Anonymous Referee #1

General comments

The authors present an experimental study of aerosols collected from Hainan Island, South China. The analysis includes absorption coefficients, mass concentrations of black carbon, organic carbon, inorganic elements, and water-soluble cations and anions. Major findings include the source apportionment of the total absorption coefficient and contribution to radiative forcing. The study shows the importance of considering ship emissions in forcing calculation. Overall, the manuscript presents interesting data and analysis shows merit.

Response: We thank the reviewer for his/her valuable suggestions, and it is useful for improving our manuscript. We have made modifications accordingly based on the reviewer's comments. Below are point-to-point responses.

Specific comments

1. This study uses AE-33 and PAX to measure absorption. AE-33 provides the mass of absorbing aerosols as final products. Previous studies have reported the calculation of absorption coefficients from AE-33 mass concentration. However, for the sake of completeness, I would recommend to include those steps in supplementary.

Response: We followed the reviewer's suggestion and added the following in the revised manuscript:

> "Since the AE33 aethalometer records the BC mass concentrations, the Abs(λ) at each wavelength were retrieved by getting the product of BC mass concentration ([BC]) and mass absorption cross-section (MAC) used in the instrument (Abs(λ) = [BC] × MAC) (Drinovec et al., 2015)."

2. This study used a Nafion dryer to reduce the RH of particles collected. These dryers are known to minimize particle concentration during the drying process. Is there any data on the % of particle loss within the dryer?

Response: We conducted a test to compare the measured light absorption coefficients

(Abs(λ)) with and without the Nafion tube. As shown in Fig. R1 below (also see Fig. S2 in the revised supporting information), the loss of Abs(λ) is little and can be ignored. We have added a sentence to show the result of this test in the revised manuscript. It reads as follows:

"As shown in Fig. S2, the loss of Abs(λ) caused by the dryer was ignored."

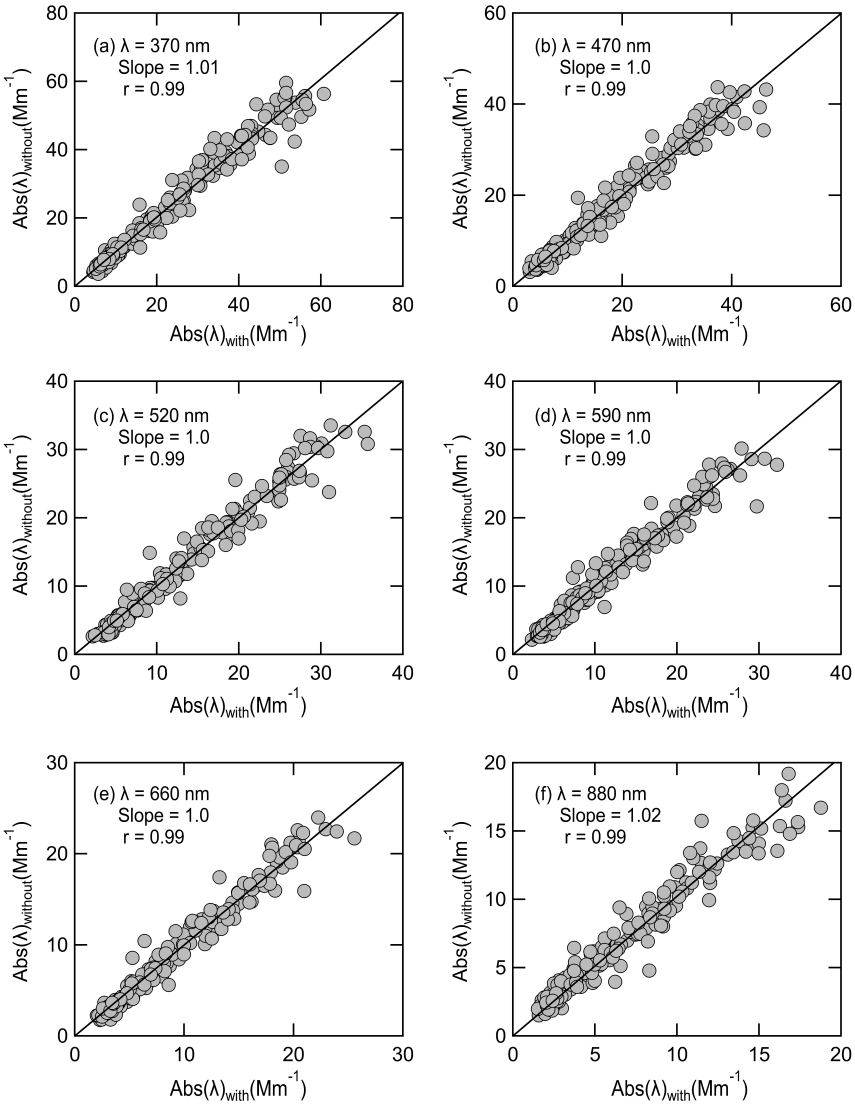

Figure R1. Scatter plot of light absorption coefficient measured with ($Abs(\lambda)_{with}$) and without ($Abs(\lambda)_{without}$) Nafion dryer (MD-700-24S-3). λ is the wavelength of 370, 470, 520, 590, 660, or 880 nm.

3. The Nafion dryers were connected to Aethalometers only? Aethalometer data is less susceptible to RH. But the PAX data can be influenced by high RH. Was there a dryer

connected to PAX?

Response: The PAX and AE33 share a same sampling tube and was set in parallel with a tee. Therefore, the PAX and AE33 were both dried by the Nafion dryer. We have clarified and added the following information in the revised manuscript:

> "It was set in parallel with the AE33 aethalometer using the same $PM_{2.5}$ cyclone and Nafion® dryer."

4. There was a $PM_{2.5}$ cyclone for Aethalometer and no cyclone for PAX. I remember the penetration efficiency of PAX reduces drastically after 1 micrometer. So, both instruments were measuring different size-cutoff particles.

Response: We apologize for our unclear description. As replied above, the PAX and AE33 were both collected the ambient aerosols using the same $PM_{2.5}$ cyclone. Therefore , the same size range of particles was measured by the PAX and AE33.

5. What is the area of quartz filters used?

Response: The area of quartz filter is 8 × 10 inch. We have added this information in the manuscript. It now reads as follows:

> "The $PM_{2.5}$ quartz-fiber filters (8 × 10 inch) (QM/A; GE Healthcare, Chicago, IL, USA) were collected during the day (from 08:00 to 20:00) and at night (from 20:00 to 08:00 the next day) using a high-volume air sampler (Tisch Environmental, Inc., USA) with a flowrate of 1.13 m^3 min^{-1}."

6. What is the flow rate of the high-volume sampler?

Response: The flowrate of high-volume sampler was 1.13 m^3 min^{-1}. We have added this information in the manuscript as shown above response.

7. One major shortcoming in this study is the absence of 'lensing effect' while calculating absorption. Studies have shown that the lensing effect can contribute to significant absorption. Since Aethalometer uses a filter tape to collect particles, one can

assume the core-shell structure of particles (the reason for lensing effect) gets destroyed. But the absorption from PAX will have contributions from the lensing effect. The slope of 2.29 in Figure S3 might include the lensing effect. Since the experimental setup used in this study does not measure the absorption of core-shell and core separately, it will be difficult to distinguish the contribution from the lensing effect. I would suggest the authors include this possibility in text.

Response: We thank the reviewer for explanation the impact of 'lensing effect' on comparison of PAX and AE33. In the revised manuscript, we have added this possible effect:

> "A slope of 2.3 was regarded as the correction factor and was comparable to the values of 2.0–2.6 reported by previous studies using a similar method (Qin et al., 2018; Tasoglou et al., 2017; Wang et al., 2019b). This difference may mainly be related to the matrix scattering and lensing effects."

8. Figure 1a – shows the apportionment of Abs, and the same is repeated as Figure 1b. Removing the repeated portion from 1a would give better visibility to it.

Response: We followed the reviewer's suggestion and modified this figure as shown in Fig. R2 below (also see Fig. 3 in the revised manuscript).

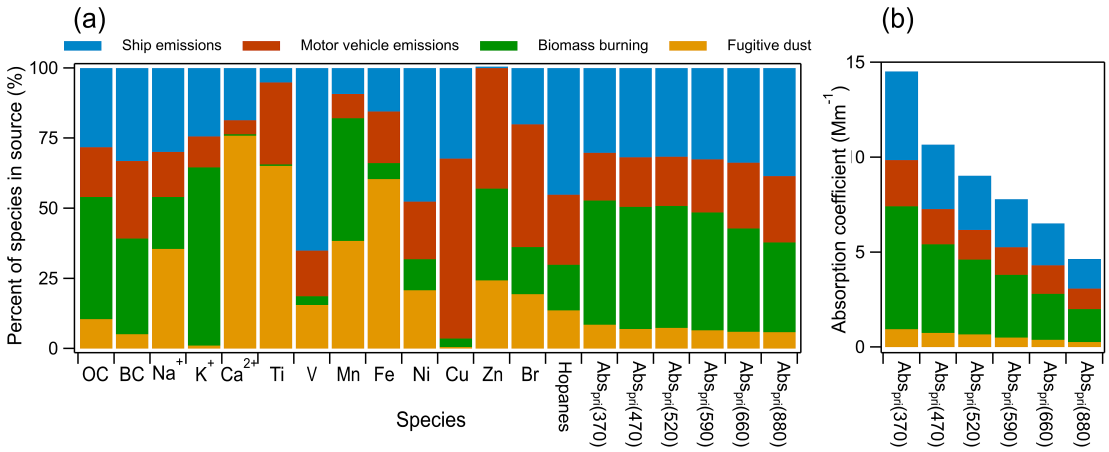

Figure R2. (a) Contributions of the four sources to each species from the positive matrix factorization model and (b) the light absorption of primary aerosols from each source at different wavelengths ($Abs_{pri}(\lambda)$, λ = 370, 470, 520, 590, 660, and 880 nm)

during the study.

9. Page 2, line 13- Optical properties of LAC is not just related to its source. It also depends on the atmospheric conditions and secondary processing.

Response: We agree with the reviewer and revised the original sentence to:

> "The optical properties of LAC aerosols are closely related to their sources as well as atmospheric conditions and secondary processing."

10. Page 4, line 3 – Educational and residential areas will have their pollution sources such as vehicles, cooking, etc.

Response: We agree with the reviewer. In the revised manuscript, we have revised the original description to:

> "The sampling site is predominantly an educational and residential area with typical urban sources of emission including vehicles and cooking appliances."

11. Page 5, Paragraph 1 – The whole paragraph is about the analysis of filters collected. It must be specified initially.

Response: Following the reviewer's suggestion, we have added a sentence to clarify this in the revised manuscript:

> "The collected quartz-fiber filters were used to analyse inorganic elements, carbonaceous matter, water-soluble ions, and organics."

12. Page 7, line 4 – Which PMF system was used for the analysis? I guess US EPA PMF 5.0! It needs to be mentioned with a reference.

Response: Yes, the version of PMF5.0 from US EPA was used in our study. We have added this information in the revised manuscript. It now reads as follows:

> "The PMF version 5.0 (PMF5.0) from the US Environmental Protection Agency (Norris et al., 2014) was applied to determine the contribution of various sources

to aerosol light absorption."

13. Page 9, line 24 – Error bars on Y-axis needed. Since the X-axis is from filters (12-hour sample) and the Y-axis is the average of the same from AE-33 Abs, the error bars are required to see the spread of data.

Response: We followed the reviewer's suggestion, and the revised version is shown in Fig. R3 and Fig. R4 below (also see Fig. S5 and Fig. S6 in the revised supporting information).

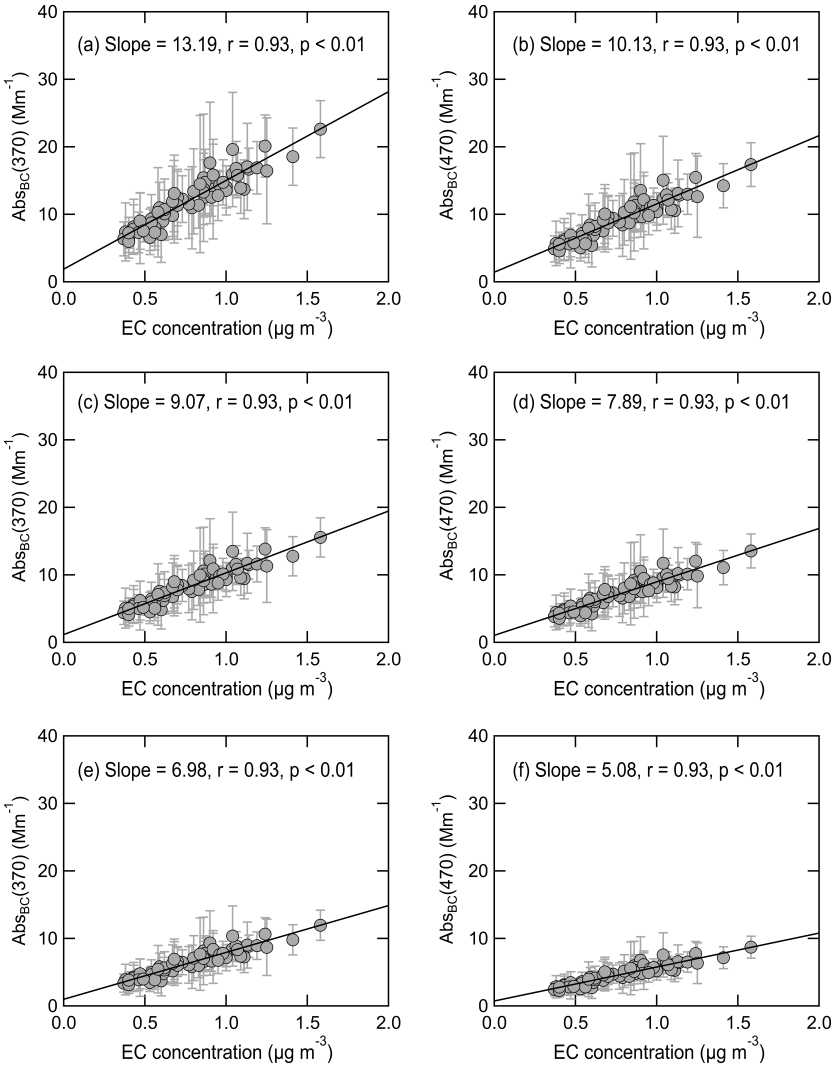

Figure R3. Scatter plots of light absorption of black carbon at different wavelengths ($Abs_{BC}(\lambda)$, λ = 370, 470, 520, 590, 660, and 880 nm) versus mass concentration of elemental carbon (EC). The black lines are the linear regression. The vertical error bars represent one standard deviation of $Abs_{BC}(\lambda)$.

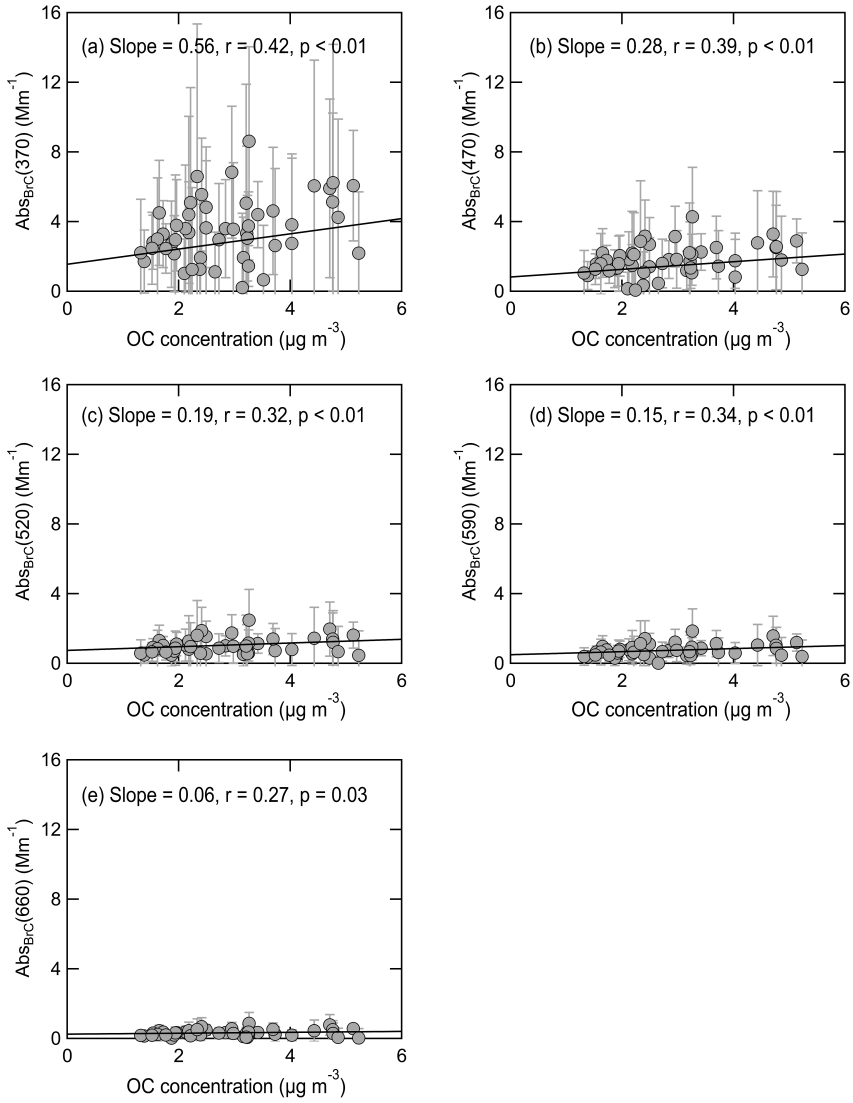

Figure R4. Scatter plots of light absorption of brown carbon at different wavelengths ($Abs_{BrC}(\lambda)$, λ = 370, 470, 520, 590, and 660 nm) versus mass concentration of organic carbon (OC). The black lines are the linear regression. The vertical error bars represent one standard deviation of $Abs_{BrC}(\lambda)$.

14. Page 12, line 1 – The cluster 2 back trajectory doesn't touch the Vietnam cost to influence the biomass burning. Was there a spread towards land for this cluster?

Response: Thanks for the reviewer pointing out this issue. In the revised manuscript, we reworked this paragraph to avoid any misunderstanding. It now reads as follows:

"Cluster #2 originated from the South China Sea near the Indochina Peninsula and accounted for 35% of the total trajectories. The $Abs_{ship}(\lambda)$ was also vital in

this cluster, accounting for 34–37% of $Abs_{pri}(\lambda)$. Fig. S10 shows that the $Abs_{pri}(\lambda)$ of Cluster #2 displayed a similar diurnal trend as that of Cluster #1. Considering that the air masses of Cluster #2 also originated from the South China Sea, the sources except for ship emissions were mainly influenced by local discharge."

Technical corrections

15. Page 2, line 26 – Don't use 'firstly'. 'First' is fine.

Response: Change made.

16. Page 4, line 10 – 'As described previously' – It is not described anywhere before.

Response: This sentence has been revised to "Afterwards, seven light emitting diodes (λ = 370, 470, 520, 590, 660, 880, and 950 nm) in the AE33 aethalometer were used to irradiate the filter deposition spot to obtain light attenuation as previously described (Drinovec et al., 2015)."