

Point-by-point response to review comments on manuscript acpd-13-17375-2013

“Seasonal variation of black carbon over the South China Sea and in various continental locations in South China”

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Reviewer III

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 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:

Response: We thank the reviewer for his/her constructive comments. Our point-by-point response to the review comments is listed below. The revised manuscript that has incorporated response to reviewer comments is submitted together with this response. Line numbers cited in our response refer to line numbers of the revised manuscript.

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.

Response: Points taken. We agree that strictly speaking the sampling periods do not necessarily represent the entire rainy and dry seasons. We have revised the paper title and the text throughout the paper to clearly indicate that the measurements were only for two periods in the wet and dry seasons.

The revised title is:

“Black carbon over the South China Sea and in various continental locations in South China

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!

Response: In terms of temporal distribution in Asia, biomass burning intensity peak in March – April and Sep (Streets et al., 2003; Deng et al., 2008). We agree that biomass burning emission intensity during our measurement period was relatively low compared to other months. We rephrased relevant contents in manuscript (see response to the previous comment).

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.

Response: We updated the introduction part by including the suggested literature.

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

Response: Suggestion taken and relevant content added (Lines 32-33).

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.

Response: Agree. Revision made (see response to comment 1 by reviewer III)

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? 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.”

Response: Dry season is sometimes also referred as “winter” for our study region. For example, Northeast monsoon in dry season as mentioned in our manuscript, is also called “South China Sea winter monsoon” in some literature (Wang et al., 2009). However, to keep the consistence and to avoid confusion we have revised the text and “dry season” is used throughout the paper .

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.”

Response: Owing to the geographical location, YX represents the northern part of the South China Sea. Classification of SCS monsoon is still controversial as a single or joint subsystem of East Asia monsoon , Indian monsoon, and Australian monsoon (Wang et al., 2009), but this is not the focus of our study. April to May is the transition period of the Northeast monsoon to the South China Sea monsoon. June to September is the South China Sea monsoon-dominated period. October is the transition period of the South China Sea monsoon to the Northeast monsoon. November to March is the Northeast monsoon-dominated period. We include these contents into manuscript to clarify the characteristic of YX.

Lines 114-115:

“YX Island, owing to its geographical location, represents the average situation of northern part of the South 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 τ_{abs} is what Aethalometers directly measure while BC concentrations by Aethalometer are derived from τ_{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).”

Response: We further elaborated the discussion on uncertainty of the conversion factor.

Lines 138-146:

“The multiplier 16.6 (specific attenuation cross-section, SACS) is an empirical conversion factor that converts attenuation to mass concentration. SACS has a different physical meaning from mass absorption efficiency (MAE), which converts absorption to mass concentration. Attenuation reported by Aethalometers suffers from sampling artifacts due to aerosol loading, filter matrix, and scattering effect (Coen et al., 2010). Therefore correction is needed to obtain absorption coefficient (σ_{abs}) from attenuation. SACS already contains information of artifacts correction and MAE. For this reason, the value of SACS is usually larger (~ 2 times higher at 550nm) than MAE and SACS cannot be directly compared with MAE reported in literature.”

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 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.

Response: While it is debatable whether BC or light absorption data are used by more researchers in the atmospheric community, we feel presentation of both in equal footing is redundant. We would like to keep the presentation of BC data in the paper while the light absorption data are still available to readers through the supplementary material document.

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.”

Response: The BC level at YX ($\sim 0.6 \mu\text{g m}^{-3}$) is higher than the Aethalometer detection limit ($\sim 0.1 \mu\text{g m}^{-3}$) (Allen et al., 1999). So detection is not a problem for Aethalometer in our study. BC loading at YX was two magnitude higher than those measurements by Aethalometer in remote areas ($0.006 \mu\text{g m}^{-3}$) (Bodhaine, 1995). The figure below (Figure R1) shows the frequency distributions of measured BC and fitted normal distributions at each site. The geometric standard deviation of the fitted normal distribution is the smallest (i.e., sharpest distribution) at YX.

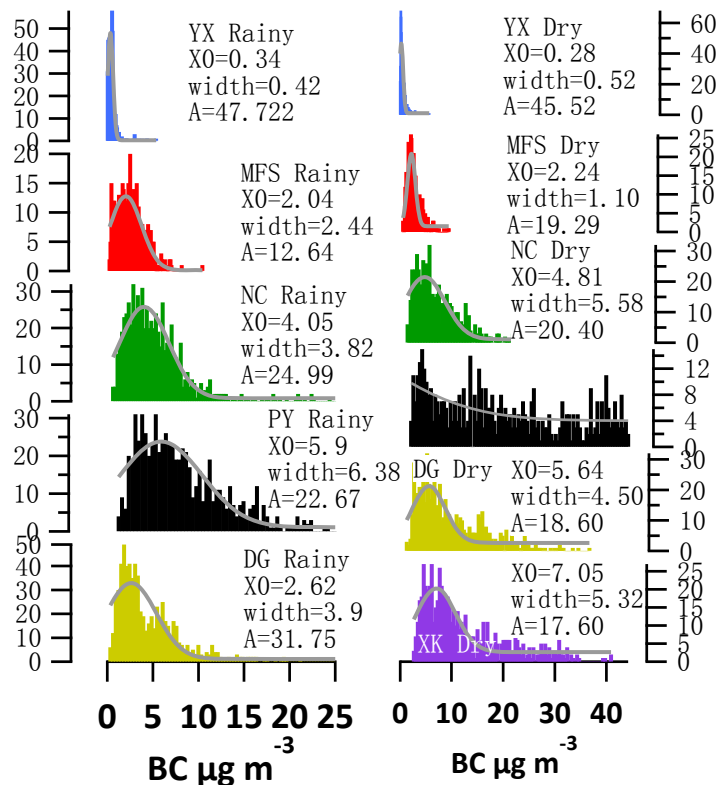


Figure R1. Frequency distributions of BC measurements and respective fitted normal distributions at individual monitoring sites

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!

Response: Which curve represent which site is described in the Figure caption, as it would make the plots too crowded if we label the legends on the plot. Wind speed information is already in the figure (the lowest plot). Solar radiation and relative humidity information are less useful for this study, therefore they are not presented. Plots of boundary layer height data at YX and PY (from balloon measurements) are now included in the revised Figure.

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, 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.

Response: We agree that the discussion pertaining to biomass burning is not a result that can be necessarily inferred from the BC measurements in this study. It’s true that the elevated AAE above unity may be due to aging (coating by non-absorbing materials) or presence of brown carbon, or both.

Information available in this study is not sufficient to differentiate these two factors. We have deleted the text related to biomass burning.

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.

Response: We have deleted this statement in the revised manuscript.

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 MFS 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 ?

Response: The average concentration during the sampling period (2.6~2.9 μgm^{-3}) is much higher than the detection limit of Aethalometer ($\sim 0.1 \mu\text{gm}^{-3}$) (Allen et al., 1999). We do not think detection limit was a problem in our study. The equation used to calculate the correlations in Table 1 is given below.

$$\text{Correl}(X, Y) = \frac{\sum (x - \bar{x})(y - \bar{y})}{\sqrt{\sum (x - \bar{x})^2 \sum (y - \bar{y})^2}}$$

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.”

Response: Thanks for pointing out. We did not pay sufficiently close attention to differentiate during and after rain period. We re-examined the data before, during and after rain events and noted that BC decreased during rain event and after rain BC could increase. Take one example: On 6-7 June 2008, there was a rain event of 78 mm from 3:00 to 6:00 next day.

- The average BC concentration in the 24-h before rain is 5.87 $\mu\text{g}/\text{m}^3$ at NC and 9.47 $\mu\text{g}/\text{m}^3$ at PY.
- During the 27-h rain event: 2.73 $\mu\text{g}/\text{m}^3$ at NC and 5.28 $\mu\text{g}/\text{m}^3$ at PY
- In the 28-h period after rain (7:00 on 7 June to 11:00 on 8 June): 6.42 $\mu\text{g}/\text{m}^3$ at NC and 9.40 $\mu\text{g}/\text{m}^3$ at PY

We have revised the relevant text.

Lines 331-336

“The BC concentration decreased during rain events (Figure 2a), indicating that wet deposition was one cause for lower BC in the rainy season sampling period. Take the rain event on 6-7 June 2008 (78 mm rainfall from 3:00 to 6:00 next day) as an example. The average BC concentration in the 24-h before rain was 5.87 $\mu\text{g}/\text{m}^3$ at NC and 9.47 $\mu\text{g}/\text{m}^3$ at PY. During the 27-h rain event, the BC concentration dropped to 2.73 $\mu\text{g}/\text{m}^3$ at NC and 5.28 $\mu\text{g}/\text{m}^3$ at PY.

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.

Response: We add mixing height from balloon measurements in the revised Figure 2.

New text added (lines 229-236)

“Variation in mixing height plays an important role in affecting BC concentrations for the urban sites. The most severe episodes at PY recorded during the two sampling periods were all associated with low mixing heights. During 30 May to 1 June, BC concentration was extremely high (peak concentration close to $40 \mu\text{g m}^{-3}$) and the mixing height was only 500 m (approximately half of those on regular days) (Figure 2a). Similar episodic events recorded during 26-29 December during the dry season sampling period were observed to coincide with low mixing height, which was only 200-300 m (Figure 2b).”

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 arteries 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.”

Response: The BC sampling site at YS was near a major road (~130m). Local transportation emissions are possible. We revise the text as below:

Lines 252-257

“The BC level in YS is unexpectedly high, considering it is mainly a tourist town. The sampling site at YS was near a major road (~130 m). Local transportation emissions could be a significant source of measured BC. Emissions from nearby mining and metallurgical industries may have an impact on BC level as well. It is not possible to further speculate the relative contributions of local and regional sources due to the short measurement period and lack of other measurements 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.”

Response: We take note of the concerns regarding back trajectories. However, the focus of this study is not modeling the transport of BC. The use of back trajectories information is to gain a general idea on the origins of the air masses during the two measurement periods.

We have now added the information on back trajectories by HYSPLIT at different heights (100 m, 300 m, 500 m, 1000 m, 1500 m) (Figure S12). The overall patterns are very similar at different altitudes within the mixing height. The ensemble of back trajectories is also consistent with the monthly average wind stream.

Lines 318-321

“Back trajectories at different heights (100 m, 300 m, 500 m, 1000 m, and 1500 m) were also calculated and shown in Figure S12. The overall patterns are very similar at different altitudes within the mixing height. The ensemble of back trajectories is also consistent with the monthly average wind stream.”

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 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.”

Response: We analyzed the monthly average wind stream pattern in each of the 12-month in 2008. As shown in Figure R2 (see below), April to May is the transition period of the Northeast monsoon to the South China Sea monsoon (Northeast wind shift to southwest over SCS). June to September is the South China Sea monsoon-dominated period (Southwest prevailing wind over SCS), and the wind pattern during this period is identical in these four months. October is the transition period of the South China Sea monsoon to the Northeast monsoon (Southwest shift to Northeast wind over SCS). November to March is the Northeast monsoon-dominated period (Northeast prevailing wind over SCS), the air flow pattern is also very similar in these five months. Thus, the two sampling periods in our study represent the typical wind pattern of corresponding season. The following text is added to the paper to address the representativeness of the air flow patterns shown in Figure 4.

Lines 292-300:

“Figure 4 shows the monthly average wind stream patterns for June and December 2008. The wind stream patterns for other months in 2008 are also examined. April to May is the transition period of the Northeast monsoon to the SCS monsoon, with wind shifting from northeast to southwest over SCS. Under the dominant influence of the SCS monsoon, June to September show highly similar wind patterns, with southwest prevailing wind over SCS. November to March, mainly under the influence of the Northeast Monsoon, share similar air flow patterns, with Northeast prevailing wind over SCS. Thus, the two sampling periods in our study represent the typical wind patterns of their corresponding seasons.”

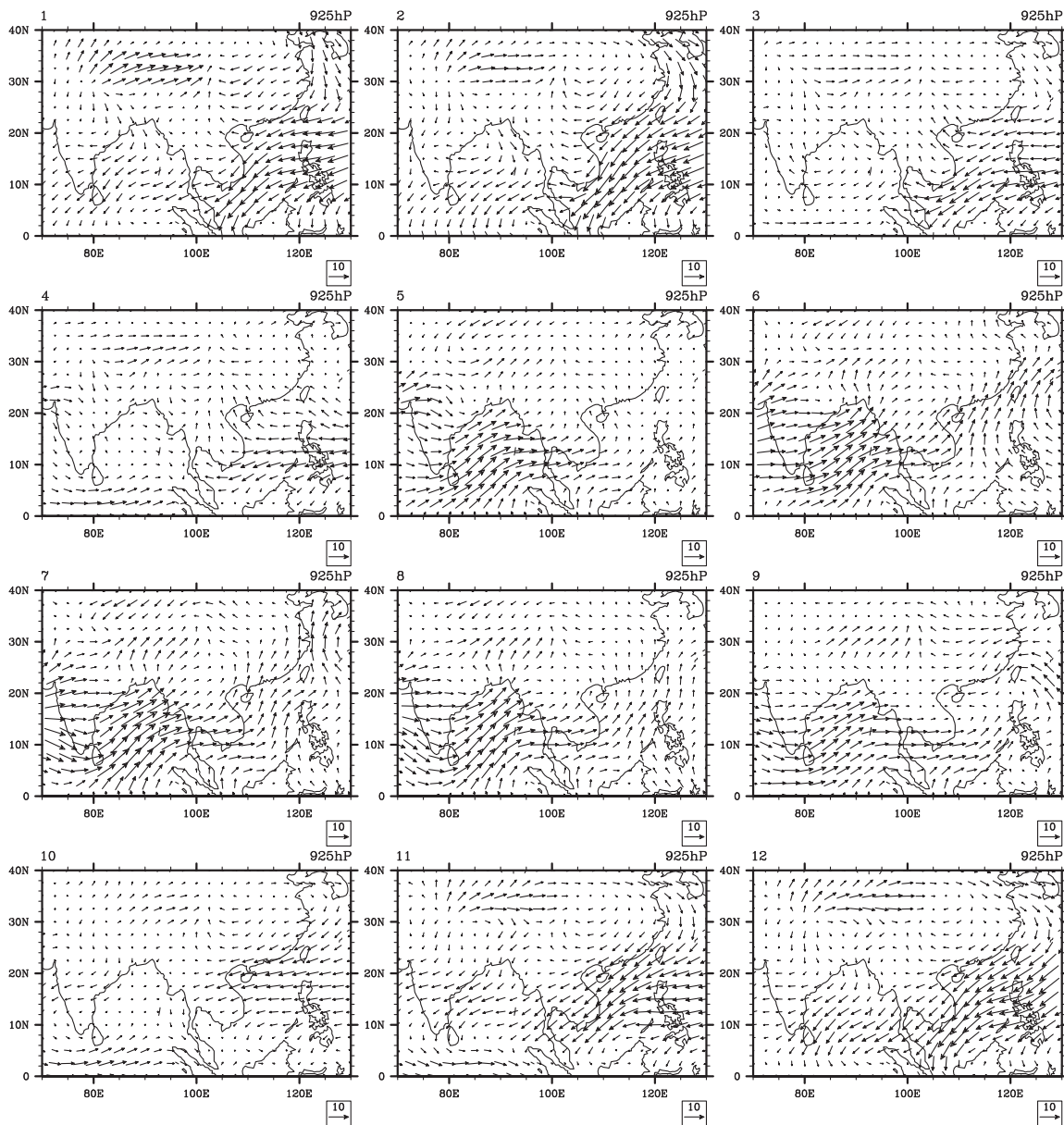


Figure R2. Monthly average wind stream patterns in 2008. The number at the upper-left corner in each plot denotes the month.

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.”

Response: Please refer to response to the previous comment.

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.

Response: We have now included mixing height data at YX and GZ from balloon measurements in Figure 2. The distributions of mixing height data in the two measurement periods are shown in

Figure S9 as supplementary material. Seen from Figure S9, the mean values of the mixing height at GZ were similar (~1070 m in the rainy season sampling period and ~1050 m in the dry season sampling period), while the most probable mixing heights in the two seasons differed by ~340 m (1420 vs. 1080 m, respectively). The mean values of the mixing height at YX were 650 m in the rainy season and ~800 m in the dry season sampling period. It appears that the seasonal variation in mixing height is not sufficient to overcome other factors to become the dominating factor for the observed seasonal difference. The following text is now added to the paper:

Lines 282-291

“For the understanding of seasonal variation of BC, variations in mixing height, monthly average wind flow patterns, and air mass origins are examined. Mixing height data by balloon sounding are available at YX and Guangzhou. The distributions of mixing height data in the two measurement periods are plotted in Figure S9. Seen from Figure S9, the mean values of the mixing height at GZ were similar (~1070 m in the rainy season sampling period and ~1050 m in the dry season sampling period), while the most probable mixing heights in the two seasons differed by ~340 m (1420 vs. 1080 m, respectively). The mean values of the mixing height at YX were 650 m in the rainy season and ~800 m in the dry season sampling period. It appears that the seasonal variation in mixing height was unlikely the driving factor for the observed BC difference in the two sampling periods (Figure 1).”

22. In much of the dry season the YX 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.

Response: We think that the reviewer meant “wet season”, instead of “dry season” in this comment, as during dry season, air mass reaching YX Island was mainly from the East China Sea (including coastal areas of China, Taiwan Strait and west Pacific area). Response below is made assuming “wet season” is the period of concern.

During the rainy season sampling period, ~25% of air masses reaching YX Island was from indo-china peninsula. As shown in the monthly average wind stream figure, part of air mass reaching SCS was affected by indo-china peninsula; this is a typical characteristic of South China Sea monsoon, not a unusual phenomenon.

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.

Response: Mixing height decreased during cold front period. As such, change in mixing height associated with the cold front was unlikely to explain the much reduced BC concentrations. Arrival of cold front is typically associated with strong wind, which effectively disperses pollutants. The long-range transported air masses encounter in our measurement periods apparently contained lower BC than the local air masses. In this work, we did not have other concurrent measurements of gas or aerosol-phase pollutants, making it impossible to further characterize the aging degree of the cold front air masses. We have re-phrased the relevant text to clarify what we mean.

Line 375:

“Passing cold fronts usually bring strong wind and ~~clean~~ air from high attitudes to the PRD region, resulting in significant reductions in BC concentrations,…”

Technical corrections:

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.”

Response: Suggestion taken.

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”

Response: This part has been rephrased as below:

Lines 66-69

“In this work, we report BC and aerosol light absorption measurements by Aethalometers in May-June 2008 in the rainy season and in December 2008-January 2009 in the dry season and their variations in these two sampling periods in a remote location over the South China Sea (SCS) and at 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.

Response: Correction made.

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.”

Response: The mainland China is conventionally divided into seven parts: Northeast China, North China, Northwest China, Southwest China, Central China, East China and South China. Chongqing belongs to Southwest China. Below are some comparisons made between Guangzhou and Chongqing.

City	Population in urban area	GDP 2012
Guangzhou	12 Million	1355 Billion (RMB)
Chongqing	8 Million	1145 Billion (RMB)

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

Response: Corrected.

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, superregional transport. Is it meso-scale, regional-scale, global-scale, etc.? Could a number be put on this?

Response: Super-regional transport refers to pollutants coming from places that are outside the PRD region. This is now clarified in the text (Line 340).

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.

Response: Thanks for altering us the modeling paper by Cohen and Prinn (2011). However, we feel this paper is not closely relevant to our work. Therefore, citation of this paper is not added.

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.

Response: We have proofread the manuscript a few times and corrected grammatical errors as much as we can.

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