Response to reviewers

We thank both reviewers for their constructive and comprehensive comments, which, as outlined below, have helped improve the manuscript. This document outlines the review comments in *plain italics***, followed by the author's response in bold, and the tracked changes in the main texts are in blue.**

Reviewer-1: ACP-2020-836-RC1

The manuscript details the analysis of SP2 data collected aboard a research ship as part of the ICARB-18 campaign with a focus on the microphysical properties of refractory black carbon (rBC)-containing particles. As the authors point out, the SP2 detects individual rBC particles through laser-induced incandescence thereby providing mass loadings and size/mass distributions. By combining the incandescence channel with the co-located SP2 scattering channel, the mixing state of individual rBC particles can also be probed. Their analysis reveals that outflow regions are characterized by higher rBC loadings and more thickly coated rBC-containing particles with a decrease in coating thickness in the oceanic regions. Studies such as this are essential in filling in measurement and information gaps in the South Asian region and thus are important for improving our quantitative understanding of the radiative forcing impacts of BC aerosols in this region. Despite the over use of very long sentences, the manuscript is clearly written. The manuscript shares a lot similarity - with a couple of notable exceptions - to that published by Brooks et al., who conducted an aircraft-based study of northern India. This Reviewer has a concern that the authors are, in some instances, over interpreting data collected by a single instrument and, in doing so, drawing potentially incomplete (or erroneous) conclusions - as exemplified by their use of rBC number fraction (which is discussed below). Therefore, it is recommended that the manuscript be revised as per comments below and resubmitted. To be clear, the findings contained in this manuscript are of good value and need to be published, but in its current state, the manuscript is not quite ready.

We thank the reviewer for the summary, the positive recommendation, and the comments for improvement. We have revised the manuscript as detailed below.

As alluded to above, 90% of this manuscript - based on rough estimate of pages dedicated to measurement type - involves only data collected by the single particle soot photometer (SP2). (The remaining 10% is on aerosol composition via an ACSM.) While the SP2 in an incredibly sensitive instrument towards rBC particles and its mixing state, it is not sensitive to changes in rBC aggregate morphology - that is rearrangement of a fractal aggregate to one that is more collapsed - and it suffers from limited detection sensitivity towards pure scatterers (i.e., non-BC particles).

We agree. We have included the following discussion in the revised manuscript.

Page 7, Line 22:

"It is recognized that the SP2 cannot provide the details of rBC aggregate morphology or the relative position of the BC within the particle, which can be determined better through microscopy-based studies (e.g., Adachi et al., 2010; Ueda et al., 2018). However, the intensity of the incandescence signal detected by the SP2 is proportional to the refractory black carbon mass present in the particle and is independent of particle morphology and mixing state (Slowik et al., 2007a; Moteki and Kondo, 2007; Schwarz et al., 2008). Again, though the SP2 has limited detection sensitivity towards pure scatterers because of the limited size range it covers, the light scattering information at 1064 has been widely used to accurately derive the size of the coated particle (Gao et al., 2007; Moteki et al., 2010; Shiraiwa et al., 2008; 2010; Laborde et al. 2013; Taylor et al., 2015; Liu et al., 2017)."

References:

Adachi, K., S. H. Chung, and P. R. Buseck (2010), Shapes of soot aerosol particles and implications for their effects on climate, J. Geophys. Res., 115, D15206, doi:10.1029/2009JD012868.

Laborde, M., Crippa, M., Tritscher, T., Jurányi, Z., Decarlo, P. F., Temime-Roussel, B., Marchand, N., Eckhardt, S., Stohl, A., Baltensperger, U., Prévôt, A. S. H., Weingartner, E., and Gysel, M.: Black carbon physical properties and mixing state in the European megacity Paris, Atmos. Chem. Phys., 13, 5831–5856, doi:10.5194/acp-13-5831-2013, 2013.

Moteki, N. and Kondo, Y.: Effects of mixing state on black carbon measurements by laser-induced incandescence, Aerosol Sci. Technol., 41, 398–417, doi:10.1080/02786820701199728, 2007.

Moteki, N., Kondo, Y., and Nakamura, S.: Method to measure refractive indices of small nonspherical particles: Application to black carbon particles, J. Aerosol Sci., 41, 513–521, doi:10.1016/j.jaerosci.2010.02.013, 2010.

Schwarz, J. P., Spackman, J. R., Fahey, D.W., Gao, R. S., Lohmann, U., Stier, P., Watts, L. A., Thomson, D. S., Lack, D. A., Pfister, L., Mahoney, M. J., Baumgardner, D., Wilson, J. C., and Reeves, J. M.: Coatings and their enhancement of black carbon light absorption in the tropical atmosphere, J. Geophys. Res., 113, D03203, doi:10.1029/2007jd009042, 2008.

Shiraiwa, M., Kondo, Y., Moteki, N., Takegawa, N., Sahu, L. K., Takami, A., Hatakeyama, S., Yonemura, S., and Blake, D. R.: Radiative impact of mixing state of black carbon aerosol in Asian outflow, J. Geophys. Res., 113, D24210, doi:10.1029/2008jd010546, 2008.

Shiraiwa, M., Kondo, Y., Iwamoto, T., and Kita, K.: Amplification of Light Absorption of Black Carbon by Organic Coating, Aerosol Sci. Technol., 44, 46–54, doi:10.1080/02786820903357686, 2010.

Slowik, J. G., Cross, E. S., Han, J.-H., Davidovits, P., Onasch, T. B., Jayne, J. T., Williams, L. R., Canagaratna, M. R., Worsnop, D. R., Chakrabarty, R. K., Moosmueller, H., Arnott, W. P.,

Schwarz, J. P., Gao, R.-S., Fahey, D. W., Kok, G. L., and Petzold, A.: An inter-comparison of instruments measuring black carbon content of soot particles, Aerosol Sci. Technol., 41, 295– 314, doi:10.1080/02786820701197078, 2007a.

First a comment on rBC morphology. For a given mass, the SP2 will report the same incandescence intensity independent of the rBC particle fractal dimension. Therefore a statement such as "The observed narrow range of mean NMD (0.10-0.11 µm) and MMD (0.19- 0.20 µm) over the entire region (Fig. 3c and 3d) reveals that the prevailing BC over the entire study region has a mixed-source origin and the particles are aged during the long advection leading to transformation processes (such as collapsing of the BC cores)" is very misleading [page 10, lines 11-12]. The authors again put forth a similar line of reasoning in the conclusion [page 23, lines 4-5] "BC size distributions indicated highly mixed BC sources, likely including a combination of fossil fuel and solid fuel sources, along with the restructuring of the cores of the aged BC particles." The authors present no supporting data that there is any restructuring of the cores and thus cannot conclude that such processes are taking place. Indeed, there is lab studies which suggest that aggregate restructuring occurs through capillary condensation at the interstitial locations between primary particles and thus such restructuring would occur very early in the coating process. The authors are encouraged to review the work by the Khalizov group (e.g., Invanova et al., AST 2020 and references therein).

Yes, we agree. Complying with the comment, the discussions and conclusion are modified in the revised manuscript as given below.

Page 11, Line 12:

"The observed mean NMD (0.10-0.11 µm) and MMD (0.19-0.20 µm) values over the entire study region (Fig. 3c and 3d) are within the range of values reported in earlier studies for aged continental outflow and a combination of sources. Aging of BC is an important factor affecting the rBC core sizes owing to transformation processes (such as collapsing of the BC cores and/or due to coagulation) taking place during the long-range transport (Shiraiwa et al., 2008; Bond et al., 2013; Ko et al., 2020). Freshly produced BC particles comprise fractal-like aggregates of spherical graphitic monomers with diameters of 10-50 nanometers (Köylü et al., 1995; Bond and Bergstrom, 2006; Bond et al., 2013; Petzold et al., 2013). However, as they evolve in the atmosphere, restructuring of these aggregates occurs due to the above processes and/or condensation of vapors. Compaction can be induced by capillary forces while vapor condensation fills the voids of the aggregates (capillary condensation) (Weingartner et al., 1995; Pagels et al., 2009; Khalizov et al., 2009; Chen et al., 2018, 2016; Invanova et al., 2020 and references therein), and/or restructuring driven by surface tension forces at the solid-liquid

interfaces during condensation of coating material (Kutz and Schmidt-Ott, 1992; Slowik et al., 2007b; Zhang et al., 2008; Zhang et al., 2016; Schnitzler et al., 2017). Recently, Invanova et al. (2020) have presented a detailed account of the above processes. As such, increased aging is more likely to result in compact cores (Liu et al., 2019; Laborde et al., 2013); however, the effectiveness of a condensable vapor to cause restructuring depends on its chemical composition also (Xue et al., 2009; Chen et al., 2016).”

Page 25, Line 10:

“BC size distributions indicated a combination of sources and the BC core sizes corresponding to aged continental outflow”.

In this study, we did not attempt to delineate the strength of each of these processes or time scales within which the restructuring of the aggregates takes place during the aging of BC particles in the atmosphere.

The following references are added in the revised manuscript:

Chen, C., Enekwizu, O. Y., Fan, X., Dobrzanski, C. D., Ivanova, E. V., Ma, Y., Gor, G.Y., and Khalizov, A.F.: Single parameter for predicting the morphology of atmospheric black carbon. Environ. Sci. Technol. , 52(24), 14169–14179, DOI: 10.1021/acs.est.8b04201, 2018.

Chen, C., Fan, X., Shaltout, T., Qiu, C., Ma, Y., Goldman, A., and Khalizov, A. F.: An unexpected restructuring of combustion soot aggregates by subnanometer coatings of polycyclic aromatic hydrocarbons, Geophys. Res. Lett., 43, 11080–11088, https://doi.org/10.1002/2016GL070877, 2016.

Invanova, E.V., Khalizov, A.F., and Gor, G.Y.: Kinetic Model for Competitive Condensation of Vapor between Concave and Convex Surfaces in a Soot Aggregate, Aerosol Science and Technology, 2020. DOI: 10.1080/02786826.2020.1846677.

Khalizov, A. F., Zhang, R., Zhang, D., Xue, H., Pagels, J., and McMurry, P. H.: Formation of highly hygroscopic soot aerosols upon internal mixing with sulfuric acid vapor. J. Geophys. Res. Atmos., 114, D05208, 1-15, doi:10.1029/2008JD010595, 2009.

Ko, J., Krasowsky, T., and Ban-Weiss, G.: Measurements to determine the mixing state of black carbon emitted from the 2017–2018 California wildfires and urban Los Angeles, Atmos. Chem. Phys., 20, 15635–15664, https://doi.org/10.5194/acp-20-15635-2020, 2020.

Kompalli, S. K., Babu, S. S., Moorthy, K. K., Manoj, M. R., Kirankumar, N. V. P., Shaeb, K. H. B., and Joshi, A. .: Aerosol black carbon characteristics over central India: temporal variation and its dependence on mixed layer height, Atmos. Res., 147-148, 27–37, https://doi.org/10.1016/j.atmosres.2014.04.015, 2014.

Köylü, Ü.Ö., Faeth, G. M., Farias, T. L., and Carvalho, M. G.: Fractal and projected structure

properties of soot aggregates, Combust. Flame, 100, 621–633, https://doi.org/10.1016/0010-2180(94)00147-K, 1995.

Kütz, S., and Schmidt-Ott, A.: Characterization of agglomerates by condensation-induced restructuring. J. Aerosol Sci., 23, 357–360, https://doi.org/10.1016/0021-8502(92)90423-S, 1992.

Pagels, J., Khalizov, A. F., McMurry, P. H., and Zhang, R. Y.: Processing of Soot by Controlled Sulphuric Acid and Water Condensation Mass and Mobility Relationship, Aerosol Sci. Technol., 43, 629–640, https://doi.org/10.1080/02786820902810685, 2009.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S.-M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., and Zhang, X.-Y.: Recommendations for reporting "black carbon" measurements, Atmos. Chem. Phys., 13, 8365–8379, doi:10.5194/acp-13-8365-2013, 2013.

Schnitzler, E. G., Gac, J. M., and Jäger, W.: Coating surface tension dependence of soot aggregate restructuring. J. Aerosol Sci., 106, 43–55, 2017. https://doi.org/10.1016/j.jaerosci.2017.01.005.

Shiraiwa, M., Kondo, Y., Moteki, N., Takegawa, N., Sahu, L. K., Takami, A., Hatakeyama, S., Yonemura, S., and Blake, D. R.: Radiative impact of mixing state of black carbon aerosol in Asian outflow, J. Geophys. Res.-Atmos., 113, 1–13, https://doi.org/10.1029/2008JD010546, 2008.

Slowik, J. G., Cross, E. S., Han, J.-H., Kolucki, J., Davidovits, P., Williams, L. R., et al.: Measurements of morphology changes of fractal soot particles using coating and denuding experiments: Implications for optical absorption and atmospheric lifetime. Aerosol Sci. Technol., 41(8), 734–750, 2007b. https://doi.org/10.1080/02786820701432632.

Weingartner, E., Burtscher H. and Baltensperger U.: Growth and structural change of combustion aerosols at high relative humidity, Environ. Sci. Technol., 29(12), 2982-2986, 1995. https://doi.org/10.1021/es00012a014.

Xue, H. X., Khalizov, A. F., Wang, L., Zheng, J., and Zhang, R.Y.: Effects of Coating of Dicarboxylic Acids on the Mass-Mobility Relationship of Soot Particles, Environ. Sci. Technol., 43, 2787–2792, doi:10.1021/es803287v, 2009.

Zhang, Y., Zhang, Q., Cheng, Y., Su, H., Kecorius, S., Wang, Z., Wu, Z., Hu, M., Zhu, T., Wiedensohler, A., and He, K.: Measuring the morphology and density of internally mixed black carbon with SP2 and VTDMA: new insight into the absorption enhancement of black carbon in the atmosphere, Atmos. Meas. Tech., 9, 1833–1843, https://doi.org/10.5194/amt-9-1833-2016, 2016.

Zhang, R., Khalizov, A. F., Pagels, J., Zhang, D., Xue, H., and McMurry, P. H.: Variability in

morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing., Proc. Natl. Acad. Sci. USA, 105, 10291–10296, doi:10.1073/pnas.0804860105, 2008.

As highlighted above, this reviewer has a major issue with the authors operational definition of the "fraction of particles containing rBC" (which the authors define as the sum of the number concentration of BC and non-BC scattering particles detected by the SP2). Using the SP2 to measure the number concentration of non-BC will severely bias the derived number fraction of rBC particles because the scattering channel has a severely limited detection range (i.e., ~200 nm - 400 nm). In contrast, the incandescence channel has a nominal detection sensitivity range, in terms of a mass equivalent diameter, from about 80 nm to ~500 nm. In short, the SP2 severely undercounts pure scattering particles and therefore deriving a rBC number fraction using the SP2 scattering data will bias the rBC number fractions high and could very well mask the impact(s) that <200 nm non-BC particles might exert in differing regions. One would think (hope) that the instrument suite deployed aboard the research ship during the campaign had a CPC measurement, which would provide an excellent measurement of aerosol number concentration for particles ranging in diameter from ~10 nm to 1000 nm, thereby greatly improving the utility of examining the rBC number fraction. Given the presence of higher anthropogenic emissions nearest the coast, it is not unreasonable to expect sub-200 nm pure scattering particles to be abundant. If no CPC, or equivalent measurement, is available then the discussion of variation in fraction of particles containing rBC is meaningless and thus should removed.

We agree with the reviewer's point that t*he SP2 severely undercounts pure scattering particles, and therefore deriving an rBC number fraction using the SP2 scattering data will bias the rBC number fractions high and could very well mask the impact(s) that <200 nm non-BC particles might exert in differing regions.* We have performed additional analysis and revised the manuscript. During the present study, the total particle concentrations are available from the condensation particle counter (with a 1-minute sampling interval). The results on particle number size distributions and new particle formation events during the ICARB-2018 are available in Kompalli et al. (2020b). As suggested by the reviewer, the fraction of BC-containing particles was calculated using the total number concentrations from the SMPS, CPC, and rBC number concentration from the SP2. Accordingly, panel (b) of figure 5 and Table-3 have been modified in the revised manuscript. Also, discussions in the manuscript are revised as below to reflect the above points.

<u>Page 8, Line 10:</u>

"Continuous measurements of the particle number size distributions in size range 10 to 414 nm

have also been carried out aboard, at the 5-minute interval, using a scanning mobility particle sizer spectrometer (SMPS; TSI Inc., USA) during the campaign (Kompalli et al., 2020b). The SMPS consists of an electrostatic classifier (TSI 3080), a long differential mobility analyzer to size segregate the particles based on their electrical motilities (Wiedensohler 1988), which are subsequently counted by using a water-based condensation particle counter (TSI 3786). Concurrent measurements of the particle number size distributions in the aerodynamic diameters range 542 to 19800 nm (which can be converted to stokes diameters using an effective particle density) have also been made using the aerodynamic particle sizer (Make: TSI, Model: 3321) that works based on 'time-of-flight' technique (Leith and Peters 2003). Though the contribution from the particles in the sizes measured by the APS to the overall aerosol number concentrations is found to be < 2 %, combining both these measurements gives the total particle number concentrations covering a wide size range (10 -10000 nm). We have used the particle number size distributions (and total particle number concentrations) from 10-1000 nm from the SMPS+APS measurements, along with the number concentration of rBC from the SP2 to estimate the fraction of rBC-containing particles."

Page 16, Lines 21-22:

"The spatial variation of number concentration (in cm^{-3}) of non-BC (i.e., purely scattering) particles detected by the SP2 and the fraction of rBC-containing particles (F_{BC}; the ratio of rBC number concentration to the total number concentration in size range 10-1000 nm from the SMPS and APS measurements) are shown in Fig. 5 (a & b)".

Page 17, Line 11:

"The rBC particles constituted about 8-12% of the total number concentration over different sub-regions, on an average. (Fig. 5b)".

Page 18, Line 2:

"The mean fraction of rBC containing particles (F_{BC}) that were in the range 0.08-0.12 over different sub-regions, occasionally decreased to very low values ~ 0.02 to 0.04, owing to a large influx of ultrafine particles (sizes < 100 nm) during the new particle formation events occurred due to substantial amounts of condensable vapors (Kompalli et al., 2020b). The highest fraction of rBC containing particles was seen over the NIO-W (0.12 ± 0.03) region, whereas the largest range of F_{BC} values (0.03-0.21) among all the regions were observed over the SEAS. The present mean F_{BC} values seen over the northern and equatorial Indian Ocean are lower than those reported over the Finnish Arctic (~0.24 across the 350 to 450 nm size range), a background site receiving aged air masses (Raatikainen et al., 2015). However, earlier studies over the continental landmass of India have shown much higher number fractions with mean F_{BC} values ~ 0.51 ± 0.02 and 0.50 ± 0.03 over two

stations, Gual Pahari (polluted site) and Mukteshwar (regional background site) in northern India (Raatikainen et al., 2017). This was attributed to the strong influence of regional anthropogenic activities on BC loading. In the present study, rBC particles constituted about 25% to 30% of the measured scattering particles over almost the entire oceanic region north of 5 °N, whereas they occasionally decreased to 15 to 20 % over the far oceanic regions. Kompalli et al. (2020a) had reported widely varying mean fractions (25-69% of the measured scattering particles in different seasons) over Bhubaneswar (eastern India) with the same instrument. They attributed it to the seasonal variation of the scattering (non-BC) particle population. Sharma et al. (2017) have reported 10-16% of particles containing rBC cores in the range of ~ 200–400 nm optical diameter over Alert, Canadian Arctic. The presence of lower F_{BC} values over the marine regions in this study, which received a strong continental outflow, is not surprising, considering the observed large number concentration of total particles (Kompalli et al., 2020b) and the non-BC scattering particles in the detection range of the SP2 (200-400 nm) (along with rBC particles)".

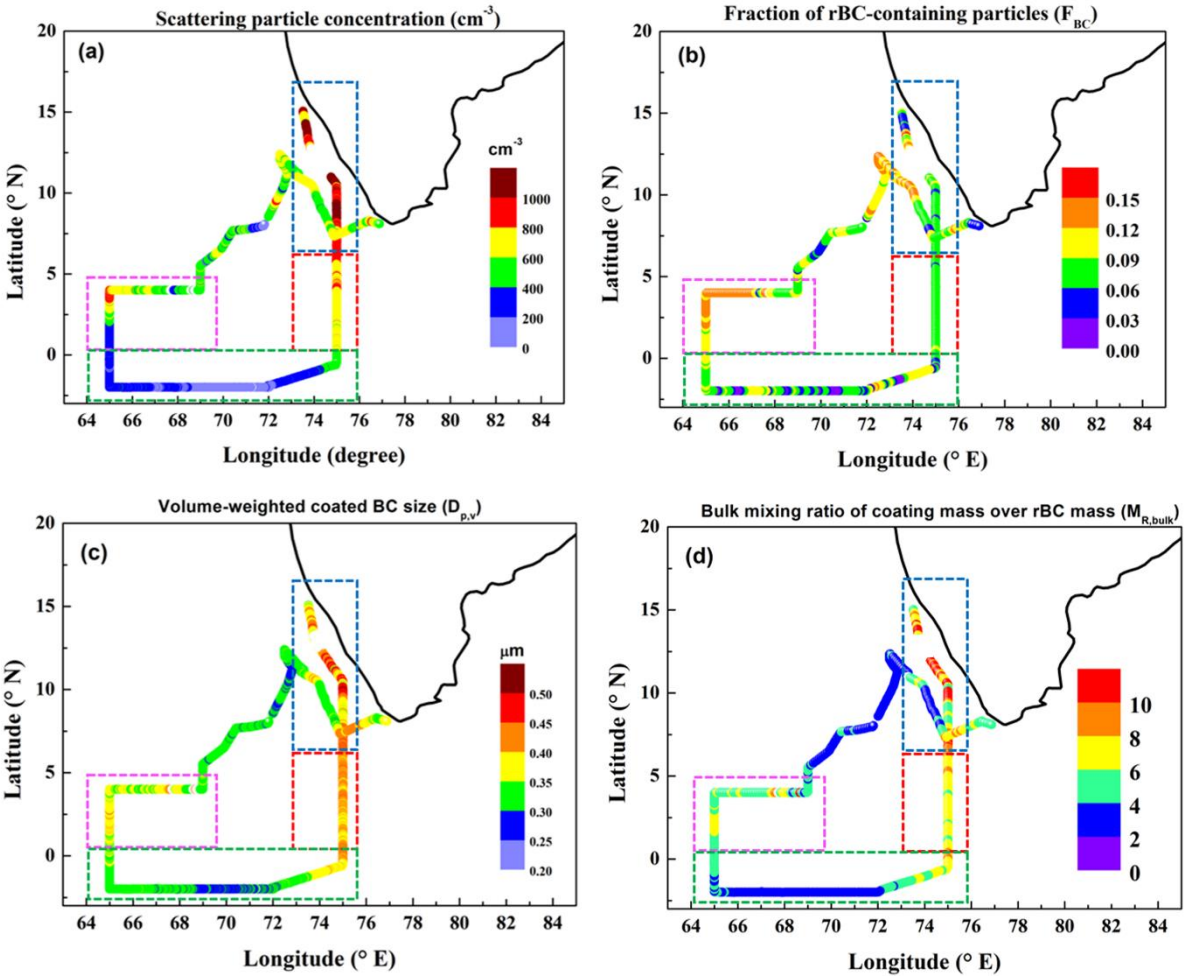


Figure 5: Spatial variation of the (a) scattering (non-BC) particle number concentration (in cm-3), (b) fraction of rBC containing particles (F_{BC}), (c) volume-weighted coated BC size ($D_{p,v}$) (in

μm), and (d) bulk mixing ratio of coating mass to rBC mass ($M_{R,bulk}$). Rectangles with dashed borders highlight different sub-regions. Table-3: A summary of regional mean values of rBC physical properties and mixing state parameters during the ICARB-2018. The values after ± are standard deviations.

Parameter	SEAS	NIO-E	EIO	NIO-W
rBC mass concentration (ng m^{-3})	938±293	546 ± 80	206 ± 114	614 ± 211
rBC number concentration (cm^{-3})	378 ± 137	191 ± 32	76 ± 38	227 ± 76
Scattering particle concentration (cm^{-3})	973 ± 187	747 ± 69	262 ± 140	580 ± 156
Mass median diameter (μm)	0.19 ± 0.01	0.20± 0.01	0.19± 0.01	0.19 ± 0.004
Number median diameter (μm)	0.10 ± 0.002	0.11± 0.003	0.11± 0.003	0.107 ± 0.002
Relative coating thickness	2.16 ± 0.19	2.05 ± 0.07	1.76 ± 0.16	1.93 ± 0.10
Absolute coating thickness (nm)	109 ± 20	104 ± 7	72 ± 17	85 ± 21
Fraction of rBC-containing particles (F_{BC})	0.08 ± 0.03	0.08 ± 0.01	0.08 ± 0.03	0.12 ± 0.03
volume-weighted coated BC size ($D_{p,v}$) (μm)	0.41 ± 0.04	0.41 ± 0.01	0.33 ± 0.04	0.37 ± 0.02
Bulk mixing ratio of coating mass over rBC mass ($M_{R,bulk}$)	8.31 ± 2.40	6.91 ± 0.71	4.24 ± 1.45	5.76 ± 1.17

Additional References:

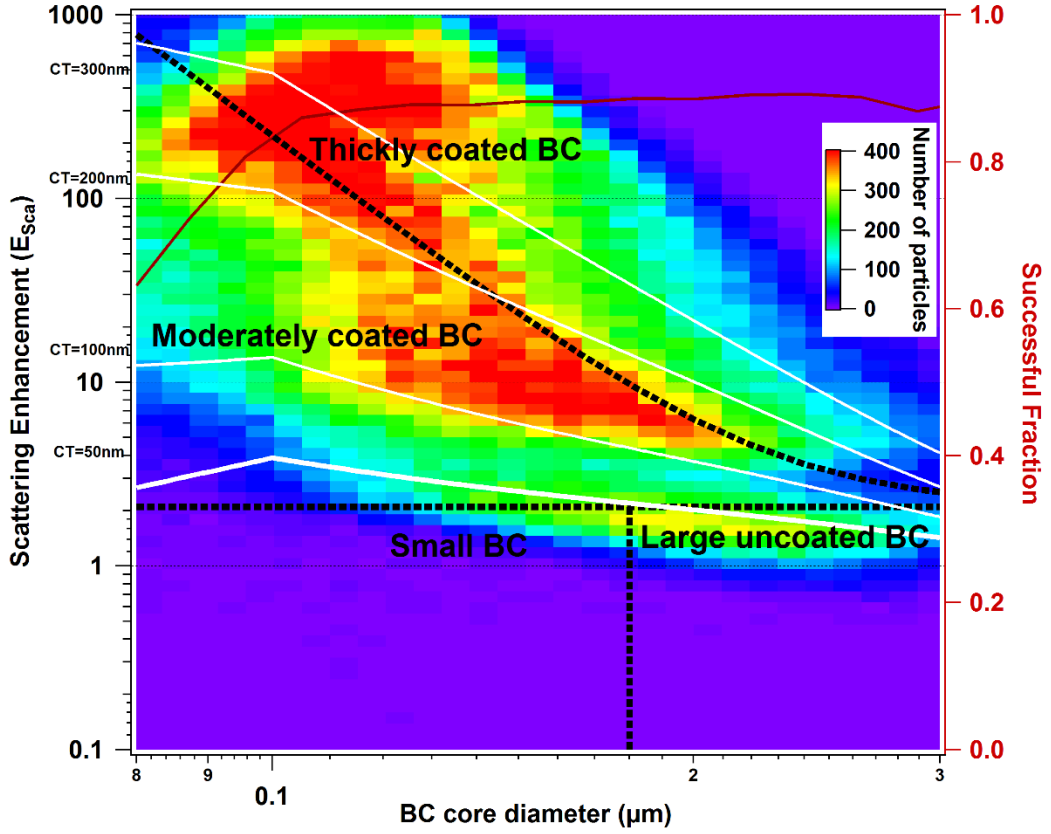
Leith, D., and Peters, T.M., 2003. Concentration Measurement and Counting Efficiency of the Aerodynamic Particle Sizer 3321.J. Aerosol Sci. 34(5), 627-634.

Wiedensohler, A. 1988. An approximation of the bipolar charge distribution for particles in the submicron size range. J. Aerosol Sci., 19, 387–389.

The authors report the observation of exceptionally thickly coated particles. Brooks et al., from which the current manuscript is nominally modeled after, reported on the rBC mixing state in northern India during the pre-monsoon and monsoon seasons. Using their Figure 6 "pre-monsoon (IGP)" plot it is estimated that a 130 nm BC core had a nominal coating thickness is ~110 nm. A similar back-of-the-envelope estimate from the current manuscript's Figure 6 suggests a coating thickness of ~250 nm (the

authors are strongly encouraged to adjust the scaling of the color mapping in Figure 6 so that more variation in the coating distribution might be seen). This difference in coating thicknesses for a 130 nm core represents about a 4.5x difference in coating volume. This is a big difference and as such, this Reviewer finds it a bit of stretch for the authors to simply state that "…the present study…..are comparable to the values reported in pollution in-plume air mass regions elsewhere (e.g., Cheng et al., 2019; Brooks et al., 2019)." [page 14, lines 19-20] Given this large difference, surly the authors could offer some discussion on why such a difference exists; that is, why are their rBC coatings so much thicker? Indeed, their reported coating thicknesses exceed what is observed for wildfires, which are known to rapidly create thickly coated rBC particles. This is perplexing and interesting finding that begs the question of why is there such a big difference, yet, no further discussion is provided by the authors beyond a cursory comparison with Brooks. The authors are encouraged to think about this finding and perhaps provide a more detailed discussion to help explain why the coating thicknesses found in this study are some much greater than that found by Brooks and in wildfires.

The color scale of Figure 6 is adjusted in the revised manuscript as below.



The point raised by the reviewer is an important and pertinent one and needs a detailed discussion. In page 14, lines 19-20, we have compared the present rBC '*bulk*' coating parameters

(bulk RCT ~1.73 to 2.15 and ACT~ 69-109 nm) (eq. 1) to the corresponding values reported by Brooks et al. (2019) (in their Fig. 2-4) over the highly polluted IGP region. We have used the bulk coating thickness because the coating thickness for individual particles is dependent on rBC core size. This approach, thus, reduces the uncertainties arising from smaller particles because of their less important contribution to the integrated volume. Further, the E_{sca}-D_c analysis is aimed at identifying the different sources. As suggested by the reviewer, we have adjusted the color scale of the figure to reflect variations in the coating distribution among rBC particles. This analysis aims not to predict the E_{sca} quantitatively or identify size segregated coating but rather to use the E_{sca} vs. D_c distributions to identify differences in rBC properties from which we can categorize different sources on a particle by particle basis. Because of this, in Page 18, lines 15 to 24, we have noted the fact that "a greater proportion of thickly coated particles with varying BC core diameters and a wide range (5-800) of scattering enhancement values were observed during the present study". The nominal ACT values shown in figure 6 of the present manuscript are to be taken in this context. We have added the following discussion in the revised according to the reviewer's comments:

Page 21, Line 6:

"Besides the normal mode similar to the one (with core diameter <0.22 µm and coating thickness of 50–200 nm) reported from the aircraft measurements of BC mixing state over the Indian continent by Brooks et al. (2019), an additional mode of BC with BC core sizes ~110-130 nm and a significant coating thickness (> 200 nm) is seen during this study. Such a mode highlights the influence of long-range transport to the ocean from the continent on BC ageing. Though the BC mass loading decreases during the long-range transport, the remaining BC cores gain a greater coating over the ocean than over land (e.g., Moteki et al., 2007). Further, it indicates a strong secondary production of aerosol components during the transport over the ocean, contributing to the BC ageing. The much thicker coatings seen during the ICABR-2018 compared to the observations from the ground-based site (Kompalli et al., 2020a) and the aircraft measurements (Brooks et al., 2019) over the Indian region are also indicative of other sources (with poor combustion efficiencies such as biomass burning) being prevalent in this region. Gong et al. (2016) have reported thick ACT (~110–300 nm) values during a biomass burning pollution episode in urban Shanghai, comparable to the present BC mode. These values are higher than the values reported from the aircraft measurements over biomass burning plumes (~ 150 nm; Ditas et al., 2018), the southeast Atlantic Ocean (~ 90 nm in the boundary layer and ~120 nm in the free troposphere; Taylor et al., 2020) and aged smoke in Amazonia (55-90 nm) (Darbyshire et al., 2019). Therefore, the measurement over the ocean thus offers an opportunity to study a

more aged BC from the continent outflow."

Additional references:

Ditas, J., Ma, N., Zhang, Y., Assmann, D., Neumaier, M., Riede, H., Karu, E., Williams, J., Scharffe, D., Wang, Q., Saturno, J., Schwarz, J. P., Katich, J. M., McMeeking, G. R., Zahn, A., Hermann, M., Brenninkmeijer, C. A., Andreae, M. O., Pöschl, U., Su, H., and Cheng, Y.: Strong impact of wildfires on the abundance and aging of black carbon in the lowermost stratosphere, P. Natl. Acad. Sci. USA, 115, E11595–E11603, https://doi.org/10.1073/pnas.1806868115, 2018.

Darbyshire, E., Morgan, W. T., Allan, J. D., Liu, D., Flynn, M. J., Dorsey, J. R., O'Shea, S. J., Lowe, D., Szpek, K., Marenco,F., Johnson, B. T., Bauguitte, S., Haywood, J. M., Brito, J. F., Artaxo, P., Longo, K. M., and Coe, H.: The vertical distribution of biomass burning pollution over tropical South America from aircraft in situ measurements during SAMBBA, Atmos. Chem. Phys., 19, 5771–5790, https://doi.org/10.5194/acp- 19-5771-2019, 2019.

Taylor, J. W., Wu, H., Szpek, K., Bower, K., Crawford, I., Flynn, M. J., Williams, P. I., Dorsey, J., Langridge, J. M., Cotterell, M. I., Fox, C., Davies, N. W., Haywood, J. M., and Coe, H.: Absorption closure in highly aged biomass burning smoke, Atmos. Chem. Phys., 20, 11201–11221, https://doi.org/10.5194/acp-20-11201-2020, 2020.

The authors report the organic contribution to the coating as being responsible for ~40% near the coast and in the remote ocean, accounting for a little over 20%, as inferred from the ACSM. What is the influence of having a mixture of sulfate and organics on the refractive index assumed in the coating calculation?

This is an important question, and sorry for the confusion created. We <u>have not quantified</u> the percentage contribution of organics or sulfate to the coating on rBC particles using the ACSM measurements. We have examined the association between the non-refractory submicron aerosol mass concentrations (NR-PM1.0) and bulk absolute coating thickness of rBC for low and high ACT observations. There is no significant association between bulk ACT and mass concentrations of NR-PM1.0 species during high ACT (ACT > 50 % of MMD) observations, suggesting complex coatings. However, for the BC population with low ACT values, a significant correlation (r ~0.62; p <0.01) was found between sulfate and rBC coating.

The value of 1.5+0i used for the coating refractive index in this study corresponds to values for ambient scattering aerosols reported in earlier studies (Metcalf et al., 2012; Laborde et al., 2013; Taylor et al., 2015). Laborde et al. (2013) have also suggested this as optimum value, which

is in the range of refractive indices of inorganic salts ($(NH_4)_2SO_4$ = 1.51; NaCl = 1.53) and secondary organic aerosol (~1.44-1.5) at λ = 1064 nm. Taylor et al. (2015) have presented a detailed assessment of the sensitivity of core/shell parameters derived using the single-particle soot photometer and reported that the refractive index of coatings has only a minor influence. They have also detailed the sources of uncertainties associated with it. Therefore, a mixture of sulfate and organics will not have a notable influence on the refractive index assumed in the coating calculation or coating parameters. We have added the following details in the revised manuscript.

<u>Page 7, Line 10:</u>

"Further, the amplitude of the scattering signal provides information about the scattering cross-section of the particles, which is used to determine the optical sizing of the particles. In the case of BC-containing particles, the scattering signal gets distorted as it passes through the laser beam because of the intense thermal heating of the particle and evaporation of the coating. Thus, the scattering signal of the BC particle is reconstructed using a leading-edge only (LEO) fitting technique, as described in Gao et al. (2007); Liu et al. (2010, 2014, 2017). This scattering cross-section is matched with the modeled values in a Mie-lookup table to derive the optical diameter of a BC particle or the coated BC size (D_p). Here, the total particle is treated as an ideal two-component sphere with a concentric core-shell morphology, with a core (rBC) refractive index value of 2.26 – 1.26i (Moteki et al., 2010; Liu et al., 2014; Taylor et al., 2015) and a coating refractive index of 1.5+0i (which is an optimum value and in the range of refractive indices of inorganic salts ($(NH_4)_2SO_4$ = 1.51; NaCl = 1.53) and secondary organic aerosol (~1.44-1.5) at λ = 1064 nm (Schnaiter et al., 2005; Metcalf et al., 2012; Lambe et al., 2013; Laborde et al., 2013; Taylor et al., 2015). These two diameters (D_p and Dc) are used to infer the coating thickness."

The following references are added in the revised manuscript:

Metcalf, A. R., Craven, J. S., Ensberg, J. J., Brioude, J., Angevine, W., Sorooshian, A., Duong, H. T., Jonsson, H. H., Flagan, R. C., and Seinfeld, J. H.: Black carbon aerosol over the Los Angeles Basin during CalNex, J. Geophys. Res., 117, D00V13, https://doi.org/10.1029/2011jd017255, 2012.

Schnaiter, M., Linke, C., Mohler, O., Naumann, K. H., Saathoff, H., Wagner, R., Schurath, U., and Wehner, B.: Absorption amplification of black carbon internally mixed with secondary organic aerosol, J. Geophys. Res., 110, D19204, doi:10.1029/2005jd006046, 2005.

Lambe, A. T., Cappa, C. D., Massoli, P., Onasch, T. B., Forestieri, S. D., Martin, A. T., Cummings,

M. J., Croasdale, D. R., Brune, W. H.,Worsnop, D. R., and Davidovits, P.: Relationship between oxidation level and optical properties of secondary organic aerosol, Environ. Sci. Technol., 47, 6349–6357, 2013. https://doi.org/10.1021/es401043j.

Additionally, while a density of 1.7 g/cc is reasonable for sulfates, it is high for organics (1.4 g/cc). Perhaps authors could conduct some sensitivity calculations to see if these factors exert influence on the derived coating thicknesses.

The coated material density was used only to derive the bulk mixing ratio of coating mass over rBC mass ($M_{R,bulk}$). The other mixing state parameters, such as coating thickness, are independent of coating density values. As described previously, for the estimation of core and coated BC particle diameters, only the core density value (1.8 g cm^{-3}) along with the refractive indices of core (2.26 – 1.26i) and coating (1.5+0i) were used. In this regard, we have revised the portion on M_{Rbulk} in the manuscript as suggested by the reviewer.

In the revised manuscript, we have used the effective dry density of ambient NR-PM1.0 based on the measured near-real-time chemical composition by assuming densities of organics and inorganics species as 1.4 (Hallquist et al., 2009) and 1.77 g cm^{-3} (Park et al., 2004), respectively. These revisions are also reflected in Figure 5 and Table-3.

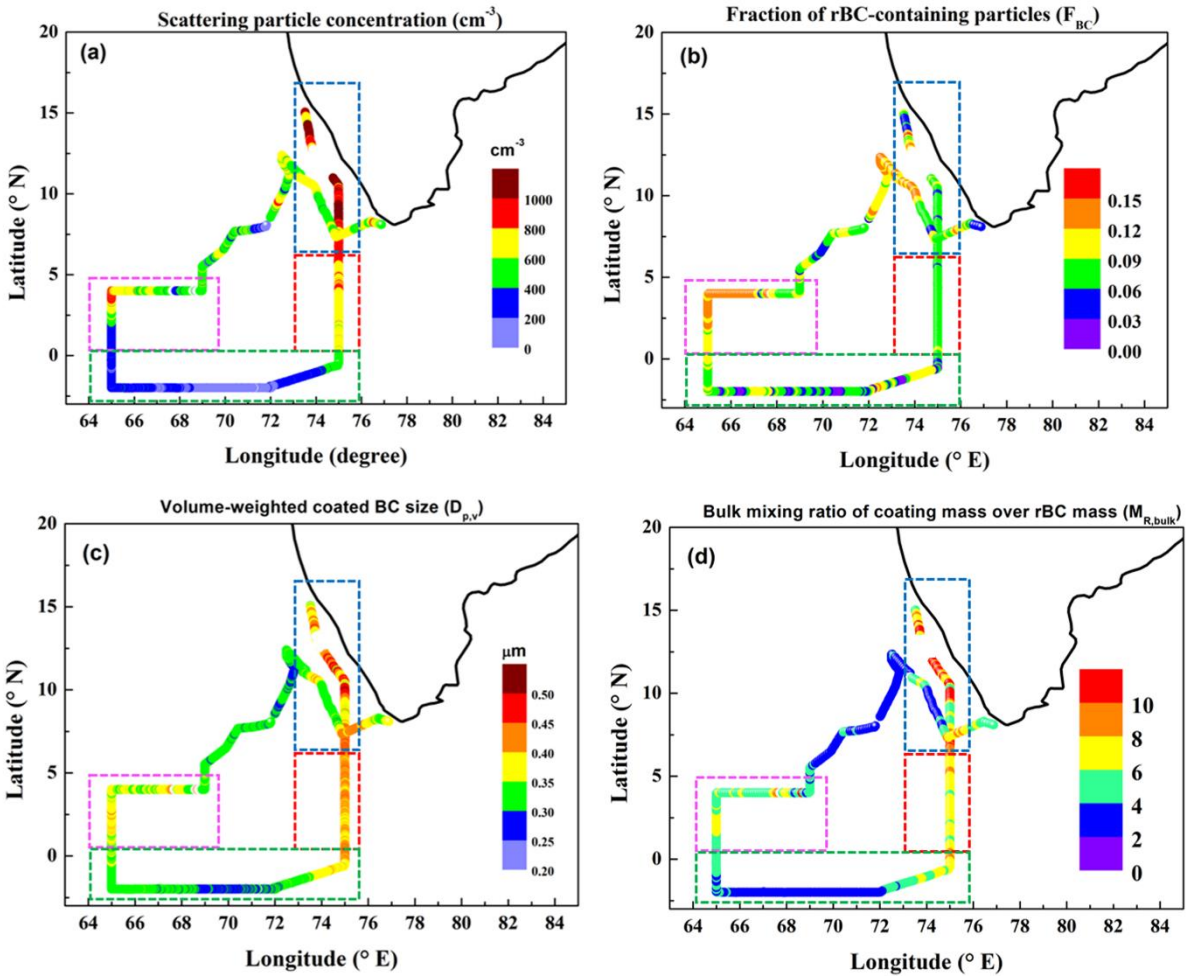


Figure 5: Spatial variation of the (a) scattering (non-BC) particle number concentration (in cm-3), (b) fraction of rBC containing particles (F_{BC}), (c) volume-weighted coated BC size ($D_{p,v}$) (in μm), and (d) bulk mixing ratio of coating mass to rBC mass ($M_{R,bulk}$). Rectangles with dashed borders highlight different sub-regions.

Table-3: A summary of regional mean values of rBC physical properties and mixing state parameters during the ICARB-2018. The values after ± are standard deviations.

Parameter	SEAS	NIO-E	EIO	NIO-W
rBC mass concentration (ng m^{-3})	938±293	546 ± 80	206 ± 114	614 ± 211
rBC number concentration (cm^{-3})	378 ± 137	191 ± 32	76 ± 38	227 ± 76
Scattering particle concentration (cm^{-3})	973 ± 187	747 ± 69	262 ± 140	580 ± 156
Mass median diameter (μm)	0.19 ± 0.01	0.20± 0.01	0.19± 0.01	0.19 ± 0.004

Number median diameter (μm)	**0.10 ± 0.002**	**0.11± 0.003**	**0.11± 0.003**	**0.107 ± 0.002**
Relative coating thickness	**2.16 ± 0.19**	**2.05 ± 0.07**	**1.76 ± 0.16**	**1.93 ± 0.10**
Absolute coating thickness (nm)	**109 ± 20**	**104 ± 7**	**72 ± 17**	**85 ± 21**
Fraction of rBC-containing particles (F_{BC})	**0.08 ± 0.03**	**0.08 ± 0.01**	**0.08 ± 0.03**	**0.12 ± 0.03**
volume-weighted coated BC size ($D_{p,v}$) (μm)	**0.41 ± 0.04**	**0.41 ± 0.01**	**0.33 ± 0.04**	**0.37 ± 0.02**
Bulk mixing ratio of coating mass over rBC mass ($M_{R,bulk}$)	**8.31 ± 2.40**	**6.91 ± 0.71**	**4.24 ± 1.45**	**5.76 ± 1.17**

We have included these points in the revised manuscript.

Page 9, Line 13:

"Since the ratio of the mass of non-absorbing coating material to the rBC core is an important parameter in determining the degree of absorption enhancement of BC, we quantified their mixing in terms of the bulk mixing ratio of coating mass over rBC mass ($M_{R,bulk}$) derived by assuming densities for the bulk coating ($\rho_{coating}$) and rBC core ($\rho_{rBCcore}$ ~1.8 g cm^{-3}) (Liu et al., 2019) as below:

$$M_{R,bulk} = \left(\left(\frac{D_p}{D_c}\right)^3 - 1\right) \times \frac{\rho_{coating}}{\rho_{rBCcore}} \quad (4)$$

Here we have used the effective dry density ($\rho_{coating}$) of ambient NR-PM1.0 based on the measured near-real-time chemical composition (Budisulistiorini et al., 2016), by assuming densities of organics and inorganics as 1.4 (Hallquist et al., 2009) and 1.77 g cm^{-3} (Park et al., 2004), respectively."

Page 18, Line 26:

"The higher bulk mixing ratio of coating mass over rBC mass values ($M_{R,bulk}$ ~2.5-15) (Fig. 5d) are seen over the adjacent marine regions, which is due to the presence of thickly coated BC particles. Though lower compared to other sub-regions, substantial $M_{R,bulk}$ (~ 4.24 ± 1.45) values were found even over the EIO region. Such high $M_{R,bulk}$ values were reported in the literature from extremely polluted environments and biomass burning source dominant regions (Liu et al., 2017; 2019). The presence of such non-absorbing coated mass on the rBC cores has significant radiative implications. Recently, Liu et al. (2017) have examined the measured and modeled optical properties of BC as a function of mass ratio ($M_{R,bulk}$) under different environments and found that significant absorption

enhancement occurs when the coating mass over rBC mass is larger than 3. They suggested that in such a scenario (i.e., $M_{R,bulk}$ >3), the core-shell model reproduces the measured scattering cross-section."

Page 25, Line 33:

"Further, high values (4.5-15) of the bulk mixing ratio of coating mass to rBC mass ($M_{R,bulk}$) were noticed in the outflow regions due to the presence of such thickly coated BC particles. Substantial mean $M_{R,bulk}$ (~ 4.24 ± 1.45) values were found even over the EIO region, which may be associated with both emissions and atmospheric processes contributing to non-refractory coatings on rBC particles."

Additional references:

Budisulistiorini, S. H., Baumann, K., Edgerton, E. S., Bairai, S. T., Mueller, S., Shaw, S. L., Knipping, E. M., Gold, A., and Surratt, J. D.: Seasonal characterization of submicron aerosol chemical composition and organic aerosol sources in the southeastern United States: Atlanta, Georgia, and Look Rock, Tennessee, Atmos. Chem. Phys., 16, 5171–5189, https://doi.org/10.5194/acp-16-5171-2016, 2016.

Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., aenhaut, W., McFiggans, G., Mentel, Th. F., Monod, A., Prévôt, A. S. H., Seinfeld, J. H., Surratt, J. D., Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmos. Chem. Phys., 9, 5155–5236, doi:10.5194/acp-9-5155-2009, 2009.

Park, K., Kittelson, D., Zachariah, M., and McMurry, P.: Measurement of Inherent Material Density of Nanoparticle Agglomerates, J. Nanopart. Res., 6, 267–272, doi:10.1023/B:NANO.0000034657.71309.e6, 2004.

The authors are sometimes using the term "aging" in a very confusing and, potentially, misleading way. For example, the authors write [page 14, lines 4-7] "While the larger BC particles are scavenged rather quickly, the smaller, less-aged, and relatively less- coated BC particles (occasionally, even bare soot particles) can persist in the outflow and be transported to the remote marine regions." This is misleading - more thinly coated BC particles are not necessarily "less-aged". Particles can undergo photolysis or heterogeneous oxidation that can bring about fragmentation leading to material loss as particles age leading to thinner coatings or larger diameter particles can be preferentially scavenged leaving behind smaller and more thinly coated particle. So to label a smaller, less-coated BC particle

- even bare soot particles - as less-aged is wrong. Correct this.

Sorry for the confusion created apparently by the adjectives being considered sequentially. We did not mean that the thinly coated particles are less aged. We have removed 'less-aged' to avoid confusion and revised the discussion using the statements suggested by the referee as below.

Page 15, Line 28:

"While the larger BC particles are scavenged rather quickly, the smaller and relatively less-coated BC particles (occasionally, even bare soot particles) can persist in the outflow and be transported to the remote marine regions (Ueda et al., 2018). As the particles spend more time in the atmosphere, they tend to gain coating material on them. Simultaneously, the loss of coating material on the particles cannot be ruled out due to photolysis or heterogeneous oxidation that can bring about fragmentation, leading to thinner coatings. Thus, preferential scavenging of larger particles leaving behind smaller and more thinly coated particles and atmospheric processes leading to loss of condensable material, explains the broad range of MMD (Fig. 3d) and lower RCT values observed over the EIO. Furthermore, in cleaner maritime regions like the equatorial Indian Ocean, the aging of BC occurs slowly due to reduced availability of coating material."

Do the authors have access to optical property measurements, specifically aerosol light absorption? If they do, they are strongly encouraged to roll that into this study so as to make this a more complete analysis.

We had an aethalometer aboard, measuring BC using optical absorption. However, the team concerned with those data are still in the process of analysing and as such, we are unable to include this in the present manuscript.

Finally, it seems to this reviewer that the authors have additional data mining that they could do. Given their bulk coating mass-to-core ratio and the mass loadings of non-refractory aerosols via the ACSM, it might prove very interesting to examine the ratio of non-refractory coating to overall non-refractory aerosol mass. Using the bulk mixing ratio of coating mass over rBC mass (Table 3) along with the rBC mass for the SEAS leg (Table 3) and the reported NR-PM mass in Figure 7, a back-of-the-envelope calculation suggests that about ~40% of the NR-PM mass is bound to BC particles, whereas for the EIO leg this ratio drops to ~15%. This suggests the preferential loss of coating and also suggests that the rBC number fraction of particles does change significantly (as per this Reviewer's comments above on this subject).

We thank the reviewer for this suggestion. Additional analyses have been carried out accordingly, and the following figure and discussion are added in the revised manuscript.

Page 23, Line 6:

"In supplementary figure S4, the ratio (expressed as %) of non-refractory coating mass on BC to the total NR-PM1.0 mass concentrations (from the ACSM) are shown. The mean ratios (varying between 23-35%) for different sub-regions are also mentioned in the figure."

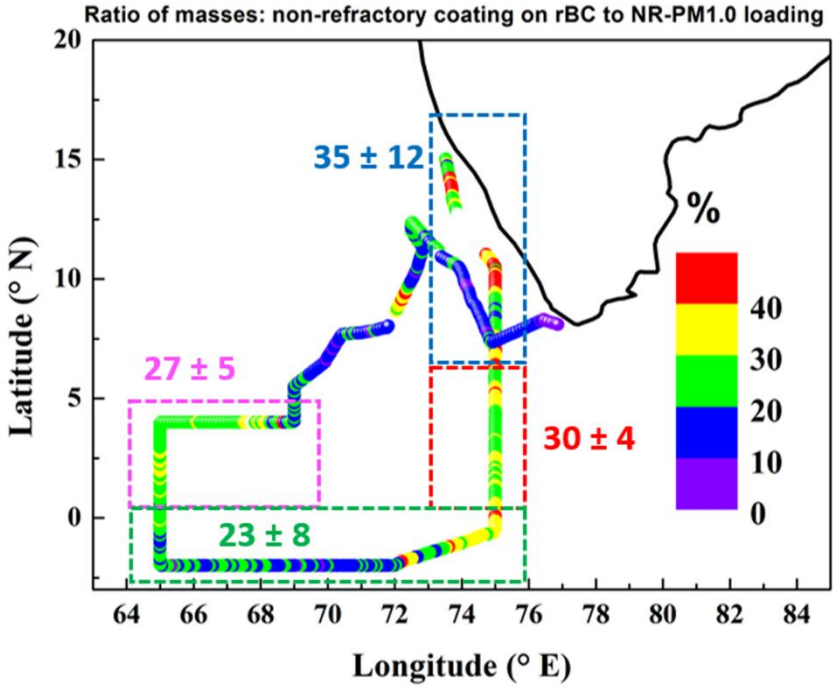


Figure S4: Spatial variation of the ratio (in %) of non-refractory coating mass on BC to the total mass concentration of non-refractory aerosols during the ICARB-2018. The mean values over different sub-regions are also written in the figure.

"It revealed that a substantial portion of the non-refractory submicron aerosol mass is bound to the BC particles over the regions with stronger outflow. While higher mean values of the ratio were observed over the SEAS (~35 ± 12 %) and NIO-E (31 ± 4) regions in the proximity of the sources, as one move away to the farther oceanic EIO, the NR-PM1.0 that is bound to BC particles dropped considerably (mean ~23 ± 8%). The values occasionally dropped to as low as 5-6%, indicating that (a) preferential loss of coating on rBC particle or (ii) a substantial contribution from non-rBC particles to the NR-PM1.0 mass loading due to new particle formation and subsequent growth events (Kompalli et al., 2020b). Though the mean fraction of r-BC containing particles remained similar (~8%) over different regions (SEAS, NIO-E and EIO), frequent new particle formation events (Kompalli et al., 2020b) resulted in lower F_{BC} (< 2%) over the EIO region which reflected in the amounts of NR-PM1.0 mass that are bound to rBC particles."

Specific issues:

***Page 3**, line 5. "Produced by the incomplete (low-temperature) combustion of hydro-carbon fuels. . ." "Low temperature" is subjective, please quantify or remove.*

Complied with. The word 'low-temperature' is removed.

***Page 3**, line 20. "The sources of BC are highly heterogeneous." What do the authors mean by "heterogenous"? There are three sources of BC: fossil fuel, biomass burning (wildfires and agricultural burns), and biofuel combustion. Please clarify what you mean by heterogeneous.*

Complied with. The sentence is now revised as "The sources of BC are highly varying, both seasonally and spatially, over the Indian region (e.g., Kompalli et al., 2014; Prasad et al., 2018 and references therein)"

References:

Kompalli, S. K., Babu, S. S., Moorthy, K. K., Manoj, M. R., Kirankumar, N. V. P., Shaeb, K. H. B., and Joshi, A. K.: Aerosol black carbon characteristics over central India: temporal variation and its dependence on mixed layer height, Atmos. Res., 147-148, 27–37, https://doi.org/10.1016/j.atmosres.2014.04.015, 2014.

Prasad, P., Ramana, R., Venkat Ratnam, M., Chen, W., Vijaya Bhaskara Rao, S., Gogoi, M. M., Kompalli, S. K., Kumar, K. S., and Babu, S. S.: Characterization of atmospheric Black Carbon over a semi-urban site of Southeast India: Local sources and long-range transport, Atmos. Res., 213, 411–421, https://doi.org/10.1016/j.atmosres.2018.06.024, 2018.

Page 3, line 20. "It has a long atmospheric lifetime." Compared to what? Certainly not CO2, which has a nominal lifetime of 100 years. The authors are encouraged to read the paper by Lund et al., (npj Climate and Atmospheric Science (2018)1:31) who report nominal BC lifetimes < 6 days. Please clarify what you mean by long atmospheric lifetime.

Complied with. The sentence is now revised as "Aerosol BC has an average atmospheric lifetime of about a week (Lund et al., 2018; Bond et al., 2013). It is prone to regional as well as long-range transport during its short atmospheric lifetime and found even over remote regions, such as the Polar Regions, albeit in lower concentrations (Raatikainen et al., 2015; Liu et al., 2015; Sharma et al., 2017; Zanatta et al., 2018)."

The following references are added:

Lund, M. T., Samset, B. H., Skeie, R. B., Watson-Parris, D., Katich, J. M., Schwarz, J. P., & Weinzierl, B.: Short black carbon lifetime inferred from a global set of aircraft observations.

npj Climate and Atmospheric Science, 1, 1-8. https://doi.org/10.1038/s41612-018-0040-x, 2018.

Sharma, S., Leaitch, W. R., Huang, L., Veber, D., Kolonjari, F., Zhang, W., Hanna, S. J., Bertram, A. K., and Ogren, J. A.: An evaluation of three methods for measuring black carbon in Alert, Canada, Atmos. Chem. Phys., 17, 15225–15243, https://doi.org/10.5194/acp-17-15225-2017, 2017.

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.

Page 8, lines 20-21. "…Esca is helpful in identifying the nature of sources." This is only true under the assumption of no material loss via oxidative and/or photochemistry that could either alter overall particle size and/or the refractive index of the coating.

Complied with. This caveat is included in the revised manuscript.

"E_{sca} is helpful in identifying the nature of sources, though under the assumption that no material loss via oxidative and/or photochemistry occurs which can either alter overall particle size and/or the refractive index of the coating."

***Page 9**, lines 13 and 15. "folds" should be singular. Please change.*

Corrected. "(which were 4-5 folds lower than..)"

***Page 10**, lines 12-18. The authors write "….the NIO-E region depicted slightly larger mean MMD (~0.20 µm) due to frequent larger values (35 % of the measurements showed MMD> 0.20 µm) compared to all the other regions (Fig. 3d). This is a result of either of two possibilities: (i) Self-coagulation of rBC cores due to enhanced atmospheric aging, which increases the rBC core diameters (at the same time, sedimentation of larger particles resulting in a large reduction in number concentration and mass concentration); (ii) The second and less likely possibility of a sizeable contribution (though not dominant) from solid fuel sources (biomass/crop residue/coal burning) in the upwind regions to the observed BC concentrations which were transported by the air masses traversing through the eastern coast of India and the Bay of Bengal" In figure 2c, the nominal rBC number concentration is ~200 /cc. Since coagulation goes a N^2, such a low concentration indicates that the rate of coagulation will be very slow. The authors should do a calculation to ascertain whether such low concentrations can result in enough self-coagulated rBC particles within the time frame of there measurements to account for ~35% of the measurements in this region showing MMD> 0.2um.*

Also, this Reviewer is quite surprised that there is very little discussion about the possible impacts of cloud processing of rBC particles that this might have on this observation (not to be confused with scavenging).

The reviewer has made a very important observation. These aspects are now included in the revised manuscript.

Page 11, Line 33:

"Notably, the NIO-E region depicted slightly larger mean MMD (~0.20 µm) due to frequent larger values (35 % of the measurements showed MMD> 0.20 µm) compared to all the other regions (Fig. 3d). This is a result of the following possibilities: (i) Self-coagulation of rBC cores due to enhanced atmospheric aging during their transport from the source regions in the east-coast to the adjacent marine regions (at the same time, sedimentation of larger particles resulting in a large reduction in number concentration and mass concentration). It may be noted that coagulation, though increases the rBC core diameters and reduces number concentrations, is a slow process. The coagulation rate depends on the square of the particle number concentrations and is the least between particles of the same size. Thus, the coagulation rates would be higher near source regions of the nascent aerosols and dropping off gradually at farther distances; (ii) the second and most important possibility is associated with the cloud processing of rBC particles. The insoluble BC particles remain within a non-precipitating cloud as interstitial particles. A cloud undergoes multiple evaporation-condensation cycles before it transforms into a precipitating system. During such cycles, interstitial BC in cloud droplets can grow larger (especially following the evaporation of cloud droplets containing multiple rBC particles) due to agglomeration with other interstitial rBC aerosols; (iii) The third possibility is the varying nature of dominant sources. A sizeable increase in the contribution from solid fuel sources (biomass/crop residue/coal burning) in the upwind regions (the eastern coast of India) through the transported air masses can lead to larger BC cores (Brooks et al., 2019; Kompalli et al., 2020a)."

Another point reviewer made is whether the nominal low rBC number concentration such as ~200 cm^{-3} can result in enough self-coagulated rBC particles within the time frame of these measurements to account for ~ 35% of the measurements in this region showing MMD > 0.2 µm. Since rBC particles originate from the regions of great source strengths (possibly, significant aerosol number concentrations compared to the values seen over the NIO-E), the rate of coagulation will be greatest near the source regions where nascent BC particles dominate. But, as the BC aerosols traverse longer distances to reach the marine regions away from the source regions, as pointed out by the reviewer, number concentration (and the rate of coagulation) is expected to decrease with

time. As such, subsequently, the contribution of the coagulation process to particle size transformation will be negligible. Apart from BC-BC coagulation, BC may coagulate with non-BC particles, which leads to increased BC coating thickness. The observed thicker coatings over the NIO-E are consistent with this.

Page 15, lines 14-17. Do the authors have access to any independent measurements of size distributions beyond solely relying on the SP2? As pointed out above, independent measurements of the microphysical properties would help in buttressing interpretations and, in some cases (e.g., rBC number fraction) enable a robust analysis to be performed.

Yes, we had independent measurements of aerosol size distributions using SMPS and used those in the revised manuscript (Figure 5b and Table-3 have been modified, and relevant discussion is revised), gratefully acknowledging the reviewer's valuable suggestions.