

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-2: ACP-2020-836-RC2

The information on various characteristics of rBC components of particles over the Indian Ocean is important. Overall, the presentation is clear, but the paper is longer than necessary for the information presented. The introduction is nicely written, but the discussion of results is verbose in many spots and should be written more succinctly. My major technical concerns relate to the use of the scattering signal from the SP2 (see specific comments on page 15 and beyond) and the non-refractory material contributing to the BC coatings. I hope the authors find these comments helpful.

We thank the reviewer for appreciating the importance of our work and the detailed comments. We have revised the manuscript considering these and another anonymous reviewer. Our responses to the specific comments of the reviewer are given below, following the same as for reviewer #1:

Page 3:

(1) Line 20 – “It has a long lifetime.” This sentence is a bit abrupt and ‘long lifetime’ needs to be defined.

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.

Line 21 - What is it about sources of BC in a clean environment that reduce its relative aging? Are BC emissions unaccompanied by fewer other emitted components?

Yes. Higher concentrations of condensable vapors (some of which are co-emitted species) contribute to faster aging of BC (Wang et al., 2014; Peng et al., 2016) in polluted environments, as compared to cleaner environments.

Lines 30-32 – “When air masses from such complex source regions are transported to remote regions devoid of any sources of BC, the aging becomes important, and the abundance of distinct species with varying lifetimes in the atmosphere differs significantly.” This statement needs some clarification: 1) why and where does the aging process suddenly become important; 2) what does the abundance of distinct species and their lifetimes have to do with BC?

This sentence is modified in the revised manuscript as below:

Page 3, Line 39:

“When air masses from such complex source regions are transported to remote regions devoid of any BC sources, the mixing state of BC becomes complicated. This is due to (a) restructuring of the BC aggregates during the transport due to different processes (Kütz and Schmidt-Ott, 1992; Weingartner et al., 1995; Slowik et al., 2007b; Pagels et al., 2009), and (ii) varying nature and amounts of coating material arising due to the different atmospheric lifetimes and microphysical processes involving different species (McFiggans et al., 2015).”

The following references are added in the revised manuscript:

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](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.**

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>.

McFiggans, G., Alfarra, M. R., Allan, J. D., Coe, H., Hamilton, J. F., Harrison, R. M., Jenkin, M. E., Lewis, A. C., Moller, S.J., and Williams, P. I. (2015). A review of the state-of-the-science relating to secondary particulate matter of relevance to the composition of the UK atmosphere. Full technical report to Defra, project AQ0732.

(2) *Lines 35-40 – This sentence is much longer than needed, especially since it does not tell us anything about the subject.*

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

“**The past** field campaigns, such as the Indian Ocean Experiment (INDOEX) during 1998-1999 (Ramanathan et al., 2001), the Integrated Campaign for Aerosols, gases, and Radiation Budget (ICARB) during March-May 2006 (phase-1); December-January 2008-2009 (phase-2) (Moorthy et al., 2008; Babu et al., 2012; Kompalli et al., 2013) have **characterized regional aerosols** over the northern Indian Ocean during different seasons.”

Page 4:

(3) *Line 5 – Everything but the AMS is referenced. Why not the AMS?*

Complied with. The following references are added for the AMS.

“...such as the aerosol mass spectrometer (AMS) (Jayne et al., 2000; Jimenez et al., 2003; Allan et al., 2003),...”.

References:

Allan, J., Jimenez, J., Williams, P., Alfarra, M., Bower, K., Jayne, J., Coe, H., and Worsnop, D.: Quantitative sampling using an Aerodyne aerosol mass spectrometer: 1. Techniques of data interpretation and error analysis, *J. Geophys. Res.-Atmos.*, 108, 4090, doi:10.1029/2003JD001607, 2003.

Jayne, J. T., Leard, D. C., Zhang, X. F., Davidovits, P., Smith, K. A., Kolb, C. E., and Worsnop, D. R.: Development of an aerosol mass spectrometer for size and composition analysis of submicron particles, *Aerosol Sci. Technol.*, 33, 49–70, 2000.

Jimenez, J. L., Jayne, J. T., Shi, Q., Kolb, C. E., Worsnop, D. R., Yourshaw, I., Seinfeld, J. H., Flagan, R. C., Zhang, X. F., Smith, K. A., Morris, J. W., and Davidovits, P.: Ambient

aerosol sampling using the Aerodyne Aerosol Mass Spectrometer, *J. Geophys. Res.- Atmos.*, 108(D7), 8425, doi:10.1029/2001JD001213, 2003.

(4) Line 8 – “such” is redundant.

Deleted.

(5) Line 15 – There is no verb. Perhaps, “what are the sources of BC and how does its mixing state evolve during transport to and over the ocean”.

This is corrected in the revised manuscript as below.

“ ... ii) **examination of** the extent of BC transport from distinct source regions and changes to its mixing state during the transport to the ocean, and (iii) **quantification of** the degree of coating on BC and **identification of** the nature of potential coating species by using concurrent chemical composition measurements during the South Asian outflow.”

(6) Line 28 - Do you know that particles smaller than 10 um were efficiently sampled or are you just assuming they were?

Prior to the experiment, we have characterized the sampling inlet system and examined the sampling losses, both theoretically and experimentally. For this purpose, the particle number size distribution measurements were used. We found that the difference between the number concentrations with and without inlet system was < 5% for sizes up to 1000 nm, 5-20% between 1000- 6000 nm, and > 30 % for sizes > 6000 nm.

Page 6:

(7) Line 7 – In recent years, the terminology for this is commonly “equivalent black carbon” or EBC (Petzold et al., Recommendations for reporting “black carbon” measurements, *Atmos. Chem. Phys.*, 13, 8356- 8379, doi:10.5194/acp-13-8365-2013, 2013.). Your oBC may be reasonable, but there is no reason to introduce your new definition, unless there has been some more recent change in terminology that I am unaware of.

We have now made the abbreviations consistent with Petzold et al. (2013). oBC is now changed to equivalent black carbon (EBC) in the manuscript.

Page 7:

(8) Lines 19-20 – It is at best questionable as to whether the ACSM truly measures PM1.0. It needs a reference.

We have provided the details as below:

“Supplementing the above, we have used the information on the mass concentration of non-refractory PM1.0 aerosols (organics, sulfate, ammonium, nitrate, and chloride) from a collocated aerosol chemical speciation monitor (ACSM; Model: 140; Aerodyne, USA; Ng et al., 2011). The objective here is to identify the possible coating material on rBC particles. The ACSM consists of a particle sampling inlet, three vacuum chambers (differentially pumped by turbopumps, backed by the main diaphragm pump), a residual gas analyzer (RGA) mass spectrometer (Pfeiffer Vacuum GmbH). The particles are drawn to an aerodynamic lens assembly having D_{50} limits (50% transmission range) of 75-650 nm and 30 to 40 % transmission efficiency at 1 μm (Liu et al., 2007) through a 100 μm critical orifice. These particles are focused into a narrow beam and transmitted to a vacuum environment where they are flash-vaporized by the thermal capture vaporizer (Xu et al., 2017; Hu et al., 2017a, 2017b) operating at 525 °C. Subsequently, these vapors are ionized via 70 eV electron impact ionization and detected with a quadrupole mass spectrometer.

Additional References:

Hu, W. W., Campuzano-Jost, P., Day, D. A., Croteau, P., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L.: Evaluation of the new capture vaporizer for aerosol mass spectrometers (AMS) through laboratory studies of inorganic species, *Atmos. Meas. Tech.*, **10**, 2897–2921, <https://doi.org/10.5194/amt-10-2897-2017>, 2017a.

Hu, W. W., Campuzano-Jost, P., Day, D. A., Croteau, P., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L.: Evaluation of the new capture vaporizer for aerosol mass spectrometers (AMS) through field studies of inorganic species, *Aerosol Sci. Technol.*, **51**, 735–754, <https://doi.org/10.1080/02786826.2017.1296104>, 2017b.

Liu, P. S. K., Deng, R., Smith, K. A., Williams, L. R., Jayne, J. T., Canagaratna, M. R., Moore, K., Onasch, T. B., Worsnop, D. R., and Deshler, T.: Transmission efficiency of an aerodynamic focusing lens system: comparison of model calculations and laboratory measurements for the Aerodyne Aerosol Mass Spectrometer, *Aerosol. Sci. Tech.*, **41**, 721–733, 2007.

Xu, W., Croteau, P., Williams, L., Canagaratna, M., Onasch, T., Cross, E., Zhang, X., Robinson, W., Worsnop, D., and Jayne, J.: Laboratory characterization of an aerosol chemical speciation monitor with PM2.5 measurement capability, *Aerosol Sci. Technol.*, **51**, 69–83, <https://doi.org/10.1080/02786826.2016.1241859>, 2017.

(9) *Line 24 – Why are so many details of the SP2 given, yet you are unwilling to give the “prescribed methodology”?* The prescribed methodology is more germane to your analysis than the details of the ACSM that have been known for over a decade.

We have updated the details on the SP2 methodology, and as suggested by the reviewer, added

more details on the ACSM data methodology in the ‘Analysis’ section in the revised manuscript.

Page 7, Line 40:

“The data is processed as per the prescribed methodology (Ng et al., 2011; Kompalli et al., 2020a). We have used software provided by the manufacturer (Aerodyne Research, ACSM Local, version 1.6.0.3, within IGOR Pro version 7.0.4.1) for processing and analysis of data. Using the default fragmentation table (Allan et al., 2004), the measured fractions of unit mass resolution spectra signals were apportioned to individual aerosol species. The required corrections for the instrument performance for the varying inlet pressures and N₂ signal were performed (Ng et al., 2011; Sun et al., 2012). Mass-dependent ion transmission efficiency correction of the residual gas analyzer was carried out using the signals from the internal diffuse naphthalene source (m/z 128). The calibrations of ionization efficiency (IE) and relative IE (RIE) calibrations were performed prior to the experiment by using monodisperse (300 nm) particles of NH₄NO₃ and (NH₄)₂SO₄ (Jayne et al., 2000; Allan et al., 2003; Jimenez et al., 2003; Canagaratna et al., 2007). The present ACSM consists of a capture vaporizer with an inner cavity to reduce the particle bounce (Xu et al., 2017), resulting in a higher collection efficiency (about unity) (Hu et al., 2017a; 2017b). Therefore, the composition-dependent collection efficiency correction prescribed by Middlebrook et al. (2012), applicable to standard vaporizer instruments, was not applied to our data. More than 1200 quality checked individual observations with a time resolution of ~30 minutes formed the database for this study.”

The following references are added:

- Allan, J., Jimenez, J., Williams, P., Alfarra, M., Bower, K., Jayne, J., Coe, H., and Worsnop, D.: Quantitative sampling using an Aerodyne aerosol mass spectrometer 1. techniques of data interpretation and error analysis, *J. Geophys. Res.*, 108, 4090, doi:10.1029/2002JD002358, 2003.
- Allan, J., Delia, A. E., Coe, H., Bower, K. N., Alfarra, R., Jimenez, J. L., Middlebrook, A. M., Drewnick, F., Onasch, T. B., Canagaratna, M. R., Jayne, J., and Worsnop, D. R.: Technical note: a generalised method for the extraction of chemically resolved mass spectra from Aerodyne aerosol mass spectrometer data, *J. Aerosol Sci.*, 35, 909–922, doi:10.1016/j.jaerosci.2004.02.007, 2004.
- Canagaratna, M., Jayne, J., Jimenez, J., Allan, J., Alfarra, M., Zhang, Q., Onasch, T., Drewnick, F., Coe, H., Middlebrook, A., Delia, A., Williams, L., Trimborn, A., Northway, M., DeCarlo, P., Kolb, C., Davidovits, P., and Worsnop, D.: Chemical and microphysical characterisation of ambient aerosols with the Aerodyne aerosol mass spectrometer, *Mass Spectrom. Rev.*, 26, 185–222, doi:10.1002/Mas.20115, 2007.
- Hu, W. W., Campuzano-Jost, P., Day, D. A., Croteau, P., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L.: Evaluation of the new capture vapourizer for aerosol mass

spectrometers (AMS) through laboratory studies of inorganic species, *Atmos. Meas. Tech.*, **10**, 2897–2921, <https://doi.org/10.5194/amt-10-2897-2017>, 2017a.

Hu, W. W., Campuzano-Jost, P., Day, D. A., Croteau, P., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L.: Evaluation of the new capture vaporizer for aerosol mass spectrometers (AMS) through field studies of inorganic species, *Aerosol Sci. Technol.*, **51**, 735–754, <https://doi.org/10.1080/02786826.2017.1296104>, 2017b.

Jayne, J., Leard, D., Zhang, X., Davidovits, P., Smith, K., Kolb, C., and Worsnop, D.: Development of an aerosol mass spectrometer for size and composition analysis of submicron particles, *Aerosol Sci. Tech.*, **33**, 49–70, 2000.

Jimenez, J. L., Jayne, J. T., Shi, Q., Kolb, C. E., Worsnop, D. R., Yourshaw, I., Seinfeld, J. H., Flagan, R. C., Zhang, X., Smith, K. A., Morris, J., and Davidovits, P.: Ambient aerosol sampling with an aerosol mass spectrometer. *J. Geophys. Res.-Atmos.*, **108**, 8425, doi:10.1029/2001JD001213, 2003.

Sun, Y., Wang, Z., Dong, H., Yang, T., Li, J., Pan, X., Chen, P. and Jayne, T.P.: Characterization of Summer Organic and Inorganic Aerosols in Beijing, China with an Aerosol Chemical Speciation Monitor. *Atmos. Environ.* **51**, 250–259, doi:10.1016/j.atmosenv.2012.01.013, 2012.

Xu, W., Croteau, P., Williams, L., Canagaratna, M., Onasch, T., Cross, E., Zhang, X., Robinson, W., Worsnop, D., and Jayne, J.: Laboratory characterization of an aerosol chemical speciation monitor with PM_{2.5} measurement capability, *Aerosol Sci. Technol.*, **51**, 69–83, <https://doi.org/10.1080/02786826.2016.1241859>, 2017.

(10)Line 37 – Page 8, line 3 – Why fit a log-normal distribution to the data when you can calculate MMD and NMD directly from the measurements? Are you sure the data always follow one log-normal mode?

Since the fairly narrow measurement range of the SP2 instrument is unlikely to represent the total ambient number and mass concentrations of BC, extrapolation of a log-normal distribution is used to predict these missing masses (below and above the detection limits), as has been done in earlier studies (McMeeking et al., 2010; Metcalf et al., 2012; Reddington et al., 2013; Liu et al., 2010, 2014). As rightly pointed by the reviewer, unimodal distribution always may not be the case for BC core size distributions. At times, an additional mode at sizes below the detection limit of the SP2 is also possible near source regions. However, such a mode (~ 50 nm) is unlikely far away from source regions, which is the case in the present study.

(11)Line 7 – What does the error indicate: experimental uncertainty in the mean, 25th percentile or standard deviation, etc.?

The values after \pm indicate standard deviation. This is now explicitly mentioned in the revised manuscript.

“The highest values and variabilities (standard deviation) in the rBC mass (mean $\sim 938 \pm 293 \text{ ng m}^{-3}$) and number ($\sim 378 \pm 137 \text{ cm}^{-3}$) concentrations...”

(12)Lines 9-11 – If you are going to suddenly toss in oBC (or EBC) measurements, then you need to discuss why they are about a factor of two higher than the rBC.

We have removed this sentence in the revised manuscript.

(13)Lines 14-15 – Concentrations of rBC reaching 200 ng/m^3 would not be defined as extremely low in other parts of the world. In the Arctic, for example, such concentrations, which can be present in the Arctic Haze, are considered high. Please replace with something like “The lower concentrations. ..”

Complied with. The sentence is now revised:

“~~Such extremely low~~ The lower concentrations ($<200 \text{ ng m}^{-3}$) highlighted...”

(14)Lines 18-21 – Since the trajectories suggest that NIO-W and NIO-E are roughly equidistant from the sources, does this mean that the source strengths on the west and east coasts are similar, and that the reduction from the SEAS region is mostly due to dispersion during transport?

Both the SEAS and NIO-W regions received continental outflow air masses from the west coast/peninsular India. Dispersion during the transport is an important factor governing the decrease in BC concentrations away from the source regions which is reflected in the spatial heterogeneity between SEAS and NIO-W. However, the strength of advection or sources between NIO-E and NIO-W cannot be inferred from this. We have referred to the earlier publications that reported the contrast between the east and west coast regions of India for this purpose. (Fig.S1 c&d; Moorthy et al., 2005; Kompalli et al., 2013)

Page 10:

(15)Lines 2-5 – References needed.

These sentences are rewritten in the revised manuscript, and references are provided.

Page 10, Line 11:

“The MMD values of the rBC size distributions are strongly influenced by the source of BC emissions (e.g., Ko et al., 2020; Cheng et al., 2018). Recently, Ko et al. (2020) have compared the MMD and NMD values of rBC size distributions from different dominant sources. Previous studies report that the MMD (and NMD) values over the regions dominated by fresh fossil fuel emissions are smaller (MMD ~100-178 nm and NMD ~ 30 to 80 nm) compared to the areas with dominant solid-fuel sources (biomass, biofuel, coal-burning) (MMD ~130-210 nm and NMD ~100 to 140 nm), whereas, well-aged and background BC particles in outflow regions have MMD values in between (MMD ~ 180-225 nm and NMD ~ 90-120 nm) (McMeeking et al., 2010; Liu et al., 2010, 2014; Kondo et al., 2011; Cappa et al., 2012; Sahu et al., 2012; Metcalf et al., 2012; Laborde et al., 2013; Reddington et al., 2013; Gong et al., 2016; Raatikainen et al., 2017; Krasowsky et al., 2018; Brooks et al., 2019; Kompalli et al., 2020a; Ko et al., 2020)”.

The following additional references are added:

Krasowsky, T. S., Mcmeeking, G. R., Sioutas, C., and Ban-Weiss, G.: Characterizing the evolution of physical properties and mixing state of black carbon particles: From near a major highway to the broader urban plume in Los Angeles, Atmos. Chem. Phys., 18, 11991–12010, <https://doi.org/10.5194/acp-18-11991-2018>, 2018.

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.

(16)Line 5 – Do you mean “have”, rather than “comprised BC particles”?

This sentence is revised.

(17)Lines 8-12 – Are you saying here that you can identify the sources contributing to the rBC based on the MMD of the rBC component of the particle?

No. Though MMD is an indicator, unequivocal identification of sources is not possible based on the MMD values of the rBC particles alone. This is explicitly stated in the revised manuscript.

Page 12, Line 3:

“It may be noted that the exact sources cannot be identified from the MMD value of rBC size distributions alone. More details on source apportionment are provided in section 3.3”.

(18) Lines 11-12 – Your MMD fall in line with the “aged continental outflow” you provide 11 references for. You refer to the continental outflow studies by saying “On the other hand”, which suggests that they are different from a combination of urban emissions and bio+coal emissions. Are you making your determination on lines 11-12 based on the 11 studies you reference, or are you assuming that your results are a combination of urban emissions and the bio+coal emissions?

Based on the supporting evidence from the previous studies, we argue that the results suggest a combination of mixed emissions and aging. Also, complying with both the reviewers’ comments, the discussion has been revised as below:

Page 11, Line 13:

“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 28, Line 10:

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

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](https://doi.org/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](https://doi.org/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](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](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>.
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- 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.

(19) Lines 13-18 – You suggest that the slight increase in MMD of the rBC over the NIO- E region is due to a complex set of processes (self-coagulation and sedimentation) occurring in those particle populations rather than differences in source types, even though you have already identified these regions as a mix of source types. Also, you give no reason why the same processes (self-coagulation and sedimentation) do not similarly affect the rBC in the other regions, in particular the NIO-W regions where the trajectories appear to have similar transport times. Why are source differences “less likely”, given the large number of sources spread out over 1000-2000 km on both coastlines (east and west)?

This is an important point, as also commented by Reviewer-1. We have revised the discussion as under:

Page 11, Line 33:

“Notably, the NIO-E region depicted slightly larger mean MMD ($\sim 0.20 \mu\text{m}$) due to frequent larger values (35 % of the measurements showed $\text{MMD} > 0.20 \mu\text{m}$) 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).”

Page 11, Line 13:

“The observed mean NMD ($0.10\text{-}0.11 \mu\text{m}$) and MMD ($0.19\text{-}0.20 \mu\text{m}$) 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).”

(20) Lines 19-20 – What do the lower MMD associated with EIO imply?

We have added more details on this in the revised manuscript.

Page 15, Line 26:

“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.**”

(21) For the above reasons (comments 16-21), this section, from lines 1-21, needs to be written with more clarity and justification.

Complied with. This portion is thoroughly revised in the manuscript.

Page 11-12:

(22) Table 2 and Lines 1-11, page 12 – There are measurements from the Arctic that could be added to this table. For example, Sharma et al. (ACP, 2017) found rBC MMD greater than 300 nm at a high

Arctic location.

Thanks for pointing this out. We have complied with and revised Table-2.

Table-2: A comparison of rBC average mass median diameters/mode of mass size distributions (MSD) reported from selected locations with distinct sources in different environments.

S.No.	Location	Type of location/ sources	MSD mode/ MMD (μm)	Reference
Aged air masses in remote/outflow regions				
1.	Southeastern Arabian Sea	Continental outflow/mixed sources	0.18-0.20 (mean~0.19 \pm 0.01)	Present study
2.	Northern Indian Ocean	Continental outflow/mixed sources	0.19-0.21	Present study
3.	Equatorial Indian Ocean	Outflow impacted remote marine/mixed sources	0.18 -0.21 (mean~0.19 \pm 0.01)	Present study
4.	Fukue island, Japan	Asian outflow	0.20-0.22	Shiraiwa et al. (2008)
5.	Suzu, Japan	Urban/east Asian outflow site	0.200	Ueda et al. (2016)
6.	Mukteshwar, the Himalayas, India	High-altitude/biofuel, crop residue outflow	0.21 \pm 0.02	Raatikainen et al. (2017)
7.	Jungfrauoch, Switzerland	High-altitude remote background / biomass burning, aged BC	0.22-0.24	Liu et al. (2010)
8.	Finnish Arctic	Remote background/aged air mass	0.15-0.20	Raatikainen et al. (2015)
9.	Alert, Nunavut, Canada (within the Arctic Circle)	Remote background/aged air mass	0.16-0.18	Sharma et al. (2017)
10.	Catalina Island, (~ 70 km southwest of Los Angeles)	Aged air masses	0.153-0.170	Ko et al. (2020)
11.	Atlantic Ocean	European continental outflow	0.199	McMeeking et al. (2010)
12.	Zeppelin, European Arctic	Remote background/aged air mass	0.24	Zanatta et al. (2018)
Urban locations				
13.	Regional average over Europe	Near source to high-altitudes (a) European continental (b) Urban outflow	0.17-0.21 (a) 0.18-0.20 (b) 0.17 \pm 0.01	McMeeking et al. (2010)
14.	Bhubaneswar, India	Urban/fresh urban emissions Urban/continental outflow, aged BC Urban/with high solid fuel emissions	0.17 \pm 0.01 0.18-0.19 0.22 \pm 0.01	Kompalli et al., (2020a)
15.	Indo-Gangetic Plain (aircraft experiment)	Urban polluted/mixed sources	0.18-0.21	Brooks et al. (2019)
16.	Gual Pahari, India	Urban polluted/ fresh biofuel, crop residue	0.22 \pm 0.01	Raatikainen et al. (2017)

17.	Shanghai, China	Urban/pollution episode with high biomass burning	0.23	Gong et al. (2016)
18.	London, England	Urban/traffic emissions Wood burning	0.119-0.124 0.170	Liu et al. (2014)
19.	Canadian oil sand mining, Canada	Urban/fresh urban emissions	0.135-0.145	Cheng et al. (2018)
20.	Catalina Island, (~ 70 km southwest of Los Angeles)	Biomass burning Fossil fuel emissions	0.149-0.171 0.112-0.129	Ko et al. (2020)

The following references are added:

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.

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, <https://doi.org/10.1029/2008JD010546>, 2008.

Raatikainen, T., Brus, D., Hyvärinen, A. P., Svensson, J., Asmi, E., and Lihavainen, H.: Black carbon concentrations and mixing state in the Finnish Arctic, *Atmos. Chem. Phys.*, **15**, 10057–10070, <https://doi.org/10.5194/acp-15-10057-2015>, 2015.

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 13:

(23)Line 8 – When you say dispersion, do you mean mixing with other air masses with smaller coating thicknesses? Reactions, unless leading to fragmentation and volatilization, are more likely to increase coating thickness than decrease it. Although there was relatively little precipitation during your study, transport across the SAS region might have impacted coatings. How might precipitation have affected the rBC size distribution and coating thickness over NIO-W? (I see you discuss this later with respect to the EIO region.)

Here dispersion and reactions refer to reduced availability of the coating material as we move away from the source regions. As the particles spend more time in the atmosphere, they tend to

gain coating material on them. But as Referee-1 also has suggested, 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 lower RCT values observed over the EIO. This aspect is now described in the manuscript.

The passage of a large-scale meteorological system (due to western disturbances) which resulted in widespread rainfall over the SAS region, has occurred at a later period. During this period, extensive precipitation also occurred over the peninsular and western parts of the Indian mainland. But the passage of this system has happened after the cruise covered the NIO-W region. Thus, the large-scale meteorological system does not appear to have impacted the measurements of the NIO-W region and the outflow air masses originating from the western coast of India remained unperturbed.

(24)Line 12 – “Nearly 95% of the SEAS measurements...”?

This sentence is revised in the manuscript as below:

“Nearly 95% of **the observational points over the SEAS region** indicated that rBC particles have an additional coating over its cores to the extent of > 90% of its size.”

(25)Line 15 – What are you contrasting with? Just start with “Over the NIO-E region...”

Complied with.

(26)Line 19 – Does that mean that sources on the east coast are stronger than on the west coast. This relates back to my comment 15 above.

Yes. We have revised this aspect in the manuscript.

Page 14:

(27)Line 9 – Smaller rather than inferior.

Complied with.

“It possibly resulted in the observed **smaller** coatings...”

(28)Lines 13-15 – It would be clearer to say “due to differences in respective coastal sources

(references) and possible transit times.” What are the relative differences in air mass transit times from the west coast to NIO-W and from the east coast to NIO-E, and what are the differences in east coast and west coast source strengths? See comment 20 above.

Complied with. We have revised the sentence as suggested by the reviewer.

“Thus, a clear contrast in the mixing state parameters is evident which is due to differences in respective coastal sources (Moorthy et al., 2008; Peng et al., 2016; Gong et al., 2016), and possible transit times over these two regions.

(29)Lines 16-18 – Repetitious.

This sentence is revised and moved to the introduction portion in the revised manuscript.

Page 3, Line 26:

“The alteration to BC mixing state depends on various factors, which include the BC size distribution, nature of sources, the concentration of condensable materials that BC encounters during its atmospheric lifetime, and processes such as photochemical aging (Liu et al., 2013; Ueda et al., 2016; Miyakawa et al., 2017; Wang et al., 2018).”

(30)Lines 24-26 – If comparisons cannot be made, then what is the point of any of the measurements?

This sentence is revised as below:

Page 16, Line 7:

“As Cheng et al. (2018) and Ko et al. (2020) have highlighted, coating parameters derived from the SP2 instruments having different system configurations (detection limits of scattering intensity and range of volume equivalent diameters covered) and different techniques used in the estimation the optical diameters from scattering amplitudes (Metcalf et al., 2012; Gong et al., 2016; Raatikainen et al., 2017; Cheng et al., 2018; Liu et al., 2019; Ko et al., 2020) can vary considerably. This caveat needs to be borne in mind when making inter-study comparisons.”

(31)Lines 27-29 – With the caveat that your study is not a Lagrangian experiment, and your “far-field” measurements are influenced by mixing with the surrounding environment.

Thanks. The point is made explicit in the revised manuscript.

Page 16, Line 13:

“Also, the earlier studies are mostly made in the ‘near-field’ situation, whereas the present study examined the coating characteristics in a ‘far-field’ scenario (far away from potential sources, especially NIO and EIO regions). The caveat here is that the present study is not a

Lagrangian experiment, and it is possible that the “far-field” measurements are influenced by mixing with the surrounding environment.”

(32)Line 32 - What is “It” that needs further investigation: coatings, enhancement, radiative forcing?

Complied with. This sentence is revised as below:

“The implication of the observed thick coatings on BC to regional radiative forcing needs further detailed investigation in future studies”.

Page 15:

(33)Line 8 – What does “very high” reference to?

Clarified as below:

“The non-BC (scattering) particle concentrations were **higher (>1000 cm⁻³) in the coastal waters (the SEAS),...”**

(34)Lines 11-13 – I question your statement that rBC particles constituted about 25% to 30% of the total number concentration. Does the scattering particle concentration measured using the SP2 represent the total number concentration, and does the use of the scattering particles from the SP2 inflate the fractional value? For this fractional estimate, do you only include rBC components that have a detectable scattering signal? If so, then you could say that rBC particles constituted about 25% to 30% of the measured scattering particles. If not, then your fractional estimates are more difficult to interpret. Please correct here and in Table 3.

This is an important point as has also been pointed out by Reviewer -1. 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.

Page 8, Line 10:

“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 $\sim 0.51 \pm 0.02$ 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)”.

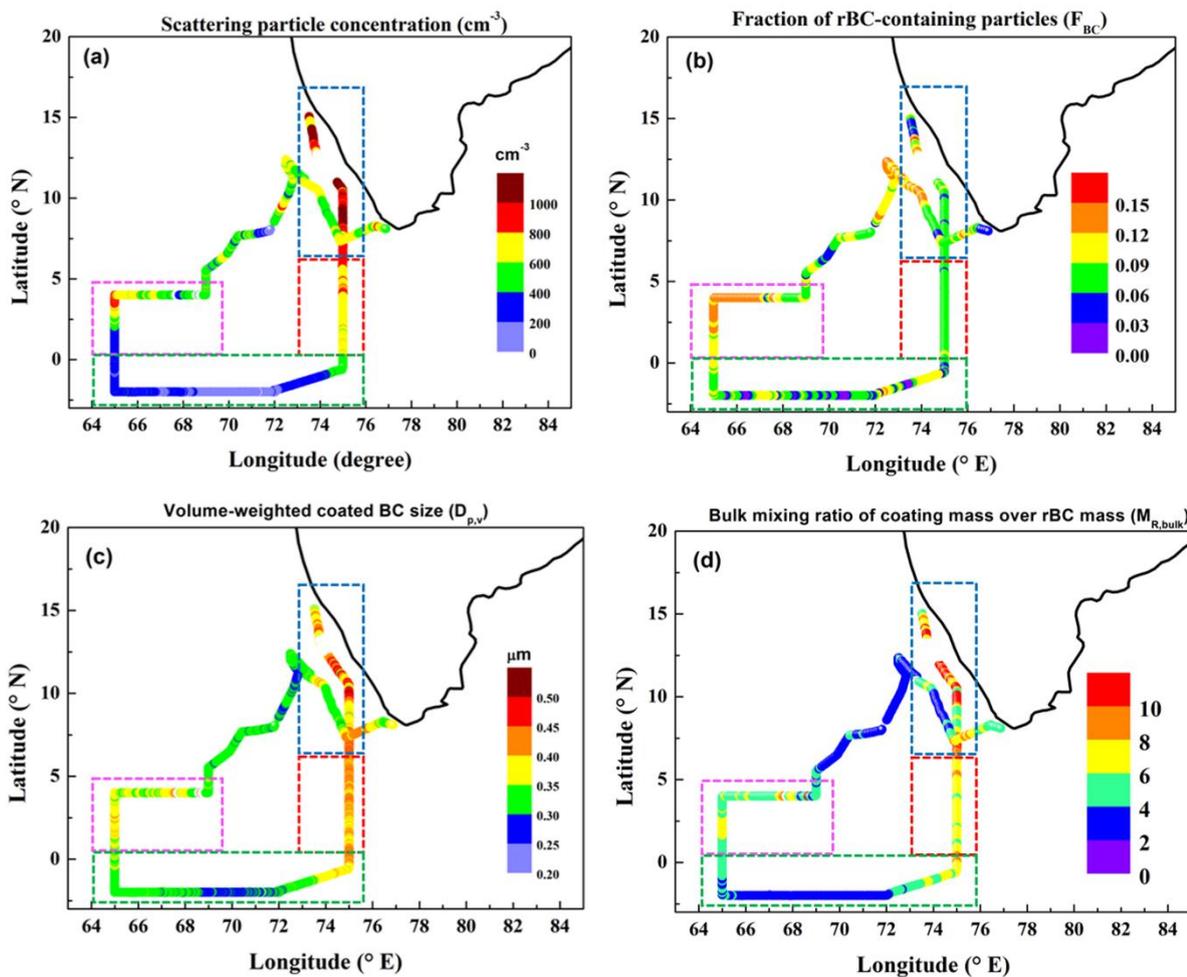


Figure 5: Spatial variation of the (a) scattering (non-BC) particle number concentration (in cm^{-3}), (b) fraction of rBC containing particles (F_{BC}), (c) volume-weighted coated BC size ($D_{p,v}$) (in μm), and (d) bulk mixing ratio of coating mass to rBC mass ($M_{R,bulk}$). Rectangles with dashed

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 \pm are standard deviations.

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

(35)Line 19 – “highER”

Complied with.

“Though $M_{\text{R,bulk}}$ values were very low over EIO, occasional higher values (> 4) are also seen (Fig. 5d)”.

(36) Lines 20–21 – *Is the lower limit of detection of scattering particles the same in this study as it was in the Kompalli et al (2020a) study?*

Yes. The same instrument has been used in both studies. This is now mentioned in the manuscript.

Page 16:

(37) Lines 3-14 – *Above (see my comment 32), you say that comparisons of coating thickness cannot be made, yet here you compare fractional estimates from a few studies with no consideration given to potential differences in the scattering estimates from the studies. Please correct.*

Complied with. This portion is rewritten in the revised manuscript.

(38) Line 20 – *Terminate this sentence at India. You are only measuring a tiny bit of air relative to a large continent. Your results suggest there are similarities between the measurements. There may be something more to that, but you can't say the strength of emissions flowing from the north and south are similar based on this little bit of information. If you had extensive measurements from an airborne platform as well as your ship platform, you might be able to draw some inferences.*

Complied with.

“...Raatikainen et al., (2017) for thickly coated BC particles in polluted outflow environments in northern India ~~highlighting the similarity of the outflow strength (concentrations of the condensable species)~~”.

(39) Lines 22-33 – *The first and last sentences of this paragraph are repetitive. The mixing ratio is just another way of representing coating thickness, and those processes have been discussed. The reference to Liu is useful, but that is all that is needed here.*

Complied with. This portion is modified in the revised manuscript.

Page 18, Line 26:

“The higher bulk mixing ratio of coating mass over rBC mass values ($M_{R,bulk} \sim 2.5-15$) (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 $M_{R,bulk}$ ($\sim 4.24 \pm 1.45$) 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 17:

(40) Lines 10-25 and Figure 6 – An interesting figure, but please indicate what data (where and when) are used in the construction of Figure 6. The last sentence (lines 23-24) is another case of repetition and too many words.

Complied with, gratefully. The portion is now revised as below.

Page 19, Line 15:

“The data collected from 16:41:24 hrs of 21-January-2018 to 18:02:46 of 22-January-2018 (Indian standard time) was used for the construction of the figure (which falls in the transition period between SEAS and NIO-E regions). The same analysis repeated for few other data sets over the other regions also yielded similar results.”

The sentence in lines 23-24 is now deleted.

~~“It appears logical, as the particles over the oceanic regions are rather aged, and away from the potential sources, which are mostly located over the continental landmass, upwind”~~

Page 18 Line 25 – I suggest writing this sentence as “The high proportion of thick coatings on BC particles may result in significant increases in absorption by the BC.”

Complied with. The sentence is rewritten in the revised manuscript.

“The high proportion of thick coatings on BC particles may result in significant increases in absorption by the BC. As reported by Brooks et al. (2019)...”

Page 19:

(41) Lines 8-11 – Re-write as “The large uncoated rBC particles (core diameters $> 0.18 \mu\text{m}$ and thin coatings of ACT $< 50 \text{ nm}$) with low scattering enhancements were also found during our measurements but in smaller quantities, consistent with the findings of Brooks et al. (2019) for the Indian region during the pre-monsoon and monsoon seasons.”

Complied with. The sentence is rewritten as suggested.

“The large uncoated rBC particles (core diameters $> 0.18 \mu\text{m}$ and thin coatings of ACT $< 50 \text{ nm}$) with low scattering enhancements were also found during our measurements but in smaller quantities, consistent with the findings of Brooks et al. (2019) for the Indian region during the

pre-monsoon and monsoon seasons.”

(42)Line 9 – smaller

Complied with.

(43)Lines 18-19 – This should already be discussed in the introduction.

Complied with. This line is moved to the introduction portion.

Page 3, Line 15:

“The information on the nature of the coating material along with the state of mixing of BC particles gives insight into the magnitude of the mixing-induced absorption enhancement for BC (Cappa et al., 2012, 2018; Peng et al., 2016; Liu et al., 2017).”

(44)Lines 21-25 – Rather than spend four lines discussing circumstantial evidence, Figure 6 directly dispels the importance of this notion?

Complied with.

Page 21, Line 41:

“Though it may be possible for BC to be mixed externally with coarse mode aerosols like dust or sea-salt aerosols in the real atmosphere, figure 6 directly dispels the importance of this notion”

Page 20:

(45)Lines 10-12 – You can't call the SEAS region “organic-rich”. Organics and sulphate are equal over SEAS in Figure 7.

Complied with. This is revised now as below:

“To summarise, the South Asian outflow plumes consist of **the more or less equal proportions of organics and sulfate aerosols** in the vicinity of west coast/peninsular India, gradually changing to a completely sulfate-rich aerosol system in the remote oceanic regions.”

(46)Lines 15-20 – You treat the change in organic composition from SEAS to EIO as a chemical loss of organics (this should be referenced). Is it not more likely that the change in organics is due to mixing of marine air and polluted continental air, as suggested by the EIO trajectories for EIO?

Complied with. The following references are added in the revised manuscript.

“Possible oxidation of primary particulate organic matter due to heterogeneous reactions involving oxidants such as OH, O₃, and NO₃ during long-range transport from anthropogenic source regions can result in their volatilization, restricting their lifetime (Molina et al., 2004; Donahue et al., 2006; DeCarlo et al., 2010). However, enhanced sulfate production is possible through gas to particle conversion in the SO₂ rich air masses (especially when ambient relative humidity is higher) (Unger et al., 2006; Meng et al., 2016) originated over the Indian region or through the dimethyl sulfide (DMS) pathway from the marine emissions (Zorn et al., 2008; Shank et al., 2012)”.

The following references are added:

DeCarlo, P.F., Ulbrich, I.M., Crouse, J., de Foy, B., Dunlea, E.J., Aiken, A.C., Knapp, D., Weinheimer, A.J., Campos, T., Wennberg, P.O. and Jimenez, J.L., 2010. Investigation of the sources and processing of organic aerosol over the Central Mexican Plateau from aircraft measurements during MILAGRO. *Atmos. Chem. Phys.* 10, 5257–5280. doi:10.5194/acp-10-5257-2010.

Donahue, N.M., Robinson, A.L., Stanier, C.O. and Pandis, S.N., (2006), Coupled partitioning, dilution, and chemical aging of semivolatile organics. *Environ. Sci. Technol.* 40, 2635–2643.

Meng, C.C., Wang, L.T., Zhang, F.F., Wei, Z., Ma, S.M., Ma, X., Yang, J., (2016), Characteristics of concentrations and water-soluble inorganic ions in PM_{2.5} in Handan City, Hebei province, China. *Atmos. Res.* 171, 133-146.

Molina, M.J., Ivanov, A.V., Trakhtenberg, S., and Molina, L.T., (2004), Atmospheric evolution of organic aerosol, *Geophys. Res. Lett.* 31, L22104. 10.1029/2004gl020910.

Shank, L. M., Howell, S., Clarke, A. D., Freitag, S., Brekhovskikh, V., Kapustin, V., McNaughton, C., Campos, T., and Wood, R., (2012), Organic matter and non-refractory aerosol over the remote Southeast Pacific: oceanic and combustion sources, *Atmos. Chem. Phys.*, 12, 557-576, <https://doi.org/10.5194/acp-12-557-2012>.

Unger, N., Shindell, D. T., Koch, D.M., and Streets D.G., (2006), Cross Influences of Ozone and Sulfate Precursor Emissions Changes on Air Quality and Climate, *PNAS*, 103, 4377–4380.

Zorn, S. R., Drewnick, F., Schott, M., Hoffmann, T., and Borrmann, S., (2008), Characterization of the South Atlantic marine boundary layer aerosol using an aerodyne aerosol mass spectrometer, *Atmos. Chem. Phys.*, 8, 4711–4728, doi:10.5194/acp-8-4711-2008.

(47) What do the details given in the paragraph starting on line 23 and ending on page 21, line 12,

have to do with BC? Most of this appears to be a rehash of Aswini et al. (2020), and could be stated much more succinctly.

Complied with. Most of this discussion is moved to supplementary information in the revised manuscript.

Page 23, Line 5:

“Further, the association between ammonium and sulfate (Fig. S3) indicated an NH_4^+ deficit environment (Aswini et al., 2020)”.

The below discussion is moved to the supplementary.

“The mean molar ratios were ~1.44, 1.47, 1.43, and 1.25 mol/mol over the SEAS, the NIO-E, the EIO, and the NIO-W regions, respectively, which indicated an NH_4^+ deficit (Aswini et al., 2020). The spatial variation of molar ratios of ammonium to sulfate is shown in the Supplement (Fig. S3, panel a). The association between submicrometre sulfate and ammonium mass concentrations depicted an excellent correlation ($r > 0.95$) during the ICARB-2018 (Fig. S3b in the Supplement) with varying slopes indicating the extent of neutralization (higher molar ratios and an increased tendency towards neutralization of sulfate near the coastal regions compared far away regions) of sulfate by ammonium. The present observation is consistent with those reported by Aswini et al. (2020), who suggested that *just-enough* ammonium was present to neutralize sulfate during the ICARB-2018 based on concurrent bulk aerosol chemistry using offline (filter paper sampling) technique. It is possible that when insufficient ammonium is present in the atmosphere, it can lead to the sulfate aerosols existing in forms other than $(\text{NH}_4)_2\text{SO}_4$. A probable reason for ammonium deficiency could be the difference in the lifetimes of gaseous ammonia and SO_2 , which are precursors for particulate sulfate and ammonium. Though dominant sources were located over the land for both the precursors, the sulfate aerosol can be formed from the oxidation of transported SO_2 over the ocean, whereas ammonia is lost rapidly away from the source regions. This is evident from the varying slopes seen in Fig. S3b.”

Page 21:

(48)Line 13 to Page 22, line 21 and Figure 8 - The figure and the associated regressions do not give any significant information other than for the case of smaller coatings: sulphate appears to an important contributor to the coating thickness. However, the role of sulphate in that case has already been established from Figure 7. What might be useful here is a plot of the coating thickness against the sum of organics, sulphate and ammonium. If the sums of those species do not represent a significant fraction of the variation in the coating thicknesses, then either there is a problem with some

of the measurements (ACSM or coating thicknesses) or there is some chemical specie(s) not measured by the ACSM that is involved in the coating.

We agree that it is not possible to establish the direct association between the coating material and submicron aerosol chemistry with the present approach. Therefore, we have not attempted to quantify the exact contribution of organics/sulfate to the coating on rBC particles. We have examined the association between the non-refractory submicron aerosol mass concentrations (NR-PM1.0) from the collocated ACSM and bulk absolute coating thickness of rBC for low and high ACT observations to infer the most probable coating material. For the same mass concentration of NR-PM1.0 species, varying ACT values were seen quite frequently.

As suggested by the reviewer, we have examined the association between absolute coating thickness with the sum of mass concentrations of organics, sulfate, and ammonium, which is shown below. Here also, we have separated the data into two categories consistent with the earlier discussion: ACT is low (ACT < 50 % of MMD) and high (ACT > 50% of MMD). This is done so because regression analysis suggested two distinct regression lines. The bulk ACT showed a reasonable association ($r > 0.48$; $p < 0.01$) with the sum of masses of organics, sulfate, and ammonium for low coating thickness scenario, whereas and the association is weaker ($r \sim 0.31$; $p < 0.01$) during high observations. Corresponding slopes indicated that ACT on BC intensified more steeply with an enhanced mass concentrations of NRPM1.0 species (slope ~ 0.73) during the low ACT scenario compared to the high ACT scenario (slope ~ 0.52). The dominance of one species (sulfate) is also one probable reason for the increased scatter in the association shown below (This discussion is not part of the manuscript and is given here only for the reviewer).

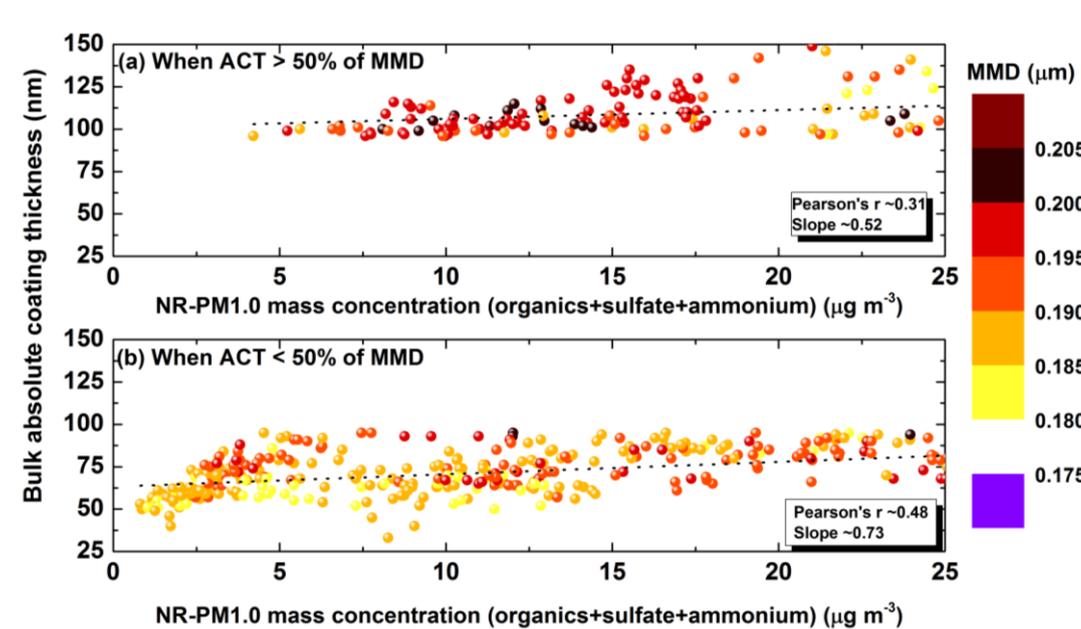


Figure. Scatter plot between the sum of mass concentrations of organics, sulfate, and ammonium aerosols and bulk absolute coating thickness during the ICARB-2018 for the observations with (a) high ACT and (b) low ACT. Color represents the corresponding mass median diameter value. Solid lines represent the linear least-squares fit to the points. Regression slopes and correlation coefficients are written in each panel.

As described in reply to Reviewer-1, we have found that a substantial portion of the non-refractory submicron aerosol mass is bound to the BC particles over the regions with stronger outflow ([Supplementary Figure S4 and corresponding discussion included in the revised manuscript](#)). While higher mean values of the ratio were observed over the SEAS ($\sim 35 \pm 12\%$) and NIO-E (31 ± 4) regions in the proximity of the sources, as we move away to the farther oceanic EIO, the NR-PM1.0 that is bound to BC particles dropped considerably (mean $\sim 23 \pm 8\%$). 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 (Kompalli et al., 2020b). Though the mean fraction of r-BC containing particles remained similar ($\sim 8\%$) 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. We have observed that new particle formation and subsequent growth to larger sizes preceded an enhancement in the mass concentrations of submicron aerosols (mostly organics and occasionally sulfate). This indicated that an increase in the mass concentration of submicron aerosols need not translate into the coating on rBC particles.

(49) I recommend removing the discussion from line 23 of page 20 to line 21 of page 22 as well as Figure 8. Alternatively, you need to significantly improve the value of this discussion.

Regarding this suggestion, we feel that this analysis highlights the complexity in the quantification of the rBC mixing state in the outflow regions such as the present study region. We feel it is appropriate to present the association between the submicron aerosols and coating on rBC to infer the probable coating material in different scenarios. Therefore, we have improved the discussion in the revised manuscript as below and prefer to retain it. The following portion is added in the revised manuscript:

Page 24, Line 21:

“As described earlier, the mean fraction of r-BC containing particles remained similar ($\sim 8\%$) over different regions (SEAS, NIO-E and EIO). But, frequent new particle formation events

(Kompalli et al., 2020b) have resulted in lower F_{BC} ($< 2\%$), which is reflected in the amounts of NR-PM_{1.0} mass that are bound to rBC particles. It was observed that new particle formation and subsequent growth to larger sizes preceded an enhancement in the mass concentrations of submicron aerosols (mostly organics and occasionally sulfate). This suggests another possibility of a reduction in the primary particulate pollution, and the available secondary condensate promotes nucleation in such cases. Also, the portion of the non-refractory submicron aerosol mass bound to the BC particles varied over a wide range of 10-40% over different regions, and especially lower values were seen during NPF periods (not shown here). In Figure S5 in the supplementary, the association between the sum of mass concentrations of organics, sulfate, and ammonium aerosols and bulk absolute coating thickness over different sub-regions covered during the ICARB-2018 is shown. This reveals a very good association ($r \sim 0.90$) over the remote EIO and a weak association over the NIO-E regions, whereas no association was found over the regions SEAS and NIO-W. It also advocates that variation in the submicron aerosol composition can explain alterations to rBC mixing state over the remote regions, whereas the rBC mixing state would be complex in the vicinity of the source regions”.

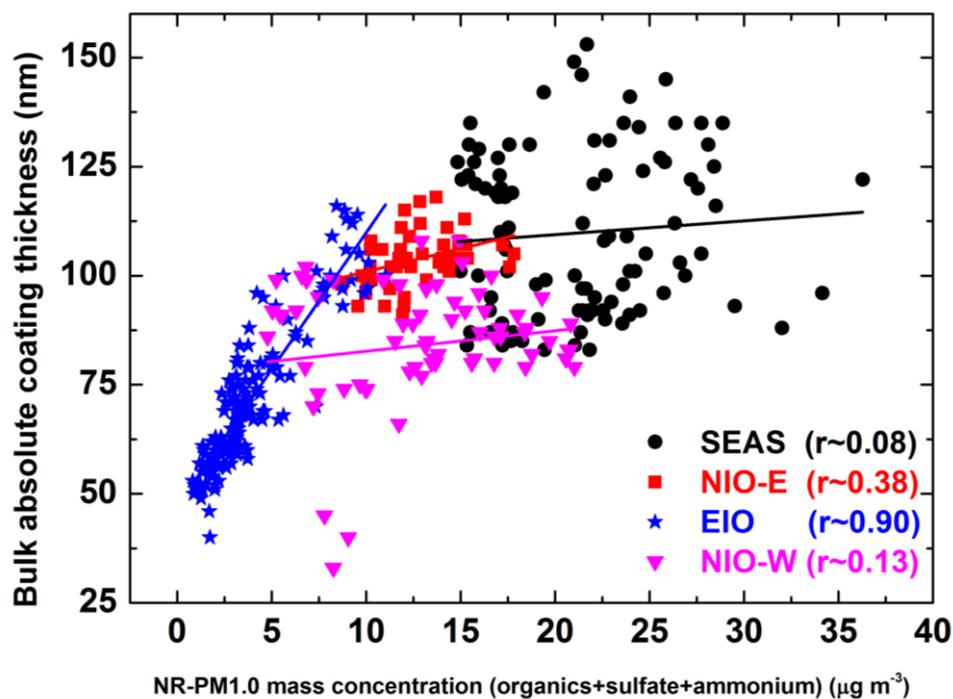


Figure S5: association between the sum of mass concentrations of organics, sulfate, and ammonium aerosols and bulk absolute coating thickness over different sub-regions covered during the ICARB-2018.”

(50) Lines 17-26 – As discussed above, the fractional analysis needs to be improved, and the associated conclusions on these lines altered accordingly.

Complied with.

(51) As above, the NR chemical composition discussion needs improvement.

Complied with.