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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal variation of black carbon over the South China Sea and in various continental locations in South China" *by* D. Wu et al.

Anonymous Referee #3

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General comments:

This paper presents a set of measurements of absorption over regions in Southeastern China and the South China Sea. These measurements are made during two of the different phases of the local Monsoon, and look at the differences in the ensuing values in these periods. While the measurements do not represent a sufficiently long record to be representative of the two periods in question, the results do present an interesting look into how these periods of time may be represented by different sources and processing of BC in-situ. The paper therefore adds to the overall literature and knowledge and I recommend eventual publication. However, the paper has many flaws

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to be addressed and improvements that must first be made. I believe, however, after a significant effort, that a paper worthwhile to the community can be achieved, and therefore want to encourage the authors to work hard to address all of the comments below.

Specific comments:

1. Since the dry and wet phases of the Monsoon in this region are far longer than 36 days and 28 days respectively, how can these limited periods of time be considered "representative" of the "wet season" and "dry season" respectively? The dates are representative of the dates only, and this should be changed throughout the paper.

2. Given the fact that many fires occur during the "dry phase" of the Monsoon and that they are responsible for a large amount of the BC loading, it would seem inappropriate for this study to consider the results from their limited sampling of the dry phase in particular as "representative". The dates are representative of the dates only, and this should be changed throughout the paper. This is especially so since so early in the dry season the impact of fires is significantly less than later in the dry season!

3. The introduction's background literature search is out of date and not comprehensive. For example two recent important papers that are not referenced are Tao et al., 2012 and Wang 2013.

4. BC is now considered possibly the second largest "global average warming agent" even ahead of methane, as given by Cohen and Wang (2013).

5. The last paragraph of chapter 1 points out that the dry season is from October to April, and therefore provides the fact that the sample period is far too short to be representative of the entire dry season. This needs to be made clear throughout the text.

6. I thought that there was a Monsoon Climate, based on the seasonality from October to April. Now the author is talking about winter? Which is it? What is the definition?

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Perhaps a climatology of the winds should be presented throughout whichever period the paper wants to emphasize and this can be presented as an additional figure. “The prevailing northerly winds in winter make XK a receptor site of pollution in Guangzhou.”

7. The South China Sea has two separate large influences, in which one has a single Monsoon overpass and the other has Two Monsoon overpasses. I am not sure which one YZ represents, but for this reason, it certainly is not an “average” area as expressed by the statement below. “YX represents the average situation of the tropical area over the Southern China Sea.”

8. The following assumption is a very strong assumption and may lead to significant errors. While it is thoughtful that it has been mentioned, perhaps an equation should be added so that it is clear to the reader. Additionally, references should be made to the fact that there is a range of attenuation cross-sections, which vary extremely widely (eg: Bond and Bergstrom, 2006). Finally that coated BC, which occurs frequently in regions which have a mix of sulfur and VOCs from urbanization as well as BC, causes additional absorption over and above the mass of BC alone, and that this lensing effect is not being taken into consideration. “BC concentration is then derived from the attenuation measurement by adopting specific values for attenuation cross section. The latter was obtained from comparison of attenuation and EC mass, which was determined to be 16.6 using a thermal analysis method”. You have alluded to this at the end of section 2, but this must be made clearer. “It is important to keep in mind that _abs is what Aethalometers directly measure while BC concentrations by Aethalometer are derived from _abs measurement assuming that Mass Absorption Efficiency (MAE) of BC aerosol is a constant during the sampling period. However, in reality MAE varies in time and space, depending on the mixing state of BC. As a result, BC concentration data have additional uncertainties due to the uncertainty introduced by the constant multiplier (MAE).”

9. Not true at all! Actually as shown in Cohen and Wang (2013) and Chung et al. (2012) many in the community are depending on light absorption measurements. “As there is

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a larger community interested in BC mass concentrations, data in this paper are mainly presented in the form of BC mass concentrations." Therefore, I strongly advise that the light absorption measurements be treated on equal footing or more footing, and be prominently presented in the paper since they are, as the authors acknowledge above, the actual piece of information being measured.

10. Looking at Figures 2a and 2b, I strongly disagree that YX has the sharpest frequency distribution. The order of magnitude between the high and low values is very small, and possibly within the order of error as given above in parts 8 and 9. Furthermore, no analytical value is given, since the ideal of "sharpest frequency distribution" is not mathematically or statistically rigorous. Perhaps a single number can be obtained to describe the variance or the variability of the frequency distribution, and then this can be compared. Remember, that the instrument error and uncertainty must also be quantified. From my perspective, I am not even sure if the change at the YX site is statistically significant. "The histograms show that YX has the sharpest frequency distribution among all sites in both the rainy and dry seasons as a result of small temporal variation in BC concentrations."

11. The meteorological parameters on Figures 2a and 2b are confusing. I have found precipitation and surface pressure, but I am not sure which curve corresponds to which site. Also, I do not see any mention of wind speed, solar radiation, boundary layer height information, or relative humidity, all also important meteorological variables!

12. The author mentions that AAE is greater than 1 and less than 1.6 for processing/coating by sulfate, nitrate, and other non-absorbers. Then states that the AAE was measured as 1.06, and then goes on to state that there is negligible biomass burning. It is not clear to me how biomass burning comes to play here. Biomass burning, as a function of the temperature, could produce more or less BC to OC and different types of OC. Hence; it is not obvious why there is any connection with biomass burning here. The AAE of 1.06 in the dry season implies that some "coating" has occurred, and does not talk one way or another about biomass burning. "The AAE values are close to unity,

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suggesting the negligible influence from biomass burning, which otherwise would lead to significantly elevated AAE due to atmospheric processing of BC particles." The next sentence explains it well in that this merely implies that there is only a moderate amount of processing, or that more of the sources are local in nature. Again, the absence of presence of biomass burning is not a result that can be gleaned from this information.

13. Given that MFS and the urban regions of the PRD are a mere 20km apart (regional to mesoscale) and the Monsoons occur over a much larger scale, you must present strong evidence to state that it is "upwind" in one season and "downwind" in another season. This data is lacking from this current version of the paper.

14. Given that the concentrations at MFS are so much lower than the other PRD regions sites, it is not at all obvious that the variations in NFS were independent of the other PRD sites in the samples taken during the raining times due to the fact that it was raining. It could be due to noise in the measurements. This needs to be explored using a more powerful and analytical technique, rather than just by "eyeball" methods. This may be true, but I am not convinced. Where is the equation used to calculate the correlations presented in Table 1?

15. I am confused. Due to the way in which rain removes BC, wouldn't the reduction be expected in absorption during the rain time, not after the rain time? "The BC reduction was not significant after rain events (Fig. 2a), implying that wet deposition was not the major cause for lower BC in the rainy season."

16. Mention of wind speed and precipitation are mentioned, but nothing about the boundary layer height. This is a critical shortcoming that makes much of the analysis confounded at best.

17. The fact that YS is a "tourist town" is deceiving. I have been there in person. I have observed a large number of vehicles transporting tourists around, including heavily polluting boats, older motorcycles, and other transport options. While I found the air generally free of local sources, there are many regions near these heavily trafficked ar-

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teries that could have a high level of local emissions. Perhaps the site location should be better explained. “The unexpectedly high level of BC in YS, considering it is mainly a tourist town, may suggest that emissions from nearby mining and metallurgical industries had a significant impact on BC level at YS.”

18. The way in which back trajectories were used is fundamentally flawed and incorrect for two reasons. I strongly recommend completely re-doing this portion of the paper, instead using a forward model. First of all, due to boundary layer mixing, one must sample all heights within the local boundary layer to obtain some idea of how local mixing will come to play when using large scale reanalysis meteorological fields as are used by HYSPLIT. Secondly, HYSPLIT is only capable of tracing dry air trajectories, and BC, with its large removal due to precipitation, will not follow the same pathway. For example, dry air passing through a rainstorm or cloud will not be impacted (other than through localized convection), whereas the impacts on the BC in the air will include wet deposition as well. “For the understanding of seasonal variation of BC, air mass back trajectories at the sampling locations were examined for all sampling days using the HYSPLIT-4 model (Draxler and Rolph, 2012). YX (16.33_ N, 112.83_ E), MFS (23.33_ N, 113.48_ E) and NC (23.00_ N, 113.36_ 5 E) are selected as the reference points for the back trajectories calculation to represent the South China Sea and the PRD region. Height of 150m is chosen to track the path of air masses which would eventually arrive at the NC and YX in the previous 72 h while for MFS the height was set as 535m to represent the real situation. Figure 3 shows the back trajectories of air masses arriving at YX, MFS, and NC in both rainy and dry seasons.”

19. The broad conclusions about the Rainy Season airflow patterns are not readily supported. Were these results found for the entire Rainy Season, or just the 28-day period being analyzed? If it is only for the 28-day period analyzed, how relevant are these results to the remainder of the rainy season? I doubt very highly relevant, although a proper statistical analysis could clear this point up. However, it would have to be done correctly, for example as mentioned in 18 above, by doing back trajectories

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over a variety of heights and initial conditions. “During the rainy season PRD was significantly affected by two different air flows. For most of the time, PRD was affected by the southerly air flow that originated from the vast ocean. On a few days PRD was affected by the northeasterly air flow, which was related to specific weather systems such as typhoons and troughs.”

20. Again, I make a similar comment with respect to the dry season meteorology. If it is only over the 36-day period, perhaps a more careful analysis can reveal the actual nature of the flow. I do not see how it can be concluded to be applicable to the entire dry season however. “During the dry season, PRD was influenced by the strong northeast monsoon, which brought polluted air masses from the more economically-developed regions in the eastern Asia.”

21. You mention wind speed reduction, but what about boundary layer height change? Often one is associated with the other. This could more than overcome the observed seasonal difference. This must be calculated and included.

22. In much of the dry season the YZ island area is located in winds, which originate from urban areas in Southeast Asia such as Hanoi and Ho Chi Minh City in Vietnam. The back trajectories seem to indicate a very different behavior, indicating that the time periods chosen are not representative of the Dry Season. Again, this point needs to be corrected throughout the entire paper.

23. Yes, the observed patterns are different during the cold front. However, a valid explanation is not given. Is it a change in the boundary layer height associated with the front? Is it a cleaner source region being transported in? Is the air generally older or younger? Just because the air is from higher in height does not mean that it is necessarily cleaner! It has to do with the source region of the air, and how long it has been separated from the source region. These explanations must be thoroughly addressed, which is not done in this case.

Technical corrections:

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1. The following is out of place and should be completely removed from the paper: “In China, a number of recent projects have focused on carbon aerosol, including projects sponsored by the National Natural Science Foundation of China and various 10 international cooperation research projects. Chinese researchers have studied various aspects of BC aerosols, such as the physical characteristics, optical properties, sources, temporal and spatial distribution and the impact on the environment and climate, using multiple approaches including field observations, laboratory investigations, numerical simulation and theoretical investigations.”

2. The following statement is not accurate, as discussed above, since the measurements are not sufficiently long to determine the seasonal characterization. They are representative of their measurement period however. “In this work, we report BC and aerosol light absorption measurements by Aethalometers and their seasonal variations in 2008 in a remote location over the South China Sea and six continental locations in South China”

3. While the PRD is indeed highly populated, and Guangzhou, Hong Kong, Shenzhen, Dongguan, Zhuhai, and Foshan are megacities, Macao certainly is too small to meet that definition.

4. The following sentence is factually incorrect and needs to be changed, since Chongqing is the biggest mega-city in Southern China, and Guangzhou is second. “Among the stations, NC, PY, XK and MFS are located in different districts in the city Guangzhou, the biggest mega-city in southern China.”

5. It is the South China Sea, not the “Southern China Sea”.

6. What does “In the dry season MFS is upwind of the PRD region, making it an indicator for super-regional transport.” mean? I am not familiar with this term, super-regional transport. Is it meso-scale, regional-scale, global-scale, etc.? Could a number be put on this?

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7. Another relevant paper that investigated the impact of the temporal variation of emissions and found a similar result to yours (and can be added to your citation list) is: Cohen and Prinn, 2011.

8. There are many grammar mistakes throughout the entire piece. Many are related to verb/noun agreement issues. The paper must be thoroughly reviewed by a native English reader/writer or another who can help successfully make such edits. The good thing is that the grammar mistakes do not distract from the overall logic or points, and hence the paper is still easy to review.

References:

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