Responses to the comments of anonymous referee #2

(Referee's comments are shown in italics and our responses are shown in normal fonts)

The manuscript by Gadhavi et al. presents a comparison of observed and model simulated equivalent black carbon (BC) concentrations. The observations were obtained with an aethalometer at a rural site in Southern India (Gadanki) during 2008 to 2012. The model simulations are based on a Lagrangian dispersion model (FLEXPART with NCEP Global Forecast Systems Final meteorological analysis data). For each day, a potential emission sensitivity (PES) field is obtained by a 10-day backward model run initialized from the receptor point. Model BC concentrations at the observation site are then calculated based on the PES using three different emission inventories. It is reported that the model simulates well the seasonal cycle of BC measurements, with highest concentrations in winter and lowest in summer. However, the model results are biased low in winter, spring, and summer. The biases appear to be correlated to fire radiative power observed by satellites. It is thus concluded that all three BC emission inventories may have under-estimated BC fluxes from open biomass burning over the Southern India. The manuscript is very interesting and well written. The work is very important for understanding the role of Indian sources of BC aerosol in global climate and regional hydrological cycle, and is suitable for publication in ACP.

We thank referee for evaluating our manuscript, providing constructive comments and considering our work interesting and suitable for publication in ACP. In following part, we are providing point by point responses to referee's comments.

Major Comments:

(1) Page 26911, lines 14-16. "The PES values in the bottom most layer (so-called footprint layer; 0-100 m a.g.l.) were multiplied by the emission fluxes to calculate the BC concentration at the receptor." This method is given without an explanation or evaluation. It may be argued that the entire planetary boundary layer (PBL) should be considered the footprint layer, because PBL height-based PES would be less sensitive to model uncertainties in surface layer mixing and dry deposition. Rapid vertical mixing of BC through the PBL is caused by turbulence in the day. Mean PBL depth retrieved by the CALIPSO satellite over India varies from 1000-1500 m in winter (DJF) to 2500-3000 m in summer (JJA) (McGrath- Spangler and Denning, 2013; Figure 3). A stable surface layer prevents vertical mixing in the night. However, this effect is often too large in models lacking a "background mixing" (intermittent mixing events are often observed at night). The PBL-based PES would be larger than estimated for 0-100 m if the model predicts a large decrease of tracer towards the surface.

It appears to us that the referee has confused our method (to multiply 0-100 PES layer with emission fluxes to get concentration) with vertical mixing of aerosol in boundary layer, probably because of insufficient details about PES (potential emission sensitivity) and the procedure in the manuscript.

Our method to multiply emission fluxes in a given grid cell to the bottom most 0-100 meter PES layer is related to assumptions about spatial distribution of emission sources within a grid box. For example, emissions from a car exhaust will take place near surface whereas emission from a factory chimney may take place at higher altitudes and the emissions from an aircraft will take place well above the boundary layer. In our case, we are assuming that the emission sources are uniformly distributed horizontally and vertically in a grid box of 100 meter height. While we know that often emission fluxes are clustered horizontally and they are far more common close to ground than at higher altitudes, however in view of other contextual information which are described next our assumption may not be major cause of concern.

(a) BC emissions are associated with burning processes and co-emitted with hot gases. The hot gases can lift BC particles at higher altitudes in very short time. Since the model does not simulate micrometeorology associated with fire temperatures and hot-gas-exhausts it is reasonable to assume 100 meter apriori vertical mixing.

(b) The model explicitly simulates the boundary layer height variation and the information is used for calculating vertical mixing of aerosols (Section 3 of Stohl et al., 2005). In other words boundary layer height variation is implicit in the PES values.

(c) Large part of BC load at the observation site is long-range transported. With the long-range transport, error due to inhomogeneities in emission fluxes in a grid-boxes is decreased.

(d) For a surface source, the height of the footprint layer should ideally be as small as possible. However, a very shallow footprint layer is not ideal from a statistical point of view, as the PES is calculated based on the mass (and, thus, approximately the number) of particles in the footprint layer. With a very shallow height (say, 10 m), one would need to release 10 times more particles than with a 100 m height of the footprint layer, to arrive at the same statistical error for the footprint PES (this assumes that the particles are well mixed over the lowest 100 m).

(e) In the sensitivity analysis carried out for two months where we varied the foot-print PES height from 100 meter to 300 meter, we have found that the simulated BC concentration at the observation site varied on average by less than 5% (Fig. 1). This indicates that very often the PBL height is higher than 300 m and particles are relatively well mixed in the PBL.

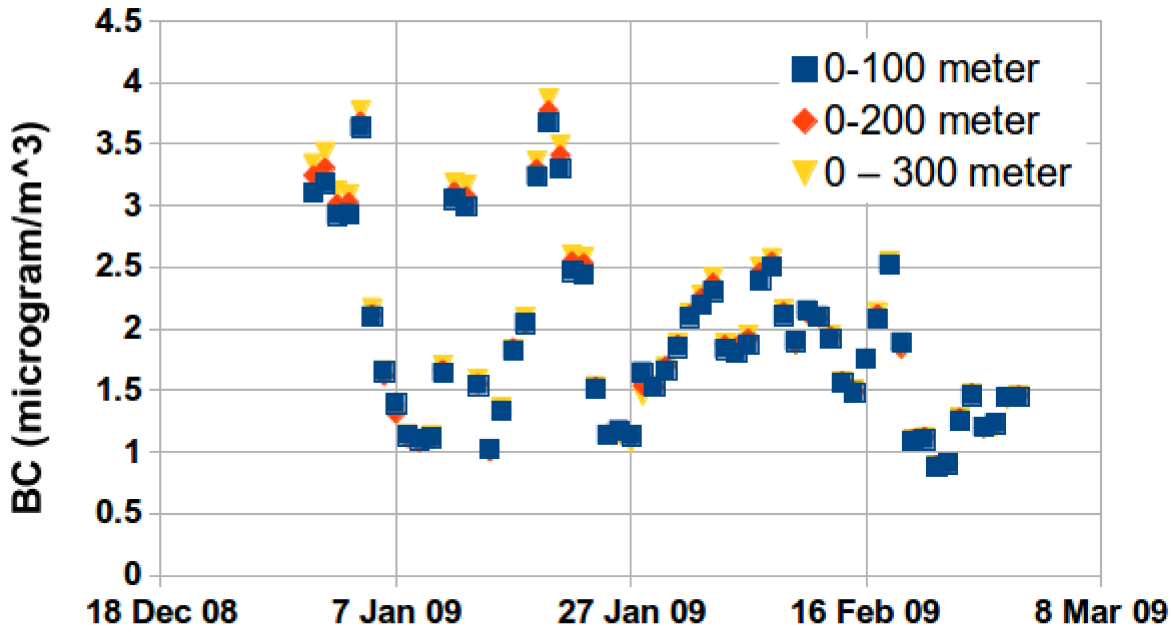

Fig. 1: Sensitivity of simulated BC concentration toward thickness of foot-print layer

In the revised manuscript, we are going to provide more details on this aspect as discussed above.

(2) Table 1. Wet deposition parameters. Wet scavenging is proportional to rain fall rate in the model, with a pre-factor A=2.E-7 per second per 1 mm/h of rain fall. This coefficient is typical for below-cloud scavenging of accumulation mode aerosols (Jung and Shao, 2006; Seinfeld and Pandis, 2006). However, in-cloud droplet nucleation occurs with hydrophilic aerosols. It is much more efficient for wet removal, and may be responsible for most of the loss of atmospheric BC aerosol (cf. Liu et al. 2011; Table 2).

We regret that we missed to mention in the manuscript that FLEXPART includes both the in-cloud and the below-cloud scavenging parametrisation. However, in-cloud scavenging parametrisation is fixed and not part of end-user settings hence they are not listed in Table 1. Indeed, in-cloud scavenging is typically more efficient than below-cloud scavenging in FLEXPART.

As noted in Table 1 the below-cloud scavenging is simulated as $\Lambda = A * prec^{B}$ where $A = 2x10^{-7}$ sec^{-1} and B=0.62. In-cloud scavenging is simulated as $\Lambda = (1.25 * prec^{0.64})/Th$ where "Th" is cloud thickness. Wet-deposition is further modified by a parametrisation of subgrid scale variability of precipitation.

As shown with an example (Fig.5e and 5f of manuscript) wet-scavenging is not underestimated as process in the model. However, unique meteorology and geography of the place viz. proximity to sources during rainy season, relatively small PES area (cf. Fig. 6c) during South-West Monsoon and short

rainy season over Northern and Western India makes the wet-deposition less important process for removal of the black carbon in our case. Wilcox and Ramanathan (2004) also reported similar conclusions based on differences in numerical simulations with and without wet-scavenging for Northern Indian Ocean.

In the revised manuscript, we are going to mention the above discussed details.

(3) Combining above comments, is it possible that the model has under-estimated both the PES and the rate of wet deposition, with (incomplete) compensating effects?

There exists a possibility that wet deposition may be overestimated since no special parametrisation is included for hydrophobic nature of BC particles, but as mentioned in reply to previous comment, it is not a major cause of concern since we are dealing with the place where wet-deposition plays a small role. While our assumptions about thickness of foot-print PES layer have their share uncertainties, it cannot be said it will result necessarily in underestimation of BC mass. It is argued in the manuscript using sensitivity analysis and age-spectra that “process” related uncertainties are unlikely to outweigh the uncertainties in emission inventories.

(4) Previously, Zhang et al. (2008) estimated the mass absorption coefficient for dust to be 1.3 mˆ2/g on average, at the wavelength (880 nm) used by the aethalometer. Are dust aerosol concentrations high enough to cause significant interference to BC measurements at Gadanki, especially when BC concentration is low and wind speed is high (in summer)?

This is an interesting observations by referee, which we have missed to discuss in the manuscript. During summer, model bias is of the order of -0.5 $\mu g/m^3$ which is nearly 50% of the observed BC mass concentration. Large region surrounding the observation site has good vegetation cover but since during summer wind speeds are high (a conducive condition for lifting dust particles) air parcel will have higher dust loading and can bring dust from medium to long-range distances to observation site. Unfortunately, we do not have concurrent measurements of dust amount in aerosol particles. However, there are other observations which we can look into to qualitatively evaluate the role of the dust.

Zhang et al. (2008) reports that BC particles have $1/\lambda$ wavelength dependence in absorption coefficient whereas absorption by the dust particles do not show significant wavelength dependence. We can characterise wavelength dependence of absorption coefficient (C) measured by aethalometer at seven wavelengths with the equation $C = \beta\lambda^{-\alpha}$ (similar to Angstrom's well-known equation for wavelength dependence of aerosol optical depth). Value of α is 1 for BC particles and 0 for dust particles according to Zhang et al. (2008). Value of α is reported greater than one for mixture of BC particles with organic carbon material (Bergstrom et al., 2004; Kirchstetter et al., 2004; Bergstrom et al., 2007; Clarke et al., 2007). In a mixture of BC and dust particles, as amount of dust particles increases values of alpha should

decrease.

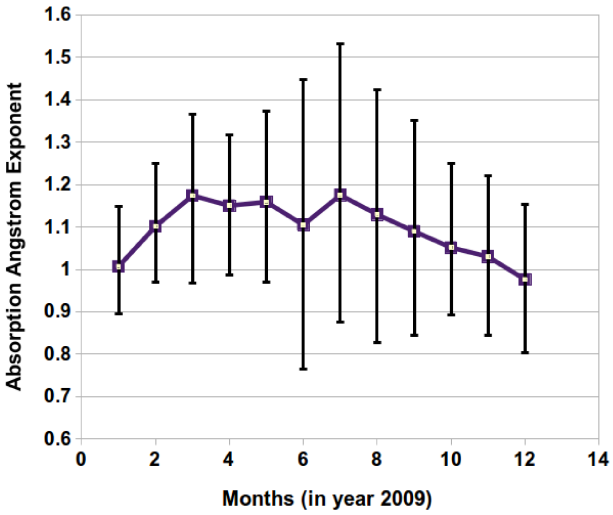

Fig. 2: Monthly median absorption Angstrom exponent for year 2009. Vertical bars are interquartile range.

Monthly median α values along with its interquartile range are shown in Fig. 2. During spring and summer there is an increase in α values, which we expect when BC particles originate from biomass burning. However, though median α values have increased during summer, the interquartile range is also extended toward both higher and smaller α values indicative of increase in episodic dust events. This suggests that though dust particles may have increased during summer but they may have small role in creating systematic bias such as seen between model and observations.

In the revised manuscript we will be including this discussion.

Minor Comments
(1) Page 26912, line 24. "due to decent" is confusing. Do you mean "due to ascent of air mass as it moves backwards (in time) from Gadanki to Arabia".

Thank you for drawing our attention to this. We will rewrite the description to avoid confusion.

(2) Figure 5. (I) The shaded circles indicating altitude is barely visible. Suggest keep the circles to indicate latitude and longitude location of the mean trajectory, and add panels to indicate altitude as a function of days before measurements. (II) The heading "Sensitivity at footprint m.a.g.l." above each panel implies that the PES is estimated for the indicated altitude, which is inconsistent with stated in the text (see above, Major comment #1).

We thank for drawing our attention to confusing title on the Figure. Since, PES fields are three dimension fields, we have used word “Footprint” to indicate that the values shown in the figures pertain to the bottom most layer (0 – 100 m.a.g.l.). In the revised manuscript, instead of the word “Footprint”

we will indicate it as 0 – 100 m.a.g.l.

Regarding altitude of mean trajectories shown with shaded black dots, we agree with the reviewer that it is difficult to view them in the figures in the main manuscript. However, we have refered to the height of mean trajectory only once in the main manuscript. Adding independent plot for height variation of the trajectories will not add further value to the text presented in the manuscript. We plan to retain dots as-it-is in the revised manuscript since they will be visible clearly in the individual day plots provided in the supporting material and the description in the manuscript will serve to explain the details in figures in supporting material.

References

Bergstrom, R. W., Pilewskie, P., Pommier, J., Rabbette, M., Russell, P. B., Schmid, B., Redemann, J., Higurashi, A., Nakajima, T. and Quinn, P. K.: Spectral absorption of solar radiation by aerosols during ACE-Asia, J. Geophys. Res.-Atmos., 109, D19S15+, doi:http://dx.doi.org/10.1029/2003JD004467, 2004.

Bergstrom, R. W., Pilewskie, P., Russell, P. B., Redemann, J., Bond, T. C., Quinn, P. K. and Sierau, B.: Spectral absorption properties of atmospheric aerosols, Atmos. Chem. Phys., 7, 5937-5943, 2007.

Clarke, A., Mcnaughton, C., Kapustin, V., Shinozuka, Y., Howell, S., Dibb, J., Zhou, J., Anderson, B., Brekhovskikh, V., Turner, H. and Pinkerton, M.: Biomass burning and pollution aerosol over North America: Organic components and their influence on spectral optical properties and humidification response, J. Geophys. Res.-Atmos., 112, D12S18+, doi:http://dx.doi.org/10.1029/2006JD007777, 2007.

Kirchstetter, T. W., Novakov, T. and Hobbs, P. V.: Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon, J. Geophys. Res.-Atmos., 109, D21208+, doi:http://dx.doi.org/10.1029/2004JD004999, 2004.

Stohl, A., Forster, C., Frank, A., Seibert, P. and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461-2474, doi:10.5194/acp-5-2461-2005, 2005.

Wilcox, E. M. and Ramanathan, V.: The impact of observed precipitation upon the transport of aerosols from South Asia, Tellus B, 56, 435-450, doi:10.1111/j.1600-0889.2004.00121.x, 2004.

Zhang, X. Y., Wang, Y. Q., Zhang, X. C., Guo, W., Niu, T., Gong, S. L., Yin, Y., Zhao, P., Jin, J. L. and Yu, M.: Aerosol monitoring at multiple locations in China: contributions of EC and dust to aerosol light absorption, Tellus B, 60, 647-656, doi:10.1111/j.1600-0889.2008.00359.x, 2008.