Response to reviewers

Observations of the reviewers are in italics, and our response is given below in bold letters.

Reviewer-1: ACP-2019-376-RC1

The size of BC clearly shows seasonal variation and emission sources of BC. They selected a suitable sampling location that gets plumes from both lands as well as from coastal/marine regions on different seasons. They used SP2 and ACMS to characterise the microphysical and chemical properties of BC and other aerosol particles. The research provides useful information on BC mixing with other aerosol components based on a diurnal and seasonal variation on the eastern coast of India. IGP is one of the regional hot spots for BC aerosol concentration in south Asia and the present research provides detail information on the mixing scenario of BC aerosol from the IGP and compares with the BC from other parts of India. They quantified the coating of BC in term of ACT and RCT and presented the influence of different coating materials, and they discussed the preferential coating in different seasons. The language used is good but can be polished, especially, long sentences are used, and it makes difficult to communicate the message. I recommend the authors to shorten/split the long sentences. I recommend the publication of this article but after addressing the concerns listed below.

We thank the reviewer for the summary comments and positive recommendation, followed by detailed evaluation, and constructive comments. We have revised the manuscript accordingly, and our responses to the specific comments of the reviewer are given below:

How relevant is the discussion of mineral dust in the introduction section (paragraphs 1 and 2)? I understand that the authors highlight the light-absorbing nature of minerals. But, I did not find any further discussion on minerals in the manuscript. In that context, the direct focus on BC mixing state is meaningful.

Complied with. This was originally included since dust constitutes a major fraction of aerosols loading over the IGP during spring and summer. However, we agree that dust is not the theme of this paper, and so we have modified the introduction section to focus mainly on the BC mixing state.

The authors mention the use of 2 kinds of aerosol characterization instruments during the study. But there is no mention of one of the two instruments throughout the introduction section.

I had to go through the method section to get information about the second instrument. Please mention a brief introduction on the second instrument used in this study.

Complied with. We have included the relevant details in the introduction section. The following sentences are included in the revised manuscript.

Page 5, Line 14: "Along with this, information on the condensable materials which act as coating substances and constantly alter the physiochemical properties of the BC containing particles, is also essential. Collocated mass spectroscopy-based high-resolution aerosol chemical composition measurements have been employed for this purpose (Liu et al., 2014; Gong et al., 2016)."

Page 5, Line 25: "To meet these objectives, state-of-the the-art instruments were installed at Bhubaneswar, which included a single particle soot photometer (SP2) for characterization of refractory BC (rBC) aerosols and an Aerosol Chemical Speciation Monitor (ACSM) for high-resolution measurements of non-refractive submicron aerosol chemical composition for long-term measurements."

Page 5: Line 31: "The contributions from distinct sources to BC concentrations and the association of coating on BC with possible condensable coating materials are examined, and the implications are discussed."

Regarding the use of SP2, a recent finding by Sedlacek III et al. (2018) cautioned about the charring of organic depending on the SP2 laser power (Sedlacek III, Arthur J., et al. "Formation of refractory black carbon by SP2-induced charring of organic aerosol." Aerosol Science and Technology 52.12 (2018): 1345-1350). It is worthy to mention this caveat as the ambient aerosol samples include organics. Clearly mention the power of the laser used during the operation of the SP2.

Thank you, and we agree. We have included this in the revised manuscript. Page 8, Line24.

"Recently, Sedlacek III et al. (2018) have cautioned that rBC may be produced by laser-induced charring of organic substances in the SP2, which depends on the laser power. Such laser-induced charring could result in an overestimate of rBC. During our measurements, the laser power varied in the range 2.1-3.7 V, which is above the threshold to detect rBC with high efficiency (> 2V) (Sedlacek III et al., 2018). Though we cannot rule out an additional rBC contribution from

charring of organic matter, it is likely this occurs in circumstances when the laser voltage is higher than that used in our study."

The following reference has been included in the revised manuscript.

Sedlacek, A. J., III, Onasch, T.B., Nichman, L., Lewis, E.R., Davidovits, P., Freedman, A., and Williams, L.: Formation of refractory black carbon by SP2-induced charring of organic aerosol, Aerosol Sci. and Technol., 52:12, 1345-1350, 2018, DOI:10.1080/02786826.2018.1531107.

Also, it is not always appropriate to assume core-shell structure for coated BC due to the complex mixing state of BC, such as the case when BC is located off-center. Though the study did not utilize single particle off-line analysis to probe the complex internal mixing state of BC, it is useful to mention the effects from such BC structures on absorption and scattering signals (e.g., Sedlacek III, Arthur J., et al. "Determination of and evidence for non-core shell structure of particles containing black carbon using the Single Particle Soot Photometer (SP2)." Geophysical research letters 39.6 (2012)). The authors should discuss how such noncore- shell particles would affect their results.

Agree. We have revised the section accordingly by adding the following:

Page 8, Line29.

"Sedlacek III et al. (2012) examined the structure of rBC containing particles using the 'lag time' technique and suggested that the core-shell model does not apply to all rBC –containing particles. A situation with non-core-shell structure (the case when BC is located off-center) arising due to the complex mixing state of BC may lead to uncertainty in determining the coating thickness of BC. Our study assumes BC to be at the centre and a uniform coating around, in the absence of other measurements to understand the complex coating. A recent study by Liu et al., (2017) demonstrated good agreement between Mie-modelled scattering values using the core-shell approximation and the SP2-measured scattering cross-section for the BC with thicker coatings as is the case for the majority of particles in this study. In addition, further the particle scattering is relatively independent of particle morphology at the SP2 wavelength 1064nm (Moteki et al., 2010)."

The following references have been included in the revised manuscript.

Sedlacek, A. J., III, Lewis, E. R., Kleinman, L., Xu,J., and Zhang, Q. : Determination of and evidence for noncore-shell structure of particles

containing black carbon using the Single-Particle Soot Photometer (SP2), Geophys. Res. Lett., 39, L06802, 2012, doi:10.1029/2012GL050905.

Moteki, N., Kondo, Y., and Nakamura, S.-i.: Method to measure refractive indices of small nonspherical particles: Application to black carbon particles, J. Aerosol Sci., 41, 513-521, 2010.

For all instruments employed, also mention the model number in addition to the manufacturing company.

Complied with.

Be consistent in the use of units such as for flow rate and BC concentration. The use of different units for the same quantity makes it difficult to infer the values. Minimize the use of numeral values for comparison all the times. It may appear to the authors that these numerals are useful for comparison, but to me, it is a source of distraction. I recommend listing the values in Tables which the authors have already done. Authors can infer Table for values and focus on their findings.

Complied with for the entire manuscript.

General comments:

Page4 line 19: Add some references on the size of monomers for nascent BC.

Cite (2017): 166, Köylü, Ümit Özgür, et al. "Fractal and projected structure properties of soot aggregates." Combustion and Flame 100.4 (1995): 621-633;

Bhandari, Janarjan, et al. "Effect of thermodenuding on the structure of nascent flame soot aggregates." Atmosphere 8.9.

Complied with. We have added the following references in the revised manuscript.

Köylü, Ü.Ö., Faeth, G.M., Farias, T.L., Carvalho, M.G.: Fractal and projected structure properties of soot aggregates, Combustion and Flame, 100, 621-633, 1995, ISSN 0010-2180, https://doi.org/10.1016/0010-2180(94)00147-K.

Bhandari, J., China, S., Onasch, T., Wolff, L., Lambe, A., Davidovits, P., Cross, E., Ahern, A., Olfert, J., Dubey, M., and Mazzoleni, C.: Effect of thermodenuding on the structure of nascent flame soot aggregates, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2016-270, 2016.

Page 4 line19: Also, it is not correct to say that the coated BC is 'spherical' after coating. Rather I would prefer to use the term like 'compact' or 'collapsed' for coated BC core, though the core-shell model treats such coated BC as the spherical core for simplicity.

We agree and have revised the relevant portion (Page 4 , Line 17)

"However, it collapses to a compact BC particle with its cores being coated with other components via coagulation among aggregates and (or) via condensation of atmospheric vapours while aging in the atmosphere."

Page 4, line 27: What are you referring to using "these"? I am not clear.

This sentence is modified in the revised manuscript.

"All the aforementioned processes have implications for direct and indirect radiative forcing of BC".

Page4 line 32: What sources? Do you mean 'aerosol sources'? Be specific.

This is now modified to read 'aerosol sources'.

Page 5, line 24: Only one instrument is revealed. But the authors mention in the abstract that they used 2 instruments. What is the second instrument used? Specify the second instrument as well in the introduction section mentioning why the instrument was selected.

Complied with. The discussion about the ACSM is added here in the revised manuscript.

Page 5, line 25: Do the authors mean to say that the working of SP2 is based on long-term measurements only? Please rephrase the sentence.

The sentence is modified in the revised manuscript as below.

"To meet these objectives, state-of-the-art instruments were installed at Bhubaneswar for long-term measurements. These included a single particle soot photometer (SP2) for characterization of refractory BC (rBC) aerosols and an aerosol chemical speciation monitor (ACSM) for high-resolution chemistry of possible coating materials. The present study provides results from a yearlong database from a combination of these instruments, perhaps for the first time over the Indian region".

Page 6, line 12: Do you mean to say 'above ground level' by acronym AGL? Please mention the full name for the first time.

Yes. Complied with.

Page 6, line 28: By 'Supplementary Figure S1' are you referring for the figures in an appendix? If so rename the figure as A1.

Complied with. Supplementary figures are now renamed as Figure S1, S2 and so on.

Page 7, lines 1-8: As the numeral values for meteorological parameters are shown in Table 1, avoid using all these numerals in the text for comparison. Please minimize the use of numeral values in the text. Include only those specific values that are striking to discuss.

Complied with. The sentence is modified in the revised manuscript as below.

"During SMS, the prevailing wind speed and temperature were moderate, and relative humidity (RH) was high (Table 1), while rainfall, associated with the monsoon, was extensive (total rainfall ~ 878 mm). Compared to the SMS, lower temperatures, winds and RH prevailed during the PoMS, with lower total rainfall (~ 201 mm). The lowest temperatures and RH of the year were seen during winter when calm wind conditions prevailed with almost no rainfall. The PMS witnessed the highest temperatures of the year (as high as 41 °C), moderately humid atmosphere and relatively higher wind speed compared to winter. During this season the region received a total rainfall of ~149 mm associated with thundershower events that led to high-velocity local winds. Details are given in Table 1"

Page 7 lines 15-17: Include the model number and company name for each instrument

Complied with. The sentence is modified in the revised manuscript as below.

"In the present study data was collected using a single-particle soot photometer (SP2) (Model: SP2-D; Droplet Measurement Technologies, Boulder, USA) and an Aerosol Chemical Speciation Monitor (ACSM) (Model: 140; Aerodyne Research Inc., USA)."

Page8 line2: It is mentioned that the RI of 2.26 – 1.26i is used for BC. Is this RI representative for ambient BC aerosol in the region? I am aware of the use of the above-mentioned RI value for BC. However, the RI of 2.26 – 1.26i looks higher than usually used value for the RI of BC.

The RI 2.26-1.26i has been widely used in the SP2 community to derive the scattering properties of rBC at the specified SP2 wavelength 1064nm (Moteki et al., 2010; Taylor et al., 2015; Laborde et al., 2012). The other RI, as commonly seen in the literature, are used for the other wavelengths, mainly for the optical properties in the visible range. We do not have a region-specific RI value for BC.

References:

Laborde, M., Schnaiter, M., Linke, C., Saathoff, H., Naumann, K. H., Möhler, O., Berlenz, S., Wagner, U., Taylor, J. W., Liu, D., Flynn, M., Allan, J. D., Coe, H., Heimerl, K., Dahlkötter, F., Weinzierl, B., Wollny, A. G., Zanatta, M., Cozic, J., Laj, P., Hitzenberger, R., Schwarz, J. P., and Gysel, M.: Single Particle Soot Photometer intercomparison at the AIDA chamber, Atmos. Meas. Tech., 5, 3077-3097, 10.5194/amt-5-3077-2012, 2012.

Liu, D., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, Dominick V., Reddington, Carly L., Kong, S., Williams, Paul I., Ting, Y.-C., Haslett, S., Taylor, Jonathan W., Flynn, Michael J., Morgan, William T., McFiggans, G., Coe, H., and Allan, James D.: Black-carbon absorption enhancement in the atmosphere determined by particle mixing state, Nat. Geosci., 10, 184-188, 10.1038/ngeo2901, 2017.

Moteki, N., Kondo, Y., and Nakamura, S.-i.: Method to measure refractive indices of small nonspherical particles: Application to black carbon particles, J. Aerosol Sci., 41, 513-521, 2010.

Taylor, J., Allan, J., Liu, D., Flynn, M., Weber, R., Zhang, X., Lefer, B., Grossberg, N., Flynn, J., and Coe, H.: Assessment of the sensitivity of core/shell parameters derived using the single-particle soot photometer to density and refractive index, Atmos. Meas. Tech., 8, 1701-1718, 2015.

Page8 line 9: As mentioned earlier, be consistent in picking unit for a given quantity. Here for flow rate, you used cm3/min while in line 21 you used liters/minute.

Complied with.

Page 8, line 15: For the 40-100 nm range, mention clearly that the size is 'aerodynamic diameter'.

Complied with.

Page 8, line 26: The sub-heading 3.1 can be made more specific. By 'Mass and number concentration' only it is not clear what is being measured.

Complied with. The sub-heading 3.1 is modified as "BC mass and number concentrations" in the revised manuscript.

Pages 8-9 section 3.1: As mentioned earlier, use as least numeral values as possible in the discussion. All the numerals can be summarized in the table. Also, use the same units for particle concentration. In some cases, ng/m3 is used in some instance, µg/m3 is used for particle concentration.

Complied with. Particle concentration is expressed in µg m^{-3} throughout the revised manuscript (relevant text is modified in abstract, discussion and conclusions). Units in figure 3(a) also are now modified.

Page 10, line 27: I am not clear about this '...reported for reported from...'. Please clarify this sentence.

Sorry; the repeated word is deleted.

Page 11, line 2: Is 'Figure S1' the same labelled as 'Figure A1' in the appendix?

In the revised manuscript, all the supplementary figures are labelled as Figure S1, S2, and so on. All these figures, corresponding text and labels are available as supplementary information in the revised manuscript.

Add label for each season in the map. In fig. A1 (a) and (b), it will be useful to mention the location of the distinct data points below south India as a note.

Complied with. Now labels are added in Figure S1.

"During SMS (and PoMS as well), a considerable amount of fire events are noticeable below south of India (over Srilankan region)"

Page 11, line 10: It is mentioned that the BC mass loading was lowest during PMS, but fire events were maximum during PMS throughout the Indian region, as shown in Fig. A (d). Is not it reasonable to expect a high concentration of BC?

The PMS is characterised by intense solar heating of the landmass and very little precipitation. As such, this season exhibits very high surface temperatures (going as high as 47-49 °C). Strong thermal convection resulting from intense solar heating of the dry land lifts the planetary boundary layer to higher altitudes, and with winds gaining speed, there is greater dispersion of the aerosols (Nair et al., 2007; Kompalli et al., 2014) leading to a substantial reduction in the surface concentrations. This stronger particle dispersion resulted in lower concentrations at the surface level, despite any increased contribution from fire events. Several studies have previously highlighted the presence of elevated aerosol layers over the Indian region during the pre-monsoon season (Satheesh et al., 2008; Babu et al., 2011).

Babu, S.S., Moorthy, K. K., Manchanda, R. K., Sinha, P. R., Satheesh, S. K., Vajja, D. P., Srinivasan, S., 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?, Geophys. Res. Lett. 38, L08803, 2011, doi:10.1029/2011GL046654.

Satheesh, S. K., Moorthy, K.K., Babu, S.S., Vinoj, V., Dutt, C. B. S.: Climate implications of large warming by elevated aerosol over India. Geophys. Res. Lett. 35, L19809, 2008, doi:10.1029/2008GL034944.

Nair V. S., Moorthy K.K., Alappattu, D. P., Kunhikrishnan, P. K., George, S., Nair, P. R., Babu, S. S., Abish, B., Satheesh, S. K., Tripathi, S. N., Niranjan. K., Madhavan, B. L., Srikanth, V., Dutt, C.B.S., Badarinath, K. V. S., Reddy, R. R.: Wintertime aerosol characteristics over the Indo-Gangetic Plain (IGP):

Impacts of local boundary layer processes and long-range transport; J. Geophys. Res. 112 D13205, 2007, doi: 10.1029/2006 JD008099.

Page 14 lines7-9: The sentence "The figure reveals. . .." is not clear to me. Please rewrite the sentence

Complied with. The sentence is modified in the revised manuscript as below.

"In addition to the typical double-humped diurnal variation of BC mass concentration, which arises due to the combined effects of atmospheric boundary layer dynamics (Kompalli et al., 2014) and diurnal variation of the anthropogenic activities, very interesting links between BC core and relative coating thickness are noticeable from the Figure".

Page 14 line11, line 13: Do you mean to say that the RCT of BC is contributed by the day-night temperature difference in different seasons? I am not clear.

The sentence is modified in the revised manuscript as below for better clarity.

"and the amplitude of the BC variation has a marked seasonality. It is caused by the seasonal change in the diurnal variation of the ABL driven by seasonal changes in surface heating and resulting thermal convection. The highest amplitude occurs in winter since the diurnal variation of the ABL is greatest due to the high variation in surface temperature; with ΔT (i.e.T_{max}-T_{min}) ~ 12 °C over a 24 hour period. Conversely, the lowest amplitude occurs during the monsoon season, when thermal convection is highly suppressed due to the overcast sky, low surface heating and the surface energy balance being dominated by latent heat (the average diurnal amplitude of temperature variation, ΔT ~ 4.9 °C)."

Page 15 lines6-9: The sentence 'Interestingly. . .occurring' is a long sentence.

The sentence is split into two sentences in the revised manuscript as below.

"Interestingly, during the morning period when the BC mass concentration peaks due to the combined effect of the boundary layer dynamics (fumigation effect) and sources (rush hour traffic contribution), RCT was at a minimum. This suggests that fresh emissions from rush hour traffic, which would push up the BC concentration and lower the RCT, outweigh the fumigation effect; though both may be occurring around the same period."

Page 15, lines 13-16: The sentence 'Not. . .role' is again a long sentence and is not clear. Simplify your statement.

The sentence is split into simpler sentences as below:

"The diurnal variations in RCT are suppressed in the SMS and PMS compared to the winter and PoMS due to the seasonality of the boundary layer dynamics that modulates the concentrations of BC and the other condensing species. In addition to this, the wet scavenging by intense rains during the SMS ensures that a greater proportion of the remaining BC in the atmosphere is likely to be freshly emitted. Such extensive precipitation also leads to a reduction in concentrations of the coating substances. During the PMS, BC particles generally have larger core sizes, and the relative coating thickness is reduced in magnitude. These effects also play a role in shaping up the diurnal pattern."

Page 16, line 28: Chlorine is shown to be present in very low concentration even when air mass arrived from marine region to the sampling site during PMS. Is this concentration normal during the PMS as well?

Since the ACSM measures only NR-PM_1 and does not detect refractory materials (sublimation temperature > 600 ^oC), which includes sea salt chloride, chloride measured by the ACSM is mainly from the sources other than of marine origin.

Page 17 lines10-13: The sentence 'Further. . .. mixed' is not clear. In "absorption condensable species", do you mean 'absorption of condensable species' and the part "which already more internally mixed" is not clear.

The sentence is modified in the revised manuscript as below.

"The concentrations of freshly produced particles with little or no coating) arising from primary as well as secondary sources are, in general, greater during day. It enables more efficient adsorption of condensable species on these particles, compared to relatively aged particles during the night which are already coated or internally mixed due to aging. A greater fractional change can occur more quickly on fresh BC particles compared to particles which are already thickly coated since a much smaller amount of condensable material is required."

Technical comments:

Page4 line 16: Correct the year for 'China et al., 2012' to make it '2013'.

Complied with.

Page4 line 27: A sentence starts with a pronoun at the beginning of a paragraph. It makes it difficult to know what you are referring to using a pronoun in the very beginning of a paragraph.

The sentence is modified in the revised manuscript as below.

“All the aforementioned processes have implications for direct and indirect radiative forcing of BC.”

Page 10 line3: What is represented by ‘A’ in equation (1)? Mention it.

Complied with.

Page 11 line1: add ‘of’ before ‘larger-sized BC particles. . .’.

Complied with.

Page 20, line 2: Stronger is already a comparative adjective. Remove ‘more’ before stronger.

Complied with.