**Summary**

Velazquez-Garcia et al. present new results from measurements taken in the 2022 ACROSS campaign to study the urban plume emitted from Paris. This paper focuses on understanding the change in the optical properties of the plume with atmospheric aging and adds to recent papers from the same group of authors focused on different campaign datasets. The paper compares various ratios of different chemical components and considers how chemical and optical properties evolve as a function of oxidation state. Finally, the authors standardize their measurements and perform PCA analysis and compare the components to the $AAE_{450\text{-}630\ nm}$ to validate their interpretation. The main conclusion is that photochemical processing causes an increase in OA/BC, an increase in Ox/BC, and a decrease in the C8 aromatics/toluene ratio with an increase in photochemical age and $AAE_{450\text{-}630\ nm}$. The authors attribute these findings to an increase photochemical oxidation and formation of light absorbing SOA generated from gas phase oxidation. The paper analyses a large amount of data and is well written, clearly presented, and addresses where more work is needed e.g. the authors did not observe photobleaching on the timescale of their measurements and therefore recommend future studies focus on longer data sets. Whilst the authors make a compelling argument for their interpretation, they should also consider how contributions from aqueous phase photochemistry can form SOA and contribute to increased absorption prior to publication.

**Major comments**

- SOA can also be formed in the aqueous phase, please add this to sentence in introduction on page 2, lines 61-62.
- 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.
- 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.

- 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?

- 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 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.

- 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?

- 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.

**Minor comments**

- 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.

- 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.