitude balloons~~-, carried out on 17th March 2010, 8th January 2011 and 25th April 2011.~~ Our study demonstrates ~~that emissions from that, the~~ high-flying aircrafts (with realistic emissions) are the most likely cause of these elevated BC layers. Furthermore, we show that such aircraft-emitted BC can get transported to even upper tropospheric/ lower stratospheric heights (~17 km) aided by the strong monsoonal convection occurring over the region, which are known to overshoot the tropical tropopause leading to injection of tropospheric air mass (along with its constituent aerosols) into the stratosphere. We show observational evidence for such an intrusion of tropospheric BC into the stratosphere over Indian region, using extinction coefficient and particle depolarization ratio data from CALIOP LIDAR on-board the CALIPSO satellite. We ~~hypothesise~~ hypothesize that such intrusions of BC to lower stratosphere and its consequent longer residence time in the stratosphere would have significant implications for stratospheric ozone, considering the already reported ozone depleting potential of BC.

1 Introduction

The important role played by atmospheric aerosol particles in global and regional climate forcing is getting increasingly accepted. The global mean net radiative forcing (including the rapid adjustments) by aerosol is estimated to be -0.9 [-1.9 to -0.1] W m⁻² (Boucher et al., 2013), after accounting for the negative forcing by most of the aerosol species (sulphate, nitrate, sea-salts etc.) and the strong positive forcing by Black Carbon (BC), which absorbs solar radiation over a wide spectral band, and to a lesser extent by dust and organics. BC, a fine particulate, is formed as a result of incomplete combustion of fossil fuels, biofuels and biomass (Kuhlbusch et al., 1996; Ramanathan and Carmichael, 2008; Bond et al., 2013). Being a strong absorber of solar and terrestrial radiation, BC heats up the atmosphere (Petzold and Schönlinner, 2004; Ramanathan et al., 2007). The

global mean radiative forcing by BC is estimated as +0.4 (+0.05 to +0.8) W m^{-2} , which is around $(1/4)^{th}$ of the global mean CO_2 radiative forcing (Boucher et al., 2013). Several earlier studies have shown that ~~such an~~ atmospheric heating by a layer of aerosol with high BC abundance, is capable of perturbing large scale atmospheric phenomenon such as the Asian monsoon system and can also affect the hydrological cycle (Chakraborty et al., 2004; Ramanathan et al., 2005; Lau et al., 2006). Also, on account of its absorbing nature, BC, if deposited on snow, would exert ‘snow-albedo forcing’ resulting from the darkening of snow and could accelerate the melting of glaciers (Lau et al., 2010; Qian et al., 2011; Yasunari et al., 2010; Nair et al., 2013; Gautam et al., 2013). Furthermore, when BC particles are located above highly reflecting surfaces (such as snow/clouds), their absorption efficiency gets amplified (Haywood and Shine, 1995; Satheesh and Moorthy, 2005; Chand et al., 2009; Nair et al., 2013). This heating can give rise to local elevated dry convection, which can further lift the BC particles to higher levels of the atmosphere (de Laat et al., 2012). The atmospheric heating by BC would be more severe at higher altitudes due to reduced air density at these altitudes, and this could amplify the heating effect. The high-altitude heating due to BC can also lead to reduced cloud cover at these elevated altitudes (Ackerman et al., 2000). Additionally, the atmospheric heating at higher ~~altitude~~ altitudes can give rise to local stable scenario below, which can affect convection and consequently impact precipitation (Fan et al., 2008). Moreover, such lifted BC and the other pollutants, upon interaction with the strong monsoonal convection, can get lifted to even higher heights (Andreae et al., 2001; Fromm et al., 2005; Randel et al., 2010; Vernier et al., 2011), and hold a potential to get lifted even beyond tropopause and intrude into the stratosphere under favorable upper tropospheric conditions. In this regard, the Tropical Tropopause Layer assumes importance. TTL is the region between the top of the major convective towers and the cold point. While forming an interface between the two dynamically different regimes, TTL acts as an entry-point to the tropical stratosphere (Fueglistaler et al., 2009). Sunilkumar et al. (2013) using radiosonde measurements, have reported thinning of the Tropical Tropopause Layer (TTL) and reduction in the Temperature Lapse Rate (TLR) within TTL, which favor such cross-tropopause transport, over the Indian region during the monsoon season. A recent study by Das et al. (2016) reports a cross tropopause transport of air-mass over Indian region during the cyclone ‘Nilam’. Once the particles enter the stratosphere, they reside for a longer period of time due to the inherent stability of stratosphere and the absence of strong removal mechanisms. Moreover, they can take part in heterogeneous chemical reactions taking place in stratosphere.

The aforementioned effects together highlight the significance of knowing the vertical distribution of BC especially over regions of strong convection (such as the Indian subcontinent especially during the pre-monsoon and the monsoon months). With this backdrop, a few studies have examined the vertical distribution of aerosols (extinction coefficient values and/or BC mass concentration values) over the Indian region using multiple observational platforms such as a ground based LIDAR (Satheesh et al., 2006), airborne and space based LIDAR (Satheesh et al., 2008; Prijith et al., 2016), instrumented aircrafts (Moorthy et al., 2004; Babu et al., 2008, 2010; Nair et al., 2016), tethered and high-altitude balloons (Babu et al., 2011). Carrying out such observations for the first time over India, Moorthy et al. (2004) have noticed an exponential decrease of BC concentration within the boundary layer and a steady value above it, over an urban station (Hyderabad) in south-central India during the month of February, 2004. Examining the vertical profile of aerosols over Bangalore (an urban station in southern part of India) using a ground based micro pulse LIDAR, Satheesh et al. (2006) have reported morning-time (night-time) vertical

lifting (confinement) of aerosols during summer days. Carrying out airborne measurements of BC and absorption coefficient over several stations across India, Babu et al. (2008, 2010); Nair et al. (2016) have reported a near-steady vertical distribution up to an altitude of 3 km above ground, with isolated peaks of higher BC concentration/ absorption coefficient during pre-monsoon period, above most of the landmass. Synthesizing multi-platform measurements Satheesh et al. (2008) have revealed the existence of elevated aerosol layers in the middle troposphere (4 to 5 km). They have shown that these elevated aerosol layers caused atmospheric heating, which exhibited a meridional gradient that varied in vertical extent and amplitude from the northern Indian Ocean to Central India, during the pre-monsoon season. Using hydrogen inflated zero pressure balloon measurements for the first time in India, Babu et al. (2011) have revealed strikingly sharp and confined layers of BC at altitude of 4.5 km and 8 km over Hyderabad during the pre-monsoon season of 2010. They also have reported a large decrease in the environmental lapse rate associated with these peaks, using concurrent measurements aboard the same balloon. ~~Analysing~~ Analyzing multi-year satellite (CALIPSO) data, Prijith et al. (2016), not only confirmed the existence of high altitude aerosol layers and their meridional gradient in the vertical extent during pre-monsoon periods over Indian region, but also have shown that mineral dust aerosol forms the dominant component of this layer. Kompalli et al. (2016) have reported spring-time enhancement in columnar Aerosol Optical Depth (AOD) and BC over ~~Hanle~~ Hanle - a western trans-Himalayan high-altitude observational location (4.5 km amsl). They attributed this enhancement to summer-time lifting of aerosols over the region west to south-west of Hanle and subsequent long-range transport. All the above studies clearly bring out the existence of high altitude aerosol layers (with significant amount of BC) over the south Asian region during the pre-monsoon periods. Such strong absorbing aerosol layers can perturb the south Asian regional monsoonal system and can also affect the glacial coverage of the south Asian region. Thus, it becomes imperative to understand the dynamics of this regional high altitude aerosol layers and investigate the exact causes behind their occurrence.

In this study, we revisit the three high-altitude balloon measurements of BC made during the years 2010, and 2011, (in different seasons) over Hyderabad (the only location in India from where such balloon ascents could be made) and investigate the potential causes behind the existence of such confined BC layers, using a regional chemistry transport model. The balloon experiments ~~and profiles are described in section 3.1~~, the model and simulation details are described section ~~2.1~~ 2 and in section 3, we discuss the results. The possible climatic implications of such high-altitude BC layers are discussed in section 4. The conclusions are presented in section 5.

~~2 High altitude measurements of BC~~ Methods

2.1 High altitude measurements of BC

The vertical profiles of BC used in this study have been obtained using an Aethalometer (Model AE-42, of Magee Scientific, USA), aboard high-altitude zero pressure balloons, launched from the National Balloon Facility at Hyderabad, (17.48°N, 78.47°E, 557 m amsl), during three ascents made on 17 March 2010, 08 January 2011 and 25 April 2011. The first and third flights corresponded to pre-monsoon conditions, while the second flight occurred in winter. The details of experimental set-up, calibration, data collection and analysis are available in Babu et al. (2011) and as such, only a brief account is given

here. The 109755 m³ zero-pressure balloon was made of 25 mm thick, linear low density polyethylene film, and was capable of carrying ~350 kg of payload up to a ceiling altitude of ~35 km. ~~Besides the aethalometer, each ascent also carried other payloads such as GPS receiver, meteorological sensors for temperature and RH, telemetry and telecommand systems.~~ The mass concentration of 'Equivalent Black Carbon' (EBC) (Petzold et al., 2013) was measured using an Aethalometer, which 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 m² 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 ~~had been 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 aethalometer 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). In addition to BC, dust also absorbs the short wave radiation, but 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.

Besides the aethalometer, each ascent also carried other payloads such as GPS receiver, meteorological sensors for temperature and RH, telemetry and telecommand systems. The entire payload was tested using a thermo-vacuum facility to ensure consistent operations up to ~9 km altitude (250 hPa pressure level and ~ -40⁰C). Beyond this altitude, as the ambient pressure drops too low to provide sufficient flow to the aethalometer, the telecommand system was used to switch off the instrument for the higher altitudes, and switch it on again at this altitude during the descent phase. A gondola, carrying the payloads, had been attached to the balloon using a parachute, which was deployed in the descent phase and enabled safe recovery of the payloads for reuse. Ballast cans attached to the gondola ensured a slow and steady ascent of ~2.6 m s⁻¹, while the descent speed was controlled by the parachute, as the ballast cans were detached after the balloon rose to the free tropospheric altitude. The scientific data along with the housekeeping data were collected during the ascent and descent phases and were continuously telemetered to the ground, besides storing on-board. The total flight duration was about 3 to 4 hours and the spatial ground spread of the flight path has been within 50 km radius of the launch site. The data have been analysed (following details in Babu et al. (2011)) and the profiles obtained were smoothed using a running mean filter. The same protocols have been followed for all the flights. ~~The vertical profiles of BC, derived from these flights are shown in fig.??, in which each profile is the average of the ascent and descent profiles for that flight. All the profiles revealed sharp and confined peaks in BC concentration in the free troposphere above a near-steady mass concentration within the planetary boundary layer (~ 2 km for pre-monsoon and ~ 1km for winter). These peaks are identified on the respective profiles in fig.?. It is also seen very clearly that the sharp and most prominent peak occurred at between 4 and 5 km in the first and last profiles (fig.??a and fig.??c); which incidentally correspond to the pre-monsoon flights, while it occurred at a slightly lower altitude (between 2 to 3 km)~~

in the winter profile. The highest altitude layer seen in the profiles were in the vicinity of 7-9 km, though the amplitudes of these are much smaller than that of the layer seen at around 4.5 km. With the help of temperature data from the collocated meteorological payloads, we evaluated the environmental lapse rate profile for each of these profiles and the mean profile for each flight is shown in fig.??, following the same order as in fig.?. It is clearly seen that in the vicinity of the prominent BC peaks (P1 to P5 in fig.??a-c) there is a large reduction in the environmental lapse rate (sometimes reaching close to zero) e.g. at 4.5 km during March 2010 (fig.??a), around 2-3 km and 7 km during January 2011 (fig.??b); and around 4-5 km and 6-7 km during April 2011 (fig.??c). The lapse rate profile during April 2011 (fig.??c) shows multiple spikes possibly due to higher number of peaks in the corresponding BC profile (fig.??c), (Since the raw temperature data is very noisy, we have smoothed the data using two points fixed and two points moving average filter). Such reductions in temperature lapse rate would affect the local stability scenario. While the occurrence of high BC below 3 km can be explained by the boundary layer dynamics, high BC values at higher levels especially at 7 to 9 km, remained a mystery. Babu et al. (2011) hypothesized that the occurrence of such BC peaks at high altitudes could be associated with the local sources of BC at those altitudes, as 4-5 km and 8-9 km are the preferred corridors for aircrafts flying over Hyderabad. With a motivation to test this hypothesis and also to estimate the climate implication of such BC layers, we in this study, investigate the causes for the occurrence of such sharp and confined BC peaks at higher altitudes, using a regional online chemistry transport model. Crucial findings from this study are reported and possible ramifications are discussed.

3 Model and simulation details

2.1 Model and Simulations Details

We used the regional chemistry transport model, WRF-Chem in this study, to simulate the observed vertical profiles of BC and to understand the causes behind the sharp layers of BC at the elevated altitudes. The WRF-Chem model ~~used in this study employed~~ was employed with horizontal grid spacing of 2 km with 70 vertical levels and 100 m vertical resolution around the layers 4-6 km, 8-9 km and 10-11 km. Such a high vertical resolution was chosen to resolve the BC peaks, if simulated by the model, accurately. Out of the remaining levels, 10 are located within the boundary layer (0-2 km). A relatively coarser resolution (500 m), is set for the remaining altitude bands i.e. 2-4 km, 6-8 km and 9-10 km. Beyond 11 km, the vertical resolution is set to be 1 km. The model domain (75.5⁰E- 80.5⁰E, 14.5⁰N- 20.5⁰N) centers at Hyderabad and spans around 330 km in each direction (See fig.?? for details.). The simulations were started 16 days prior to the balloon flight days (flight day 1: 17 March, 2010, flight 2 : 8 January 2011 and flight day 3: 25 April 2011) and were run till the end of the flight day. In these simulations, the convective processes were not parameterized due to the fine grid spacing; cloud microphysical processes were parameterized using the Thompson scheme (Thompson et al., 2004), boundary layer processes were parameterized using the YSU scheme (Hong et al., 2006; Hong, 2010), and the surface processes were modeled using RUC-LSM scheme (Smirnova et al., 1997, 2000). The short-wave radiation processes were parameterized using Dudhia scheme (Dudhia, 1989) while the long-wave processes were parameterized using RRTM (Mlawer et al., 1997). Gas-phase chemistry in these simulations was handled by MOZART mechanisms (Emmons et al., 2010), while the aerosol-phase chemistry was parameter-

ized using GOCART bulk aerosol scheme (Chin et al., 2002) with Fast-J scheme for photolysis (Wild et al., 2000).

The model takes into account the following aerosols species: BC1 (Hydrophobic), BC2 (Hydrophilic), OC1 (Hydrophobic), OC2 (Hydrophilic), Dust (5 bins: effective diameters from 0.5 to 8 μm), Sea-salts (4 bins: effective diameters from 0.1 to 7.5 μm) and sulphate. The model simulates aerosol transport processes like emissions, advection, diffusion and deposition (dry and wet). The aerosol direct effects are modeled by coupling the aerosol scheme with the radiation scheme. The NCEP FNL (Final) Operational Global Analysis data, (interpolated to model resolution) has been used for setting up the initial and boundary conditions (updated every 6 h) for meteorological variables in the model, while the global chemistry transport model MOZART-4 (Emmons et al., 2010) has been used for creation of initial and boundary conditions for chemistry variables. The near-surface emissions of gas-phase as well as aerosol-phase species are formulated using the standard emission pre-processor software PREP-CHEM-SRC (version 1) (Freitas et al., 2011). The RETRO database (Schultz et al., 2007) is used for various greenhouse and precursor gases, EDGAR (Olivier et al., 1996) for emissions of CO, NO, NH₃ and VOCs and the GOCART (Chin et al., 2002) database is used for the emissions OC, BC and SO₂ over the region. The necessary modifications in BC emissions from GOCART database over this region (Govardhan et al., 2016) have been incorporated. We conducted two sets of simulations for each of the events. The first set included only surface emissions (NoACEM/Ctrl (Control) run: [simulations without the prescription of Aircraft Emissions of BC](#)) and the second set ([ACEM- simulations with the prescription of Aircraft Emissions of BC](#)), included surface and elevated emissions (~~ACEM-~~. The details of elevated emissions ([BC emissions from aircrafts](#)) are discussed in section 2.2)

2.2 [Prescription of aircraft BC emissions](#)

[To understand the role of BC emissions from aircrafts in formation of the sharp layers of BC at elevated altitudes, we prescribed aircraft BC emissions from the MACCity global anthropogenic emissions inventory \(Lamarque et al., 2010\) in the WRF-Chem simulations. The inventory provides aircraft BC emissions at \$0.5^0 \times 0.5^0\$ grids and 23 vertical levels from 0.305 km to 13.725 km, with a vertical resolution of 610 m. The vertical profile of BC emissions from aircrafts, averaged over a \$1^0 \times 1^0\$ grid box centering Hyderabad \(fig.1, red line\) and that over another grid box \(fig.1, blue line\) of the same dimension just \$2^0\$ south-west of the Hyderabad grid box, are shown in fig.1. The vertical locations of the emission peaks is seen to vary depending upon the proximity to airport \(Hyderabad, in this case\). The near-surface peak could be related to Landing-Take-Off \(LTO\) emissions over the airport, while the upper level peaks could be due to the emissions from the planes which are overpassing the location without landing.](#)

[The basic information this inventory considers are: global aircraft movements, performance characteristics of the different aircraft types, the actual paths of aircrafts during their journey and the emission factor for BC per kg of aviation fuel burnt. This global inventory for aircraft BC emissions inherently have several uncertainties, especially over a region like India mainly due to uncertainties associated with a\) amount of total aircraft fuel used across the country by domestic and international fleets; b\) quality of aircraft fuel; c\) age of aircrafts and hence the degradation in performance; d\) exact number of civil aircrafts flying over the region e\) unaccounted military aircrafts and f\) exact routes followed by the aircrafts over this region. Additionally, a major source of uncertainty in the estimates of BC emission from aircrafts arises due to the uncertainties in the estimations of](#)

emission factor for BC 'EI(BC)' for the particular aircraft fuel being used. EI(BC) is the amount of BC emitted (in g) per kg of aircraft fuel burnt and it depends on engine type, load conditions, engine conditions etc.. The measurement of EI(BC) by several studies (Herndon et al., 2008; Onasch et al., 2009; Timko et al., 2010) indicate that EI(BC) spans around four orders of magnitude. Stettler et al. (2013) discuss and quantify the underestimation of EI(BC) observed in currently available aircraft BC emissions inventories. In a previous study (Stettler et al., 2011) have stated: "The First-Order Approximation (FOA3) - currently the standard approach used to estimate particulate matter emissions from aircraft - is compared to measurements and it is shown that there are discrepancies greater than an order of magnitude for 40% of cases for both organic carbon and black carbon emissions indices".

Thus, 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., 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). Depending upon such evaluation of the MACCity emissions database over Hyderabad region, we have modified the inventory emissions by a corresponding scaling factor.

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.

2.3 Computation of atmospheric heating rate

The atmospheric heating rates due to the vertical profiles of BC measured as well as simulated, are computed using the Discrete Ordinate Radiative Transfer Model (Santa Barbara DISORT Atmospheric Radiative Transfer - SBDART (Ricchiazzi et al., 1998)). SBDART solves radiative transfer equations considering a plane-parallel atmosphere. Aerosol are specified in the model through total aerosol optical depth, single scattering albedo (SSA) and the Legendre moments of the scattering phase function for the assumed aerosol mixture; at every vertical layer. To compute the heating rate associated with the observed BC profiles, the required layer-wise total AODs were computed using vertical profile total extinction coefficient from CALIOP (Cloud Aerosol Lidar with Orthogonal Polarization) LIDAR on-board the CALIPSO (Cloud Aerosol Lidar Pathfinder Satellite Observation) satellite. The level 2 extinction coefficient data from CALIPSO was cloud-screened using the standard, recommended cloud-screening algorithm (https://www-calipso.larc.nasa.gov/resources/calipso_users_guide/tools/index.php) which makes use of the flags like AVD (Atmospheric Volume Description), CAD-Score (Cloud Aerosol Detection- Flag), extinction coefficient uncertainty and extinction coefficient quality control to separate aerosol from clouds. The final vertical profile of extinction coefficient considered was the mean all such cloud-screened vertical profiles which fall within $\pm 1.5^{\circ}$ of the balloon flight location in x-y direction and also ± 20 days of the balloon flight date. Such an averaging was done to get a mean picture of the aerosol loading over the balloon flight region. To get the required single scattering albedo we made use of the observed vertical profile

of BC and the OPAC (Optical Properties of Aerosol And Clouds) model (Hess et al., 1998). OPAC provides optical properties like extinction coefficient, absorption coefficient, scattering coefficient, single scattering albedo, asymmetry parameter, phase function etc. for the prescribed mixture of aerosol species. In OPAC, we prescribed the measured vertical distribution of BC to get the corresponding absorption coefficients. These absorption coefficients along with extinction coefficients from CALIPSO were used to get SSA. Finally, in order to compute the Legendre moments of the scattering phase function, we execute OPAC with one of its preset aerosol mixture- 'urban'; as it is best-suited for our observational location. This aerosol mixture has water soluble species (28000 \# cm^{-3}), insoluble species (1.5 \# cm^{-3}) and soot ($130000 \text{ \# cm}^{-3}$). For the 'urban' aerosol mixture we derived the scattering phase function and computed 8 Legendre moments of the scattering phase function. Along with the aforementioned primary inputs, SBDART requires some additional inputs such as mean state of atmospheric variables viz. temperature and pressure on every vertical level; and vertical profile of specific humidity and ozone. These inputs were taken from SBDART database under 'tropical' category, which suits our observation location (Hyderabad). Additionally, spectrally varying surface albedo values for Hyderabad were taken from MODIS surface reflectance product (MOD09CMG). This daily level 3 product provides surface reflectance for 7 bands (469 nm, 555 nm, 645 nm, 858.8 nm, 1240 nm, 1640 nm and 2130 nm) at 0.05° resolution. With all these inputs SBDART model was executed with 8 streams in the radiative transfer calculations with 7.5° resolution for solar zenith angle varying from 0 to 180. The net radiative fluxes were computed at top and bottom of every vertical layer starting from surface to 100 km. A similar run of SBDART was conducted without the prescription of aerosols. Using radiative fluxes from these 2 runs, radiative forcing within each layer was estimated using

$$\Delta F = Flux_{NA} - Flux_A \quad (1)$$

where, $Flux_{NA}$ = Radiative flux in absence of aerosols (W m^{-2})

$Flux_A$ = Radiative flux in presence of aerosols (W m^{-2})

The change in radiative fluxes in each layer due to aerosol is the amount of energy absorbed in the layer due to aerosols. Corresponding atmospheric heating rates were computed using Liou (2002)

$$dT/dt = (g/C_p)(\Delta F/\Delta p) \quad (2)$$

where dT/dt is the heating rate (K s^{-1}), g is the acceleration due to gravity, C_p the specific heat capacity of air at constant pressure ($C_p=1006 \text{ J kg}^{-1} \text{ K}^{-1}$) and p is the atmospheric pressure. The heating rates (K day^{-1}) were then averaged for 24 hours.

The vertical extents of heating rate profiles from the measurements were limited due to the availability of extinction coefficient data from CALIPSO over the region of interest. Such a limitation does not exist for model data, hence we have computed the atmospheric heating rates using the model results. To compute the atmospheric heating rates using model data we follow exactly the same aforementioned procedure. The only differences are: we use extinction coefficient data from model simulations instead of the CALIPSO satellite and we use SSA from model simulations instead of deriving it from the observed BC.

3 Results and Discussions

3.1 Observed vertical profiles of BC

The vertical profiles of BC, derived from flight measurements are shown in fig.2a-c, in which each profile is the average of the ascent and descent profiles for that flight. All the profiles revealed sharp and confined peaks in BC concentration in the free troposphere above a near-steady mass concentration within the planetary boundary layer (~ 2 km for pre-monsoon and ~ 1 km for winter). These peaks are identified on the respective profiles in fig.2a-c. It is also seen very clearly that the sharp and most prominent peak occurred at between 4 and 5 km in the first and last profiles (fig.2a and fig.2c); which incidentally correspond to the pre-monsoon flights, while it occurred at a slightly lower altitude (between 2 to 3 km) in the winter profile. The highest altitude layer seen in the profiles were in the vicinity of 7-9 km, though the amplitudes of these are much smaller than that of the layer seen at around 4.5 km. With the help of temperature data from the collocated meteorological payloads, we evaluated the environmental lapse rate profile for each of these profiles and the mean profile for each flight is shown in fig.2d-f, following the same order as in fig.2a-c. It is clearly seen that in the vicinity of the prominent BC peaks (P1 to P5 in fig.2a-c) there is a large reduction in the environmental lapse rate (sometimes reaching close to zero) e.g. at 4.5 km during March 2010 (fig.2d), around 2-3 km and 7 km during January 2011 (fig.2e); and around 4-5 km and 6-7 km during April 2011 (fig.2f). The lapse rate profile during April 2011 (fig.2f) shows multiple spikes possibly due to higher number of peaks in the corresponding BC profile (fig.2c), (Since the raw temperature data is very noisy, we have smoothened the data using two points fixed and two points moving average filter). Such reductions in temperature lapse rate would affect the local stability scenario. While the occurrence of high BC below 3 km can be explained by the boundary layer dynamics, high BC values at higher levels especially at 7 to 9 km, remained a mystery. Babu et al. (2011) hypothesized that the occurrence of such BC peaks at high altitudes could be associated with the local sources of BC at those altitudes, as 4-5 km and 8-9 km are the preferred corridors for aircrafts flying over Hyderabad. With a motivation to test this hypothesis and also to estimate the climate implication of such BC layers, we in this study, investigate the causes for the occurrence of such sharp and confined BC peaks at higher altitudes, using a regional online chemistry transport model. Crucial findings from this study are reported and possible ramifications are discussed.

3.2 Meteorological evaluation of the model

Meteorological processes like advection, diffusion, deposition etc. play a significant role in controlling the concentration of pollutants near the surface and their vertical distribution in the troposphere (Govardhan et al., 2015). Hence, before inspecting the model simulated vertical profile of BC, we first examine the performance of the model in simulating meteorology over the region of interest. We show such comparisons for March 2010 NoACEM simulations as a representative one. The model simulated meteorological parameters are averaged over the balloon flight region and then compared with the corresponding observations from the balloon flight. The three-point running mean smoothing is used to smoothen observational and simulated vertical profiles. Firstly, we have compared the model simulated vertical profile of horizontal wind speed (Blue line, fig.3 a) and direction (Blue line, fig. 3b) with the corresponding observations (Red line, fig.3a and Red line, fig.3b) available from the balloon flight measurements (deduced from the GPS data on board the balloon). The figures depict relatively weak (less than 10

m s⁻¹) low-level northerly-northeasterly winds upto around 5 km. Beyond this altitude, the winds change direction gradually and become westerlies above 8 km. The wind speeds also increase drastically beyond 8 km and reach a value of 20-25 m s⁻¹ at a height of 9 km and beyond. While the model simulated vertical variation of wind speed (Blue line, fig.3a) and direction (Blue line, fig.3b) agree broadly with the measurements, they differ in details and magnitudes. In model simulations, the change in wind direction starts occurring at an altitude ~ 1 km lower than in observations. The large increase in wind speed beyond 8 km altitude is satisfactorily captured by the model. Thus, with some disagreements in the actual magnitudes, the model captures broad features of horizontal advection strength (wind speed) and nature (wind direction) over the balloon flight domain vis-a-vis the measurements. We next examine the model simulations of vertical stability over the flight domain by comparing the model-simulated vertical profiles of potential temperature (θ) with the corresponding profiles from the balloon data. The observed profile (Red line, fig.3c) shows a stable layer (θ increases with height) up to ~~1st~~ first 2 km of the lower troposphere. Above this layer, there occurs a thick, well-mixed layer upto a height of 4 km, in which θ remains almost height-invariant. Above this height, the atmosphere remains largely stable with increase in θ with height. The modeled altitude variation of θ (Blue line, fig.3c) in general, matches with the measurements, barring a few discrepancies such as the extent of stable and well mixed layer in the lower atmosphere, magnitude of θ in the vicinity of the primary BC maxima in observations (around 4-4.5 km, fig. ~~??a~~ ete.2a). The model captures the stable layer lying up to around 1.5 km, similar to that seen in observations (up to 2 km, red line, fig.3c). A convectively unstable, well-mixed layer (θ constant) extending upto a height of 3.5 km, occurs above this layer, akin to observations (up to 4 km, red line, fig.3c), with an underestimation in θ values. Beyond this height, model also does not show any sign of instability, while θ increases with height, in agreement with the observations, yet with differences in the magnitudes. Thus, the model simulated vertical profile of θ also appears to be broadly comparable to the observed profile, with some differences. Examining the vertical thermal structure, we have also compared the model simulations of vertical profile of temperature with the corresponding measurements from the flight. While showing in general reduction in temperature with height similar to the observations (Red line, fig.3d), the model (Blue line, fig.3d) shows discrepancies in actual magnitudes vis-a-vis the observations (-1.7 to +3.4 K), as in case of the other meteorological variables. Possibly, owing to the existence of the primary BC peak between 4 to 5 km, the observations (Red line, fig.3d) depict a near-steady temperature within the particular altitude layer. The model (Blue line, fig.3d) on the other hand, fails to capture this feature. Thus, along with some differences in the details, the model simulations capture some of the large-scale features of the meteorology over the balloon flight region vis-a-vis the observations.

3.3 Simulated vertical profile of BC

With the above broad agreements in the meteorological fields, we proceeded to examine and evaluate the model simulated vertical profile of BC ~~over in the vicinity of~~ the balloon flight domain (~~fig.??a~~ blue line, fig.2g-i) in the NoACEM configuration. For 17th March 2010, the model simulations ~~though successfully capture the nature of the altitude distribution of BC, there are considerable differences in the finer details.~~ (NoACEM) show considerable differences vis-a-vis the ~~observations observed vertical profile of BC~~ (fig. ~~??a~~ ete.2a). The magnitudes of the simulated BC ~~significantly match~~ (blue line, fig.2g) show a relatively good comparison with the observations only over the lower most altitudes i.e. below 3.5 km. Above that altitude, the 2 profiles

completely differ from each other; while the observations (fig.??2a) show an increase in BC followed by a sharp peak at 4.5 km ($BC > 12 \mu\text{g m}^{-3}$), the model (fig.??a2g) displays a rapid decrease in BC concentration with altitude, without even a sign of the ‘elevated’ (high altitude) layers of high BC concentration seen in the measurements. Thus, though the model satisfactorily simulates the meteorology over the flight domain, it does not simulate the observed vertical profile of BC beyond 4 km altitude. The results for the other two flights are shown in fig.??b-2h and fig.??e2i. During January 2011, the model simulations (fig.??bNoACEM) (blue line, fig.2h) show rapid reduction in BC beyond within first 1 km. There is no sign of sharp and confined peaks beyond this height unlike the corresponding observations (fig.??2b). During April 2011, the model simulations (fig.??eNoACEM) (blue line, fig.2i) show sharp reduction in BC in within first 1 km. Beyond this height, the BC concentration remains relatively steady with height upto 3 km, above which a small peak is indicated between 3-4 km a sharp peak in BC is seen with a maxima near 2 km. A relatively gradual reduction in BC occurs beyond this height upto 4 km, followed by a flat steady BC profile with very low values above that height. Thus, even during April 2011, though the model satisfactorily captures the BC variations upto 3-4 km, it fails to do so at higher altitudes (blue line, fig.2i). Thus, none of the model simulations capture the sharp-confined BC peaks (or the elevated BC layers) occurring at higher altitudes. This strongly suggests that the meteorological factors alone cannot be responsible for the existence of the elevated BC layers. To throw more light on this, we considered other hypothesis.

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 (Babu et al., 2011), one has to look for local injection of BC aerosols at mid and upper troposphere because, surface based emissions would not lead to elevated BC layers at altitudes of 4-5 km or 7-9 km. In this context, emissions from commercial air traffic (that overfly Hyderabad, as well land and take off from there) assume significance. In their first reporting of the elevated layers, Babu et al. (2011) have hypothesized the role of such emissions, where they have obtained an approximate estimate of air traffic over the study location; about 200 aircrafts that overfly Hyderabad in the corridor 8-10 km and another 250 to 300 km that use the corridor 4 to 5 km, in the course of landing and taking off from the airport. We next evaluate elevated emissions due to aircrafts as a likely candidate for these high concentrations.

3.4 Prescription of aircraft BC emissions

For this, we prescribed aircraft BC emissions from the MACCity global anthropogenic emissions inventory (Lamarque et al., 2010) in the WRF-Chem simulations. The inventory provides aircraft BC emissions at $0.5^0 \times 0.5^0$ grids and 23 vertical levels from 0.305 km to 13.725 km, with a vertical resolution of 610 m. The basic information this inventory considers are: global aircraft movements, performance characteristics of the different aircraft types, the actual paths of aircrafts during their journey and the emission factor for BC per kg of aviation fuel burnt. This global inventory for aircraft BC emissions inherently have several uncertainties, especially over a region like India mainly due to uncertainties associated with a) amount of total aircraft fuel used across the country by domestic and international fleets; b) quality of aircraft fuel; c) age of aircrafts and hence the degradation in performance; d) exact number of civil aircrafts flying over the region e) unaccounted military aircrafts and f) exact routes followed by the aircrafts over this region. Additionally, a major source of uncertainty in the estimates of BC emission from

aircrafts arises due to the uncertainties in the estimations of emission factor for BC 'EI(BC)' for the particular aircraft fuel being used. EI(BC) is the amount of BC emitted (in g) per kg of aircraft fuel burnt and it depends on engine type, load conditions, engine conditions etc.. The measurement of EI(BC) by several studies (Herndon et al., 2008; Onasch et al., 2009; Timko et al., 2010) indicates that EI(BC) spans around four orders of magnitude. Stettler et al. (2013) discuss and quantify the underestimation of EI(BC) observed in currently available aircraft BC emissions inventories. In a previous study (Stettler et al., 2011) have stated: "The First Order Approximation (FOA3) — currently the standard approach used to estimate particulate matter emissions from aircraft — is compared to measurements and it is shown that there are discrepancies greater than an order of magnitude for 40 of cases for both organic carbon and black carbon emissions indices". 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.

3.4 Simulated vertical profile of BC with aircraft BC emissions

We examine the outcome of prescription of BC emissions from aircrafts on the modeled vertical profile of BC in fig. 22, averaged over a region 8 km^2 2g-i, in the vicinity of the balloon flight location domain, for the 3 balloon flight days. It is quite interesting to note that, upon the prescription of BC emissions from aircrafts, the model simulations (ACEM- simulations with the prescription of AirCRAFT EMissions) show sharp layers in the vertical profiles akin to the observed BC profiles (fig. 22, red line, fig. 2g-i), which are not simulated otherwise (NoACEM/Ctrl (control) run- simulations without the prescription of AirCRAFT EMissions). Though the actual altitudes and the magnitude of the BC layers differ in comparison with the observations, the nature of the layers (sharp and confined) looks very similar to those seen in the measurements sharpness of the modeled BC layers make them look similar to the observed BC layers. The two peaks in BC profile during March (red line, fig. 2g), the lower and the upper level BC peaks during January (red line, fig. 2h) and the clustered BC peaks during April (red line, fig. 2i) are well simulated by the model only after the prescription of BC emissions from aircrafts. Thus, even for the model with realistic meteorology, the high altitude BC peaks/ layers are captured only when the high altitude sources of BC are prescribed. This clearly highlights the role played by aircraft emissions of BC in creation of the high altitude BC peaks, which remained as a hypothesis earlier. Additionally, as a consequence of such high altitude BC emissions, the modelled modeled vertical profile of temperature lapse rate over the balloon flight region shows a reduction in the magnitude in ACEM case as compared to the NoACEM case, indicating the warming due to BC (fig. 4a). The corresponding differences in the temperature lapse rate ($d(T/dz) = (dT/dz)_{ACEM} - (dT/dz)_{NoACEM}$) appear to be higher especially at higher altitudes of 4 km and beyond 7km (fig. 4b). Such a reduction in the magnitude of the temperature lapse rate values, results in a better match between model simulations and the corresponding observations of temperature lapse rate especially at higher altitudes (from 6 to 8 km and from 9 to 9.5 km)(fig. 4a). Additionally, we carried out one more model simulation, in which we prescribed the emissions of BC from biomass burning activities using FINN-version the Fire INventory from NCAR (FINN) version 1.5 inventory (Wiedinmyer et al., 2011) biomass burning data. This Fire INventory from NCAR (FINN) The FINN provides high resolution, global emission estimates from such open burning activities. The temporal resolution of the inventory is 1 day, while the spatial resolution is 1 km^2 . In our simulations, we allowed such emissions from biomass burning to lift vertically following the online

plume-rise module (Freitas, 2007). However, the model could not simulate the observed sharp and confined BC peaks (figure not shown), if we switch off the prescription of BC emissions from aircrafts. This clearly shows the important role of aircraft BC emissions in causing high altitude BC peaks.

3.5 Sensitivity of the vertical profile of BC to the surface level emissions

5 To test the robustness of the simulated high altitude BC peaks, we have also examined the impact of near-surface emissions of BC on the elevated layers. For this, we have done a similar simulation with aircraft BC emissions, in which we have turned-off the near-surface fossil fuel emissions of BC over the model domain. We have then examined the effect of this on the simulated vertical profile of BC in fig.5. It can be clearly seen from this figure that the vertical profiles simulated by the 2 runs (SE- the run with prescribed near-surface anthropogenic emissions of BC and N-SE - the run with near-surface anthropogenic BC emissions turned off) differ only in the lower altitude region, upto a height about 4 km (probably the altitude up to which BC could be lofted by the convection). Beyond 4 km, the profiles are ~~identical~~largely similar, implying that the elevated BC layers are insensitive to surface BC emissions. Beyond 4 km, the profiles look similar. 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 $0.1 \mu\text{g m}^{-3}$. Thus, our model simulations indicate that, the high altitude peaks in BC are not a result of convective-lifting of near-surface BC, but they are indeed caused due to the BC emissions (injection of BC) at higher altitudes by the aircrafts.

3.6 ~~Atmospheric heating rate due to the observed and modelled BC vertical profile~~Being a strong absorber of radiation over a wide spectral band, Model simulated elevated sharp BC layers absorb incoming solar, which would then heat up the atmosphere locally and can alter the vertical stability. In this section, we compute the atmospheric heating caused by the observed and modelled BC vertical profiles. The methodology that we follow to compute the atmospheric heating rates due to BC is similar to that followed by Babu et al. (2002) and Babu et al. (2011). Seasonal Scenario

We noticed that the model, WRF-Chem, produces the elevated sharp peaks of BC akin to the observations upon the prescription of realistic emissions of BC from aircrafts. We now proceed to examine the seasonal behavior such elevated sharp BC layers, within the model simulations. For this, we have carried out 1 month long model simulations during each season of the year 2010 i.e. January, March, July and October, representative of winter, pre-monsoon, monsoon and post-monsoon seasons. Keeping in mind, the limited width and the dynamic behavior of the horizontal location of the aircraft emitted trail, instead of plotting a monthly mean vertical profile of BC, averaged over a region, we have computed seasonal probability of occurrence of a high altitude BC peak at every point within the domain.

3.6.1 ~~Atmospheric heating rates due to observed BC~~

The atmospheric heating rates due to the observed vertical profiles of BC are computed using a Discrete Ordinate Radiative Transfer Model (Santa Barbara DISORT Atmospheric Radiative Transfer – SBDART (Ricchiazzi et al., 1998)). SBDART solves radiative transfer equations considering a plane parallel atmosphere. Aerosol are included in SBDART by specifying total aerosol optical depth, single scattering albedo (SSA) and the Legendre moments of the scattering phase function for the assumed aerosol mixture; at every vertical layer. The required layer wise total AODs were computed using vertical profile total extinction coefficient from CALIOP (Cloud Aerosol Lidar with Orthogonal Polarization) LIDAR on board the CALIPSO (Cloud Aerosol Lidar Pathfinder Satellite Observation) satellite. The level 2 extinction coefficient data from CALIPSO was cloud screened using the standard, recommended cloud screening algorithm (https://www-calipso.larc.nasa.gov/resources/calipso_users_guide/tools/index.php) which makes use of the flags like AVD (Atmospheric Volume Description), CAD-Score (Cloud Aerosol Detection Flag), extinction coefficient uncertainty and extinction coefficient quality control to separate aerosol from clouds. The final vertical profile of extinction coefficient considered was the mean all such cloud screened vertical profiles which fall within $\pm 1.5^{\circ}$ of the balloon flight location in x-y direction and also ± 20 days of the balloon flight date. Such an averaging was done to get a mean picture of the aerosol loading over the balloon flight region. To get the required single scattering albedo we made use of the observed vertical profile of BC and the OPAC (Optical Properties of Aerosol And Clouds) model (Hess et al., 1998). OPAC provides optical properties like extinction coefficient, absorption coefficient, scattering coefficient, single scattering albedo, asymmetry parameter, phase function etc. for the prescribed mixture of aerosol species. In OPAC, we prescribed the measured vertical distribution of BC to get the corresponding absorption coefficients. These absorption coefficients along with extinction coefficients from CALIPSO were used to get SSA. Finally, in order to compute the Legendre moments of the scattering phase function, we execute OPAC with one of its preset aerosol mixture ‘urban’, as it is best suited for our observational location. This aerosol mixture has water soluble species (28000 \# cm^{-3}), insoluble species (1.5 \# cm^{-3}) and soot ($130000 \text{ \# cm}^{-3}$). For the ‘urban’ aerosol mixture we derived the scattering phase function and computed 8 Legendre moments of the scattering phase function. Along with the aforementioned primary inputs, SBDART requires some additional inputs such as mean state of atmospheric variables viz. temperature and pressure on every vertical level; and vertical profile of specific humidity and ozone. These inputs were taken from SBDART database under ‘tropical’ category, which suits our observation location (Hyderabad). Additionally, spectrally varying surface albedo values for Hyderabad were taken from MODIS surface reflectance product (MOD09CMG). This daily level-

3.6.1 Probability of occurrence of sharp BC peak within 9-11km

For every point within the domain, the probability of occurrence of sharp BC peak is computed by examining the maximum value of the simulated BC within 9-11 km altitude band and that within 7-9 km altitude band. When the ratio of maximum BC magnitude within 9-11 km band to that within 7-9 km band, is more than 3 product provides surface reflectance for 7 bands (469 nm, 555 nm, 645 nm, 858.8 nm, 1240 nm, 1640 nm and 2130 nm) at 0.05° resolution. With all these inputs SBDART model was executed with 8 streams in the radiative transfer calculations with 7.5° resolution for solar zenith angle varying from 0 to 180. The net radiative fluxes were computed at top and bottom of every vertical layer starting from surface to 100 km. A similar run of SBDART was conducted without the prescription of aerosols. Using radiative fluxes from these 2 runs, radiative

~~forcing within each layer was estimated using-~~

$$\Delta F = Flux_{NA} - Flux_A$$

, we considered the occurrence of high altitude sharp BC peak within 9-11 km altitude band. The probability of occurrence of sharp BC peak within 9-11 km band for the entire month is computed as follows,

$$5 \quad p_{(9-11)} = \frac{T_{(Peak:9-11)}}{T_{Total}} \quad (3)$$

where, ~~Flux_{NA} = Radiative flux in absence of aerosols (W m⁻²)-~~

~~Flux_A = Radiative flux in presence of aerosols (W m⁻²)-~~ p₍₉₋₁₁₎ = probability of occurrence of a sharp BC peak within 9-11 km altitude band

T_(Peak:9-11) = Number of instances when a sharp BC peak occurs at 9-11 km altitude band

10 T_{Total} = 240 for the entire month with 3 hourly output frequency

~~The change in radiative fluxes in each layer due to aerosol is the amount of energy absorbed in the layer due to aerosols. Corresponding atmospheric heating rates were computed using Liou (2002),-~~

$$\underline{dT/dt = (g/C_p)(\Delta F/\Delta p)}$$

~~where dT/dt is the heating rate (K s⁻¹), g is the acceleration due to gravity, C_p the specific heat capacity of air at constant pressure (C_p = 1006 J kg⁻¹ K⁻¹) and p is the atmospheric pressure. (Satheesh and Ramanathan, 2000).~~ Such computation of p₍₉₋₁₁₎ is done for all the months of the model simulations and is shown in fig.6. During the month of January 2010 (fig.6a) and March 2010 (fig.6b), the probability of getting an upper level (9-11 km) sharp peak in BC within the model domain goes as high as 60% over the area in the south-west part of the domain. This looks to be primarily related to the location of the aircraft emitted trail in the emissions inventory. Around 40% chances of getting a sharp peak at upper level (9-11 km) are seen at various locations which roughly occur over the location of the aircraft emissions in the inventory and are further controlled by the direction of the prevailing upper level winds. For the monsoonal period i.e. during the month of July 2010 (fig.6c), such probability values reduce, with most of the region showing less than 10% probability. Such a reduction in p₍₉₋₁₁₎ over the domain looks to be a result of convective lifting of BC during monsoon (which is relatively lesser during pre-monsoon and post-monsoon) resulting into more uniform concentration of BC at the elevated altitudes causing removal of such sharp peaks in BC, but increased abundance of BC at those altitudes as shown in fig.7. During the month of October 2010 (fig.6d), the p₍₉₋₁₁₎ once again reaches to as high as 50%, with the specific aircraft emission zones showing higher values. The spatial pattern of the probability appears to be roughly contrast of that during January or March 2010. This behavior could be due to the difference between the direction of the prevailing winds during January/March and that during October. Thus, in summary, probability of getting a BC peak at 9-11 km altitude in the model simulations, looks to be dependent on the season and the location.

3.6.2 Probability of occurrence of sharp BC peak within 4-5km

Following a similar exercise, we have estimated the probability of occurrence of BC peak within 4-5 km altitude band. The lower level (4-5 km) peak in BC is identified when the maxima of BC magnitude within the 4-5 km altitude band goes more than twice of that of the neighboring altitude bands. The spatial plot of such probability values for each month is shown in fig.8. In-general, it could be seen that the probability of getting a lower level (here 4-5 km) peak in the simulated BC (fig.8) is lower than that for the upper-level (9-11 km) peak (fig.6), within the model simulations. This could possibly be linked with the differences associated with, the altitude variation and the magnitudes, of BC emissions from aircrafts, over 9-11 km altitude band vis-a-vis 4-5 km altitude band, within the inventory. During July 2010, the monsoonal convection reduces the p_{4-5} values by vertically lifting BC throughout the column, getting rid of the sharp BC peaks as shown in fig.7.

3.7 Atmospheric heating rate due to the observed and modeled BC vertical profile

Being a strong absorber of radiation over a wide spectral band, elevated BC layers absorb incoming solar, which would then heat-up the atmosphere locally and can alter the vertical stability. In this section, we compute the atmospheric heating caused by the observed and modeled BC vertical profiles. The methodology that we follow to compute the atmospheric heating rates due to BC is similar to that followed by Babu et al. (2002) and Babu et al. (2011)

3.7.1 Atmospheric heating rates due to observed BC

Following the ~~aforementioned procedure~~ procedure outlined in section 2.3, we computed the atmospheric heating rates due to the measured BC profiles for the 3 balloon flights. The heating profiles are presented in fig.??9a, fig.??9b and fig.??9c for the March, January and April flights respectively. The vertical extent of the profiles is limited to the availability of the extinction coefficient data from CALIPSO satellite. During March 2010 (fig.??9a), the heating rates are seen to be within 0.5 to 1 (K day⁻¹) upto 2-2.5 km of the atmosphere. The denser atmosphere could also be one of the factors responsible for such low values of heating rates at lower altitudes. Beyond this height, a large increase in heating rates could be seen up to ~5 km. The heating rate maxima (~5 K day⁻¹) is seen to occur around the height of BC maxima (fig.??2a). Such a large heating rate could cause the observed reduction in temperature lapse rate around those altitudes (fig.4a2d). During the winter flight (January 2011, fig.??9b), the observed BC ~~profile causes~~ profiles along with extinction coefficient profiles from CALIPSO, cause more heating near the surface ~~than that at higher heights~~. The corresponding heating rates at near-surface levels are even higher than that during the summer months (March 2010, fig.??9a and April 2011, fig.??9c). The heating rate profile during April 2011 (fig.??9c) shows similar features as that of March 2010 (fig.??9a). The maxima in heating rate also occurs around the elevated BC layers, with comparable magnitudes. As expected, since the observed BC profile during April 2011 (fig.??2c) shows more number local peaks as compared to that during March 2010 (fig.??2a), the heating rate profile during April 2011 also shows more pronounced spikes vis-a-vis March 2010. The ~~identical~~ 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.

Thus, during the summer months the study region is characterized with maxima in the heating rates ($\sim 5 \text{ K day}^{-1}$) at around 4-5 km, while during the winter months the maxima ($\sim 1 \text{ K day}^{-1}$) lies near the surface.

3.7.2 Atmospheric heating rates due to ~~modelled~~ modeled BC

The ~~vertical extents of heating rate profiles from the measurements were limited due to the availability of extinction coefficient data from CALIPSO over the region of interest. Such a limitation does not exist for model data, hence we now compute the atmospheric heating rates using the model results. To compute the atmospheric heating rates using model data we follow exactly the same procedure that was used earlier. The only differences are: we now use extinction coefficient data from model simulations instead of the CALIPSO satellite and we use SSA from model simulations instead of deriving it from the observed BC.~~ The corresponding atmospheric heating rate profiles in model simulations are shown respectively in fig. ~~??a,b and c~~ 9d,e and f. The heating rates are computed for the corresponding BC profiles (ACEM) shown in fig. ~~??a,b and c~~ 2g,h and i (red line). The profiles are ~~averaged for a region 8 km^2 and are~~ representative of all such profiles, which show the effect of aircraft emissions. During March 2010, the model heating-rate profile (fig. ~~??a~~ 9d) shows similar features vis-a-vis the corresponding observations (fig. ~~??a~~ 9a). Though the actual magnitudes of heating rates are lower for the model simulated profiles, it captures the major and important features such as steady increase in heating rates from surface to 3 km and maxima in heating rates at around 3-4 km. Interestingly, a secondary peak in heating rates can be seen around 11 km. This peak corresponds to the peak in BC as seen in fig. ~~??a~~ 2a. During January 2011 (fig. ~~??b~~ 9e), the model also shows high values of heating rates nearer to the surface similar to the observations (fig. ~~??b~~ 9b). Beyond this height the heating rates reduce. A secondary peak in heating rates at around 10-12 km is seen also during January 2011, with the magnitude as high as 0.5 K day^{-1} . During April 2011, the model heating rate profile (fig. ~~??e~~ 9f) shows a maximum close to the surface corresponding to the maximum in BC (fig. ~~??c~~ 2c), and multiple maxima at higher heights (3km, 6km) corresponding to the presence of BC layers. The upper level maxima at around 11 km is seen to be relatively weaker during April (fig. ~~??e~~ 9f) vis-a-vis March (fig. ~~??a~~ 9d) and January (fig. ~~??b~~ 9e) simulations. The heating rate profiles shown in fig. ~~??a-e~~ 9d-f, are a representative of all those locations which show an existence of the aircraft emitted BC trails at higher altitudes, but are located away from the ~~starting point of the trail (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 area with high emissions (the region, with high magnitude of emissions compared to the rest of the trail, where two or more such trails cross each other).~~ We term these generic heating rate profiles as 'normal profiles'. Additionally, we inspected the heating rate profiles for those locations within the trail which lie in the vicinity of the ~~starting point of the trails area with relatively high emission intensity. Such regions are formed when two or more such trails cross each other.~~ We term ~~these the corresponding~~ heating rate profiles as 'extreme profiles'. Such 'extreme profiles' of heating rates are shown in fig. ~~??d-f~~ 9g-i, for March 2010, January 2011 and April 2011 respectively. It can be noticed from fig. ~~??a-f~~ 9d-i that, while the extreme profiles show similar heating rate magnitudes within lower part of the atmosphere as that of the normal profiles; they produce very high heating rates at the higher heights (10-12 km). Such heating rates even cross the near-surface maxima for the April 2011 profile (fig. ~~??f~~ 9i). The rarer atmosphere could also contribute to such a high values of heating rates at those altitudes. Such large heating rate values at those heights can affect the stability of the atmosphere. The resulting vertical mixing can lift the BC particles even

higher. This continuous warming-lifting cycle can result in further vertical transport of the BC particulates, which is termed as self-lifting of BC (de Laat et al., 2012). In addition to the self-lifting mechanism, the BC emitted at high altitudes can get transported vertically up in presence of strong underlying convection. In the next section, we show, with the help of model simulations, that such convective transport of the high-altitude BC could occur in tropics during periods of strong convection.

5 3.8 Convective lifting of BC

To examine the convective lifting of high-altitude BC, we have carried out WRF-Chem simulations with the prescription of aircraft BC emissions over Indian region for July 2010. July month was selected for this purpose, in view of the known prevalence of deep convection over Indian region, associated with the summer monsoon and the tropical tropopause layer being thinnest at this time of the year (Meenu et al., 2010; Sunilkumar et al., 2013). The ~~tropical tropopause layer is the region between the top of the major convective towers and the cold point. While forming an interface between the two dynamically different regimes, this layer acts as an entry point to the tropical stratosphere (Fueglistaler et al., 2009).~~ The animation (Mov.1 in supplementary material) shows height-longitude plots of maximum value of BC for a latitude belt of 2.25° over Hyderabad (X-axis: Longitude, Y-axis: Height (m)). The animation clearly reveals the capability of strong convections to lift BC to heights beyond 14 km. On a few occasions, these layers are seen to get transported even beyond 17 km, across the tropopause. Similarly, we compute the difference in the upper tropospheric BC mass concentration values in ACEM (runs with the prescription of aircraft BC emissions) simulations vis-a-vis the NoACEM case (runs without prescription of aircraft BC emissions) ($\Delta BC = BC_{ACEM} - BC_{NoACEM}$), averaged for the entire duration of model simulations carried out during July 2010. The maximum of such time-averaged ΔBC values across the latitude belt of 2.25° centered over Hyderabad, for every longitude of the model domain is shown in fig.10. The time-averaged increment in UTLS (Upper Troposphere and Lower Stratosphere) BC load due to aircraft BC emissions is seen to occur over the entire longitude belt, though more pronounced over a few longitude bands ($79^{\circ}E-80^{\circ}E$). Higher ΔBC values extend vertically even beyond 17.6 km, highlighting the mean vertical transport of aircraft emitted BC in the UTLS region. These provide a strong evidence for the intrusion of high altitude aircraft emitted BC into the upper-tropospheric-lower-stratospheric heights over the Indian region during strong convective periods. ~~Similar lifting of pollutants due to convection to stratospheric heights has also been reported by some of the earlier studies (Andreae et al., 2001; Fromm et al., 2005; Randel et al., 2010; Vernier et al., 2011).~~ The intrusion of such tropospheric air into the stratosphere will be favored under certain conditions like thinner tropopause layer, unstable conditions within the tropopause layer etc. ~~Examining the long term records of tropopause characteristics over Indian region using the vertical profiles of temperature obtained from radiosonde measurements at thirty-four different India Meteorological Department (IMD) stations, for the period 1994-2011, Sunilkumar et al. (2013) have reported thinning of the Tropical Tropopause Layer (TTL) over the region during monsoon season occurring mainly due to strong monsoonal convection. The TTL thickness goes down to ~ 2.5 km during July-August, from ~ 3.5 km during December-January. Also, during monsoon season, the Temperature Lapse Rate (TLR) within TTL increases to ~ 6 K/km from its value of ~ 5.4 K/km during the other months. Such a rise in TLR make TTL more convectively unstable and favorable for vertical lifting of air parcels. Thus, the reduction in TTL thickness and increase in TLR within TTL during monsoon season create favorable conditions for cross-tropopause transport of air and pollutants. A~~

~~recent study by Das et al. (2016) reports a cross tropopause transport of air mass over Indian region during the cyclone 'Nilam'. Such a cross tropopause transport coupled with convective transport/self lifting of high altitude aircraft emitted BC to higher heights, lends strong support to the stratospheric intrusion of BC particles. Once the particles enter the stratosphere, they reside for a longer period of time due to the inherent stability of stratosphere and the absence of strong removal mechanisms.~~ In the

5 next section, we show observations supporting such intrusion.

3.9 Occurrence of BC at high altitudes: Observational evidence

There exist several observational reports of presence of BC at upper tropospheric and the stratospheric altitudes (Chuan and Woods, 1984; Okada et al., 1992; Pusechel et al., 1992; Sheridan et al., 1994; Blake and Kato, 1995; Pusechel et al., 1997; Strawa et al., 1999; Baumgardner et al., 2004; Schwarz et al., 2006; Kremser et al., 2016). One of the ways to ascertain the occurrence of

10 such layers in the stratosphere is to examine the CALIOP LIDAR (on board CALIPSO satellite) extinction coefficient data (at 550 nm). We have examined all such vertical profiles for 3 consecutive years (from January 2010 to December 2012), over 5 specifically chosen regions ($5^{\circ} \times 5^{\circ}$) over and around India (fig.11). Since, we want to focus on the presence of aerosols at stratospheric altitudes, we have plotted the vertical profiles of extinction coefficient in the stratospheric altitudes (i.e. from altitude 20 km to ~ 30 km). The results are shown in fig.12.

15 To our surprise, we notice that the aerosol extinction coefficient values over the stratospheric altitudes are more than 0.02 km^{-1} over all these regions; occasionally values greater than 0.06 km^{-1} are also noticeable (fig.12a). The corresponding appended time-series of stratospheric (i.e. from altitude 20 km to ~ 30 km) Aerosol Optical Depth over the regional boxes under consideration from 2010 to 2012 is plotted in fig.12b. The AODs are seen to be higher than 0.01 on most of the occasions with frequent spikes reaching as high as 0.16. We considered different scenarios to explain such high AOD. These included, (i)

20 volcanic perturbation to stratospheric AOD, and (ii) high altitude cirrus clouds at TTL region. We first compared these extinction coefficient and AOD values with the corresponding area-averaged values for the entire tropical belt ($\sim 20^{\circ}\text{S}$ to $\sim 20^{\circ}\text{N}$) (Kremser et al., 2016). The maximum value of stratospheric AOD averaged for the entire tropical belt (Kremser et al., 2016) for the period ~~2000-2010~~ to 2012 (which is considered to be the background tropical stratospheric AOD in this study), is marked with the dotted red-line in fig.12b. It can be seen that, the stratospheric AODs from our analysis over the 5 regional boxes are

25 roughly 4-6 times higher than the background tropical stratospheric AOD (dotted red line, fig.12b), same is the case for aerosol extinction coefficient values as well (comparison not shown). ~~This signifies~~ The large differences in the background tropical stratospheric AOD and the stratospheric AODs over the Indian region, signify the enhancement in the stratospheric aerosol burden over the regions under consideration, above the background stratospheric aerosol loading. One of the major controllers of stratospheric AOD is the in-flux of particulates namely sulphates, organics and ash, and gases like SO_2 from volcanic eruptions

30 into the stratosphere (Kremser et al., 2016). An examination of the occurrence of global volcanic eruptions from 2008 to 2012 (Kremser et al., 2016) suggests that, out of the 11 major volcanic eruptions that occurred during that period, 7 were centered away from the tropics. Also, the flux of SO_2 in the stratosphere from the most intense of the 11 events, was merely $(1/10)^{\text{th}}$ of that of the mount Pinatubo eruption (Kremser et al., 2016). Thus, this proposes that the enhancement in AOD and the extinction coefficient over the regions under consideration in this study may not be linked to volcanic eruptions. The monsoon season

(June to September) appears to be the favorite period for the lifted layers in the stratosphere (fig.12a). This looks to be linked to the vertical lifting associated with severe convection (as seen in our model simulations fig.10 and Mov.1 in supplementary material) and thin tropopause layer as explained earlier (Fu et al., 2006; Randel et al., 2010; Vernier et al., 2011; Thampi et al., 2012; Uma et al., 2012). Additionally, these layers seem to be present during the other seasons (pre-monsoon and winter) as well. The Indian region experiences a large scale ascending motion above 10 km during winter months (Rao et al., 2008). Such an ascending motion can cause intrusion of tropospheric air (lying at 8-10 km) into the stratosphere (Rao et al., 2008). Thus, while the monsoonal convection and the related thermodynamics appears to be a cause for the summer time higher extinction coefficient values at stratospheric heights over the Indian region, the transport of tropospheric air mass into the stratosphere due to the large-scale ascent beyond 10 km looks to be responsible for the winter time high values.

To throw light on the aerosol species responsible for such values of extinction coefficient, we examined the Particle Depolarization Ratios (at 550nm, henceforth PDR) at these altitudes (fig.12c). PDR is the ratio of attenuated backscatter coefficient in perpendicular direction to that in the parallel direction. We computed the PDR values from the corresponding backscatter coefficient values. The PDR values are seen to be higher than 0.3 on most of the occasions. This suggests the presence of non-spherical aerosol species at those altitudes. One of the major non-spherical aerosol specie over this region is mineral dust. ~~But dust is relatively too heavy and coarse to be lifted up to such altitudes. On the other hand~~ 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 previously, 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 μ m) (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) , 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. Additionally, the maximum cloud top altitude occurring over the Indian region during the monsoon season is 20 km (Meenu et al., 2010). This suggests that, the non-spherical ice crystals emanating from the anvils of the convective towers over the Indian region could be found only around 20 km and not above 21-22 km. These together eliminate the possibility of presence of non-spherical mineral dust particles and ice crystals at these heights. A few previous studies have noticed presence of BC chain agglomerates at the upper tropospheric and lower stratospheric heights (Blake and Kato, 1995; Bekki, 1997).

Such a transport of BC to the stratospheric heights could be a result of convective-lifting (as seen in our model simulations fig.10 and Mov.1) and 'self-lifting' (de Laat et al., 2012) associated with them. Though pure and nascent BC has relatively lower value of PDR, the aged BC particles being porous (Chand et al., 2005; Dusek et al., 2005) and capable of forming long chain agglomerates are non-spherical and would depict higher values of PDR. Such BC agglomerates even when mixed with stratospheric sulfate could still give rise to non-spherical shapes and thus higher values of PDR. This indicates the possible existence of BC at the stratospheric altitudes over the Indian region. Thus, the model simulated convective transport of high altitude aircraft emitted BC layers to UTLS region, favorable conditions for cross-tropopause transport of air-mass over Indian region during monsoonal months, large scale winter-time ascending motion above 10 km over Indian region and the presence of local BC emission sources (aircrafts) at high altitudes (~ 10km) throughout the year together indicate the possibility of aircrafts emissions being primarily responsible for the possible presence of BC in stratosphere over the Indian region.

4 Possible implications of presence of BC at stratospheric heights

Our model simulations suggest that, aircrafts emissions are capable of producing the observed elevated aerosol layers in free troposphere (middle and upper) and the BC particles in these layers can intrude into the stratosphere, especially during periods of thin TTL, aided by strong convective lofting (associated with summer monsoon) and self-lifting mechanisms. The average residence time of stratospheric aerosol is about 1 year (Snetsinger et al., 1987). BC aerosols, owing to their porosity and fractal shape (Blake and Kato, 1995; Bekki, 1997), can provide larger surface area to support heterogeneous chemical reactions in the upper troposphere and lower stratosphere. One of such reactions is the ozone decomposition on BC (Akhter et al., 1985; Bekki, 1997; Schurath and Naumann, 1998; Disselkamp et al., 2000; Chughtai et al., 2003; Satheesh et al., 2013), which may result in depletion of stratospheric ozone. Besides, if BC is transported to the stratospheric heights, the high altitude cirrus clouds existing around the tropical tropopause layer, which reflect back most of the solar radiation incident on them, would consequently induce increased interaction between solar radiation and the lifted BC layer (Satheesh and Moorthy, 2005). This would substantially enhance BC-induced warming of lower stratosphere. Since the atmosphere becomes increasingly thinner with altitude, for a given amount of absorbed energy, the warming would be higher at upper levels than that near the surface. Kamm et al. (1999), based on laboratory experiments, have reported that reaction rate of loss of ozone in presence of BC has positive temperature dependence. Thus, the aircraft emitted BC could possibly contribute to the depletion of the stratospheric ozone layer. The continued negative trend in stratospheric ozone over 40⁰S-40⁰N from 1984 to 2014 (Bourassa et al., 2014) could possibly be related to the ever increasing aircraft traffic (Paraschis and Gittens, 2014; Piccolo and Gittens, 2015, 2016). The observed delays in recovery of stratospheric ozone hole (Kane, 2008, 2009), could also be influenced by such interactions of lifted BC with the stratospheric ozone.

Thus, with the help of a regional chemistry transport model, our study showed that the aircrafts (with realistic emissions) appear to be one of the primary causes behind the occurrence of the the sharp elevated layers of BC over the Indian region. This study utilized high altitude measurements of BC carried out using a zero-pressure balloon. Though, the balloon measurements were carried out in 3 different seasons to capture the seasonal picture, they are limited in number. In future, more number of

such high altitude balloon measurements of BC would be needed to confirm the occurrence of such elevated sharp BC peaks. Also, though the high altitude balloon launching facility is currently available only at 1 station in India, more number of such balloon measurements from different locations, using mobile facility would be useful to generalize the results. Additionally, such high altitude BC measurements could also be carried out in the vicinity of busier airport locations across the world to throw more light on such implications of aircraft emissions. The BC emissions from aircraft, in the MACCity inventory were scaled in this study, to match realistic emissions of BC from aircrafts over the Indian region. This highlights the large uncertainty associated with BC emissions from aircrafts, especially regarding the emission factor for BC (EI(BC)) and possibly with actual air traffic data. This uncertainty needs to be examined in future to constrain the implications of emissions from aircrafts.

5 Conclusions

The altitude distribution of BC in atmosphere plays a crucial role in deciding the BC-induced warming of the atmosphere. Such an atmospheric warming due to BC gets amplified when BC lies above strongly reflecting surface like clouds. The enhanced warming of BC can give rise to local instability and subsequent vertical lifting. Such absorption-warming-convective cycle can transport BC to higher altitudes. On account of its absorbing nature, high altitude BC can burn-off cirrus clouds. Realizing the importance of the vertical profile of BC, Babu et al. (2011) measured vertical profile of BC over strongly convecting Indian region during pre-monsoon periods (March 2010) using high altitude zero pressure balloon. The authors reported 2 sharp and confined high altitude layers of BC at 4.5 km and 8.3 km. The existence of the high altitude BC layers was confirmed by two subsequent balloon flights conducted during the subsequent winter and pre-monsoon seasons. While the high values of BC within ~~1st~~ first 3 km of atmosphere could be explained by boundary layer mixing, the higher BC values aloft appeared to be a mystery. The present study used the regional chemistry transport model - WRF-Chem, to understand the causes behind the observed high altitude sharp and confined BC layers. Firstly the simulations (incorporating surface BC emissions) of the meteorological parameters within the model were examined. Broad agreements of model simulated meteorological variables with their corresponding observations from the balloon measurements were found. Then the model simulated vertical profile of BC were compared with the corresponding observed profile, but it was found that the model could not replicate the high altitude BC layers as seen in observations. Thus reasonable simulations of meteorology within the model could not give rise to the high altitude BC layers. Then the emission of BC from high altitude sources i.e. aircrafts were prescribed in our simulations. The aircraft BC emissions from MACCity inventory, interpolated to our model grids with necessary modifications were used for this purpose. Upon the prescription of realistic BC emissions from aircrafts, the model simulated vertical profile of BC started showing the mysterious high altitude BC peaks as seen in the observations. The sharp and confined nature of the simulated BC peaks showed large similarities with the observations, though their exact altitude and the magnitude differed vis-a-vis the observations. The fact that, aircraft emissions of BC being the primary cause behind the high altitude BC peaks, was re-confirmed when the peaks remained undisturbed, even after shutting down the surface level emissions of BC. The probability of occurrence of such high altitude (9-11 km) BC peak is found to be more than 40% over many locations within the model domain, during all the seasons except monsoon. During the monsoon season, the strong convection mixes

BC throughout the atmospheric column, resulting into removal of such elevated sharp peaks. The corresponding atmospheric heating rates due to such observed and simulated profiles of BC were then computed. It was seen that such BC profiles could give rise to local instability and subsequent vertical mixing even at higher altitudes (~10-11 km). Such high altitude BC peaks would cause warming which could be amplified in the presence of highly reflecting clouds beneath them. This absorption-warming-convection cycle could possibly lift BC to even higher altitudes. In addition to such a 'self-lifting', it was seen in a separate model simulation done for the monsoonal month of July that, such high altitude BC were transported vertically up by the underlying strong convection over the Indian region. The transport was seen to be as high as up to 17.6 km i.e. the UTLS region. Many previous studies have reported that the near-tropopause conditions over the Indian region especially during monsoonal months are more favorable for the cross-tropopause transport of the tropospheric air-mass. Thus this could act as a pathway for the lifted aircraft-emitted BC layers to reach the stratosphere. Many previous studies have also reported presence of BC in stratosphere. To understand this further, the vertical profiles of aerosol extinction coefficient from satellite measurements over the stratospheric heights across the Indian region were examined. The extinction coefficient values as high as 0.02 km^{-1} , which were an order of magnitude greater than the average tropical stratospheric aerosol extinction coefficients were noticed. Though such layers were more visible during the monsoonal months, their presence during winters hinted at the possible co-operation between self-lifting and large-scale ascent over Indian region, in bringing the layers in the UTLS region. On the further examination, it was found that the particle depolarization ratio corresponding to the high values extinction coefficient were higher than 0.3. Such high values of PDR signify non-spherical nature of the involved species. Eliminating the possibility of mineral dust particles and ice crystals being responsible for such high values of PDR and finding from previous studies about the presence of non-spherical BC chain agglomerates in the UTLS region, it was concluded that such BC structures would possibly be responsible for the high values of PDR over the stratospheric altitudes. Once in stratosphere, BC can reside for a longer period of time due to the inherent stability of stratosphere and the absence of strong removal mechanisms. One of the possible implications of presence of BC in stratosphere is the reaction involving decomposition of ozone on BC, which would result in depletion of ozone. So potentially, the lifted upper tropospheric BC in stratosphere could harm the earth's protective blanket. Thus, while aircrafts look to be the cause for the sharp and confined high altitude layers of BC, such layers when lifted to stratosphere (under favorable conditions), can potentially affect the ozone layer and can have significant implications for health of all living organisms. Realizing the potential of emissions from aircrafts, a recent article (Editorial, 2016) also advocates for regulations on aircraft emissions. More observational studies using satellite and stratospheric balloons, and modeling studies are required to address this important phenomena especially over regions of high aircraft activity.

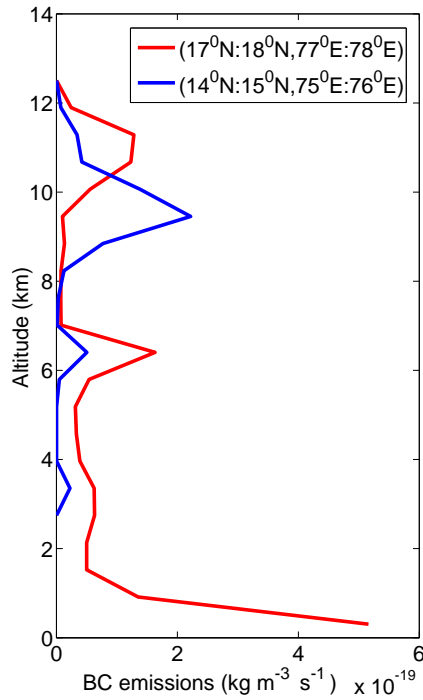


Figure 1. The vertical profile of BC emissions ($\text{kg m}^{-3} \text{s}^{-1}$) from aircrafts in the MACCity inventory over $1^\circ \times 1^\circ$ grid boxes a) centered over Hyderabad (red line) b) located 2° south-west of Hyderabad (blue line) during the month of March, 2010.

~~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 (a). during 17 March 2010 (b). during 8 January 2011 (c). during 25 April 2011. The locations corresponding to high altitude BC peaks in fig.?? are identified in profile by letters P1 to P5.~~

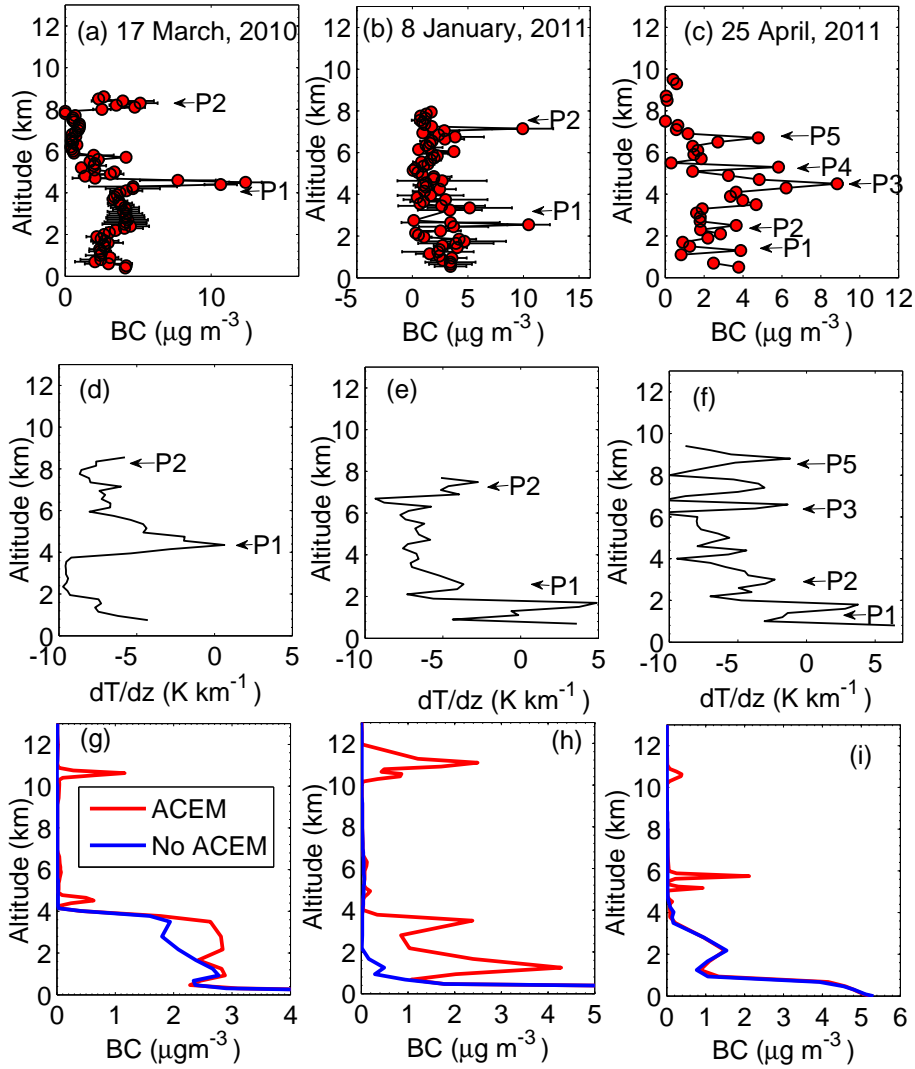


Figure 2. Observed vertical profile of BC over Hyderabad obtained from balloon measurements (a). during 17 March 2010 (b). during 8 January 2011 (c). during 25 April 2011. The high altitude BC peaks in each profile are identified by letters P1 to P5. Observed vertical profile of dT/dz ($K km^{-1}$) over 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.

-Model domain, Hyderabad is marked with a white dot. The blue area in the map indicates a part of Bay of Bengal.

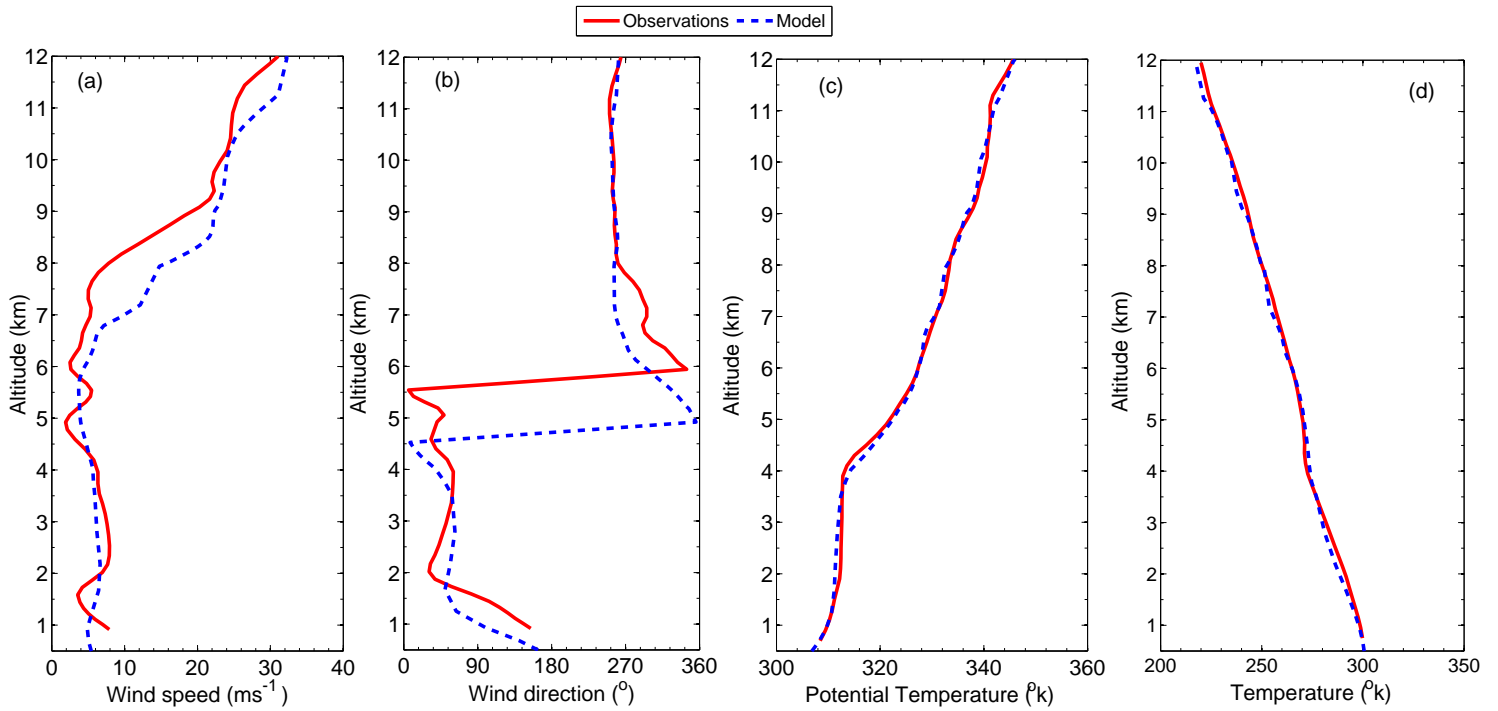


Figure 3. Comparison between the model simulated meteorological variables in NoACEM/Ctrl (control) run and the corresponding observations over the balloon flight region on 17, March 2010 for (a) wind speed (b) wind direction (c) Potential temperature (d) Temperature

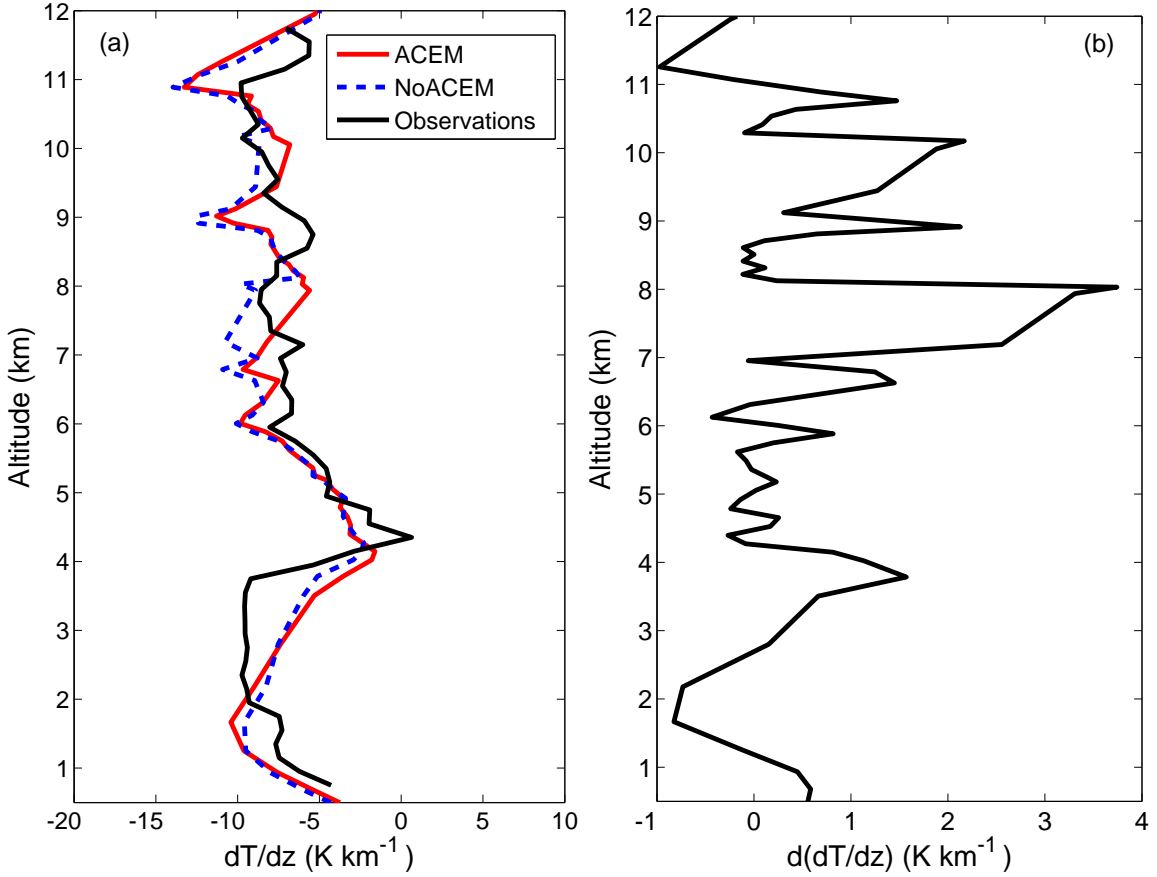


Figure 4. ~~Model simulated vertical~~ (a) Vertical profile of ~~BC~~ dT/dz (K km⁻¹) over the balloon flight region ~~in the on 17, March 2010 from:~~ (black-line)- Observations, (blue dashed line)- Simulations without BC emissions from aircrafts (NoACEM/Ctrl) and (control red line) ~~run~~ ~~during~~ Simulations with BC emissions from aircrafts (eACEM). ~~17 March 2010,~~ (b) ~~8 January 2011 and~~ Vertical profile of difference in dT/dz values for 2 model simulations, ~~(e~~ $d(dT/dz)$. ~~25 April 2011,~~ $= dT/dz)_{ACEM} - (dT/dz)_{NoACEM}$.

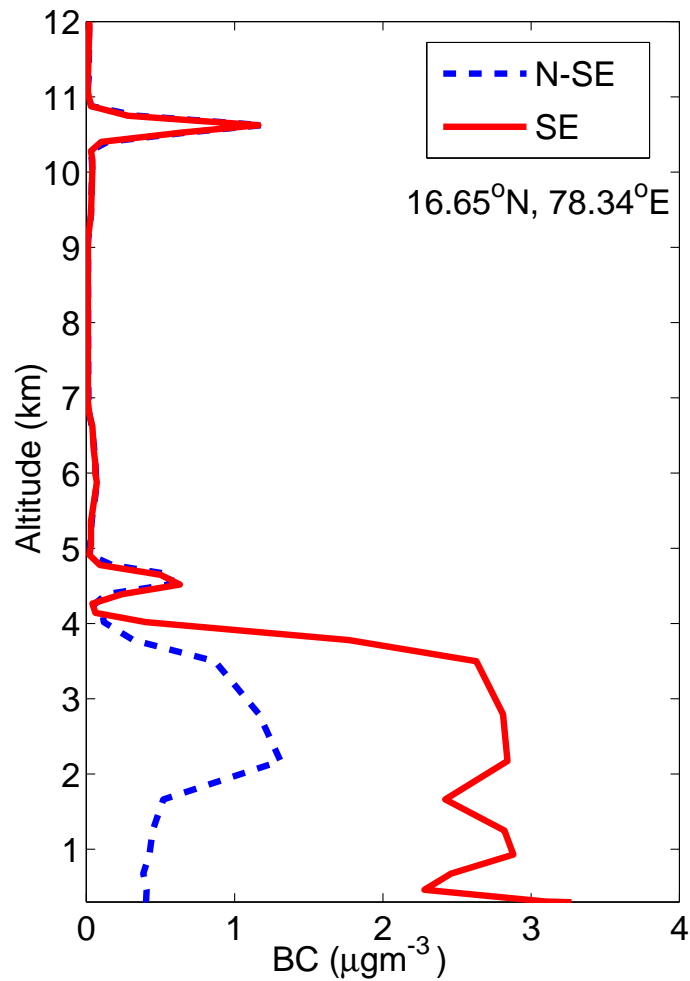


Figure 5. Model simulated vertical profile of BC over the area in the vicinity of the balloon flight region during 17 March 2010 for (blue line) NoACEM/Ctrl (control run) and (red line) ACEM (N-SE runs with (model simulations without prescription of aircraft surface level anthropogenic BC emissions) during (a)missions) -17 March 2010, (b). 8 January 2011 and SE runs (model simulations with prescription of surface level anthropogenic BC emissions). 25 April 2011.

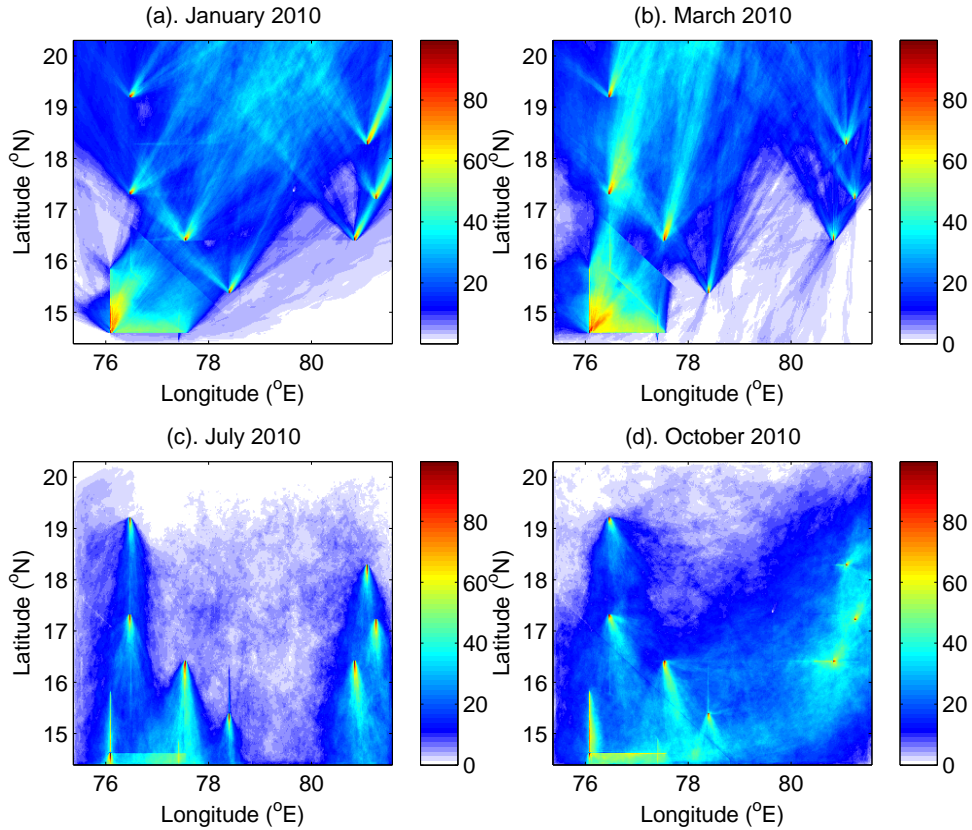


Figure 6. Spatial plot of probability (%) of capturing a peak in simulated BC within 9-11km for the model simulations carried out during a). January 2010, b). March 2010, c). July, 2010 and d). October 2010

- (a) Vertical profile of dT/dz ($K\ km^{-1}$) over the balloon flight region on 17, March 2010 from: (black line) Observations, (blue dashed line) Simulations without BC emissions from aircrafts (NoACEM) and (red line) Simulations with BC emissions from aircrafts (ACEM).
- (b) Vertical profile of difference in dT/dz values for 2 model simulations, $d(dT/dz) = dT/dz_{ACEM} - dT/dz_{NoACEM}$

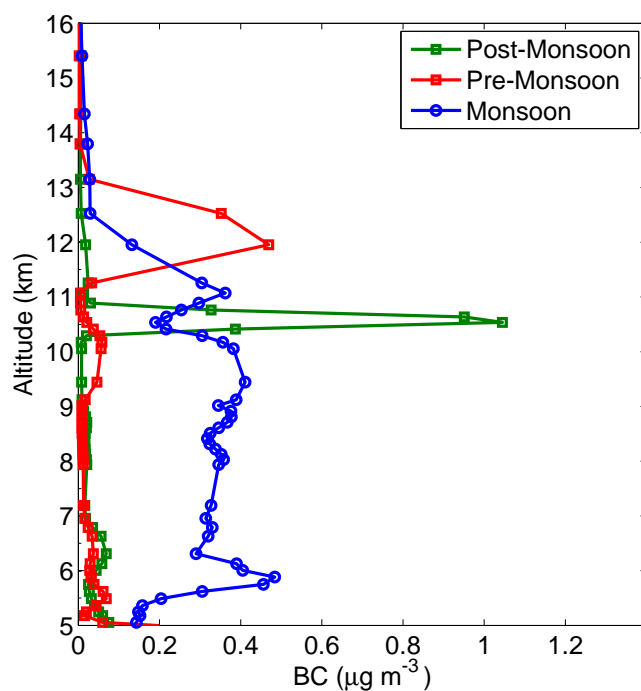


Figure 7. The model simulated vertical profile of BC mass concentration ($\mu\text{g m}^{-3}$) in the vicinity of balloon flight domain during different seasons, Pre-Monsoon (red line), Monsoon (blue line) and Post-Monsoon (green line), of the year 2010.

~~Model simulated vertical profile of BC during 17 March 2010 for (a). N-SE runs (model simulations without prescription of surface level anthropogenic BC emissions) (b). SE runs (model simulations with prescription of surface level anthropogenic BC emissions).~~

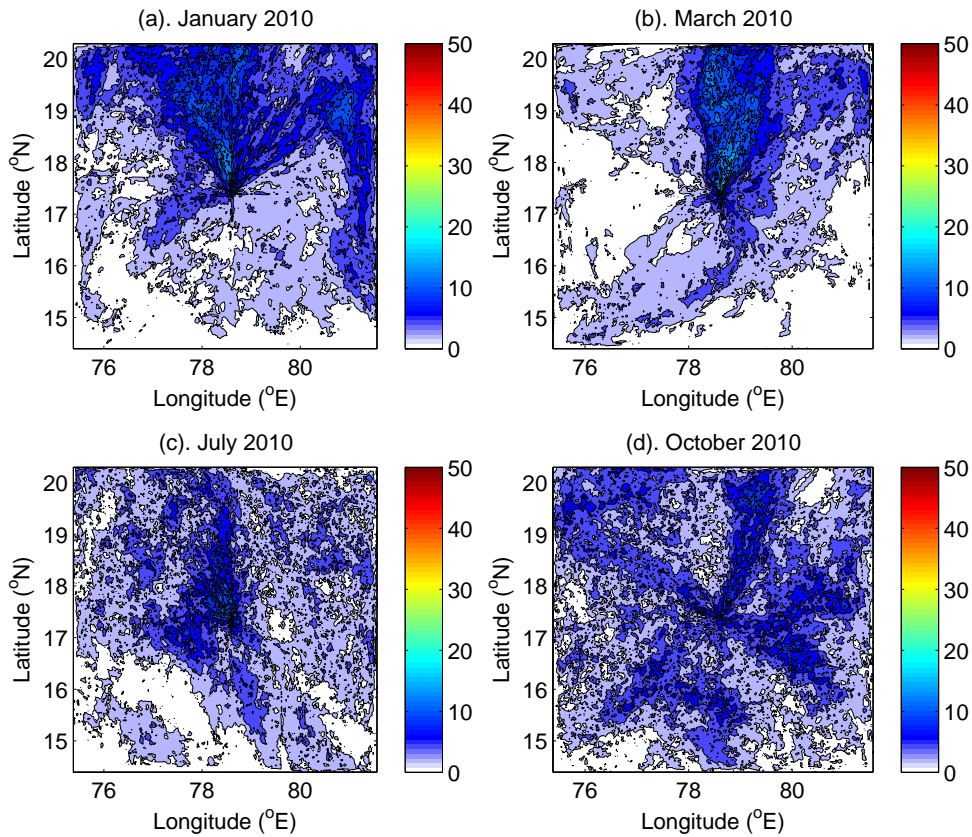


Figure 8. Spatial plot of probability (%) of capturing a peak in simulated BC within 4-5km for the model simulations carried out during a). January 2010, b). March 2010, c). July, 2010 and d). October 2010

~~Vertical profiles of atmospheric heating rates for the observed BC profiles during (a). March 2010, (b). January 2011 and (c). April 2011.~~

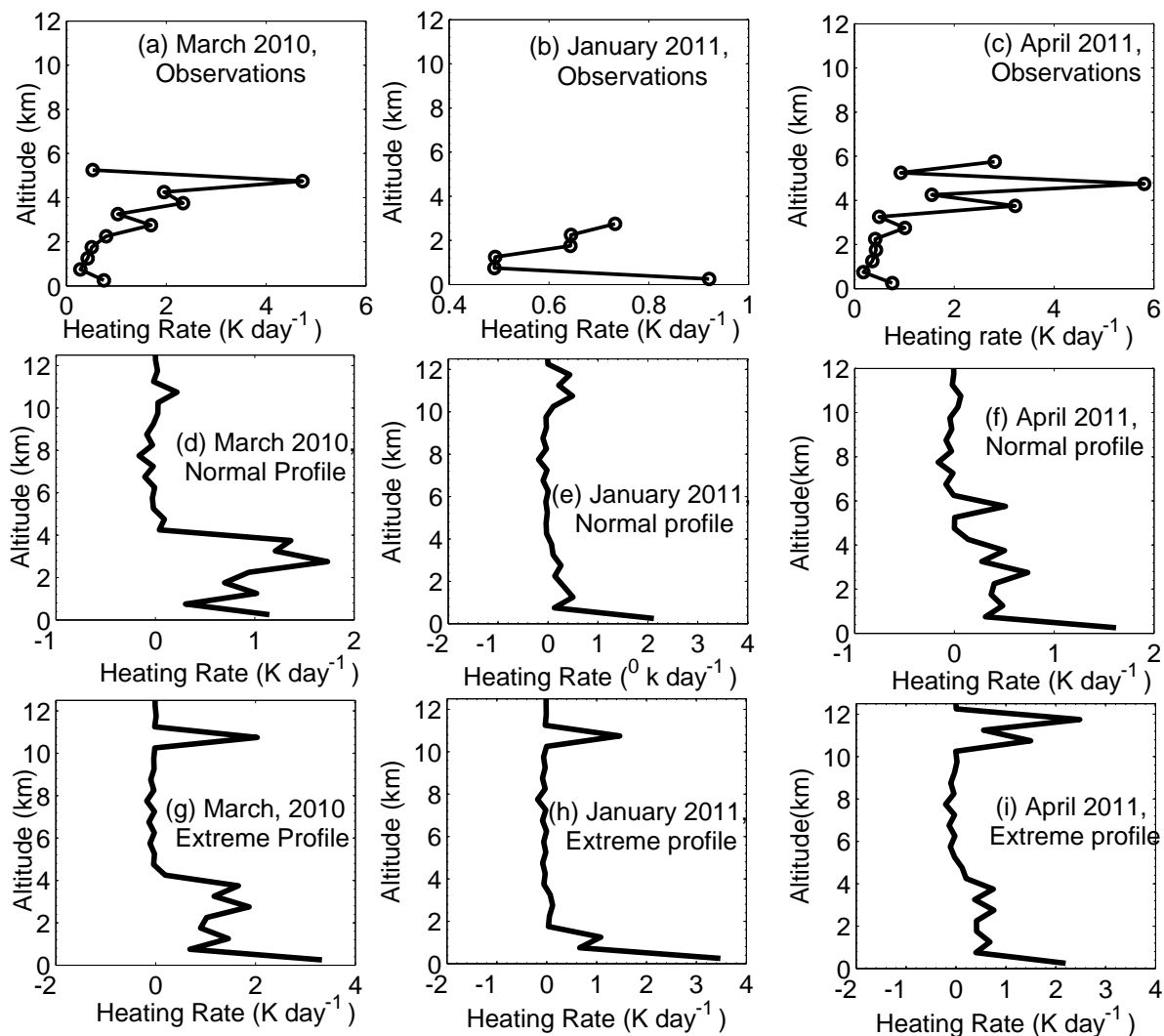


Figure 9. Vertical profiles of atmospheric heating rates for the observed BC profiles during (a).March 2010, (b). January 2011 and (c). April 2011. Vertical profiles of atmospheric heating rates corresponding to the observed BC profiles during (a).March 2010, (b). January 2011 and (c). April 2011. Vertical 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.

Vertical profiles of atmospheric heating rates for the model simulated BC profiles during (a and d). 17 March 2010 (b and c). 8 January 2011 (e and f). 25 April 2011. Please refer section 3.7.2 for the meaning of the terms ‘normal profile’ and ‘extreme profile’.

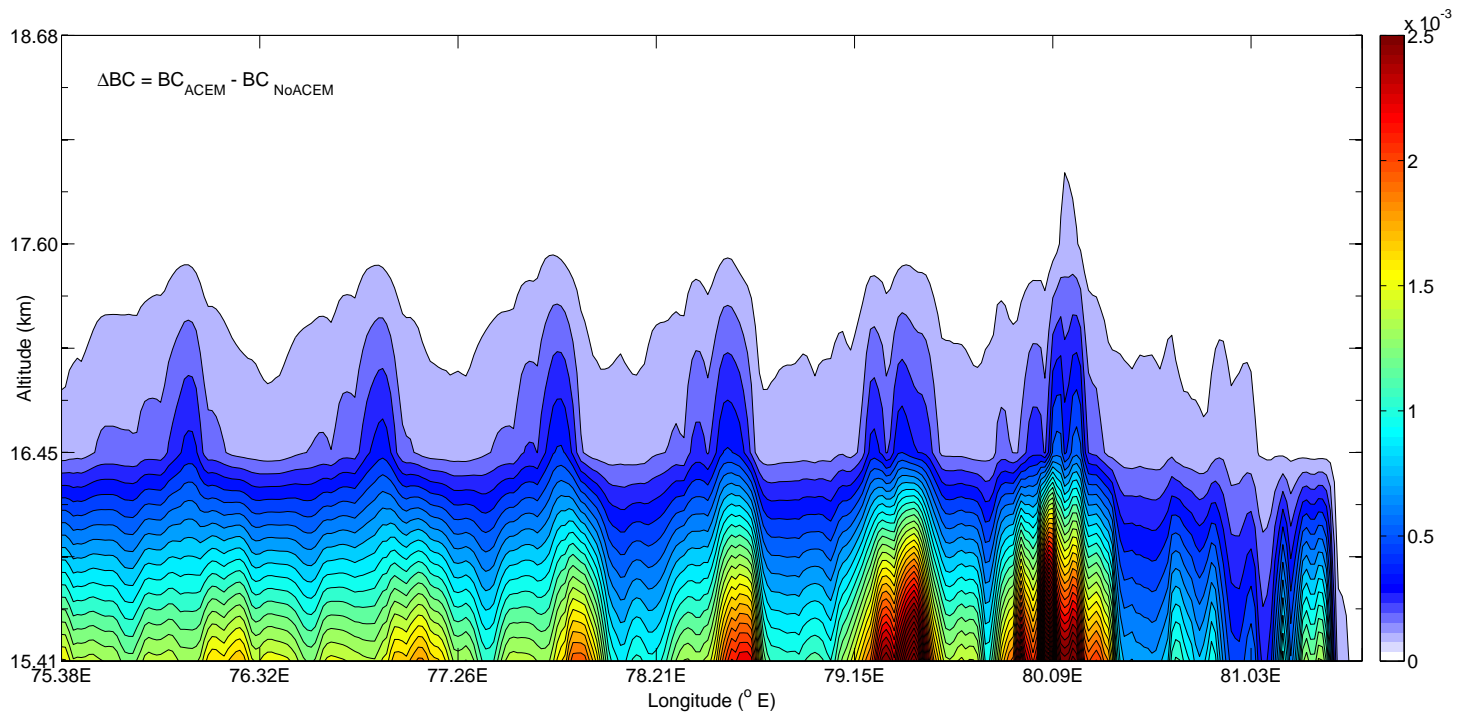


Figure 10. Vertical profile of the difference in BC mass concentration ($\mu\text{g m}^{-3}$) in ACEM runs vis-a-vis NoACEM run, averaged for the entire duration of the model simulation, carried out during July 2010. Here, $\Delta BC = BC_{ACEM} - BC_{NoACEM}$.

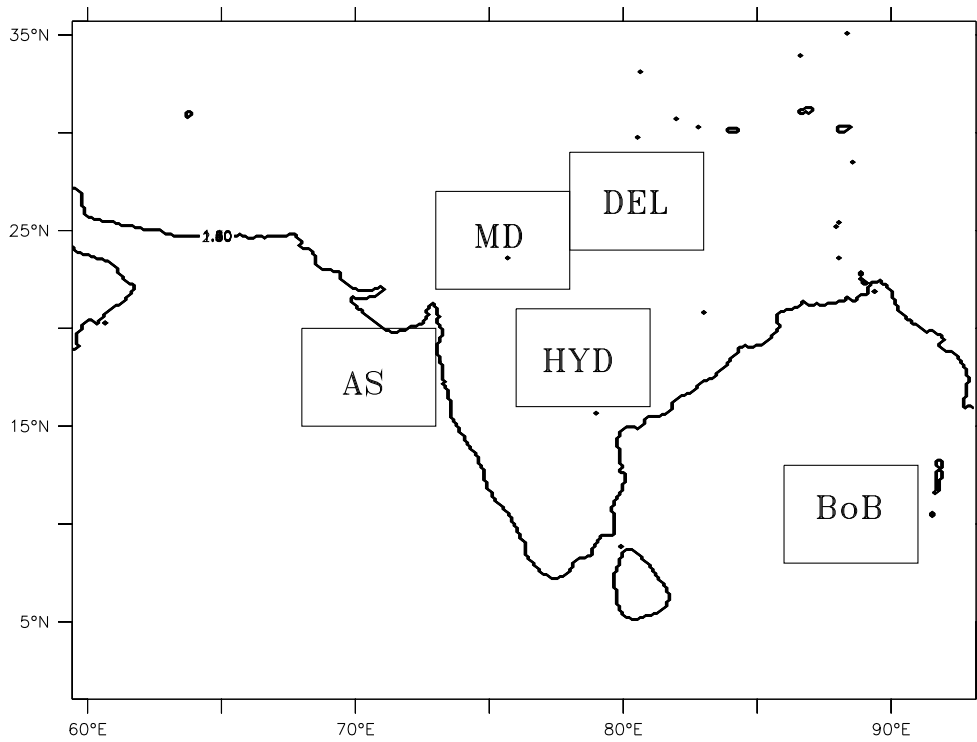


Figure 11. $5^0 \times 5^0$ regional boxes considered for the CALIPSO analysis (AS-Arabian sea, BoB-Bay of Bengal, HYD- Hyderabad, MD- Mumbai-Delhi and DEL-Delhi).

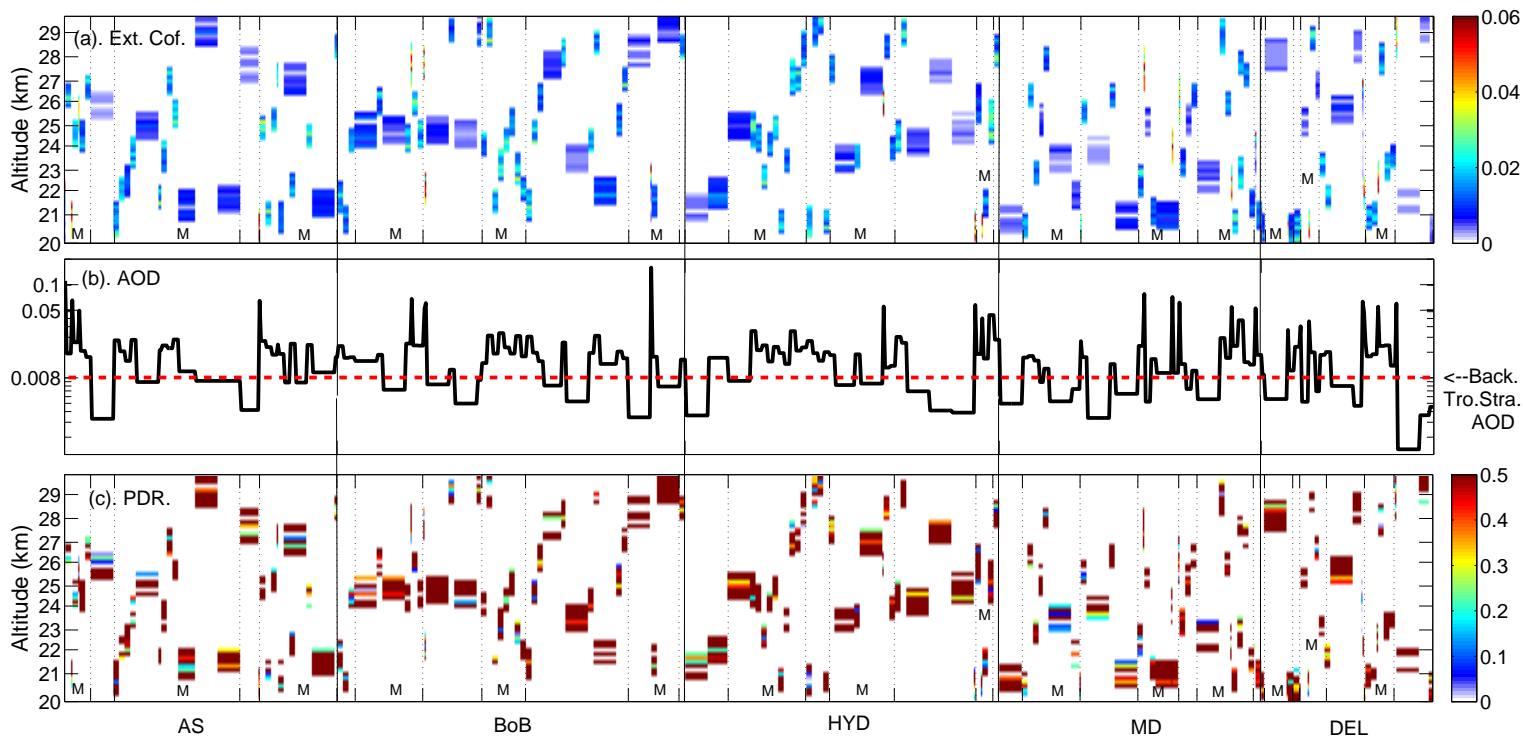


Figure 12. Time series of vertical profile of (a). Extinction coefficient over 5 regions (AS-Arabian sea, BoB- Bay of Bengal, HYD- Hyderabad, MD- Mumbai-Delhi and DEL-Delhi) over and around India (b). Corresponding stratospheric (from 20 km to ~30 km) AOD, the background stratospheric AOD for the entire tropical belt (Kremser et al., 2016) is shown by the dotted red line (c). Particle Depolarization Ratio (PDR) over the same 5 regions from January 2010 to December 2012. The individual time series of Ext. Cof., AOD and PDR. of each region are appended together in the figure one after the other. The letter M signifies the Monsoon (JJAS) season during a year under consideration

6 Code availability

The simulations are done using an open source online regional chemistry transport model WRF-Chem, which is freely available at <http://www2.mmm.ucar.edu/wrf/users/>.

7 Data availability

- 5 The extinction coefficient level 2 data from CALIPSO is available freely on https://eosweb.larc.nasa.gov/HBDOCS/langley_web_tool.html.

Competing interests. The authors declare that they have no conflict of interest

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References

- Ackerman, A. S., Toon, O. B., Stevens, D. E., Heymsfield, A. J., Ramanathan, V., and Welton, E. J.: Reduction of Tropical Cloudiness by Soot, *Science*, 288, 1042–1047, doi:10.1126/science.288.5468.1042, <http://science.sciencemag.org/content/288/5468/1042>, 2000.
- Akhter, M. S., Chughtai, A. R., and Smith, D. M.: The Structure of Hexane Soot I: Spectroscopic Studies, *Applied Spectroscopy*, 39, 143–153, doi:10.1366/0003702854249114, <http://asp.sagepub.com/content/39/1/143.abstract>, 1985.
- Andreae, M. O., Artaxo, P., Fischer, H., Freitas, S. R., Grégoire, J.-M., Hansel, A., Hoor, P., Kormann, R., Krejci, R., Lange, L., Lelieveld, J., Lindinger, W., Longo, K., Peters, W., de Reus, M., Scheeren, B., Silva Dias, M. A. F., Ström, J., van Velthoven, P. F. J., and Williams, J.: Transport of biomass burning smoke to the upper troposphere by deep convection in the equatorial region, *Geophysical Research Letters*, 28, 951–954, doi:10.1029/2000GL012391, <http://dx.doi.org/10.1029/2000GL012391>, 2001.
- 10 Arkoudeas, P., Kalligeros, S., Zannikos, F., Anastopoulos, G., Karonis, D., Korres, D., and Lois, E.: Study of using JP-8 aviation fuel and biodiesel in {CI} engines, *Energy Conversion and Management*, 44, 1013 – 1025, doi:[https://doi.org/10.1016/S0196-8904\(02\)00112-7](https://doi.org/10.1016/S0196-8904(02)00112-7), <http://www.sciencedirect.com/science/article/pii/S0196890402001127>, 2003.
- Arnott, W. P., Hamasha, K., Moosmüller, H., Sheridan, P. J., and Ogren, J. A.: Towards Aerosol Light-Absorption Measurements with a 7-Wavelength Aethalometer: Evaluation with a Photoacoustic Instrument and 3-Wavelength Nephelometer, *Aerosol Science and Technology*, 39, 17–29, doi:10.1080/027868290901972, <http://dx.doi.org/10.1080/027868290901972>, 2005.
- 15 Babu, S. S., Satheesh, S. K., and Moorthy, K. K.: Aerosol radiative forcing due to enhanced black carbon at an urban site in India, *Geophysical Research Letters*, 29, 27–1–27–4, doi:10.1029/2002GL015826, <http://dx.doi.org/10.1029/2002GL015826>, 1880, 2002.
- Babu, S. S., Satheesh, S. K., Moorthy, K. K., Dutt, C. B. S., Nair, V. S., Alappattu, D. P., and Kunhikrishnan, P. K.: Aircraft measurements of aerosol black carbon from a coastal location in the north-east part of peninsular India during ICARB, *Journal of Earth System Science*, 20 117, 263–271, doi:10.1007/s12040-008-0030-1, <http://dx.doi.org/10.1007/s12040-008-0030-1>, 2008.
- Babu, S. S., Moorthy, K., and Satheesh, S.: Vertical and Horizontal Gradients in Aerosol Black Carbon and Its Mass Fraction to Composite Aerosols over the East Coast of Peninsular India from Aircraft Measurements, *Advances in Meteorology*, 2010, doi:10.1155/2010/812075, <http://dx.doi.org/10.1155/2010/812075>, 812075, 2010.
- Babu, S. S., Moorthy, K. K., Manchanda, R. K., Sinha, P. R., Satheesh, S. K., Vajja, D. P., Srinivasan, S., and Kumar, V. H. A.: Free tropospheric black carbon aerosol measurements using high altitude balloon: Do BC layers build “their own homes” up in the atmosphere?, *Geophysical Research Letters*, 38, doi:10.1029/2011GL046654, <http://dx.doi.org/10.1029/2011GL046654>, 108803, 2011.
- Baumgardner, D., Kok, G., and Raga, G.: Warming of the Arctic lower stratosphere by light absorbing particles, *Geophysical Research Letters*, 31, doi:10.1029/2003GL018883, <http://dx.doi.org/10.1029/2003GL018883>, 106117, 2004.
- Bekki, S.: On the possible role of aircraft-generated soot in the middle latitude ozone depletion, *Journal of Geophysical Research: Atmospheres*, 102, 10 751–10 758, doi:10.1029/97JD00134, <http://dx.doi.org/10.1029/97JD00134>, 1997.
- 30 Blake, D. F. and Kato, K.: Latitudinal distribution of black carbon soot in the upper troposphere and lower stratosphere, *Journal of Geophysical Research: Atmospheres*, 100, 7195–7202, doi:10.1029/94JD03118, <http://dx.doi.org/10.1029/94JD03118>, 1995.
- Blakey, S., Rye, L., and Wilson, C. W.: Aviation gas turbine alternative fuels: A review, *Proceedings of the Combustion Institute*, 33, 2863 – 2885, doi:<https://doi.org/10.1016/j.proci.2010.09.011>, <http://www.sciencedirect.com/science/article/pii/S1540748910003950>, 2011.
- 35 Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T.,

- Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, *Journal of Geophysical Research: Atmospheres*, 118, 5380–5552, doi:10.1002/jgrd.50171, <http://dx.doi.org/10.1002/jgrd.50171>, 2013.
- Boucher, O., Randall, D., Artaxo, P., Bretherton, C., G., F., P., F., V.-M., K., Y., K., H., L., U., L., P., R., S.K., S., S., S., B., S., and X.Y., Z.: Clouds and Aerosols. In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)], Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 571–658, doi:10.1017/CBO9781107415324.016, 2013.
- Bourassa, A. E., Degenstein, D. A., Randel, W. J., Zawodny, J. M., Kyrölä, E., McLinden, C. A., Sioris, C. E., and Roth, C. Z.: Trends in stratospheric ozone derived from merged SAGE II and Odin-OSIRIS satellite observations, *Atmospheric Chemistry and Physics*, 14, 6983–6994, doi:10.5194/acp-14-6983-2014, <http://www.atmos-chem-phys.net/14/6983/2014/>, 2014.
- Chakraborty, A., Satheesh, S. K., Nanjundiah, R. S., and Srinivasan, J.: Impact of absorbing aerosols on the simulation of climate over the Indian region in an atmospheric general circulation model, *Annales Geophysicae*, 22, 1421–1434, doi:10.5194/angeo-22-1421-2004, 2004.
- Chand, D., Schmid, O., Gwaze, P., Parmar, R. S., Helas, G., Zeromskiene, K., Wiedensohler, A., Massling, A., and Andreae, M. O.: Laboratory measurements of smoke optical properties from the burning of Indonesian peat and other types of biomass, *Geophysical Research Letters*, 32, doi:10.1029/2005GL022678, <http://dx.doi.org/10.1029/2005GL022678>, 112819, 2005.
- Chand, D., Wood, R., Anderson, T. L., Satheesh, S. K., and Charlson, R. J.: Satellite-derived direct radiative effect of aerosols dependent on cloud cover, *Nature Geosci*, 2, 181–184, doi:10.1038/ngeo437, http://www.nature.com/ngeo/journal/v2/n3/supinfo/ngeo437_S1.html, 2009.
- Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B. N., Duncan, B. N., Martin, R. V., Logan, J. A., Higurashi, A., and Nakajima, T.: Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and Sun photometer measurements, *Journal of the Atmospheric Sciences*, 59, 461–483, doi:10.1175/1520-0469(2002)059<0461:TAOTFT>2.0.CO;2, 2002.
- Chuan, R. L. and Woods, D. C.: The appearance of carbon aerosol oarticles in the lower stratosphere, *Geophysical Research Letters*, 11, 553–556, doi:10.1029/GL011i005p00553, <http://dx.doi.org/10.1029/GL011i005p00553>, 1984.
- Chughtai, A. R., Kim, J. M., and Smith, D. M.: The Effect of Temperature and Humidity on the Reaction of Ozone with Combustion Soot: Implications for Reactivity near the Tropopause, *Journal of Atmospheric Chemistry*, 45, 231–243, doi:10.1023/A:1024250505886, <http://dx.doi.org/10.1023/A:1024250505886>, 2003.
- Cookson, D. J. and Smith, B. E.: Calculation of jet and diesel fuel properties using carbon-13 NMR spectroscopy, *Energy & Fuels*, 4, 152–156, doi:10.1021/ef00020a004, <http://dx.doi.org/10.1021/ef00020a004>, 1990.
- Corr, C. A., Ziemba, L. D., Scheuer, E., Anderson, B. E., Beyersdorf, A. J., Chen, G., Crosbie, E., Moore, R. H., Shook, M., Thornhill, K. L., Winstead, E., Lawson, R. P., Barth, M. C., Schroeder, J. R., Blake, D. R., and Dibb, J. E.: Observational evidence for the convective transport of dust over the Central United States, *Journal of Geophysical Research: Atmospheres*, 121, 1306–1319, doi:10.1002/2015JD023789, <http://dx.doi.org/10.1002/2015JD023789>, 2015JD023789, 2016.
- Corrigan, C. E., Ramanathan, V., and Schauer, J. J.: Impact of monsoon transitions on the physical and optical properties of aerosols, *Journal of Geophysical Research: Atmospheres*, 111, n/a–n/a, doi:10.1029/2005JD006370, <http://dx.doi.org/10.1029/2005JD006370>, d18208, 2006.

- Das, S. S., Venkat Ratnam, M., Uma, K. N., Patra, A. K., Subrahmanyam, K. V., Girach, I. A., Suneeth, K. V., Kumar, K. K., and Ramkumar, G.: Stratospheric intrusion into the troposphere during the tropical cyclone Nilam (2012), *Quarterly Journal of the Royal Meteorological Society*, 142, 2168–2179, doi:10.1002/qj.2810, <http://dx.doi.org/10.1002/qj.2810>, 2016.
- de Laat, A. T. J., Stein Zweers, D. C., Boers, R., and Tuinder, O. N. E.: A solar escalator: Observational evidence of the self-lifting of smoke and aerosols by absorption of solar radiation in the February 2009 Australian Black Saturday plume, *Journal of Geophysical Research: Atmospheres*, 117, doi:10.1029/2011JD017016, <http://dx.doi.org/10.1029/2011JD017016>, d04204, 2012.
- Disselkamp, R. S., Carpenter, M. A., Cowin, J. P., Berkowitz, C. M., Chapman, E. G., Zaveri, R. A., and Laulainen, N. S.: Ozone loss in soot aerosols, *Journal of Geophysical Research: Atmospheres*, 105, 9767–9771, doi:10.1029/1999JD901189, <http://dx.doi.org/10.1029/1999JD901189>, 2000.
- Dudhia, J.: Numerical Study of Convection Observed during the Winter Monsoon Experiment Using a Mesoscale Two-Dimensional Model, *Journal of Atmospheric Science*, 105, 3077–3107, 1989.
- Dusek, U., Frank, G. P., Helas, G., Iinuma, Y., Zeromskiene, K., Gwaze, P., Hennig, T., Massling, A., Schmid, O., Herrmann, H., Wiedensohler, A., and Andreae, M. O.: “Missing” cloud condensation nuclei in peat smoke, *Geophysical Research Letters*, 32, doi:10.1029/2005GL022473, <http://dx.doi.org/10.1029/2005GL022473>, l11802, 2005.
- Editorial: Green-sky thinking, *Nature*, 530, 253–253, doi:10.1038/530253a, <http://www.nature.com/news/green-sky-thinking-1.19376>, 2016.
- Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geoscientific Model Development*, 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010.
- Fadnavis, S., Semeniuk, K., Pozzoli, L., Schultz, M. G., Ghude, S. D., Das, S., and Kakatkar, R.: Transport of aerosols into the UTLS and their impact on the Asian monsoon region as seen in a global model simulation, *Atmospheric Chemistry and Physics*, 13, 8771–8786, doi:10.5194/acp-13-8771-2013, <http://www.atmos-chem-phys.net/13/8771/2013/>, 2013.
- Fan, J., Zhang, R., Tao, W.-K., and Mohr, K. I.: Effects of aerosol optical properties on deep convective clouds and radiative forcing, *Journal of Geophysical Research: Atmospheres*, 113, n/a–n/a, doi:10.1029/2007JD009257, <http://dx.doi.org/10.1029/2007JD009257>, d08209, 2008.
- Freitas, S.: Including the sub-grid scale plume rise of vegetation fires in low resolution atmospheric transport models, *Atmospheric Chemistry Physics*, 7, 3385–3398, 2007.
- Freitas, S. R., Longo, K. M., Alonso, M. F., Pirre, M., Marecal, V., Grell, G., Stockler, R., Mello, R. F., and Sánchez Gácita, M.: PREP-CHEM-SRC- 1.0: a preprocessor of trace gas and aerosol emission fields for regional and global atmospheric chemistry models, *Geoscientific Model Development*, 4, 419–433, doi:10.5194/gmd-4-419-2011, <http://www.geosci-model-dev.net/4/419/2011/>, 2011.
- Fromm, M., Bevilacqua, R., Servranckx, R., Rosen, J., Thayer, J. P., Herman, J., and Larko, D.: Pyro-cumulonimbus injection of smoke to the stratosphere: Observations and impact of a super blowup in northwestern Canada on 3–4 August 1998, *Journal of Geophysical Research: Atmospheres*, 110, doi:10.1029/2004JD005350, <http://dx.doi.org/10.1029/2004JD005350>, d08205, 2005.
- Fu, R., Hu, Y., Wright, J. S., Jiang, J. H., Dickinson, R. E., Chen, M., Filipiak, M., Read, W. G., Waters, J. W., and Wu, D. L.: Short circuit of water vapor and polluted air to the global stratosphere by convective transport over the Tibetan Plateau, *Proceedings of the National Academy of Sciences*, 103, 5664–5669, doi:10.1073/pnas.0601584103, <http://www.pnas.org/content/103/15/5664.abstract>, 2006.
- Fueglistaler, S., Dessler, A. E., Dunkerton, T. J., Folkins, I., Fu, Q., and Mote, P. W.: Tropical tropopause layer, *Reviews of Geophysics*, 47, doi:10.1029/2008RG000267, <http://dx.doi.org/10.1029/2008RG000267>, rG1004, 2009.
- Gautam, R., Hsu, N. C., Lau, W. K.-M., and Yasunari, T. J.: Satellite observations of desert dust-induced Himalayan snow darkening, *Geophysical Research Letters*, 40, 988–993, doi:10.1002/grl.50226, <http://dx.doi.org/10.1002/grl.50226>, 2013.

- Gorkavyi, N., Rault, D. F., Newman, P. A., da Silva, A. M., and Dudorov, A. E.: New stratospheric dust belt due to the Chelyabinsk bolide, *Geophysical Research Letters*, 40, 4728–4733, doi:10.1002/grl.50788, <http://dx.doi.org/10.1002/grl.50788>, 2013.
- Govardhan, G., Nanjundiah, R. S., Satheesh, S. K., Krishnamoorthy, K., and Kotamarthi, V. R.: Performance of WRF-Chem over Indian region: Comparison with measurements, *Journal of Earth System Science*, 124, 875–896, doi:10.1007/s12040-015-0576-7, <http://dx.doi.org/10.1007/s12040-015-0576-7>, 2015.
- Govardhan, G. R., Nanjundiah, R. S., Satheesh, S., Moorthy, K. K., and Takemura, T.: Inter-comparison and performance evaluation of chemistry transport models over Indian region, *Atmospheric Environment*, 125, Part B, 486 – 504, doi:<http://dx.doi.org/10.1016/j.atmosenv.2015.10.065>, <http://www.sciencedirect.com/science/article/pii/S1352231015304829>, south Asian Aerosols And Anthropogenic Emissions: Regional And Global Climate Implications, 2016.
- Hansen, A.: Magee Scientific Aethalometer User's Guide, Magee Sci., Berkeley, California, 56pp, 1996.
- Hansen, A.: The Aethalometer, Magee Sci., Berkeley, California, 2005.
- Haywood, J. M. and Shine, K. P.: The effect of anthropogenic sulfate and soot aerosol on the clear sky planetary radiation budget, *Geophysical Research Letters*, 22, 603–606, doi:10.1029/95GL00075, <http://dx.doi.org/10.1029/95GL00075>, 1995.
- Herndon, S. C., Jayne, J. T., Lobo, P., Onasch, T. B., Fleming, G., Hagen, D. E., Whitefield, P. D., and Miake-Lye, R. C.: Commercial Aircraft Engine Emissions Characterization of in-Use Aircraft at Hartsfield-Jackson Atlanta International Airport, *Environmental Science & Technology*, 42, 1877–1883, doi:10.1021/es072029+, <http://dx.doi.org/10.1021/es072029+>, PMID: 18409607, 2008.
- Hess, M., Koepke, P., and Schult, I.: Optical Properties of Aerosols and Clouds: The Software Package OPAC, *Bulletin of the American Meteorological Society*, 79, 831–844, doi:10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2, [http://dx.doi.org/10.1175/1520-0477\(1998\)079<0831:OPOAAC>2.0.CO;2](http://dx.doi.org/10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2), 1998.
- Hong, S., Noh, Y., and Dudhia, J.: A New Vertical Diffusion Package with an Explicit Treatment of Entrainment Processes, *Monthly Weather Review*, 134, 2318–2341, doi:10.1175/1520-0493(2004)132<0519:EFOWPU>2.0.CO;2, 2006.
- Hong, S.-Y.: A new stable boundary-layer mixing scheme and its impact on the simulated East Asian summer monsoon, *Quarterly Journal of the Royal Meteorological Society*, 136, 1481–1496, doi:10.1002/qj.665, <http://dx.doi.org/10.1002/qj.665>, 2010.
- Kamm, S., Möhler, O., Naumann, K.-H., Saathoff, H., and Schurath, U.: The heterogeneous reaction of ozone with soot aerosol, *Atmospheric Environment*, 33, 4651 – 4661, doi:[http://dx.doi.org/10.1016/S1352-2310\(99\)00235-6](http://dx.doi.org/10.1016/S1352-2310(99)00235-6), <http://www.sciencedirect.com/science/article/pii/S1352231099002356>, 1999.
- Kane, R.: Is ozone depletion really recovering?, *Journal of Atmospheric and Solar-Terrestrial Physics*, 70, 1455 – 1459, doi:<http://dx.doi.org/10.1016/j.jastp.2008.04.012>, <http://www.sciencedirect.com/science/article/pii/S1364682608001107>, 2008.
- Kane, R.: Ozone depletion, worst not yet over, *Indian J. Radio Space Phys.*, 38, 313–316, <http://nopr.niscair.res.in/handle/123456789/6898>, 2009.
- Klekociuk, A. R., Brown, P. G., Pack, D. W., ReVelle, D. O., Edwards, W. N., Spalding, R. E., Tagliaferri, E., Yoo, B. B., and Zagari, J.: Meteoritic dust from the atmospheric disintegration of a large meteoroid, *Nature*, 436, 1132–1135, doi:<http://dx.doi.org/10.1038/nature03881>, http://www.nature.com/nature/journal/v436/n7054/supinfo/nature03881_S1.html, 10.1038/nature03881, 2005.
- Kompalli, S., Babu, S., N.B. B., and K.K., M.: Spring-time enhancement in aerosol burden over a high-altitude location in western trans-Himalaya: results from long-term observations, *Current Science*, 111, 117 – 131, doi:10.18520/cs/v111/i1/117-131, <http://www.currentscience.ac.in/Volumes/111/01/0117.pdf>, 2016.
- Kremser, S., Thomason, L. W., von Hobe, M., Hermann, M., Deshler, T., Timmreck, C., Toohey, M., Stenke, A., Schwarz, J. P., Weigel, R., Fueglistaler, S., Prata, F. J., Vernier, J.-P., Schlager, H., Barnes, J. E., Antuña-Marrero, J.-C., Fairlie, D., Palm, M., Mahieu, E., Notholt,

- J., Rex, M., Bingen, C., Vanhellemont, F., Bourassa, A., Plane, J. M. C., Klocke, D., Carn, S. A., Clarisse, L., Trickl, T., Neely, R., James, A. D., Rieger, L., Wilson, J. C., and Meland, B.: Stratospheric aerosol—Observations, processes, and impact on climate, *Reviews of Geophysics*, 54, 278–335, doi:10.1002/2015RG000511, <http://dx.doi.org/10.1002/2015RG000511>, 2015RG000511, 2016.
- 5 Kuhlbusch, T. A. J., Andreae, M. O., Cachier, H., Goldammer, J. G., Lacaux, J.-P., Shea, R., and Crutzen, P. J.: Black carbon formation by savanna fires: Measurements and implications for the global carbon cycle, *Journal of Geophysical Research: Atmospheres*, 101, 23 651–23 665, doi:10.1029/95JD02199, <http://dx.doi.org/10.1029/95JD02199>, 1996.
- Kwan, I. and Rutherford, D.: Transatlantic airline fuel efficiency ranking, 2014, The International Council on Clean Transportation, <http://www.theicct.org/transatlantic-airline-efficiency-2014>, 2015.
- Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, *Atmospheric Chemistry and Physics*, 10, 7017–7039, doi:10.5194/acp-10-7017-2010, <http://www.atmos-chem-phys.net/10/7017/2010/>, 2010.
- 10 Lau, K., Kim, M., and Kim, K.: Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau, *Climate Dynamics*, 26, 855–864, doi:10.1007/s00382-006-0114-z, <http://dx.doi.org/10.1007/s00382-006-0114-z>, 2006.
- Lau, W. K. M., Kim, M.-K., Kim, K.-M., and Lee, W.-S.: Enhanced surface warming and accelerated snow melt in the Himalayas and Tibetan Plateau induced by absorbing aerosols, *Environmental Research Letters*, 5, 025 204, <http://stacks.iop.org/1748-9326/5/i=2/a=025204>, 2010.
- Liou, K.-N.: An introduction to atmospheric radiation, vol. 84, Academic press, 2002.
- 20 Mackinnon, I. D. R., McKay, D. S., Nace, G., and Isaacs, A. M.: Classification of the Johnson Space Center stratospheric dust collection, *Journal of Geophysical Research: Solid Earth*, 87, A413–A421, doi:10.1029/JB087iS01p0A413, <http://dx.doi.org/10.1029/JB087iS01p0A413>, 1982.
- Meenu, S., Rajeev, K., Parameswaran, K., and Nair, A. K. M.: Regional distribution of deep clouds and cloud top altitudes over the Indian subcontinent and the surrounding oceans, *Journal of Geophysical Research: Atmospheres*, 115, doi:10.1029/2009JD011802, <http://dx.doi.org/10.1029/2009JD011802>, d05205, 2010.
- 25 Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave, *Journal of Geophysical Research: Atmospheres*, 102, 16 663–16 682, doi:10.1029/97JD00237, 1997.
- Moorthy, K. K., Babu, S. S., Sunilkumar, S. V., Gupta, P. K., and Gera, B. S.: Altitude profiles of aerosol BC, derived from aircraft measurements over an inland urban location in India, *Geophysical Research Letters*, 31, doi:10.1029/2004GL021336, <http://dx.doi.org/10.1029/2004GL021336>, l22103, 2004.
- 30 Nair, V., Babu, S. S., Moorthy, K. K., Sharma, A., Marinoni, A., and ., A.: Black carbon aerosols over the Himalayas: direct and surface albedo forcing, *Tellus B*, 65, doi:<http://dx.doi.org/10.3402/tellusb.v65i0.19738>, <http://www.tellusb.net/index.php/tellusb/article/view/19738>, 2013.
- 35 Nair, V. S., Babu, S. S., Gogoi, M. M., and Moorthy, K. K.: Large-scale enhancement in aerosol absorption in the lower free troposphere over continental India during spring, *Geophysical Research Letters*, 43, 11,453–11,461, doi:10.1002/2016GL070669, <http://dx.doi.org/10.1002/2016GL070669>, 2016GL070669, 2016.

- Okada, K., Ikegami, M., Uchino, O., Nikaidou, Y., Zaizen, Y., Tsutsumi, Y., and Makino, Y.: Extremely high proportions of soot particles in the upper troposphere over Japan, *Geophysical Research Letters*, 19, 921–924, doi:10.1029/92GL00487, <http://dx.doi.org/10.1029/92GL00487>, 1992.
- Olivier, J. G. J., Bouwman, A., Berdowski, J., Veldt, C., Bloos, J., Visschedijk, A., Zandveld, P., Haverlag, J., et al.: Description of EDGAR
5 Version 2.0: A set of global emission inventories of greenhouse gases and ozone-depleting substances for all anthropogenic and most natural sources on a per country basis and on 1 degree x 1 degree grid, Rijksinstituut voor Volksgezondheid en Milieu RIVM, 1996.
- Onasch, T. B., Jayne, J. T., Herndon, S., Worsnop, D. R., Miake-Lye, R. C., Mortimer, I. P., and Anderson, B. E.: Chemical Properties of Aircraft Engine Particulate Exhaust Emissions, *Journal of Propulsion and Power*, 25, 1121–1137, doi:10.2514/1.36371, <http://dx.doi.org/10.2514/1.36371>, 2009.
- 10 Outcalt, S., Laesecke, A., and Freund, M. B.: Density and Speed of Sound Measurements of Jet A and S-8 Aviation Turbine Fuels, *Energy & Fuels*, 23, 1626–1633, doi:10.1021/ef800888q, <http://dx.doi.org/10.1021/ef800888q>, 2009.
- Paraschis, Y. and Gittens, A.: Annual Report 2013, The Voice of the World's Airports,, Airports Council International, <http://www.aci.aero/Publications/Full-Publications-Listing>, 2014.
- Petzold, A. and Schönlinner, M.: Multi-angle absorption photometry—a new method for the measurement of aerosol light absorption and atmospheric black carbon, *Journal of Aerosol Science*, 35, 421 – 441, doi:<http://dx.doi.org/10.1016/j.jaerosci.2003.09.005>, <http://www.sciencedirect.com/science/article/pii/S0021850203004403>, 2004.
- 15 Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S.-M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrl, C., Wiedensohler, A., and Zhang, X.-Y.: Recommendations for reporting "black carbon" measurements, *Atmospheric Chemistry and Physics*, 13, 8365–8379, doi:10.5194/acp-13-8365-2013, <http://www.atmos-chem-phys.net/13/8365/2013/>, 2013.
- 20 Piccolo, F. and Gittens, A.: Annual Report 2014, The Voice of the World's Airports,, Airports Council International, <http://www.aci.aero/Publications/Full-Publications-Listing>, 2015.
- Piccolo, F. and Gittens, A.: Annual Report 2015, The Voice of the World's Airports,, Airports Council International, <http://www.aci.aero/Publications/Full-Publications-Listing>, 2016.
- Prijith, S., Babu, S. S., Lakshmi, N., Satheesh, S., and Moorthy, K. K.: Meridional gradients in aerosol vertical distribution over Indian Mainland: Observations and model simulations, *Atmospheric Environment*, 125, Part B, 337 – 345, doi:<http://dx.doi.org/10.1016/j.atmosenv.2015.10.066>, <http://www.sciencedirect.com/science/article/pii/S1352231015304830>, south Asian Aerosols And Anthropogenic Emissions: Regional And Global Climate Implications, 2016.
- Pueschel, R. F., Boering, K. A., Verma, S., Howard, S. D., Ferry, G. V., Goodman, J., Allen, D. A., and Hamill, P.: Soot aerosol in the lower stratosphere: Pole-to-pole variability and contributions by aircraft, *Journal of Geophysical Research: Atmospheres*, 102, 13 113–13 118, doi:10.1029/96JD03061, <http://dx.doi.org/10.1029/96JD03061>, 1997.
- 30 Pusechel, R. F., Blake, D. F., Snetsinger, K. G., Hansen, A. D. A., Verma, S., and Kato, K.: Black carbon (soot) aerosol in the lower stratosphere and upper troposphere, *Geophysical Research Letters*, 19, 1659–1662, doi:10.1029/92GL01801, <http://dx.doi.org/10.1029/92GL01801>, 1992.
- Qian, Y., Flanner, M. G., Leung, L. R., and Wang, W.: Sensitivity studies on the impacts of Tibetan Plateau snowpack pollution on the Asian hydrological cycle and monsoon climate, *Atmospheric Chemistry and Physics*, 11, 1929–1948, doi:10.5194/acp-11-1929-2011, <http://www.atmos-chem-phys.net/11/1929/2011/>, 2011.
- Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon, *Nature Geosci*, 1, 221–227, doi:10.1038/ngeo156, <http://dx.doi.org/10.1038/ngeo156>, 2008.

- Ramanathan, V., Chung, C., Kim, D., Bettge, T., Buja, L., Kiehl, J., Washington, W., Fu, Q., Sikka, D., and Wild, M.: Atmospheric brown clouds: Impacts on South Asian climate and hydrological cycle, *Proceedings of the National Academy of Sciences of the United States of America*, 102, 5326–5333, doi:10.1073/pnas.0500656102, 2005.
- Ramanathan, V., Ramana, M. V., Roberts, G., Kim, D., Corrigan, C., Chung, C., and Winker, D.: Warming trends in Asia amplified by brown cloud solar absorption, *Nature*, 448, 575–578, doi:10.1038/nature06019, http://www.nature.com/nature/journal/v448/n7153/supinfo/nature06019_S1.html, 2007.
- Randel, W. J., Park, M., Emmons, L., Kinnison, D., Bernath, P., Walker, K. A., Boone, C., and Pumphrey, H.: Asian Monsoon Transport of Pollution to the Stratosphere, *Science*, 328, 611–613, doi:10.1126/science.1182274, <http://science.sciencemag.org/content/328/5978/611>, 2010.
- 10 Rao, T., Uma, K. N., Narayana Rao, D., and Fukao, S.: Understanding the transportation process of tropospheric air entering the stratosphere from direct vertical air motion measurements over Gadanki and Kototabang, *Geophysical Research Letters*, 35, doi:10.1029/2008GL034220, <http://dx.doi.org/10.1029/2008GL034220>, 115805, 2008.
- Ricchiuzzi, P., Yang, S., Gautier, C., and Sowle, D.: SBDART: A Research and Teaching Software Tool for Plane-Parallel Radiative Transfer in the Earth's Atmosphere, *Bulletin of the American Meteorological Society*, 79, 2101–2114, doi:10.1175/1520-0477(1998)079<2101:SARATS>2.0.CO;2, [http://dx.doi.org/10.1175/1520-0477\(1998\)079<2101:SARATS>2.0.CO;2](http://dx.doi.org/10.1175/1520-0477(1998)079<2101:SARATS>2.0.CO;2), 1998.
- 15 Satheesh, S. and Moorthy, K. K.: Radiative effects of natural aerosols: A review, *Atmospheric Environment*, 39, 2089 – 2110, doi:<http://dx.doi.org/10.1016/j.atmosenv.2004.12.029>, <http://www.sciencedirect.com/science/article/pii/S135223100500018X>, 2005.
- Satheesh, S. and Ramanathan, V.: Large differences in tropical aerosol forcing at the top of the atmosphere and Earth's surface, *Nature*, 405, doi:10.1038/35011039, <http://dx.doi.org/10.1038/35011039>, 2000.
- 20 Satheesh, S., Moorthy, K. K., and Srinivasan, J.: New Directions: Elevated layers of anthropogenic aerosols aggravate stratospheric ozone loss?, *Atmospheric Environment*, 79, 879 – 882, doi:<http://dx.doi.org/10.1016/j.atmosenv.2013.07.052>, <http://www.sciencedirect.com/science/article/pii/S1352231013005839>, 2013.
- Satheesh, S. K., Vinoj, V., and Moorthy, K. K.: Vertical distribution of aerosols over an urban continental site in India inferred using a micro pulse lidar, *Geophysical Research Letters*, 33, n/a–n/a, doi:10.1029/2006GL027729, <http://dx.doi.org/10.1029/2006GL027729>, 120816, 2006.
- 25 Satheesh, S. K., Moorthy, K. K., Babu, S. S., Vinoj, V., and Dutt, C. B. S.: Climate implications of large warming by elevated aerosol over India, *Geophysical Research Letters*, 35, n/a–n/a, doi:10.1029/2008GL034944, <http://dx.doi.org/10.1029/2008GL034944>, 119809, 2008.
- Schultz, M., Backman, L., Balkanski, Y., Bjoernsdalseter, S., Brand, R., Burrows, J., Dalsoeren, S., de Vasconcelos, M., Grodtmann, B., Hauglustaine, D., et al.: REanalysis of the TROpospheric chemical composition over the past 40 years (RETRO)—A long-term global modeling study of tropospheric chemistry, Final Report, Jülich/Hamburg, Germany, 2007.
- Schurath, U. and Naumann, K.: Heterogeneous processes involving atmospheric particulate matter, *Pure and Applied Chemistry*, 70, 1353–1361, doi:<https://doi.org/10.1351/pac199870071353>, 1998.
- Schwarz, J. P., Gao, R. S., Fahey, D. W., Thomson, D. S., Watts, L. A., Wilson, J. C., Reeves, J. M., Darbeheshti, M., Baumgardner, D. G., Kok, G. L., Chung, S. H., Schulz, M., Hendricks, J., Lauer, A., Kärcher, B., Slowik, J. G., Rosenlof, K. H., Thompson, T. L., Langford, A. O., Loewenstein, M., and Aikin, K. C.: Single-particle measurements of midlatitude black carbon and light-scattering aerosols from the boundary layer to the lower stratosphere, *Journal of Geophysical Research: Atmospheres*, 111, doi:10.1029/2006JD007076, <http://dx.doi.org/10.1029/2006JD007076>, d16207, 2006.
- 35

- Sheridan, P. J., Brock, C. A., and Wilson, J. C.: Aerosol particles in the upper troposphere and lower stratosphere: Elemental composition and morphology of individual particles in northern midlatitudes, *Geophysical Research Letters*, 21, 2587–2590, doi:10.1029/94GL01387, <http://dx.doi.org/10.1029/94GL01387>, 1994.
- Sheridan, P. J., Arnott, W. P., Ogren, J. A., Andrews, E., Atkinson, D. B., Covert, D. S., Moosmüller, H., Petzold, A., Schmid, B., Strawa, A. W., Varma, R., and Virkkula, A.: The Reno Aerosol Optics Study: An Evaluation of Aerosol Absorption Measurement Methods, *Aerosol Science and Technology*, 39, 1–16, doi:10.1080/027868290901891, <http://dx.doi.org/10.1080/027868290901891>, 2005.
- Smirnova, T. G., Brown, J., and M.Benjamin, S. G.: Performance of Different Soil Model Configurations in Simulating Ground Surface Temperature and Surface Fluxes, *Monthly Weather Review*, 125, 1870–1884, doi:10.1175/1520-0493(1997)125<1870:PODSMC>2.0.CO;2, 1997.
- 10 Smirnova, T. G., Brown, J. M., Benjamin, S. G., and Kim, D.: Parameterization of cold-season processes in the MAPS land-surface scheme, *Journal of Geophysical Research: Atmospheres*, 105, 4077–4086, doi:10.1029/1999JD901047, 2000.
- Snetsinger, K. G., Ferry, G. V., Russell, P. B., Pueschel, R. F., Oberbeck, V. R., Hayes, D. M., and Fong, W.: Effects of El Chichón on stratospheric aerosols late 1982 to early 1984, *Journal of Geophysical Research: Atmospheres*, 92, 14 761–14 771, doi:10.1029/JD092iD12p14761, <http://dx.doi.org/10.1029/JD092iD12p14761>, 1987.
- 15 Stettler, M., Eastham, S., and Barrett, S.: Air quality and public health impacts of {UK} airports. Part I: Emissions, *Atmospheric Environment*, 45, 5415 – 5424, doi:<http://dx.doi.org/10.1016/j.atmosenv.2011.07.012>, <http://www.sciencedirect.com/science/article/pii/S135223101100728X>, 2011.
- Stettler, M. E. J., Boies, A. M., Petzold, A., and Barrett, S. R. H.: Global Civil Aviation Black Carbon Emissions, *Environmental Science & Technology*, 47, 10 397–10 404, doi:10.1021/es401356v, <http://dx.doi.org/10.1021/es401356v>, PMID: 23844612, 2013.
- 20 Strawa, A. W., Drdla, K., Ferry, G. V., Verma, S., Pueschel, R. F., Yasuda, M., Salawitch, R. J., Gao, R. S., Howard, S. D., Bui, P. T., Loewenstein, M., Elkins, J. W., Perkins, K. K., and Cohen, R.: Carbonaceous aerosol (soot) measured in the lower stratosphere during POLARIS and its role in stratospheric photochemistry, *Journal of Geophysical Research: Atmospheres*, 104, 26 753–26 766, doi:10.1029/1999JD900453, <http://dx.doi.org/10.1029/1999JD900453>, 1999.
- Sunilkumar, S. V., Babu, A., and Parameswaran, K.: Mean structure of the tropical tropopause and its variability over the Indian longitude sector, *Climate Dynamics*, 40, 1125–1140, doi:10.1007/s00382-012-1496-8, <http://dx.doi.org/10.1007/s00382-012-1496-8>, 2013.
- 25 Thampi, B. V., Parameswaran, K., and Sunilkumar, S.: Semitransparent cirrus clouds in the upper troposphere and their contribution to the particulate scattering in the tropical {UTLS} region, *Journal of Atmospheric and Solar-Terrestrial Physics*, 74, 1 – 10, doi:<http://dx.doi.org/10.1016/j.jastp.2011.09.005>, <http://www.sciencedirect.com/science/article/pii/S1364682611002598>, 2012.
- Thompson, G., Rasmussen, R. M., and Manning, K.: Explicit forecasts of winter precipitation using an improved bulk microphysics scheme. Part I: Description and sensitivity analysis, *Monthly Weather Review*, 132, 519–542, doi:10.1175/1520-0493(2004)132<0519:EFOWPU>2.0.CO;2, 2004.
- 30 Timko, M. T., Onasch, T. B., Northway, M. J., Jayne, J. T., Canagaratna, M. R., Herndon, S. C., Wood, E. C., Miake-Lye, R. C., and Knighton, W. B.: Gas Turbine Engine Emissions—Part II: Chemical Properties of Particulate Matter, *Journal of Engineering for Gas Turbines and Power*, 132, 061 505–061 505–15, doi:10.1115/1.4000132, <http://dx.doi.org/10.1115/1.4000132>, 2010.
- 35 Turco, R. P., Toon, O. B., Hamill, P., and Whitten, R. C.: Effects of meteoric debris on stratospheric aerosols and gases, *Journal of Geophysical Research: Oceans*, 86, 1113–1128, doi:10.1029/JC086iC02p01113, <http://dx.doi.org/10.1029/JC086iC02p01113>, 1981.

- Uma, K. N., Kumar, K. K., Shankar Das, S., Rao, T. N., and Satyanarayana, T. M.: On the Vertical Distribution of Mean Vertical Velocities in the Convective Regions during the Wet and Dry Spells of the Monsoon over Gadanki, *Monthly Weather Review*, 140, 398–410, doi:10.1175/MWR-D-11-00044.1, <http://dx.doi.org/10.1175/MWR-D-11-00044.1>, 2012.
- Vernier, J.-P., Thomason, L. W., and Kar, J.: CALIPSO detection of an Asian tropopause aerosol layer, *Geophysical Research Letters*, 38, doi:10.1029/2010GL046614, <http://dx.doi.org/10.1029/2010GL046614>, 107804, 2011.
- Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B., and Baltensperger, U.: Absorption of light by soot particles: determination of the absorption coefficient by means of aethalometers, *Journal of Aerosol Science*, 34, 1445 – 1463, doi:[https://doi.org/10.1016/S0021-8502\(03\)00359-8](https://doi.org/10.1016/S0021-8502(03)00359-8), <http://www.sciencedirect.com/science/article/pii/S0021850203003598>, intercomparison of Soot Measurement Techniques, 2003.
- 10 Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning, *Geoscientific Model Development*, 4, 625–641, doi:10.5194/gmd-4-625-2011, <http://www.geosci-model-dev.net/4/625/2011/>, 2011.
- Wild, O., Zhu, X., and Prather, M.: Fast-J: Accurate Simulation of In- and Below-Cloud Photolysis in Tropospheric Chemical Models, *Journal of Atmospheric Chemistry*, 37, 245–282, doi:10.1023/A:1006415919030, 2000.
- 15 Yasunari, T. J., Bonasoni, P., Laj, P., Fujita, K., Vuillermoz, E., Marinoni, A., Cristofanelli, P., Duchi, R., Tartari, G., and Lau, K.-M.: Estimated impact of black carbon deposition during pre-monsoon season from Nepal Climate Observatory - Pyramid data and snow albedo changes over Himalayan glaciers, *Atmospheric Chemistry and Physics*, 10, 6603–6615, doi:10.5194/acp-10-6603-2010, <http://www.atmos-chem-phys.net/10/6603/2010/>, 2010.