Response to egusphere-2023-2441 reviews for RC2

Referee comments are marked in black bold and are numbered as R2Cx with x the comment number. Author (AC) responses are marked in black directly below the comments. The original text from the manuscript is repeated in blue and corrected text in revised manuscript is typed in red. Previous line numbers given in "()" following the updated line numbers.

Testa et al. comprehensively analyze the aviation engine-emitted soot aerosol particles to understand their ice nucleation ability at cirrus cloud temperatures. Overall, the experiments were well-designed, and the results support their conclusions. Moreover, this study can fill the knowledge gap in the indirect climate effects of soot emitted from aviation engines, which might be an unignore source of soot at high altitudes. I only have some minor comments and questions that can help improve the manuscript. Thus, I recommend publishing it with minor revisions. Please see my comments below:

We thank Reviewer 2 for their comments and respond to questions and concerns individually below.

Minor comments:

R2C1: Could the authors comment on how high-altitude ambient conditions might affect the results since the experiments were conducted at the ground level?

The ice nucleation experiments were designed to mimic the atmospheric conditions, i.e., T and RH_w relevant to ice nucleation, and represent by definition high-altitude conditions. The presence of highaltitude background aerosols (dust, haze droplets) would likely reduce the aviation soot AF due to more water vapor competition; this has been explored with air parcel model in Kärcher et al. (2021) and Kärcher et al. (2023) and the quantification of this effect at global scale is planned for future modeling paper using the results of this work.

Then, as mentioned in R1C3 and R1C4, the morphology of the soot primary particles (their size and overlap) is fixed in the combustor. Temperature and pressure in the combustor are high (~2000 K and tens of bar, respectively, Dakhel et al., 2007; Starik, 2007) and largely driven by the engine design and thrust, hence uncorrelated to the ambient conditions. Yet, due to coagulation in the aerosol reservoir, aggregates sampled in our study are larger and lacier (open-branched structure) than uncoagulated particles, such as for high-altitude aviation soot (as discussed in the atmospheric implication section and above). Besides, we acknowledge that the conditions experienced by the soot aggregates before entering the cloud chamber (HINC) differ from high-altitude conditions. In our aerosol reservoir, the temperature and pressure are higher, no nucleation mode particle (H₂SO₄ and organic droplets [e.g., oil droplets]) is allowed to form and no dilution with ambient air takes place. As discussed in the paper's Atmospheric implication section, these factors/processes would likely affect the soot mixing state, presumably decreasing the coating over the soot particles (Kärcher et al., 2007; Onasch et al., 2009; Peck et al., 2014; Timko et al., 2013; Wong et al., 2014; Wong et al., 2008) for soot sampled in our study.

R2C2: I am curious to see the morphology change with and without CS based on the SMPS and Tandem DMA-CPMA measurements.

Measurements of size change with and without CS with SMPS have been conducted in a companion study (Testa et al., 2024; their Figure 4; simplified in Figure 1 at the end of this document). Those measurements

were conducted at the same maintenance and testing aircraft engine facility and with a similar set up (but focusing on another research question and for different engine types). SMPS measurements from that study (Figure 1 at the end of this document) show that CS-soot aggregate sizes are comparable (within measurement uncertainty) to unCS-soot sizes and hence corroborate the TEM analysis conducted in the present study (Fig. F1).

The analysis of the TEM images of unCS- and CS-soot (Fig. F1) show no change in morphology, and hence we expect fractal dimension of CS-soot to be similar to unCS-soot. Yet, due to the limited time of aerosol sampling from our aerosol reservoir, measurements of CS aggregate mass were conducted only at 200 and 400 nm (as opposed to unCS-soot for which measurements have been conducted at several size points), hence no morphology information (e.g., fractal dimension) can be extracted from the mass measurements due to having only two data points.

R2C3: Do you expect any physical (e.g., partition on the soot and cause compression) or chemical reaction (oxidation) to happen inside the tank?

We expect oxidation to be considerably inhibited once exiting the engine combustor (Dakhel et al., 2007) due to the low temperature in the lines and in the aerosol reservoir. However, condensation and evaporation of exhaust gas onto/from the soot particles can occur in the tank due to the drop in temperature from the line to the tank (433 K to 298 K) and the dilution of tank gas with air during the ice nucleation experiment. These changes in particle mixing state were however not characterized, however we acknowledged in the manuscript (see R2C1) that high-altitude soot mixing state likely differs from the soot sampled in our study.

R2C4: Please note that C, N, and O are semiquantitative in EDX. Moreover, some C and O signal might come from substrates.

We agree with the reviewer that EDX is only semiquantitative. EDX is nonetheless useful for comparing element concentration of different samples collected on similar grids with the same microscope, as done in this study.

In lines 175-177 (167) we have added: "We point out that although EDX is semiquantitative, it is nonetheless useful for comparing elemental concentrations of different samples collected on similar grids with the same microscope, as done in this study. We further note that sulfur might get vaporized [...]"



Figure 1: Mode diameter change ΔD_m upon processing of the particles with the catalytic stripper measured by SMPS for the given engines. This figure is a modified version of Fig. 4 from Testa et al. (2024)

- Dakhel, P. M., Lukachko, S. P., Waitz, I. A., Miake-Lye, R. C., & Brown, R. C. (2007). Postcombustion Evolution of Soot Properties in an Aircraft Engine. *Journal of Propulsion and Power*, 23(5), 942-948. <u>https://doi.org/10.2514/1.26738</u>
- Kärcher, B., Mahrt, F., & Marcolli, C. (2021). Process-oriented analysis of aircraft soot-cirrus interactions constrains the climate impact of aviation. *Communications Earth & Environment*, 2(1), 113. <u>https://doi.org/10.1038/s43247-021-00175-x</u>
- Kärcher, B., Marcolli, C., & Mahrt, F. (2023). The role of mineral dust aerosol particles in aviation soot-cirrus interactions. *Journal of Geophysical Research: Atmospheres, n/a*(n/a), e2022JD037881. <u>https://doi.org/https://doi.org/10.1029/2022JD037881</u>
- Kärcher, B., Möhler, O., DeMott, P. J., Pechtl, S., & Yu, F. (2007). Insights into the role of soot aerosols in cirrus cloud formation. *Atmospheric Chemistry and Physics*, 7(16), 4203-4227. <u>https://doi.org/10.5194/acp-7-4203-2007</u>
- Onasch, T. B., Jayne, J. T., Herndon, S., Worsnop, D. R., Miake-Lye, R. C., Mortimer, I. P., & Anderson, B. E. (2009). Chemical Properties of Aircraft Engine Particulate Exhaust Emissions. *Journal of Propulsion* and Power, 25(5), 1121-1137. <u>https://doi.org/10.2514/1.36371</u>
- Peck, J., Yu, Z., Wong, H.-W., Miake-Lye, R., Liscinsky, D., Jennings, A., & True, B. (2014). Experimental and Numerical Studies of Sulfate and Organic Condensation on Aircraft Engine Soot. ASME Turbo Expo 2014: Turbine Technical Conference and Exposition,
- Starik, A. M. (2007). Gaseous and Particulate Emissions with Jet Engine Exhaust and Atmospheric Pollution. Advances on Propulsion Technology for High-Speed Aircraft, Paper 15(Educational Notes RTO-EN-AVT-150), pp. 15-11 – 15-22.

- Testa, B., Durdina, L., Edebeli, J., Spirig, C., & Kanji, Z. A. (2024). Contrail processed aviation soot aerosol are poor ice nucleating particles at cirrus temperatures. *EGUsphere*, 2024, 1-22. <u>https://doi.org/10.5194/egusphere-2024-151</u>
- Timko, M. T., Fortner, E., Franklin, J., Yu, Z., Wong, H. W., Onasch, T. B., Miake-Lye, R. C., & Herndon, S. C. (2013). Atmospheric Measurements of the Physical Evolution of Aircraft Exhaust Plumes. *Environmental Science & Technology*, 47(7), 3513-3520. <u>https://doi.org/10.1021/es304349c</u>
- Wong, H.-W., Jun, M., Peck, J., Waitz, I. A., & Miake-Lye, R. C. (2014). Detailed Microphysical Modeling of the Formation of Organic and Sulfuric Acid Coatings on Aircraft Emitted Soot Particles in the Near Field. Aerosol Science and Technology, 48(9), 981-995. https://doi.org/10.1080/02786826.2014.953243
- Wong, H.-W., Yelvington, P. E., Timko, M. T., Onasch, T. B., Miake-Lye, R. C., Zhang, J., & Waitz, I. A. (2008).
 Microphysical Modeling of Ground-Level Aircraft-Emitted Aerosol Formation: Roles of Sulfur-Containing Species. *Journal of Propulsion and Power*, 24(3), 590-602.
 https://doi.org/10.2514/1.32293