

We thank the reviewer for the questions and suggestions for improvement. We have revised the manuscript accordingly as shown in these point-by-point responses.

The original referee comments are given in *blue, italic font*. Our responses are given in black. The numbers in the responses refer to the revised text (without changes visible).

Manuscript quotations are given in the intended sections as in the revised text.

*Line 79: It may be more precise to acknowledge certain constraints, as the light-absorbing properties vary among different types of organic aerosols and are not universally significant.*

Both the light absorbing and light scattering properties of organic aerosols may indeed vary depending on the aerosol composition. This is now noted in the Introduction (l. 77-79):

“Optical properties can vary greatly among different types of organic aerosols, and ultimately the direct radiative forcing caused by aviation particles is impacted by absorption and scattering caused by both the primary BC and the potentially BrC-containing organic aerosol formed during the exhaust cooling or in the atmosphere.”

*Line 97: Is the combustion chamber used in the study representative enough to make the results broadly applicable to real-world aviation emissions?*

With our setup, we aimed to generate aircraft engine -like exhaust emissions using a laboratory scale combustion chamber in order to estimate the tendencies in photoaging. These results are not broadly applicable to real-world aviation emissions, where the emissions also differ due to the technical design and operating conditions between different engine technologies and generations (see, e.g., the ICAO database at <https://www.easa.europa.eu/en/domains/environment/icao-aircraft-engine-emissions-databank>).

We have improved the Conclusions of our work when highlighting the need for further studies with different engine models and operation conditions (l. 662-664):

“This work focused on mimicking the airport operation by replicating the average emissions of an LTO cycle using a laboratory model system, to simulate on- or near-ground exhaust emissions originating from an airport. The influence of engine model and operation conditions on the contents of aging aircraft exhaust emissions should be thoroughly studied in the future.”

*Line 176: “However, but the photolysis was slightly higher compared to tropospheric conditions, which may have influenced the formation of the oxidation products.” Given this, could the authors provide some indication of how this might have influenced their results? Are the findings still considered representative of actual atmospheric aging?*

Photolysis and OH-radical initiated oxidation reactions of organic compounds in the atmosphere are known to lead to different peroxy radical (RO<sub>2</sub>) reaction pathways and reaction products (see, e.g., Peng & Jimenez, 2020). In general, the fates of RO<sub>2</sub> radicals (which are very relevant in atmospheric oxidation processes) are influenced by the extent of OH radical initiated oxidation reactions compared to other reaction pathways. These reaction pathways can vary strongly between different compound groups. The kerosene combustion exhaust used in this study is a complex and partly unresolved mixture of organic compounds, which makes it impossible to evaluate the role of potential

variance in the radical chemistry (such as the over-emphasized photolysis) in the oxidation flow reactor.

Conditioning of a comprehensive flow reactor study requires compromises when adjusting photochemical aging conditions and suitable concentrations for all instruments and concurrent cell exposures. We utilized the framework described by Peng & Jimenez (2017) to guarantee sufficiently representative flow reactor conditions. This is mentioned on l. 166-167:

“The ratio of photolysis to OH exposure remained between 0.6 and  $2 \times 10^6 \text{ cm s}^{-1}$  (Table S2), indicating that in total, the flow reactor conditions were in the ‘risky’ regime as defined by Peng and Jimenez (2017).”

*Line 225: What collection efficiency was adopted for AMS measurements? Also, what is the degree of agreement (or correlation) between the AMS + aethalometer and SMPS mass measurements?*

AMS CE of 1 was used. This was validated by measuring standard ammonium nitrate and ammonium sulfate particles. This is now mentioned at l. 223:

“Ionization efficiencies of the HR-ToF-AMS were obtained by measuring 350 nm ammonium nitrate and ammonium sulfate particles. Collection efficiency of 1 was applied.”

Mass emission indices derived from the AMS and SMPS measurements are shown side by side in Figure 6. The mass-EIs agreed well for the DR50 experiments, where the particles were in a size range measurable by the AMS. For the DR200 experiments, there was a slight underestimation by the AMS, caused largely by the particle size being below the instrument detection range.

Aethalometer doesn’t measure mass but light attenuation. The ‘equivalent’ masses are often calculated using a known/set mass absorption efficiency (MAE). In this work, we use the aethalometer measurements of attenuation together with the SMPS-based mass estimation to derive the MAEs of the particles deposited on the filter (l. 262-266). The light absorption at 880 nm was minor, and the ‘eBC’ concentrations likely biased by the high organic loading. All in all, we expect that the mass of refractory particles was negligible in both the fresh and aged exhaust, compared to the organic aerosol.

*Line 269: “Where PMSMPS is the SMPS-based mass estimate with the assumption that all the mass measured by SMPS is organic, which was a sound assumption considering the AMS results on particle composition.” This sentence is unclear. Why not use the AMS-measured organic mass directly, if the assumption is that all mass is organic? Also, how is the assumption justified based on AMS results on particle composition, given that AMS only detects non-refractory species?*

We used the SMPS-derived mass due to the small size of the particles, as the AMS doesn’t efficiently capture the sub-50nm particles. For example, in the fresh exhaust the AMS failed to measure the organics altogether due to the small particle size (see Fig. 5 for particle size distributions).

We have edited the sentence on l. 265-266 to better indicate the dominant role of organics to the total particulate emission:

“This was considered a sound assumption based on the low eBC concentrations and AMS results on non-refractory particle composition.”

*Line 450: For the aged emissions, the authors reported EIs of particle mass for the DR 50 experiments.*

*What are the corresponding values for the DR 200 conditions? Would the higher dilution ratio be more atmospherically relevant?*

Particle mass EIs for the DR200 experiments are available in Figure 6. The EIs were similar to the DR50 experiments around 2 equivalent days, and did not increase by elongated photochemical processing. This is now mentioned in text on lines 456-457:

“The formed particle mass was, however, only slightly higher for the DR50 experiments (Fig. 6a) at similar degree of exposure. Particle mass EI were not found to increase by enhanced photochemical aging after 2 eqv.d.”

The choice of dilution ratio is a crucial parameter, but often a compromise especially when extended aging times are considered. In reality, the exhaust plume would be gradually diluted within its atmospheric residence, but this cannot be considered in our experiments due to obvious technical restrictions. The results from two dilution ratios show that dilution does matter when particle composition and amounts are considered, but that the absolute mass-EIs and optical properties of the forming particles might still be close to each other.

*Line 568: Could the authors elaborate on the large difference in BC EIs between fresh and aged exhaust emissions? Can the later-mentioned loading effects fully account for this discrepancy?*

Elemental carbon structures will not form via photochemical processing of organic vapors. Organic coatings can enhance light absorption at 880 nm (Lack and Cappa 2010), which is interpreted as increased ‘eBC’ when a constant mass-absorption coefficient is applied. Also purely scattering particles can enhance the attenuation when the loading is excessive (Collaud Coen et al. 2010, Virkkula et al. 2015). This is now explained on l. 570:

“For the aged exhaust measurements, the increase of aerosol scattering due to enhanced OA concentrations likely resulted in positive bias in assessing light absorption from attenuation at 880 nm.”

We cannot discern whether the discrepancy between fresh and aged BCs arises purely from the Aethalometer filter loading effects, or is some non-soluble but absorbing particulate matter also formed.

## References

Collaud Coen, M., Weingartner, E., Apituley, A., Ceburnis, D., Fierz-Schmidhauser, R., Flentje, H., Henzing, J. S., Jennings, S. G., Moerman, M., Petzold, A., Schmid, O., and Baltensperger, U.: Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms, *Atmos. Meas. Tech.*, 3, 457–474, <https://doi.org/10.5194/amt-3-457-2010>, 2010.

Lack, D. A. and Cappa, C. D.: Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon, *Atmos. Chem. Phys.*, 10, 4207–4220, <https://doi.org/10.5194/acp-10-4207-2010>, 2010.

Peng, Z., and Jimenez, J. L.: Modeling of the chemistry in oxidation flow reactors with high initial NO. *Atmos. Chem. Phys.*, 17(19), 11991–12010. <https://doi.org/10.5194/acp-17-11991-2017>, 2017.

Peng, Z., and Jimenez, J. L.: Radical chemistry in oxidation flow reactors for atmospheric chemistry research. In *Chemical Society Reviews* (Vol. 49, Issue 9, pp. 2570–2616). Royal Society of Chemistry. <https://doi.org/10.1039/c9cs00766k>, 2020.

Virkkula, A., Chi, X., Ding, A., Shen, Y., Nie, W., Qi, X., Zheng, L., Huang, X., Xie, Y., Wang, J., Petäjä, T., and Kulmala, M.: On the interpretation of the loading correction of the aethalometer, *Atmos. Meas. Tech.*, 8, 4415–4427, <https://doi.org/10.5194/amt-8-4415-2015>, 2015.