Response to Reviewer-1

This paper presents altitude profiles of aerosol size distribution and Black carbon obtained through in situ on-board research aircraft as a part of South-West Asian Aerosol Monsoon Interaction (SWAAMI) experiment conducted jointly under Indo-UK project over three distinct locations (Jodhpur, Varanasi, and Bhubaneswar) just prior to the onset of Indian Summer Monsoon. Simultaneous measurements from Cloud Aerosol Transportation System (CATS) on-board International Space Station and OMI measurements are also used as supporting information.

Major results include an increase in coarse mode concentration and coarse mode mass-fraction with increase in altitude across the entire IGP, especially above the well-mixed region. Further authors found increase with altitude in both the mode radii and geometric mean radii of the size distributions. Near the surface the features were specific to the different sub-regions ie., highest coarse mode mass fraction in the western IGP and highest accumulation fraction in the Central IGP with the eastern IGP coming in-between. The elevated coarse mode fraction is attributed to mineral dust load arising from local production as well as due to advection from the west which is further verified using CATS measurements. Existence of a well-mixed BC variation up to the ceiling altitude (3.5 km) is reiterated in this manuscript and match well with those obtained using previous aircraft and balloon platforms.

Results presented in the manuscript are in general unique and apt for the prestigious journal like ACP. Manuscript is written preciously and concise except at few places. The results present also add new understanding on the size distribution of aerosol concentration in both altitudinal and longitudinally which are very important in understanding their role on precipitation processes besides radiative forcing estimates. Though major part of manuscript is written well, at some place revision is required. Manuscript may be acceptable after satisfactory revising the following.

We appreciate the summary evaluation of the reviewer and agree to the observations. Following the valuable comments and fruitful suggestions for improving the quality of the manuscript, we have revised it incorporating the review comments of all the reviewers. Our point wise response to each of the comment is given below in bold letters, below the respective comments.

Major comments/suggestions:

It is not clear from where and how rainfall and relative humidity measurements presented in Figure 2e are obtained. Further they are not discussed at all in the rest of the manuscript. Same for the profiles of temperature presented in figure 2f.

I suggest providing profiles of temperature and relative humidity (if obtained from aircraft) as a separate figure in Supplementary material and add related discussion the manuscript. This information may be useful while dealing with hygroscopic nature of aerosol.

Response: The values of surface meteorological parameters were obtained from the meteorological observations at the respective airports during the period of flight operations. In addition, ambient temperatures at different altitude levels of the atmosphere were obtained from the aircraft sensor. However, we did not have dedicated meteorological sensors and data loggers aboard for continuous recording of ambient RH and T. Hence, we are unable to show the profiles. In view of this, we have modified Fig-2 in the revised manuscript and kept the relevant meteorological information (as numerical values) in the appropriate places.

It is not clear what is the source of the data (TPP) presented in Figure 12a. Further (b) and (c) are inter-changed. Note that SO2 is presented in (c) and NO2 in (b).

Response: Thanks for the suggestion. We have included the source of data (https://www.ntpc.co.in/en/power-generation/coal-based-power-stations) used to geo-locate the coal based TPP distributed over the Indian region. The inadvertent error in the figure caption (Figure-13-R1) is also corrected in the revised manuscript.

Measurements of Black Carbon with Aethalometer: Though authors made correction to the data obtained from Aethalometer, it is not clear how they have taken care of it in the un-pressurized air craft?

Response: We have elaborated the discussion (as given below) on the estimation of true BC concentrations from the unpressurised operation of aethalometer in the aircraft in the revised manuscript.

Line nos. 189-204: "In the present study, BC mass concentrations were obtained at 1-minute interval by operating the aethalometer at 50% of the maximum attenuation and a standard mass flow rate of 2 LPM corresponding to standard temperature (T_O, 293 K) and pressure (P_O, 1013 hPa). As the unpressurised aircraft climbed higher, the instrument experienced ambient pressure (P) and temperature (T). In order to maintain the set mass flow, the pumping speed of the instrument was automatically increased (through internal program) to aspire more volume of air. However, the volume of air aspirated at ambient pressure and temperature requires to be corrected to standard atmospheric condition for the actual estimate of BC (Moorthy et al., 2004). Thus, the actual volume of air aspirated by the Aethalometer at different atmospheric level is,

$$V = V_o \frac{P_o T}{P T_o}$$

Thus, true BC mass concentration (M_{BC}) is

$$M_{BC} = M^*_{BC} \left[\frac{P_O T}{P T_O}\right]^{-1} \quad (1)$$

Here, M^*_{BC} is the instrument measured raw mass concentration of BC at ambient pressure and temperature."

It will be good to show heating rates due to BC profiles at these three different regions. Some discussion is needed on how the results presented in the manuscript are linked with the main objective of the SWAAMI experiment.

Response: We comply with suggestion with thanks and have included the heating rate profiles due to BC at all the three distinct regions of the IGP. The methodology of deriving BC forcing and atmospheric heating rate due to BC is included in the revised manuscript as given below:

Line nos. 469-504: "To quantify the climatic implications of BC, the heating rate profiles of BC are examined based on the estimation of shortwave direct radiative forcing (DRF) due to BC alone. The DRF due to BC represents the difference between the DRF for aerosols with and without the BC component. The in-situ values of scattering (σ_{sca}) and absorption (σ_{abs}) coefficients measured on-board the aircraft were used to estimate spectral values of AOD (layer-integrated σ_{sca} + σ_{abs}), single scattering albedo (SSA) and asymmetry parameter (g) for each level, assuming a well-mixed layer

of 200 m above and below the measurement altitude (details are available in Vaishya et al., 2018). The layer mean values of AOD, SSA and Legendre moments of the aerosol phase function (derived from Henyey–Greenstein approximation) are used as input in the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART, Ricchiazzi et al., 1998) model to estimate diurnally averaged DRF (net flux with and without aerosols) at the top (DRF_{TOA}) and bottom (DRF_{SUR}) of each of the layers. The atmospheric forcing (DRF_{ATM}) for each of the levels is then estimated as

$$DRF_{ATM} = DRF_{TOA} - DRF_{SUR}$$

In order to estimate the forcing due to BC alone, optical parameters for aerosols were deduced again. For this, values of σ_{abs} were segregated to the contributions by BC (σ_{BC}) and OC (σ_{OC}), where σ_{BC} were estimated following inverse wavelength dependence of BC (e.g., Vaishya et al., 2017). Based on this, a new set of AOD and SSA for BC-free atmosphere is calculated and fed into SBDART for estimating DRF_{ALL-BC} without the BC component. Thus, DRF due to BC is

$$DRF_{BC} = DRF_{ALL} – DRF_{ALL-BC}$$

Here, DRF_{ALL} represents forcing due to all the aerosol components, including BC. Change in AOD from total to BC-free atmosphere is not significant (< 2%), whereas SSA changes to a greater extent which actually participates in the DRF_{BC} estimation.

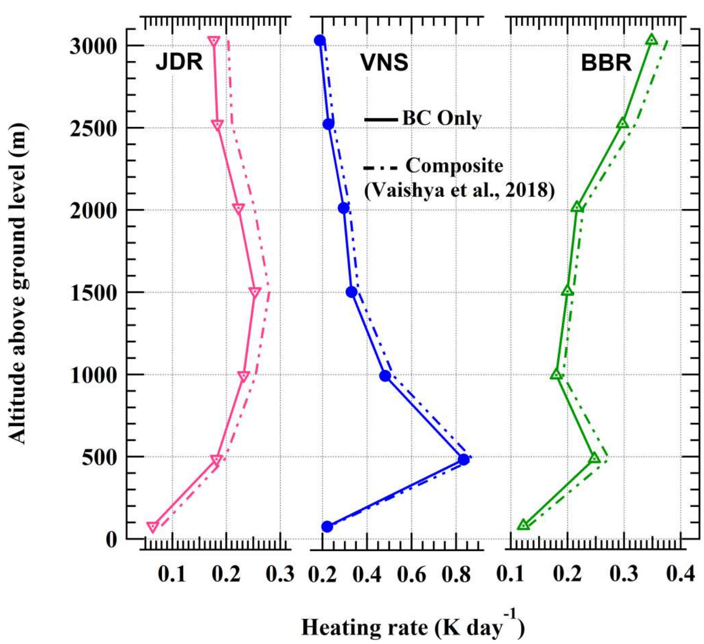

Figure-12: Vertical profiles of atmospheric heating rate due to BC (solid lines) and composite (dashed lines) aerosols for the regions of the IGP: (a) JDR in western IGP, (b) VNS in central IGP and (c) BBR in eastern IGP. Data for the composite heating rate profiles are from Vaishya et al., 2018.

The vertical profiles of atmospheric heating rate (HR, estimated based on the atmospheric pressure difference between top and bottom of each layer and aerosol induced forcing in that layer) due to BC alone shows (Figure-12) maximum influence of BC in trapping the SW-radiation at VNS, followed by BBR and JDR. Interestingly, the altitudinal profiles of heating rate are distinctly different over the regions, BBR showing an increase with altitude, while VNS shows the opposite pattern with

maximum heating (~ (~ 0.81 K day^{-1}) near the surface. Enhanced heating at 500-2000 m altitude is seen at JDR. These results indicate the dominant role of absorbing aerosols near the surface at VNS, while the atmospheric perturbation due to elevated layers of absorbing aerosols is conspicuous at BBR (HR ~ 0.35 K day^{-1} at the ceiling altitude). The column integrated values of atmospheric forcing due to BC alone are 7.9 Wm^{-2}, 14.3 Wm^{-2} and 8.4 Wm^{-2} at JDR, VNS and BBR respectively."

Regarding the linkage of the results to the objectives of SWAAMI, we add (line nos. 78-88) the following.

"The information on aerosol size distribution is important for accurately describing the phase function, which describes the angular variation of the scattered intensity. The knowledge of its vertical variation would thus improve the accuracy of ARF estimation and hence heating rates. Such information is virtually non-existing over this region. Further, the knowledge of the variation of size distribution with altitude would be useful better understanding the aerosol-cloud interactions and CCN characteristics, during the evolving and active phase of the Indian monsoon. This was among the important information aimed to be obtained under SWAAMI - RAWEX (https://gtr.ukri.org/projects?ref=NE%2FL013886%2F1 and http://www.spl.gov.in/SPL/index.php/arfs-research/field-campaigns/asfasf) - a joint Indo-UK field experiment involving airborne measurements using Indian and UK aircrafts during different phases of the Indian monsoon, right from just prior to the onset of monsoon (i.e. in the beginning of June)."

Minor issues:

Results presented in Page 4 at lines 90-94 and 111-121 are mostly repeating. Both can be clubbed and rewrite to the point.

Response: Complied with.

Figure 11 caption does not match with the information presented in the figure. I am unable to see (b) Daily profiles of MBC during each of the flight sorties on different days.

Response: Sorry for the oversight. We have corrected the figure caption in the revised manuscript.

Figure 14. Figure caption need to be changed as per the information presented in the Figure (It should be 18 and 20 May but not 19 and 20 May).

Response: Complied with.

I do not see any logic for presented vertical velocities for 2012, 2013 and 2016.

Response: We have shown the vertical velocities to support the role of changing dynamical processes during three distinct seasons (Winter, represented by December), Spring (represented by May) and just to prior to onset of Indian Summer Monsoon (represented by June), which very well support the vertical profiles of BC at the respective seasons. We have made this clear in the legends of the figure in the revised manuscript.

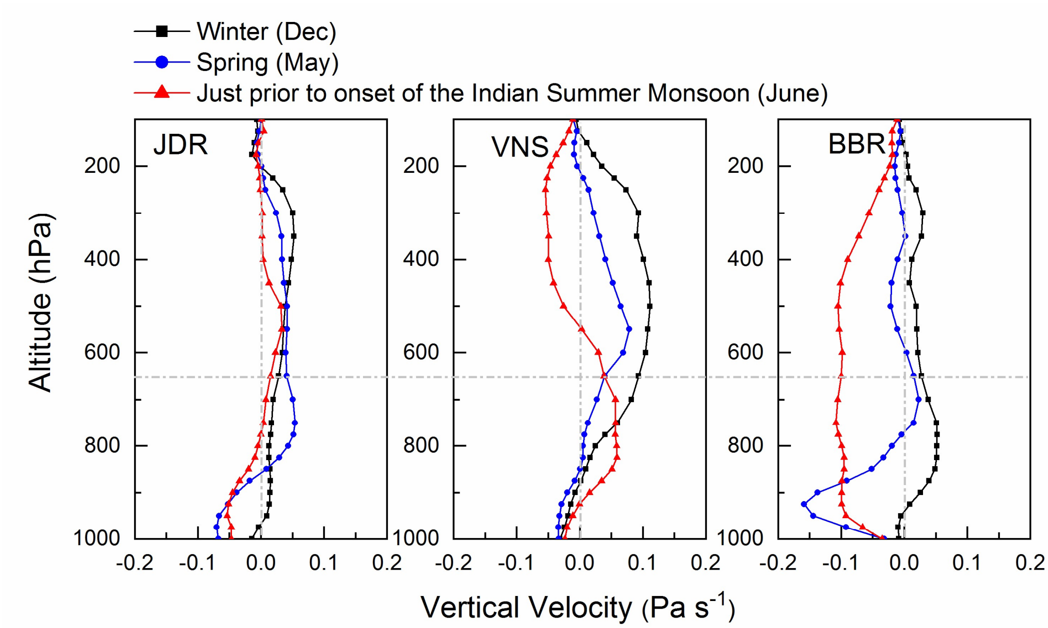

Figure-16: Vertical profiles of vertical velocity (Pa s^{-1}) over the study locations representing Winter (December, 2012), Spring (May, 2013) and just prior to the onset of the Indian Summer Monsoon (June, 2016) at different pressure levels from 1000 to 100 hPa. The positive and negative values are indicative of the descending and ascending motions respectively. The horizontal dashed line indicated the ceiling altitude (~ 3.5 km above ground level) of aircraft measurements while the vertical dashed lines mark the boundary of vertical velocity (= 0) changing from positive to negative and vice versa.

References:

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Vaishya, A., Singh, P., Rastogi, S., and Babu, S. S.: Aerosol black carbon quantification in the central Indo-Gangetic Plain: Seasonal heterogeneity and source apportionment, Atmos. Res., 185,13–21, https://doi.org/10.1016/j.atmosres.2016.10.001, 2017.

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