Response to Reviewer 1

Comments:

The manuscript *Morphological and optical properties of carbonaceous aerosol particles from ship emissions and biomass burning during a summer cruise measurement in the South China Sea* investigated the morphological and absorption properties of BC particles in South China Sea and found that the size and mixing state of BC particles and tar balls differs during ship navigation and stop period, indicate the different aging degrees. Meanwhile, this study revealed biomass burning and fossil fuel combustion contributed respectively to 18–22% and 78–82% of all the BC light absorption, showed that biomass burning was predominantly from the Philippines and Southeast Asia before and after the summer monsoon during the cruise campaign. Generally, the study is interesting and meaningful. The study still needs some improvements. The manuscript needs some revision in order to be published:

We thank the reviewer for valuable comments and suggestions. We have revised the manuscript accordingly. All revised points are indicated in red in the manuscript. The point-by-point responses are given below. Note that we have rearranged the Results and Discussion section per the reviewer #2's suggestion.

1. What's the difference between Feret diameters and geometrical diameters?

Feret diameter and geometrical diameter are different. Feret diameter or Feret's diameter, is a measure of a particle size along a specific direction. We have revised the main text in lines 194-197, "In the analysis of particle size, the Feret diameter is defined as the distance between the parallel tangential lines that constrain the particle perpendicularly. In this study, we applied the Feret diameter as the longest distance between any two points along the boundary of the selected particles."

The term "geometrical diameter" signifies the distance between two points located on the surface of a geometric shape, with this line passing through the center of the shape. In this study, we utilized this concept in the analysis of transmission electron microscopy (TEM) images to quantify the size of tar balls with circular shapes. Specifically, we employed TEM data acquisition software to measure the geometrical diameters of the tar balls.

Why you use the former one to describe BC particles and use the latter to describe tar balls?

The Feret diameter is utilized to describe BC particles in this study because it allows for efficient particle counting capabilities before and after coating vaporization under electron beams using the ImageJ software.

The reason for using geometrical diameter is that bare tar balls are not found within the analyzed samples. Instead, tar balls mixed with other components (e.g., sea salt, organic matter, BC, and sulfate) were observed. Therefore, it is appropriate to quantify the size of the observed tar balls which excluded any coatings or additional materials using geometrical diameter.

We have revised the main text in lines 197-201, "Moreover, we utilized geometrical diameter to describe the size of tar balls with circular shape, which signifies the distance between two points located on the surface of a geometric shape, with this line passing through the center of the shape. The usage of geometrical diameter is reasonable for measuring the size of the observed tar balls which excluded any coatings or additional materials. Specifically, we employed TEM data acquisition software to measure the geometrical diameters of the observed tar balls."

The *Abstract* part is too long, maybe it will be better just listing the most important results in abstract.

The abstract has been revised according to the reviewer's suggestion. Specifically, the methodology section was condensed through the revision of lines 21-22, 26-27, 30 and the removal of the following sentences:

"Single particle samples were classified into two modes: "stop" when the ship was anchored and "navigation" when the ship sailed at high speed."

"The median OC/EC ratios were 8.14, 5.20, 6.35, and 2.63 during BMP, TMP, AMP, and SPP, respectively, showing higher OC/EC ratios for biomass burning emissions than for fossil fuel emissions. Additionally,"

"This study provides information about the morphology and the optical properties of carbonaceous aerosols which can be used to evaluate their effects on light absorption and hence the climatic radiative forcing in the SCS region."

2. In line 54, what's the meaning of onion-like graphite layer microstructures? From the TEM image, the BC particles don't look much like onions.

The term "onion-like" was originally used to describe the microstructure of nano-soot particles. To avoid the ambiguity, it has been revised to "graphene-like layers" and a reference has also been cited (Adachi et al., 2019) in line 48.

3. As for Figure 6, why just chose some BC particles not all BC particles? Since not all BC particles are included in the discussion, the conclusion that small-sized BCs are more easily encapsulated is not very convincing (In line 295).

We should point out that the particles in Figure 6 include pure BC and BC without thick coatings. We chose specific BC particles instead of all BC particles for two primary reasons: (1) A comprehensive investigation of all BC particles (BC-containing particles) has recently been addressed by others (Pang et al., 2022). In their paper, all BC particles were discussed, including fresh soot, partly coated soot and embedded soot particles. It was found that the number fraction of embedded soot particles at the rural sites was higher, and these particles had the highest fractal dimension (D_f), implying that aged BC particles became more compact after long-range transport. However, the characteristics of bare BC or pure BC exposed to other composition have not been comprehensively investigated. We hence focus on the pure or bare BC particles to explore their roles during aging in this study. (2) We convey that most aged BC particles were small after long-range transport, regardless they were initially small or became smaller due to the collapse of large BC aggregates. We have now revised the main text in lines 371-374, "Most BC particles were below 1 μm in Feret diameter during navigation (Figure 7), while their sizes cover a wide range below 3 μm during stop, implying that the aged BC particles become smaller after long-range transport. Despite only a total of 134 BC data points shown in Figure 7, the results are still statistically meaningful due to the wide range of BC sizes covered in our analysis. Note that the size change of a BC particle cannot be determined because the original size of the particle is unknown before the removal of the coatings."

In line 285, *among which were emitted from the own ship (e, f)*: Why only mention e/f, isn't d also from own ship's emissions?

The emissions from the own ship (research vessel) are much easily distinguished from other ships. For example, the BC particles (e, f in Figure 6) are emitted directly from the research vessel, showing the presence of large BC aggregates in the freshly emitted BC particles. In contrast, aged BC particles (d in Figure 6) were thickly coated, which may originate from long-range transport of emissions from distant ships. We have now revised the relevant description in lines 354-359, "Comparatively, a mixture of aged BC particles and much larger fresh BC particles as well as smaller scattered BC particles during stop were found (Fig. 6d-f), which were likely emitted from other ships

(Fig. 6d) and the research vessel (e, f). These TEM images showed that the compressed BC particles are typically more aged and atmospherically processed, while the fractal BC particles are fresh. Moreover, EDS analysis showed that sulfate formed from aqueous processes and less viscous organic coating indicate an aging process. Those BC particles with Feret diameters larger than 2 µm during stop were fractal aggerates which could unlikely survive due to deposition during long-range transport."

4. In line 300， since Tar balls were frequently observed during the campaign, then what's the number fraction of tar balls in all particles?

We estimated the fraction of tar balls to be approximately 11.8% through the number of observed samples containing tar balls divided by the total number of analyzed samples, including both Navigation and Stop samples. We have now included this information in the main text in line 377, "Tar balls were frequently observed during the campaign with an estimated fraction of 11.8%."

5. There are mismatches between the appendix images and the image numbers mentioned in the main text: (1) Fig S5 is Map of the ship route, but line 287 says Fig S5 demonstrate "heavily coated internal BC particles were found during stop"; Fig S6 is titled "particles taken during navigation", but line 306 says Fig S6 contains tar ball mix with BC taken during stop. There are many more descriptions that don't match up.

We have thoroughly double checked and corrected all the mismatches/discrepancies both in the main text and the Supplementary Information (SI).

6. In line 335, *EC concentrations during SPP, ranging from 0.15 to 22.8 µg m^{-3}*, But the EC concentration range for SPP in Fig 9 is around 1.7, why is that?

The EC concentrations ranged from *0.15 to 22.8 µg m^{-3}* with a median concentration of 1.7 µg m^{-3} during SPP (Figure 9). We have revised Figure 9 and added more discussion in lines 399-402, "Compared with Figure 9d, the higher and more scattered OC/EC ratios in Figure 9a, b, c are caused by the very low EC concentrations. The presence of extremely low EC concentrations, often falling below or close to the detection limit, can introduce discrepancies in the calculation of the OC/EC split, ultimately resulting in inaccurate EC concentrations (Bauer et al., 2009)."

References

Adachi, K., Sedlacek, A. J., III, Kleinman, L., Springston, S. R., Wang, J., Chand, D., Hubbe, J. M., Shilling, J. E., Onasch, T. B., Kinase, T., Sakata, K., Takahashi, Y., and Buseck, P. R.: Spherical tarball particles form through rapid chemical and

physical changes of organic matter in biomass-burning smoke, Proc. Natl. Acad. Sci. U.S.A., 116, 19336-19341, https://doi.org/10.1073/pnas.1900129116, 2019.

Bauer, J. J., Yu, X.-Y., Cary, R., Laulainen, N., and Berkowitz, C.: Characterization of the Sunset semi-continuous carbon aerosol analyzer, J. Air Waste Manag. Assoc., 59, 826-833, https://doi.org/10.3155/1047-3289.59.7.826, 2009

Pang, Y., Wang, Y., Wang, Z., Zhang, Y., Liu, L., Kong, S., Liu, F., Shi, Z., and Li, W.: Quantifying the fractal dimension and morphology of individual atmospheric soot aggregates, J. Geophys. Res. Atmos., 127, 10.1029/2021jd036055, 2022.