Response to reviewers

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

Reviewer-2: ACP-2019-376-RC2

This paper describes measurements of black carbon (BC) containing particles and of non-refractory PM1 over 9-10 months at an urban site on the east coast of India. The data show some interesting seasonal trends and are worth publishing.

We thank the reviewer and appreciate the summary evaluation of the merit of our work.

However, the paper itself needs significant work. There are many missing words, misused words, confusing sentences and repetitive sections. It should not be such hard work to figure out what the authors are trying to say. Since the paper has several UK co-authors, I am surprised that it was not carefully edited by a native English speaker before submission. This paper needs major revisions before it is accepted for publication.

We are sorry. Now we have revised the manuscript thoroughly, considering all the suggestions of the reviewer, including those on the language.

Specific comments:

1- The introduction is rather repetitive and does not mention the second instrument, the ACSM. Please tighten up the language.

Complied with. Also commented by reviewer 1. We have removed the reduntant statements and also added the details of the second instrument. The following sentences are included in the revised manuscript.

Page5, 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)."

Page5, 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 characterisation 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."

Page5: 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."

2- page 2, lines 17 -25. It would be helpful to mention which months are included in each season.

Complied with.

3- page 2, lines 28 - 33: The statement "The diurnal pattern of sulphate resembled that of the RCT" and the statement "the coating on BC showed a negative association with sulphate" contradict each other. The association plot in Figure 11 is an interesting way to present the data, but the conclusions are tenuous and don't belong in the abstract. I would delete the sentence starting "Though the pre-monsoon: : :" and ending ": : :mixing state of BC."

Complied with. The sentence is deleted.

The following sentence is added in the revised manuscript (Page 2, Line29) "Seasonally, the coating on BC showed a negative association with the mass concentration of sulphate during the pre-monsoon season and with organics during the post-monsoon season."

4- page 4, line 6: VOCs are not an aerosol species.

Agreed. We have rewritten the sentence.

"…phosphates, and secondary organic aerosols (SOA) originating from volatile organic compounds (VOC)".

5- page 4, line 19: A diameter of a few tens of nanometers is more likely for the primary spherules than for the chain agglomerates. Please cite a reference or correct the text.

Complied with. We have included the following references in the revised manuscript. (Page 4, Line 17)

References:

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.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Karcher, 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, J. Geophys. Res. Atmos., 118, 5380–5552, doi:10.1002/jgrd.50171, 2013.

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.

6- *page 4, line 27: Paragraph starts with the word "These," but it is not clear what "these" is referring to.*

This sentence is modified in the revised manuscript as below:

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

7- *page 6, line 9: Figure 1b can be removed. No-one needs to see a picture of a shipping container.*

Complied with. Revised figure 1 is shown below:

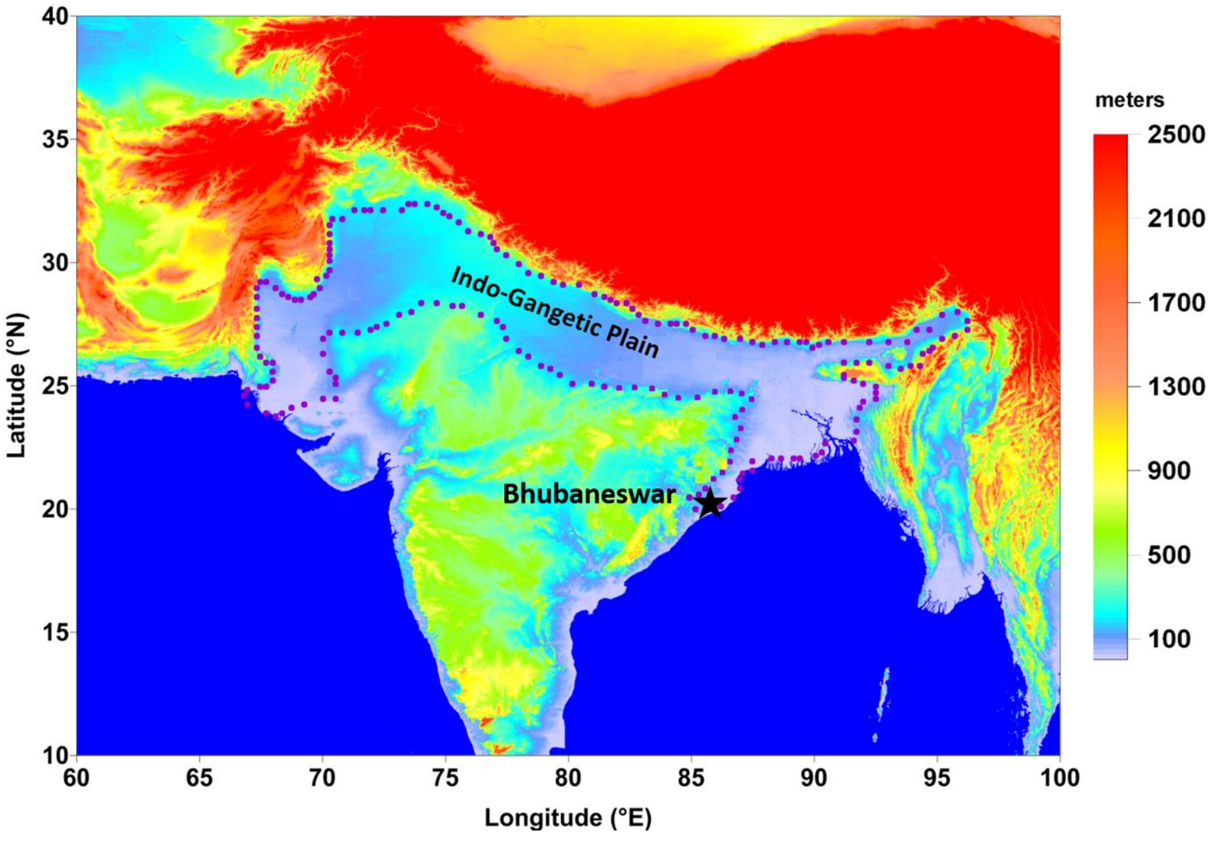


Figure 1: Geographic location of Bhubaneswar marked by a star symbol on the topographic map; the boundary of the Indo-Gangetic Plains (IGP) region is indicated with dotted lines.

8- page 6, line 11: Example of poor English usage "away from the proximity of." Just say "not near."

Complied with.

9- page 6, line 25: You refer to Figure S1, but the extra figures are in an appendix. Please label extra material consistently.

Complied with. Supplementary figures are renamed as Figure S1, S2 and so on in the revised manuscript.

10- page 6, line 30: I don't understand what the words in parentheses are conveying. If you mean that there are more fire events in those regions, please rephrase the sentence.

Complied with. The sentence is now rephrased and split into simpler sentences as below: (Page 6, Line 30)

"Figure S1 depicts the seasonal variation in the distribution of fires. The greatest number of fire events across the Indian region occur during the PMS. However, during other seasons, less intense fires are noticeable at the sub-regional scale, which are confined mostly to the northwest IGP during the PoMS, and to western, northeastern regions during winter".

11- page 7, lines 10-14: What about the 6-week gap in November-December? That is more than a brief gap and needs some explanation.

It has been explained in the revised manuscript as below:

"Only major gap in the data occurred during 11-November to 27-December, 2016, when measurements were paused due to logistical issues at the experimental site."

12- page 7, lines 17-18: Why mention instruments that are not relevant to this study?

Complied with. These sentences have been removed in the revised manuscript.

13- page 7, lines 31 – page 8, line 4: What do you mean by scattering enhancement? I think you are deriving the optical diameter of the coated particle from the scattering signal using a Mie Scattering model, but this description is garbled. Please phrase this more clearly.

Complied with. This discussion is rephrased in the revised manuscript as below:

"This signal is reconstructed using the leading edge only (LEO) fitting technique, which uses the leading edge of the unperturbed scattering signal before volatilization of the coating material becomes significant. This is used to reconstruct the full scattering signal (Liu et al., 2014). The reconstructed scattering signal and the BC core size (D_c) are used to derive the optical diameter of the BC particle or the coated BC size (D_p) by employing Mie calculations, where the whole particle is idealized as a two-component sphere with a concentric core-shell morphology".

14- page 8, line 15: Specify that this size range is vacuum aerodynamic diameter. The range is really more like 80 nm – 800 nm.

Complied with.

15- page 8, lines 19-20: It really doesn't seem necessary to discuss the pumps on the ACSM.

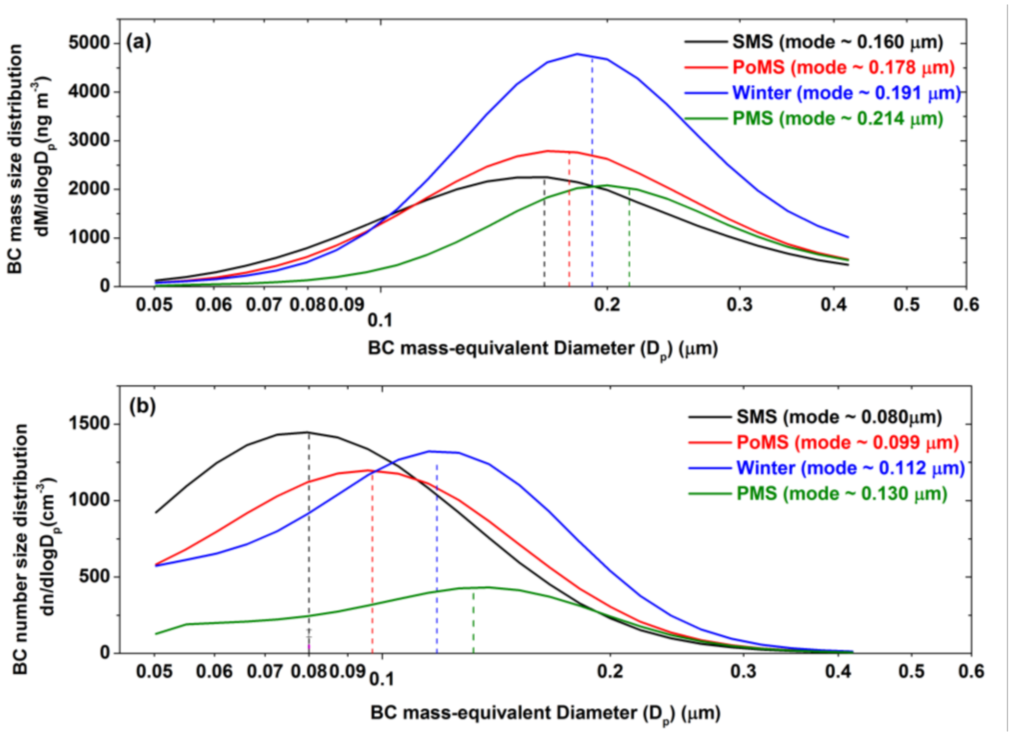
Complied with.

16 – page 10, lines 6-7 and elsewhere: Please pick one term, either counts or number and use it consistently throughout the paper including the figures.

Complied with. We have used 'number' instead of 'count' and count median diameter (CMD) is modified to number median diameter (NMD) throughout the manuscript, including figures.

17- page 10, first paragraph: Why is there a distinct jog at 40 nm in the number size distributions?

Thanks to the reviewer for this observation. In the present data analysis, the sum of the masses of all the single particle rBC detected formed the total rBC mass loading. A certain amount of rBC mass exists at core sizes, too small to be detected by the SP2, or too large, thus saturating the detector. In the present analysis, masses of such BC particles are predicted based on the extrapolation of a log-normal fit on the D_c mass distribution (Liu et al., 2014). The values below 50 nm are obtained from such extrapolation and in the revised manuscript, the particles with D_c < 50 nm are omitted, and the figure is modified (Supplementary figure S2).



18- page 16, line 16: What does "evolving least-squares fitting" mean?

This sentence has been modified in the revised manuscript as below:

"These size distributions were parameterized by least-squares fitting to an analytical monomodal log-normal distribution".

19- pages 10-11: I find this discussion extremely confusing and repetitive. The most important data is displayed in Figures 3 and 4 (and Table 4). I do not understand the point of averaging the size and number distributions over a season, taking the mode and getting a slightly different number than the average of the mode for individual data points. This does not add any new information and leads to repetitive discussion of the results. I would remove Figure 5, Table 2 and the associated discussion. Similarly, with the peak of the seasonal number size distributions – what new information does this give you beyond what you already know from the BC mass loading?

We partly agree. The idea of averaging over seasons is to provide inputs into models being developed in our own group and elsewhere, where the seasonal averages are needed. However, the repetitive discussions are avoided, and Figure 5 (and Table 2) is moved to supplementary section as supplementary figure S2 (and supplementary table-1) in the revised manuscript.

The discussion about previous work is split between page 10, lines 18-22 and page 11, line 29 – page 12, line 4. Please consolidate.

Complied with.

Finally, if you think you can tell the difference between local emissions during SMS with a smaller size and continental outflow with a larger size, why not make two entries in Table 3 for this study?

Complied with. These entries are made in the Table (Table-2 in the revised manuscript).

20- Pages 12-13: The discussion of RCT and ACT is confusing and repetitive. You are making a major assumption of core-shell morphology in order to calculate Dp and therefore RCT and ACT. Really all you can say is that you have a ratio that represents the amount of non-BC material associated with BC – you don't know the morphology of the particles nor how it changes with season. Morphology is likely to be quite different between fresh emissions during SMS and aged emissions during other seasons. I would not interpret RCT and ACT as literal diameters and coating thicknesses. In fact, I do not think ACT adds to the discussion. I would rewrite this section to present only the RCT data and include enough caveats that it is clear RCT is a representation, not an actual ratio of diameters.

We agree with the reviewer. It is correct that both RCT and ACT are used to represent the amount of non-BC material associated with BC and not an actual ratio of diameters. The information on the morphology of the BC, which may vary seasonally, is not available during our study period. The morphology would also be different for fresh and aged emissions. The coating thickness for individual particles is dependent on core sizes. However, the coating parameters estimated here are the bulk coating thicknesses in a given time window. It is calculated as the total volume of coated BC particles divided by the total volume of the rBC cores following Liu et al., (2014), which was used by subsequent studies (Liu et al., 2019, Brooks et al., 2019). As described by Liu et al., (2019), as the contribution from smaller particles to the integrated volume is very less, the bulk coating thickness values are generally independent of the uncertainties arising due to the presence of smaller particles. Some of the studies reported coating on rBC in terms of absolute coating thickness (ACT) in nm (Gong et al., 2016; Li et al., 2019; Cheng et al., 2018; Zanatta et al., 2019). Thus we used both these parameters for comparison of our values with the other regions. The RCT and ACT come from derived parameters that require Mie calculations based on a core-shell model that may not bear relation to reality. The caveat is that we assume the morphology of the particles ; they are

spherical and coating is uniform (coated particle also is spherical). We agree that this is an over-simplification and the true morphology could be different. The RCT (and ACT) parameter provides a qualitative measures of the amount of condensed material that is present on the same particle as the rBC core. We are using this to examine the extent of rBC mixing with other components in different seasons and compared to different regions. Further, using correlations with the bulk NR-PM1 composition, we can obtain some insights into the coating material associated with rBC in different periods. Thus, we have used both the volume-weighted bulk RCT (D_p/D_c) and ACT ($(D_p-D_c)/2$) in this study as representative diagnostics for the overall mixing state of the whole population of BC particles. We have modified the text in the revised manuscript to reflect the above discussion.

Page 8; Line 7

"These are calculated as the total volume of coated BC particles divided by the total volume of the rBC cores in a given time window (5 minutes) following Liu et al., (2014), which has been used by subsequent studies (Liu et al., 2019, Brooks et al., 2019a). It may be noted that the RCT and ACT used in this study come from derived parameters that require Mie calculations based on a core-shell model that may not bear relation to reality, and the RCT (and ACT) is not an actual ratio of diameters. The coating thickness for individual particles is dependent on core sizes. However, we have used the volume-weighted bulk RCT and ACT as representative diagnostics for the overall mixing state of the whole population of BC particles (Gong et al., 2016; Cheng et al., 2018; Liu et al., 2019). As described by Liu et al., (2019), since the contribution from smaller particles to the integrated volume is very less, the bulk coating thickness values are generally independent of the uncertainties arising due to the presence of smaller particles. Further, the information on the morphology of the BC, which would be different for fresh and aged emissions, is not available in this study.. The important caveat here is that we assume the morphology of the particles; they are spherical and coating is uniform (coated particle also is spherical). The RCT (and ACT) parameter provides a qualitative measure of the amount of condensed material that is present on the same particle as the rBC core. We are using this to examine the extent of rBC mixing with other components in different seasons and compared to different regions. Further, using correlations

with the bulk NR-PM1.0 composition, we intend obtain some insights into the coating material associated with rBC in different periods".

References:

Brooks, J., Liu, D., Allan, J. D., Williams, P. I., Haywood, J., Highwood, E. J., Kompalli, S. K., Babu, S. S., Satheesh, S. K., Turner, A. G., and Coe, H.: Black carbon physical and optical properties across northern India during pre-monsoon and monsoon seasons, Atmos. Chem. Phys., 19, 13079–13096, https://doi.org/10.5194/acp-19-13079-2019, 2019.

Cheng, Y., Li, S.M., Gordon, M., and Liu, P.: Size distribution and coating thickness of black carbon from the Canadian oil sands operations. Atmos. Chem. Phys., 18, 2653–2667, 2018.

Li, K., Ye, X., Pang, H., Lu, X., Chen, H., Wang, X., Yang, X., Chen, J., and Chen, Y.: Temporal variations in the hygroscopicity and mixing state of black carbon aerosols in a polluted megacity area, Atmos. Chem. Phys., 18, 15201–15218, https://doi.org/10.5194/acp-18-15201-2018, 2018.

Liu, D., Joshi, R., Wang, J., Yu, C., Allan, J. D., Coe, H., Flynn, M. J., Xie, C., Lee, J., Squires, F., Kotthaus, S., Grimmond, S., Ge, X., Sun, Y., and Fu, P.: Contrasting physical properties of black carbon in urban Beijing between winter and summer, Atmos. Chem. Phys., 19, 6749–6769, https://doi.org/10.5194/acp-19-6749-2019, 2019.

Zanatta, M., Laj, P., Gysel, M., Baltensperger, U., Vratolis, S., Eleftheriadis, K., Kondo, Y., Dubuisson, P., Winiarek, V., Kazadzis, S., Tunved, P., and Jacobi, H.-W.: Effects of mixing state on optical and radiative properties of black carbon in the European Arctic, Atmos. Chem. Phys., 18, 14037–14057, https://doi.org/10.5194/acp-18-14037-2018, 2018.

I also don't understand the point of Figure 7. You already have the information about the width of the distributions in Table 4. You mention multiple maxima, but have no interpretation. The discussion of Figure 7 on page 13 repeats the same information about sources and processing as on page 12, making this section very repetitive. I would remove Figure 7 and the associated discussion.

We have complied with the reviewer's suggestion. Figure 7 has been moved to supplementary information in the revised manuscript (Supplementary Figure S3) and the repetitive discussion is removed.

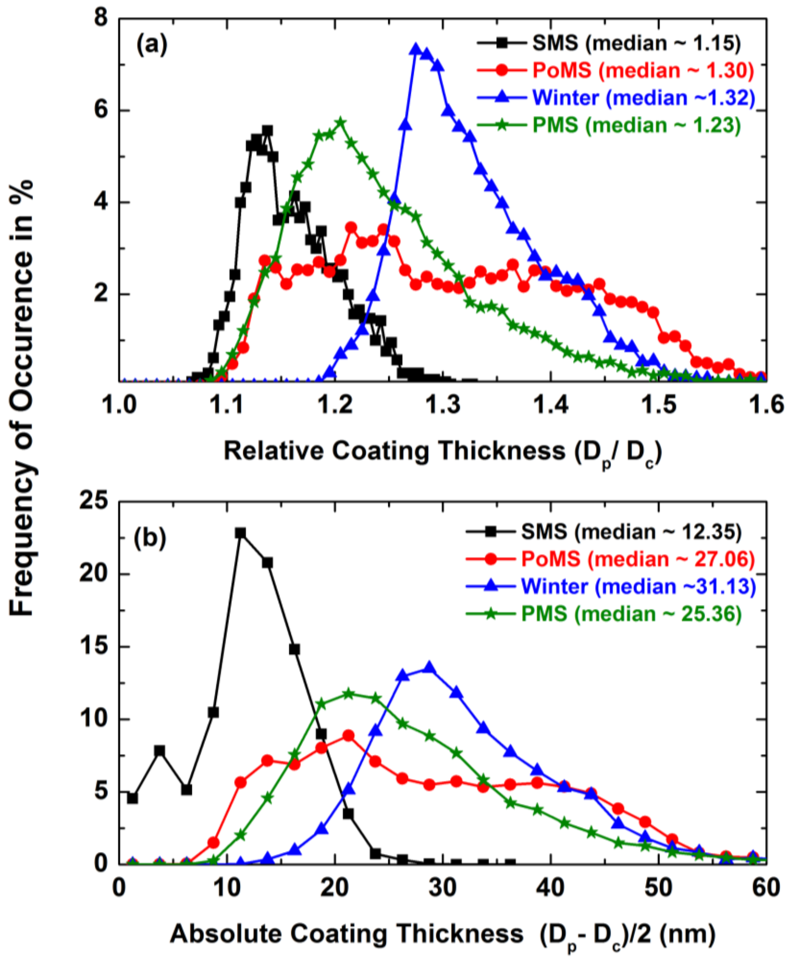
21- Page 12, lines12-14: "Both of these parameters: : :mixing state of the particles." I do not understand this sentence. Does "both" refer to RCT and ACT or to Dp and Dc? Either way, how can Dp not depend on the mixing state?

Yes. Both refer to RCT and ACT. We have modified the sentence in the revised manuscript accordingly.

22- Page 13, lines 1-2: "Intra-seasonal variability: : :Figure 6) is also higher during PoMS." I do not understand this sentence. Figure 6 shows daily values, not seasonal values. By eye, the variation in the daily points and the spread of the error bars looks very similar across PoMS, Winter and PMS. There are very few points during SMS, so it is hard to draw conclusions for that season.

The sentence is modified in the revised manuscript.

"Intra-seasonal variability (as highlighted by the wide range of frequency of occurrence of RCT and ACT values during the PoMS seen in the supplementary figure S3) is also higher during the PoMS".



23- Pages 15-17, Section 3.5: I have several questions about the ACSM data analysis. Why is ammonia so low? Was the aerosol not neutralized and do you have corroborating evidence? Or was the RIE_NH_4 incorrect for this instrument? Why not estimate OOA and HOA using the parameterization in Ng et al. (EST 2011)? This would give you additional information about local and regional sources.

We agree with the reviewer that the estimation of OOA and HOA provides information on the nature of sources. Detailed factorization of organics forms the scope for the future study, and is not attempted here. Periodic ionization efficiency calibrations were performed using ammonium nitrate, ammonium sulphate, and corresponding RIE_NH_4 values were updated in the DAQ of the ACSM. The reviewer made a good observation about the concentration of ammonium. We have estimated the aerosol neutralization ratio (ANR) (this information is not available in the present manuscript) in terms of the ACSM measured (m) NH_4^+ to predict (p) NH_4^+ ratio for different seasons and found a seasonal variability in the ANR values indicating ammonium deficit to fully neutralized aerosol system. A detailed analysis on this is being carried out. Earlier, Mahapatra et al., (2013) estimated chemical compostion of total suspended particulate (TSP) matter at Bhubaneswar using year-round filter-based sampling and have reported that both the acidic and basic components have significant seasonal variability. From the recent filter-based offline chemistry data there is evidence for seasonally varying ANR which indicated dominance of acidic (NO_3^- and SO_4^-) over basic (NH_4^+, Mg^{2+} and Ca^{2+}) atmospheres in different seasons at Bhubaneswar (unpublished data). One of the reasons for ammonium deficiency is possible heterogeneous reactions during the presence of a high number of pre-existing large particles and very high concentrations of acidic species (Pathak et al., 2009; Hsu et al., 2014). Collocated measurements of number size distributions of ultrafine and fine particles during the present study period at the site have also revealed the absence of new particle events due to high condensation sink (unpublished data) corroborating the above. The consolidated analysis of the aerosol chemistry from a combination of the size segregated off-line and online data methods is in progress to understand these aspects in detail.

References:

Mahapatra, P.S., Ray, S., Das, N., Mohanty, A., Ramulu, T.S., Das, T., Chaudhury, G.R., Das, S. N.: Urban air-quality assessment and source apportionment studies for Bhubaneshwar, Odisha, Theor.Appl. Clim.,112,243-25, 2013.

Hsu, S.-C., C. S. L. Lee, C.A. Huh, R. Shaheen, F.-J. Lin, S. C. Liu, M.-C. Liang, Tao, J.: Ammonium deficiency caused by heterogeneous reactions during a super Asian dust episode, J. Geophys. Res. Atmos., 119, 2014, 6803–6817, doi:10.1002/2013JD021096.

Pathak, R.K., Wu, W.S., Wang, T.: Summertime PM2.5 ionic species in four major cities of China: nitrate formation in an ammonia-deficient atmosphere, Atmos. Chem. Phys., 9, 1711–1722, 2009. DOI: 10.5194/acp-9-1711-2009.

24- Page 17, lines 1-7: "Even though: : :coating." I do not understand this sentence, partly because it is too long and convoluted, but also because the two parts contradict each other. You say in part a that concurrent peaks in RCT and sulphate suggest that sulphate is mixed with BC, but in part b, you say the opposite. You can't have it both ways. Or are you saying that the ACSM detects sulphate when it is mixed with BC, but not organic? That does not make sense.

This confusion has been cleared. We have rewritten this in the revised manuscript as below: (Page17, Line1)

"It is challenging to determine the exact coating material on the atmospheric BC particles in a multi-component system containing organic and inorganic aerosols, and gaseous vapours. The association between the diurnal variations of organics and sulphates and BC mixing state as represented by RCT presents two possibilities of having different coating material on BC during a day. Similar diurnal variations in RCT (as seen in Figure 6) and sulphate suggest the possibility of sulphate serving as the most probable material. However, organic matter can also contribute to the BC coating material due to its huge abundance in particles of submicron sizes. This is particularly true during the late evening periods, when concurrent peaks in the mass fraction of organics and rBC mass loading occur, a significant fraction of which could be secondary in nature. The extent of contribution of each species depends on processes such as gas-phase chemistry and production of condensable vapours and strength of the condensation sink".

25- Pages 17-18, Section3.7: Figure 11 is another way of comparing the diurnals for RCT and MF. While it is a nice visualization, I don't think it needs a new section repeating much of the discussion as in Section 3.6. I would combine the discussion in Sections 3.6 and 3.7.

Complied with. We have combined the sections 3.6 and 3.7, which describe the association between rBC relative coating thickness and NR-PM1 chemical species in diurnal and seasonal scales.

I also wonder if you have thought about the fraction of particles containing BC (i.e., BC number con./(BC num conc. + scatt num conc.))? This fraction is much higher in SMS than in PoMS or Winter and is lowest in PMS. The low value in PMS might be part of the reason that the association in Figure 11d and h is so poor since there is less overlap between the particle population detected with the ACSM and the population detected with the SP2.

Agreed with thanks. We have modified the text in the revised manuscript to include this possibility. (Page18, Line 28)

"It may be noted that it is difficult to decipher the exact coating on BC with the present approach, since the SP2 retrieves black carbon mass and provides a measure of co-existing material within the same particles (as measured by RCT) whereas the ACSM measures the mass of refractory material in the total submicron population. An examination of coating material can only be directly achieved by employing the instruments such as the soot particle aerosol mass spectrometer (Aerodyne SP-AMS) (Liu et al., 2018). However, the SP2 can determine both the rBC content of single particles and the optical size by light scattering for diameters between 200 and 400 nm. The coating thickness estimated within this range represents most of the particles which contribute significantly to the light extinction. A comparison of the proportion of rBC containing particles within the total population as a function of season sheds some light on interpreting variation throughout the year. In our study, the fraction of particles containing BC, i.e., the ratio of BC number concentration and total number concentration (BC number concentration + scattering number concentration) showed a clear seasonal variation. The fraction of BC containing particles was highest during the SMS (mean ~ 0.69 ± 0.11) and decreased through winter (~0.44 ± 0.16), PoMS (~0.36 ± 0.11) to reach the lowest value (~0.25 ± 0.10) during the PMS. This shows a gradual decrease in the overlap between the particle population detected with the ACSM and the population detected with the SP2 with changing seasons from SMS to PMS. This should be

borne in mind while examining the association between the ACSM detected particle mass concentrations and the SP2 derived coating parameters."

Reference:

Liu, D., Taylor, J. W., Crosier, J., Marsden, N., Bower, K. N., Lloyd, G., Ryder, C. L., Brooke, J. K., Cotton, R., Marenco, F., Blyth, A., Cui, Z., Estelles, V., Gallagher, M., Coe, H., and Choularton, T. W.: Aircraft and ground measurements of dust aerosols over the west African coast in summer 2015 during ICE-D and AER-D, Atmos. Chem. Phys., 18, 3817–3838, https://doi.org/10.5194/acp-18-3817-2018, 2018.

26- Figure 1: It is very hard to see the circle indicating the IGP in panel (a). I would delete panel (b). There is no need for a picture of a shipping container.

Complied with. Panel (b) has been deleted, and Figure 1 has been modified in the revised manuscript.

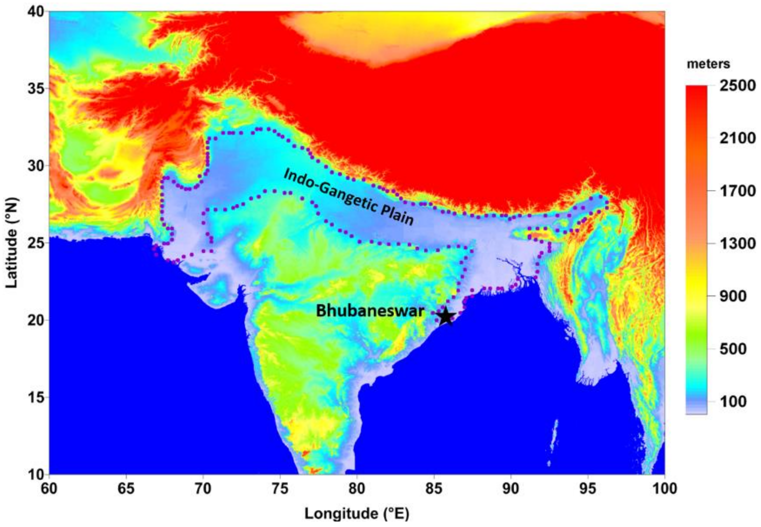
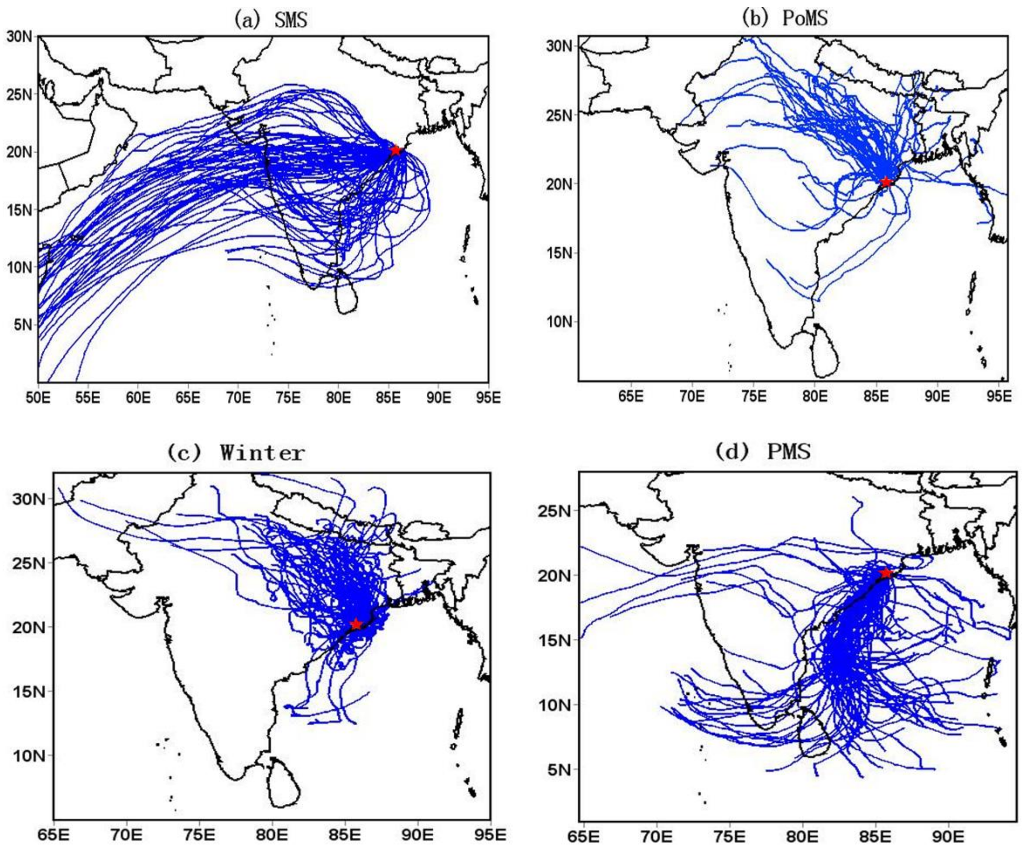


Figure 1: Geographic location of Bhubaneswar marked by a star symbol on the topographic map; the boundary of the Indo-Gangetic Plains (IGP) region is indicated with dotted lines.

27- Figure 2: The star symbol is not visible.

Figure 2 has been updated in the revised manuscript.



28- Figure 3: Indicate the seasons in panels (a) and (b).

Figure 3 has been updated in the revised manuscript as per the suggestion.

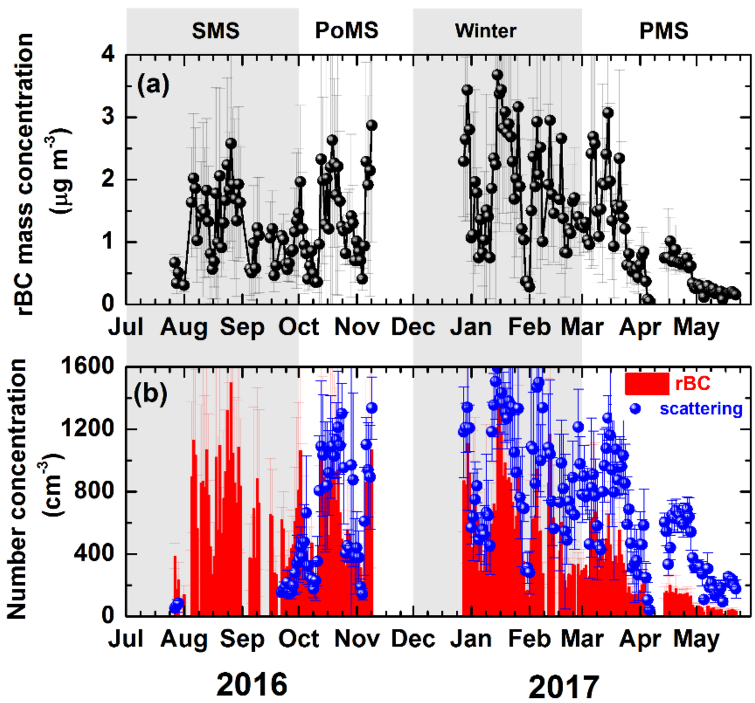
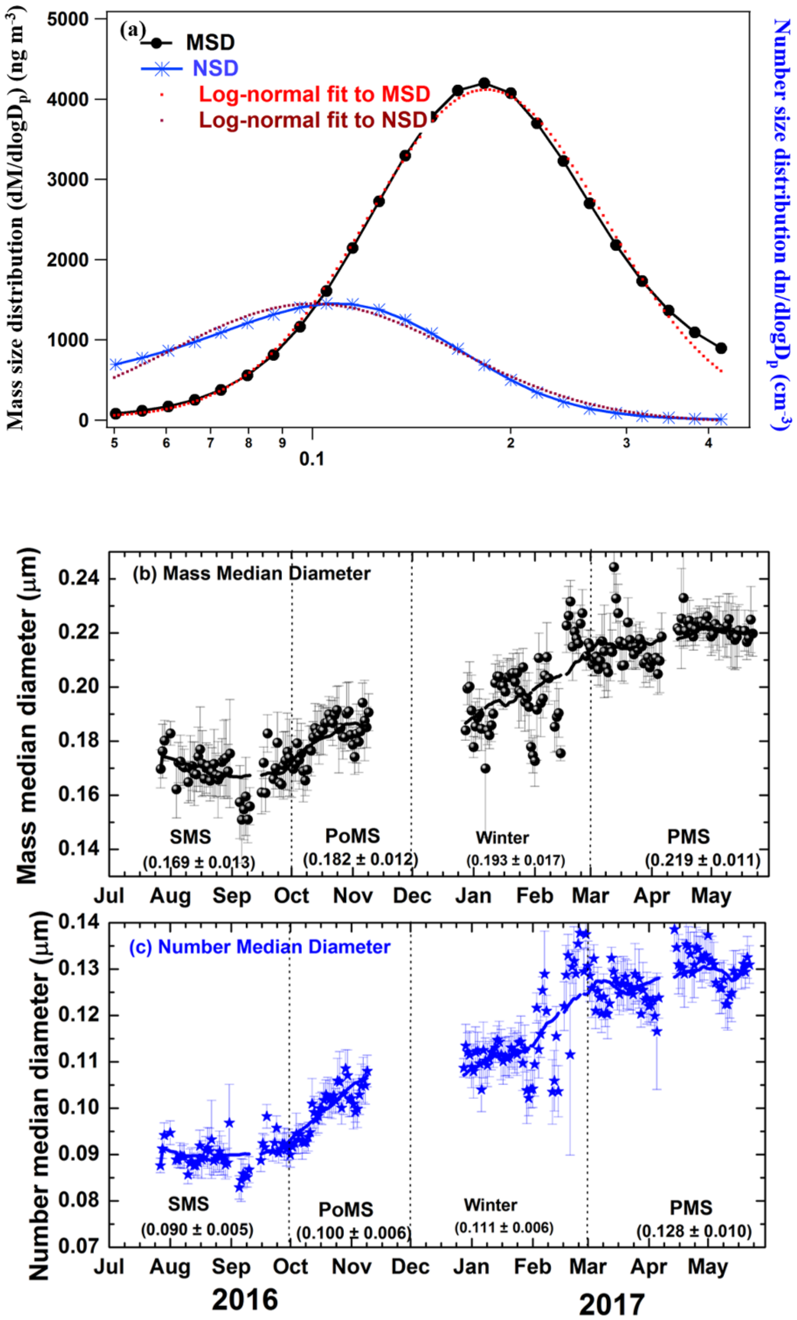


Figure 3: Temporal variation of daily mean (a) r_{BC} mass concentration; and (b) number concentration of BC (bars) and non-BC scattering particles (filled circle). The vertical line passing through them is the standard deviation. The shaded portions demarcate the seasons.

29- Figure 4: Include the dashed lines in the legend. Panel (b) has circles not triangles. Please use either number or count, but not both.

Complied with. Figure 4 is modified in the revised manuscript.



Do you have any data covering the gap between end of May and August? Do the MMD and CMD really drop from PMS to SMS values over 6 weeks? Or could you have some kind of instrumental drift that causes both to increase over the displayed 10 months of data?

Unfortunately, no data is available covering the gap between the end of May and August during the present study period due to technical issues with the SP2 optics (a drop in the SP2 laser power due to contamination of the optics owing to heavy particle loading). We rule out any instrumental drift as it has been periodically calibrated to account for any variation in the laser power and detector response. Notably, the present MMD values during the PMS are consistent with the values reported by Brooks et al., (2019) based on the aircraft experiments over the same region. They have reported that core MMD values which were 0.22 µm during the PMS (flights on 11–12 June 2018) dropped down to 0.20 µm with the onset of monsoon (flights on 30 June–11 July 2018; which were temporally just 2-4 weeks away from the pre-monsoon flights). They attributed it to change in the nature of air masses. The large scale changes in the air mass characteristics combined with the widespread precipitation across the Indian region associated with monsoon circulation contributes to changes in both the nature and strength of BC sources.

Reference:

Brooks, J., Liu, D., Allan, J. D., Williams, P. I., Haywood, J., Highwood, E. J., Kompalli, S. K., Babu, S. S., Satheesh, S. K., Turner, A. G., and Coe, H.: Black carbon physical and optical properties across northern India during pre-monsoon and monsoon seasons, Atmos. Chem. Phys., 19, 13079–13096, https://doi.org/10.5194/acp-19-13079-2019, 2019.

30- Figure 11: "Speices" is mis-spelled in the x-axis label.

It has been corrected in the revised manuscript.

31- Table 1: "metrological" should be "meteorological"

It has been corrected in the revised manuscript

32- Table 3: "Shangai" is mis-spelled.

It has been corrected in the revised manuscript

33- Table 4: Are these averages of the daily values shown in the figures, or averages of all the underlying data? Please specify. Also, somewhere in the text you should state the time-base of the SP2 data.

The values tabulated are averages of all the underlying data. Table-4 Table-3 in the revised manuscript) has been slightly modified as per the suggestion from reviewer-1. Time-base of the processed SP2 data is 5 minutes, and it is specified in the revised manuscript.

34- Figure A1 (or S1?): Please decide if this section is an appendix or supplemental information.

In the revised manuscript, Figure S1, S2, S3 and S4 are available as supplementary figures, and the corresponding discussion is available as supplementary information.

Please label the panels with the season. It's not clear what the words in parentheses in the last sentence are supposed to mean.

Complied with. Now labels are added in Figure S1. Figure caption has been revised.

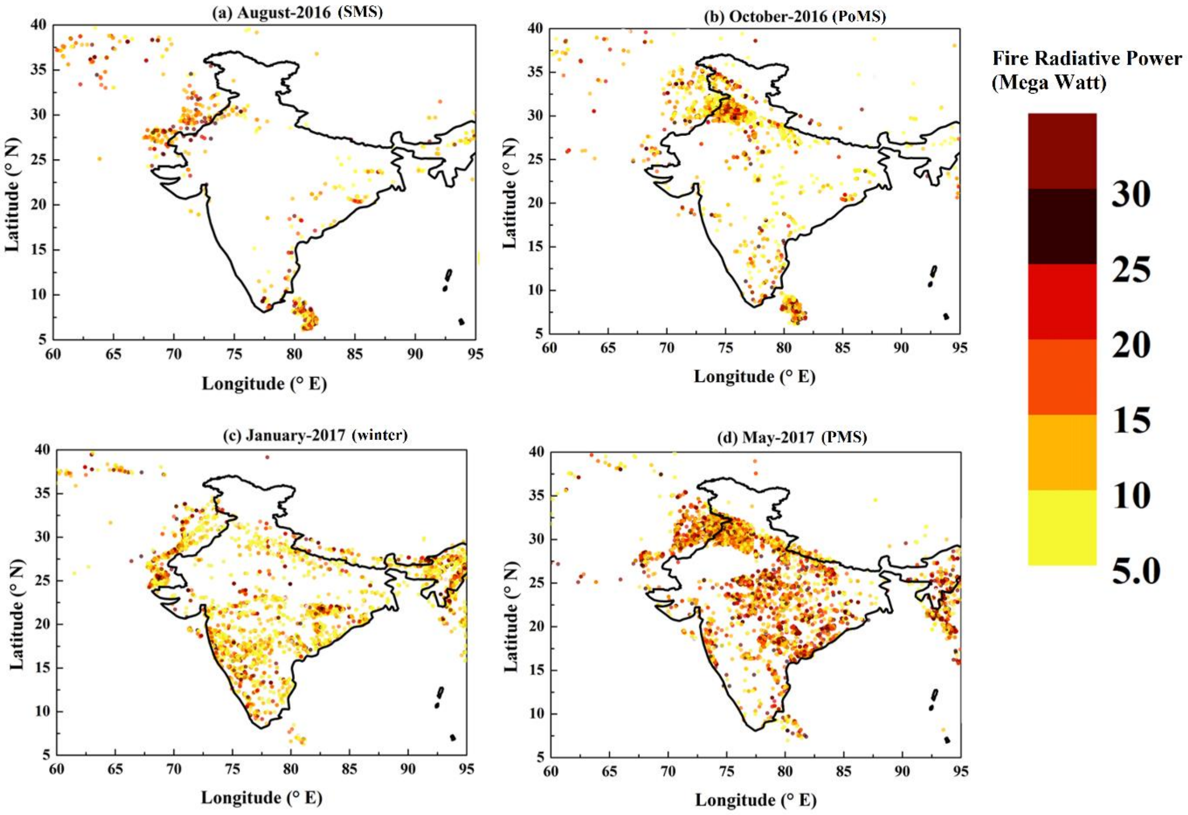


Figure S1: Spatial distribution of Moderate Resolution Imaging Spectroradiometer (MODIS) fire radiative power (MODIS Thermal Anomalies / Fire locations Collection 6 product obtained from https://earthdata.nasa.gov/firms) for the representative months of different seasons; (a) August -2016 (SMS), (b) October -2016 (PoMS), (c) January -2017 (winter) and (d) May -2017 (PMS). A significant amount of fire events during PMS are seen over the Indian region. During the PoMS (fire events to confined to northwest IGP) and winter (fire events to confined to western, northeastern regions of India) less intense regional fire events are noticeable. During SMS (and PoMS as well), a considerable amount of fire events are noticeable below south of India (over Srilankan region).