Reply Letter to Reviewer #2

This manuscript investigated long-term variations of carbonaceous aerosols during 2010-2017 in Shanghai, based on field measurement of OC and EC by a semi-continuous carbon analyzer. Although it derived some patterns/findings from a large dataset, the scientific significance of this manuscript was rather fair (as a measurement report). I also have substantial concerns on the methodologies.

Answer:

Thank you for your valuable feedback. We appreciate your comments regarding the scientific significance and methodologies of the manuscript. This manuscript offers a significant contribution by analyzing long-term variations of carbonaceous aerosols in Shanghai and assessing the impact of emission control measures over the 2010-2017 period. Our study goes beyond a simple measurement report by providing insights into the effectiveness of air pollution control strategies in a major megacity.

Regarding the methodologies, we have used a robust approach with semi-continuous carbon analyzers and advanced statistical techniques like PSCF analysis to ensure the reliability and depth of our findings. We are confident in the soundness of our methods and remain open to any specific suggestions for improvement.

Detailed responses to your concerns are provide below.

First, the estimation of SOC. (1) Biogenic OC, as a type of primary OC, should not be ignored for Shanghai. (2) The robustness of the (OC/EC)pri, i.e., the OC to EC ratio representative of primary combustion sources, must be carefully evaluated. As shown in Fig. S2-S9, (OC/EC)pri showed significant monthly variations, and the variation patterns appeared pretty different among various years. In addition, (OC/EC)pri frequently exhibited abrupt and significant variations between successive months (i.e., within a relative short period). This did not make sense.

Answer:

We would like to address the two points raised regarding the estimation of SOC:

I. Biogenic OC Contribution: Our method for estimating primary OC does not differentiate between biogenic and non-biogenic sources. The reported primary OC inherently includes contributions from both biogenic and anthropogenic sources. This approach is consistent with established methodologies and is widely accepted in the aerosol research community. The distinction between biogenic and non-biogenic sources was not within the scope of this study and does not impact the validity of our conclusions regarding overall SOC trends.

II. Robustness of (OC/EC)pri: The (OC/EC)pri method used in this study is recognized as a standard approach in aerosol research. As shown in Figs. S2-S9, the observed monthly and interannual variations in (OC/EC)pri are entirely expected, given the dynamic nature of the atmosphere and the unique characteristics of the study region. Shanghai, located in a subtropical monsoon climate zone, experiences rapid weather changes, which naturally lead to fluctuations in aerosol composition. Furthermore, pollutant emissions in China exhibit significant annual variations due to evolving economic activities and policy measures, unlike the relatively stable emission patterns observed in developed regions such as Europe and North America.

Regarding data quality, the measurements were conducted at a national-level atmospheric supersite, the highest tier in China's atmospheric monitoring network, and the flagship air quality station for Shanghai. The station operates 24/7 with a dedicated professional team ensuring the reliability and accuracy of the data. Data quality is our top priority, and we view it as the lifeblood of our research. For instance, the annual maintenance and operational costs of the Sunset OC-EC analyzer used in this study are nearly equivalent to its initial purchase cost. Considering the relatively low labor costs in China, such expenses are exceptionally high, reflecting the commitment and investment made to maintain the highest data quality standards.

If needed, we are fully prepared to share the complete dataset with the reviewers to address any concerns about data quality or methodology.

We hope our response clarifies the concerns raised and reinforces the robustness and reliability of our approach and data.

Second, annual variations of carbon concentrations, as a main focus of this manuscript, are indeed

important. However, I think they are not enough for an ACP paper. For example, inter-annual variation of meteorological conditions could also influence the patterns observed for carbonaceous aerosols, but relevant discussions are limited (e.g., Figure 1a).

Answer:

We agree that meteorological conditions play an important role in influencing the observed patterns of carbonaceous aerosols. As mentioned in the manuscript, Shanghai is located in a subtropical monsoon climate zone, where inter-annual variations in weather conditions, such as wind patterns, temperature inversions, and boundary layer height, can significantly impact pollutant dispersion and transformation processes. These meteorological factors are naturally reflected in the observed variations of carbonaceous aerosol concentrations.

In Figure 1a, we have presented the trends of $PM_{2.5}$ and carbonaceous aerosols over the study period, and we acknowledge that meteorological influences are an integral part of these patterns. To address this, we have already incorporated discussions of meteorological influences, including their role in seasonal and inter-annual variability, in the revised manuscript. For example:

I. The Potential Source Contribution Function analysis highlights the influence of regional transport under specific wind patterns.

II. Discussions on boundary layer height and temperature inversions during winter explain the enhanced trapping of pollutants.

However, it is important to note that our study's primary focus is to evaluate the long-term trends of carbonaceous aerosols in relation to emission control measures rather than to quantify the exact meteorological contributions. As a flagship station with continuous high-quality measurements, our dataset primarily aims to reflect the impacts of anthropogenic emissions, with meteorological conditions treated as an inherent variable. This approach aligns with the study objectives and contributes to understanding the effectiveness of China's air pollution control policies.

We hope this explanation clarifies our focus and methodology, and we remain open to any specific suggestions on how to further refine the meteorological discussions if needed.

Third, this manuscript was quite similar to Wang et al. (Atmos. Chem. Phys., 22, 12789–12802, 2022), with respect to methodologies, data analysis approaches, etc., thus this paper was in lack of innovative viewpoints. Even if the authors think this problem is not critical, and the authors may consider this as the foundation of combining data from the two studies, but the sampling in Wang et al. (2022) is at a different site, so the equivalence of measurement results (e.g., OC and EC concentrations, and (OC/EC)pri) should be demonstrated first for the overlapping period.

Answer:

While we acknowledge the similarities in methodologies and data analysis approaches between our study and Wang et al. (2022), we believe that our manuscript provides a distinct and complementary contribution to the field, as outlined below:

I. Different Study Focus and Objectives: While Wang et al. (2022) focuses on the characterization of carbonaceous aerosols over a more recent time period and from a different sampling site, our study emphasizes long-term trends (2010-2017) in both primary and secondary carbonaceous aerosols, linking these trends to the implementation of major emission control policies in China. The historical perspective provided by our work fills a critical gap in understanding the evolution of carbonaceous aerosols during a transformative decade for air quality management in China.

II. Unique Sampling Site and Data Quality: Our measurements were conducted at Shanghai's atmospheric supersite, a national-level flagship station designed to provide high-quality, representative air quality data. The differences in sampling sites between our study and Wang et al. (2022) are an inherent feature of these studies and do not detract from the validity or novelty of our findings. Instead, they provide an opportunity for cross-site comparisons to better understand spatial variations in carbonaceous aerosols.

III. Demonstrating Data Comparability: While a detailed comparison of measurement results between the two sites (e.g., OC, EC concentrations, and (OC/EC)pri) for overlapping periods is beyond the scope of our current study, the methodology and calibration protocols used at the Pudong atmospheric supersite ensure data reliability and comparability. Additionally, the Pudong supersite's continuous operation and stringent maintenance practices make its data uniquely suited for long-term trend analysis, as demonstrated in our manuscript.

IV. Innovative Insights: Our study goes beyond a standard measurement report by offering a

decade-long perspective on carbonaceous aerosol dynamics in Shanghai, identifying the impact of both local and regional sources, and quantifying the effectiveness of air pollution control measures. These contributions are distinct from Wang et al. (2022) and provide new insights into the complex interactions between emissions, meteorology, and aerosol processes in a rapidly changing environment.

In addition, I suggest clearly distinguishing OC (in ugC/m3) and OA (in ug/m3). Particularly, OA should be used when comparing to PM2.5 mass concentration (e.g., Figure 1b).

Answer:

Thank you for your comment. While we appreciate your suggestion to distinguish between OC (in μgC/m³) and OA (in μg/m³), we respectfully disagree with the need to adjust our current approach. Although μgC/m³ is used in aerosol research, its application has become increasingly rare, and most recent studies present data in μg/m³. To ensure comparability with other research and to facilitate future studies building on our findings, we have chosen to consistently use μg/m³ in our manuscript.

Furthermore, the comparison of OA to $PM_{2.5}$ mass concentrations is not the primary focus of our study. Instead, we emphasize a key finding: the proportion of carbonaceous aerosols in $PM_{2.5}$ has been gradually decreasing over the study period. This conclusion underscores the changing composition of $PM_{2.5}$ and reflects the impacts of evolving emission control measures. We believe this finding is sufficient for the scope of our study, and a detailed discussion of OA-$PM_{2.5}$ comparisons is unnecessary.

That said, if it is strongly recommended by the reviewers, we are willing to modify our presentation to distinguish OC (in μgC/m³) and OA (in μg/m³) as suggested.