

Response to Review Comments by Anonymous Referee #2 on “Bayesian Inference-Based Estimation of Hourly Primary and Secondary Organic Carbon at Suburban Hong Kong: Multi-temporal Scale Variations and Evolution Characteristics during PM_{2.5} episodes” by S. Wang et al.

Review of “Bayesian Inference-Based Estimation of Hourly Primary and Secondary Organic Carbon at Suburban Hong Kong: Multi-temporal Scale Variations and Evolution Characteristics during PM_{2.5} episodes” by Wang et al.

General comments

The manuscript focuses on understanding primary and secondary organic carbon (POC and SOC) in PM_{2.5} and their driving factors. The study was conducted in suburban Hong Kong from July 2020 to December 2021. It employs a novel Bayesian inference approach to differentiate between POC and SOC, using sulfate as a tracer for SOC. The study explores the temporal characteristics of POC and SOC, including diurnal, weekly, and seasonal variations, and their evolution during PM_{2.5} episodes. The methodology developed offers practical guidance for similar studies elsewhere, providing valuable insights for atmospheric models and understanding PM pollution processes. The study’s results indicate distinct SOC formations under different seasonal and pollution conditions, influenced by factors like temperature, relative humidity, and atmospheric oxidants. This research contributes to refining atmospheric models and developing strategies for air quality improvement and climate change mitigation. The results are presented effectively, with appropriate statistical analysis and visual aids. This paper is within the scope of ACP and might be of great interest to the broad atmospheric science community.

We thank the anonymous reviewer for the detailed comments. Below is our point-by-point response to each comment, marked in blue. The related text in the manuscript is copied here for reference.

However, there are areas for improvement in terms of clarity and depth in the discussion of certain results, particularly the implications of the findings for broader atmospheric science and policy-making. Currently, it tends to read more as a data-centric measurement report. Further elaboration on the Bayesian inference method used is recommended for accessibility to readers less familiar with this approach. I agree with the concerns raised by Anonymous Referee #1, particularly regarding the use of sulfate as the best tracer for SOC for all seasons and pollution episodes.

Response:

Please see our response to comment 1 raised by reviewer #1.

Additionally, I have a few specific questions and comments that should also be addressed before the manuscript can be considered for publication.

Specific comments:

1. Figure 1b: Could you explain the noticeable decrease in K1 observed between 6 and 7 am during winter, spring, and fall?

Response: We thank the reviewer for pointing out this missing piece of information. In short, such a drop in K1 can be attributed to the increase of traffic emissions during rush hours. The definition of K1 is POC/EC, which, in other words, is the average of OC/EC ratios from all primary sources weighted by their contributions. It is foreseeable that for vehicular emissions, the OC/EC ratio should be much lower than those from non-vehicular emissions, considering the significantly elevated EC level from engines. According to our PMF results, the OC/EC from vehicular emission is about 0.5, whereas for other primary sources (e.g., industrial emission, cooking emission, residual oil combustion), the OC/EC ratio is 0.85-1.18. Therefore, as the contribution from vehicular emissions to the primary pollution increases, the POC/EC ratio, i.e., K1, is bound to decrease towards the OC/EC of vehicular emission. We add some explanations in this regard.

The following sentence will be added to describe the noticeable decrease in K1 observed between 6 and 7 am.

“The diurnal variations of K1 in Figure 1b align closely with the local rush hours, when vehicular emissions exert a dominant influence among all primary sources. In comparison to non-vehicular primary sources, the

EC amount from vehicular sources is much higher, resulting in a lower OC/EC. During periods of heavy traffic, the overall POC/EC ratio decreases, approaching the typical OC/EC ratio of vehicular emissions.”

2. Figure 2a: Please clarify the term “corrected PM2.5.” What does this correction entail?

Response: The reasons to correct SHARP PM_{2.5} mass concentrations are included in the updated manuscript and supplement. Please refer to our reply in response to comment 3 raised by reviewer #1.

3. Figure S3: The error bar (uncertainty) in Figure S3a should be defined for clarity. Additionally, in Figure S3b, the legend should be corrected to read “SOC_u/SOC_c”.

Response: Revised as suggested. Please find the revised Figure S3 (new Figure S5 in the revised manuscript).

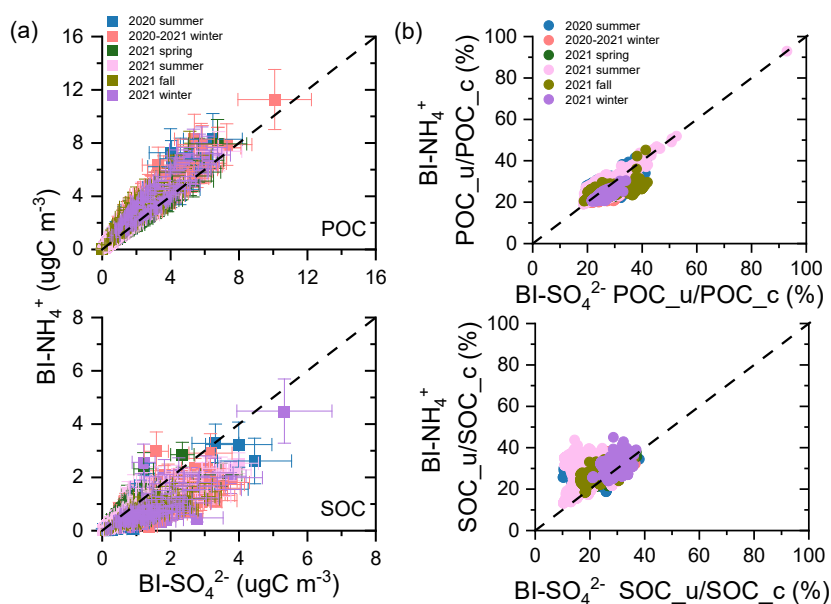


Figure R1 (New Figure S5). Comparison of (a) absolute concentration and (absolute uncertainties represented in error bar), and (b) relative uncertainty of POC and SOC between BI with sulfate and ammonium as tracers for SOC.

4. Line 290: Based on Figure 3, it appears that SOC values are slightly higher on weekends, particularly in winter and spring. Could you provide any insights into this observation?

Response: It is well documented that secondary organic aerosol formation is influenced by various factors, such as the organic precursors and atmospheric oxidants levels, as well as meteorological conditions. As shown in Figure 3, SOC values are slightly higher on weekends, which is similar to O₃ patterns. The slightly higher O₃ levels on weekends than weekdays could be due to the weak titration effects due to the reduced NO_x from vehicle emissions or other anthropogenic emissions during weekends. Thus, the slightly higher SOC levels on weekends would be attributed to the stronger atmospheric oxidation capacity.

The following sentence will be added to describe the difference of SOC variations on weekdays and weekend.

“The weekday-weekend patterns of POC and SOC displayed notable distinctions. Specifically, SOC was slightly higher on weekends, whereas enhancement of POC was found on weekdays throughout different seasons. Similar higher weekend levels were found for O₃, which could be due to the weak titration effects due

to the reduced NO_x from vehicle emission or other anthropogenic emissions during weekends. This observation suggests that anthropogenic emissions exerted a stronger influence on POC levels, while SOC levels appeared to be more influenced by the active photochemistry VOCs emissions from the nearby broadleaf woods rather than the anthropogenic sources.”

5. Line 298: The reference to daily ozone patterns appears to be incorrectly cited as Figure S4b; it should be Figure 3b.

Response: Revised as suggested.

6. Line 372: The term “ensuring analysis” seems unclear to me. Could you provide a more detailed explanation or rephrase it for clarity?

Response: To clarify, the sentence has been revised as follows and will be incorporated in the revised manuscript.

“In this work, PM_{2.5} episodes were identified as periods of hourly concentrations exceeding 25 µg m⁻³ and lasting 6 consecutive hours or longer at more than three monitoring stations.”

7. Figure 6: For the non-episode data, is the representation limited to the hours at the start of each season, or does it encompass all non-episode hours throughout the respective season?

Response: The non-episode data in Figure 6 is the combination of all non-episode hours in individual season. The caption of Figure 6 is revised to clarify the statement. The revised caption is copied here for easy reference.

“Figure 6. Comparison of select pollutant levels during episodes and non-episodes for individual episodes. The comparison parameters include concentrations of (a) O₃, (b) NO_x, (c) PM_{2.5}, (d) POC, and (e) SOC, (f) POC and SOC percentage contributions, and mass increment ratios of (g) O₃ and NO_x, (h) PM_{2.5}, and (i) POC and SOC. In panels (a)-(e), the filled squares represent during-episode concentrations while the empty circles represent the combination of all non-episode hours throughout the individual season. In panels (g)-(i), the light-yellow shaded zone marks the mass increment ratios (calculated as mass concentration during the episode divided by that during the non-episode hours in the same season) values of less than 1.