Response to reviewer 3

The authors thanks the reviewers for their thoughtful evaluation of our manuscript and for their constructive comments and suggestions. We appreciate the time and effort devoted to reviewing our work, which have helped us improve its quality and clarity. Below, we provide point-by-point response addressing the concerns raised.

Main comments

1.SOA can also be formed in the aqueous phase, please add this to sentence in introduction on page 2, lines 61-62.
We thank the reviewer for pointing out this mechanism. We have revised the sentence in the introduction (lines 64-65) to explicitly mention that SOA can also be formed through aqueous-phase processing.

2. Related to the above point, photochemical processing of organic aerosol can also induce browning through reactions in the condensed phase e.g. https://doi.org/10.1021/acsearthspacechem.7b00123, https://doi.org/10.1016/j.atmosenv.2021.118401, etc. The increased absorption in the study could be due to contributions from the formation of light absorbing SOA originating from reactions in both the gas and condensed phase. These reactions can also occur without light e.g. https://doi.org/10.1021/es102933x, https://doi.org/10.1021/es4038325, etc. Please address this in the paper (introduction, discussion and conclusion) and discuss how it could affect the interpretation of results including formation of nitrated aromatics.
We thank the reviewer for raising this aspect and sharing the references. We have addressed aqueous-phase reactions in the introduction (lines 82-84), in the discussion section 3.4 (lines 475-485) and conclusion (lines 541-548).

3. The final sentence in the third paragraph on page 15 (lines 392-393) supports the initial statements in the paragraph and therefore should be moved from the end of the paragraph to after the second sentence i.e. move the final sentence to follow "…with the photochemical formation of BrC from precursor gases (e.g., aromatics). However, also related to the above points, some discussion regarding contributions of SOA in the aqueous phase and how this could increase the AAE should be added here.
We thank the reviewer for this constructive suggestion. We have restructured the manuscript following the suggestions of reviewer #2. The sentence to follow the statement of the photo-chemical formation of BrC from aromatics precursors is now in section 4 (lines 540-541). Additionally, we discussed in the paragraph how aqueous-phase SOA processing could impact our results (lines 541-548).

4. Pages 9-10, lines 261-271: why does formation of SOA lead to increased SSA? What is the cause of this? How does increased scattering align with an increase in AAE?.
We thank the reviewer for raising these questions regarding the physical and chemical mechanisms linking SOA formation, $SSA_{aerosol}$, and $AAE_{aerosol}$. As remarked by reviewers #1 y #2 in their comments, increases in $SSA_{aerosol}$ and $AAE_{aerosol}$ are not necessarily solely attributable to SOA formation. Therefore, in the revised version of the manuscript, we have reworded the text (section 3.1, lines 335-337, section 3.2 lines 365-366, section 4 lines 560-561) acknowledging the various mechanisms that could explain the values observed in our study.

5. Aim 3) at the end of the introduction on page 4 states "…(3) provide recommendations for incorporating BrC dynamics into climate models." However, the authors do not fully address this aim in the conclusion. On page 16, the conclusion states the importance of including time-

dependent changes in BrC absorption as a function of photochemical processing and aerosol composition but does not make clear recommendations as to what should be included in climate models. Please add clear recommendations or edit aim 3), if this is not possible given that the timescale of the study i.e. no photobleaching observed.

In the revised version of the manuscript, we have removed objective 3. Additionally, because we could not be derive a linear fit between BrC production rate and OH-equivalent photochemical age (in absolute terms and corrected for dilution) as the Reviewer #2 requested to report a parameterization in the life cycle of BrC within the observed urban plume, we have reworded the objectives in the manuscript.

6. In Figure 3, what is the change in concentration in BC with photochemical age? It is diluted? Is the increase in the radio of Ox/BC because of an increase in Ox or because of a decrease in BC or a balance of the two? This data is not shown, please comment.

We thank the reviewer for this constructive question. To address whether the increase in the O_x/BC ratio is driven by O_x production, BC dilution or both, it is important to consider the role of BC as a conservative tracer of plume dilution. As the plume expands during transport, physical dispersion reduces species concentrations. Because BC is chemically inert (i.e. does not volatilize or react to form secondary species) on the timescales examined in this study, its absolute mass concentration decreases with plume age primarily due to physical dilution and atmospheric dispersion. If O_x were also a purely passive scalar undergoing only dilution at the same rate as BC, the ratio O_x/BC would remain constant over time. However, as shown in Figure 4, the O_x/BC ratio exhibits a statistically significant increase with OH-equivalent photochemical age (increasing from ~35 to 60 ppbv μg^{-1} m^3). This net increase suggests that active photochemical production of O_x occurs within the urban outflow. To make this explicit for the reader, we have included the time series of O_x in Figure 1b.

7. In Figure S2 a) there is one data point at a ratio of 6 ng m-3 ppbv-1 for BC/CO. If this point wasn't there, then the fit would be quite different. What is the uncertainty in the individual data point and is this representative for the fit?

We thank the reviewer for highlighting the potential influence of the individual data point in Figure S2a. As shown by the statistical tests requested by Reviewer #2 described in section 2.6 (lines 305-309), the correlations changed to not significant for either condition. Therefore, the sentence in lines 328-329 has been reworded.

8. It would be useful to see a plot of AAE vs OA/BC as discussed in the final paragraph on page 15 i.e. lines 395-398.

We appreciate the suggestion of the reviewer. However, we have excluded the analysis of linear changes between AAE and OA/BC (slope of d(AAE)/d(ΔOA/ΔBC) = 0.83) after incorporating the remarks from Reviewer #2. In the revised version of the manuscript, we report the linear relationship between BrC absorption and OA in absolute terms (section 3.2 lines 382-385).

Minor comments

9. The C8aromatics/toluene ratio decreases with increasing photochemical age. In the caption of Figure 4. b) on page 14 it states "Relationship between organic aerosol enhancement and the depletion of toluene". Should it be depletion of C8 aromatics/toluene ratio? The same phrase is also repeated in the text underneath the figure "…and the depletion of toluene with ageing…". If talking about decrease in ratio then this is not correct as for ratio to decrease, toluene should increase. Please check

We thank the reviewer for pointing out this inconsistency in our manuscript. The ratios and interpretations have been revised throughout the revised manuscript and the caption in figure 5 has been rewritten in accordance with the recommendations of Reviewer #1

10. There is a lot of scatter in Figure S6 (b). It's difficult to make any real conclusions from this figure in terms of the AAE, please comment.

We acknowledge the reviewer's concern regarding the high variability depicted in Figure S10b (before Figure 6b). In the revised manuscript (Section 3, lines 514-518) we have explicitly added a note of caution regarding the interpretation of these data, framing the observed scatter as a reflection of competing atmospheric processes rather than a growth trajectory.

References

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