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Interactive comment on "Mixing state of refractory black carbon aerosol in the South Asian outflow over the northern Indian Ocean during winter" *by* Sobhan Kumar Kompalli et al.

Anonymous Referee #2

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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.

Page 3:

1. Line 20 – "It has a long lifetime." This sentence is a bit abrupt and 'long lifetime'

needs to be defined.

2. 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?

3. 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?

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

Page 4:

5. Line 5 – Everything but the AMS is referenced. Why not the AMS?

6. Line 8 – "such" is redundant.

7. 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".

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

Page 6:

9. 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.

Page 7:

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

11. 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.

12. 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?

Page 9:

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

14. 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.

15. Lines 14-15 – Concentrations of rBC reaching 200 ng/m3 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. . ."

16. 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?

Page 10:

17. Lines 2-5 – References needed.

18. Line 5 – Do you mean "have", rather than "comprised BC particles"?

19. 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?

20. 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?

21. 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)?

22. Lines 19-20 – What do the lower MMD associated with EIO imply?

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

Page 11-12:

24. 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.

Page 13:

25. 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.)

26. Line 12 – "Nearly 95% of the SEAS measurements. . ."?

27. Line 15 – What are you contrasting with? Just start with "Over the NIO-E region. . ."

28. 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.

Page 14:

29. Line 9 – Smaller rather than inferior.

30. 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.

31. Lines 16-18 – Repetitious.

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

33. 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.

34. Line 32 - What is "It" that needs further investigation: coatings, enhancement,

radiative forcing?

Page 15:

35. Line 8 – What does "very high" reference to?

36. 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.

37. Line 19 – "highER"

38. 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?

Page 16:

39. 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.

40. 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.

41. 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.

Page 17:

42. 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.

43. 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."

Page 19:

44. Lines 8-11 – Re-write as "The large uncoated rBC particles (core diameters > 0.18 μm and thin coatings of ACT < 50 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."

45. Line 9 – smallER

46. Lines 18-19 – This should already be discussed in the introduction.

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

Page 20:

48. Lines 10-12 – You can't call the SEAS region "organic-rich". Organics and sulphate are equal over SEAS in Figure 7.

49. 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?

50. 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.

Page 21:

51. 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.

52. 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.

Page 23:

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

54. As above, the NR chemical composition discussion needs improvement.