We would like to thank Dr. Stettler for taking the time to thoroughly read our submission and offer his thoughtful *comments* that help us to bring forward our opinion even more clearly. We hope our itemized replies below and proposed text changes address his concerns but if anything remains, we would welcome any input.

This opinion paper aggregates literature in a useful way, however there are several gaps that need addressing, detailed below. Furthermore, it is not entirely clear what the opinion is - is this paper supposed to put forward a view that soot emissions should be minimised? It currently appears that this is more of a review article.

Indeed, after first reviewing in some detail the state of the art in the field, we expressed our opinion that realistic and science-based descriptions of aircraft soot emissions are needed highlighting thus opportunities for contributions by aerosol scientists. For example, even in our abstract we stressed the issues stemming from oversimplification of jet fuel soot characteristics (lines 11-13). At its end, we stress that existing technologies for reducing jet fuel soot emissions through combustor and fuel design are reviewed to identify strategies that eliminate aircraft soot emissions (lines 19-20). Isn't this a loud and clear opinion? Nevertheless, in the revised version of our manuscript we will stress further that realistic descriptions of jet fuel soot structure and composition are needed rather than the simplistic ones in today's otherwise very sophisticated models of fluid and energy dynamics in the operation of jet turbines for elimination of their emissions. Such realistic descriptions have, for example, been used quite effectively to describe black carbon (BC) formation and growth from a variety of combustion sources and even facilitate monitoring of BC emissions by aerosol (e.g., particle mobility and mass analyzers), laser (e.g., light extinction) diagnostics and fire detectors by accounting for BC morphology and limiting the current uncertainty regarding BC mass and particle size. In addition, by capitalizing on the accurate description of the high temperature residence time during enclosed combustion synthesis of nanomaterials and the latest advances in soot structure and composition, more than 99% of the emitted soot mass and concentration from enclosed jet fuel combustion was removed. Even though something like this had been stated explicitly in our p. 7, lines 327 - 329, this will be expanded as above. Similarly, a realistic description of BC allowed for the first time to determine conditions for synthesis of carbon black (CB) with closely controlled structure and size that is crucial for its diverse applications where for tire reinforcement hard agglomerates consisting of large primary particles (PP) are needed as fillers while for battery electrodes such agglomerates should consist of much finer PP and for inks or paints the CB agglomerates should be soft ones. Clearly such an understanding should be incorporated into the design of aircraft engines burning fossil and/or sustainable aviation fuels as it greatly facilitates engine design and operation for complete oxidation of any soot formed before its emission. This point will be emphasized in our abstract, text and conclusions in the revised paper version.

The comments below represent significant omissions and I do not recommend publication of this article in it's current form.

Major comments:

la. The authors have used the ICAO emissions databank to show data on nvPM emissions indices for different combustor types. It would have been useful to show this as a trend in time in addition to with respect to engine rated thrust.

We agree that a trend with time would be useful, however, the only 'time' that is given in the ICAO database are the initial and final test dates. We have plotted this in Fig. R1 below showing the nvPM number (#/kg) emissions at a) idle and b) take-off. There appears to be no trend with the initial test

date. This is likely because it does not account for the production or design date of the engines so older engines could have been tested at a later date.



Figure R1: The nvPM number as a function of the initial test date at (a) idle/taxi (7% thrust), (b) take-off (100%). Combustor types represented in the database include SAC (squares), DAC (diamonds), RQL (circles), LDI (inverted triangles), LPP (triangles). The total nvPM number is normalized by the fuel flow (kg).

1b. Furthermore, a discussion of the nvPM mass could also be shown.

The nvPM mass is also important and it is shown in Fig. R2 below with the nvPM mass (mg/kg) at a) idle and b) take-off. As we had stated in the paper (p. 9 lines 409 - 410), the nvPM mass shows a similar trend to the nvPM number. The main differences between nvPM mass and nvPM number are the LPP values falling closer to the other combustors with mass-based emissions compared to number based emissions. This suggests that the LPP produces fewer but larger particles than other combustors on average. This will be stated in the revised paper on the current page 9 around line 410.



Figure R2: The nvPM number as a function of the rated thrust at (a) idle/taxi (7% thrust), (b) take-off (100%). Combustor types represented in the database include SAC (squares), DAC (diamonds), RQL (circles), LDI (inverted triangles), LPP (triangles). The total nvPM mass (mg) is normalized by the fuel flow (kg).

1c. There is no mention on whether this data shown has been corrected for line-losses. Discussion on suggestions on improving or adding to the regulatory measurement procedure would be a welcome addition.

Data submitted to the ICAO database should be collected following the procedure outlined in the ICAO Annex 16, Vol. II (ICAO, 2017). Briefly, particles are sampled at the engine exhaust with a no more than 35 m long (from probe tip to instrument inlet) heated sampling line to the measurement devices. This relatively long line, paired with the small size of aircraft soot may result in significant diffusional and thermophoretic losses due to temperature gradients as the sample cools from the exhaust temperature to sample line temperature. Since 2017, the nvPM mass and number diffusion and thermophoretic losses must be accounted for with the methods outlined in the ICAO Annex 16, Vol. II (ICAO, 2017). However, it is important to note that these losses are size-dependent, but the regulations do not require particle size measurements. Therefore, the estimate of the line loss correction may not be accurate for all engines. This will be stated in the revised version of our paper in current pg. 9, around line 407.

1d. Discussion on how ground-level measurements scale to cruise conditions would also be welcome, e.g. <u>https://egusphere.copernicus.org/preprints/2023/egusphere-2023-724/</u>

We had stated (pg. 2, line 50): As these emissions are measured only at ground level for the LTO cycle, the emissions most relevant for climate considerations are only indirectly estimated (Stettler et al., 2013). Subsequent to this sentence in the revised version of our paper, we will add: Estimates of emissions inventories must convert values measured at the ground to account for the drastically different atmospheric conditions at cruise (Teoh et al., 2023 Preprint).

2. There is extremely limited discussion on the role of other aerosol particles in contrail formation. This is literature going back more than a couple of decades looking at the effect of sulphur, and there is emerging evidence that lubrication oil particles might play a role in the case of low soot conditions (https://egusphere.copernicus.org/preprints/2023/egusphere-2023-1264/). Consideration should be given to the potential contrail impacts under low soot conditions (https://www.nature.com/articles/s41467-018-04068-0).

Of course, other aerosols may affect contrail formation as discussed in the cited pertinent literature (i.e. Kärcher, 2018). This is an active field of research especially in low soot conditions. As our paper is on eliminating aircraft soot emissions rather than on contrail formation, we only report the current understanding on the role of soot in contrail formation. Nonetheless, in the revised paper we will state around current line 60 (p. 2) that the role of soot in contrail formation is still unclear.

3. There is no mention of aerosol cloud interactions. These is the most uncertain contribution of aviation to climate change and might be the largest contribution to RF, however both the sign and magnitude the RF is extremely uncertain

(https://www.sciencedirect.com/science/article/pii/S1352231020305689). Emerging evidence suggests that the role of soot particles might be less important than ambient particles

(https://egusphere.copernicus.org/preprints/2023/egusphere-2023-2441/;

https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2022JD037881). It is critical that this is covered in the article.

We had already mentioned aerosol cloud interactions on pg. 1, line 37 "Soot emissions can impact the climate by warming the atmosphere through direct Radiative Forcing (RF) and indirectly by altering cloud processes and decreasing snow albedo (Bond et al., 2013)." Further detail was not given as this is an active area of research with large uncertainties that deviates from the topic of eliminating aircraft soot emissions. To highlight this, we will state in the revised version of our paper on pg. 2, around line

60 that there is high uncertainty in the RF of aviation aerosol-cloud interactions (i.e. indirect RF) and therefore no best estimate is given by Lee et al. (2021).

As with contrail formation, aerosol-cloud interactions is an active field of research deserving an opinion paper. Perhaps Prof. Stettler might be interested in contributing one highlighting opportunities for aerosol scientists as we tried to do here for eliminating soot emissions in which we had some first-hand experience in soot formation, growth, oxidation and interaction with light over the last 40 years, in contrast to aerosol-cloud interaction or contrail formation where we are eagerly looking forward to their experts' opinions.

References:

Teoh, R., Engberg, Z., Shapiro, M., Dray, L., and Stettler, M. E. J.: A high-resolution Global Aviation emissions Inventory based on ADS-B (GAIA) for 2019-2021, 2018, 1–27, 2023. Prepint.