Measurements of particle emissions of an A350-941 burning 100% sustainable aviation fuels in cruise

Author's Response to Referees

RC1: 'Comment on egusphere-2024-1224', Anonymous Referee #1, 08 May 2024

We would like to thank the Referee for reviewing our manuscript and helping to improve the presentation of our results. We enlarged the symbols of some plots, have modified the coloring and used a colorblind safe palette to improve readability. Answers to specific comments are discussed in the following.

> **Comment 1):** Line 40: What is meant by a "positive effect" on contrail properties? Is it "positive" in the climate sense that it reduces contrail occurrence and radiative effect, or "positive" in the sense of a positive feedback in abundance?

Answer comment 1): HEFA-SPK is free of aromatics and sulfuric compounds, which can have a positive effect on contrail properties leading to a reduced radiative forcing. We clarified this in the manuscript.

> **Comment 2):** Line 42: A "seed droplet" is not the right description for a solid soot particle serving as an ice-forming nucleus.

Answer comment 2): We clarified this sentence: Soot particles are emitted in large numbers and are sufficiently large to serve as condensation nuclei that can be readily activated as seed droplets for ice formation.

> **Comment 3):** Line 102: How well could the fuel tanks and lines be emptied? It would be worthwhile to state what the level of contamination from residual fuel might be (<1%?).

Answer comment 3): It is difficult to make a statement as contamination can occur throughout the entire supply chain. Assuming that the underlying HEFA contained 0% sulfur, the world average Jet A-1 sulfur content is 0.046% and the two HEFA fuels contained 0.0003-0.0007% (see Table 2), the contamination can be roughly estimated to be around 1%.

> **Comment 4):** Line 122: What is meant by "low" sulfur levels--do you mean "measurable"? How much is present in the HEFA-SPK fuel after the aircraft has been fueled?

Answer comment 4): Although the HEFA should not contain any sulfur, the fuel samples show a sulfur content of 0.0003-0.0007% sulfur by mass. Results of the analysis of fuel properties of the fuels from the aircraft tank is shown in Table 2. Fuel samples were taken from the tank before the measurement flights. Samples for GCxGC analysis of ECLIF3-2 HEFA-SPK fuel were collected during loading of the ISO container. We have specified this in the table caption.

> **Comment 5):** Line 181: What is a "simplified mission" in the context of the MEEM methodology?

Answer comment 5): The wording was misleading and we removed it.

Comment 6): Line 189: Define "T30" and "P30" here.

Answer comment 6): The combustor inlet temperature T30 is already defined in line 85 and the corresponding pressure at the combustor inlet P30 is defined in line 87.

Comment 7 and 8): Line 197: Define "AFR". Line 204: Define ICAO and provide a reference for the Aircraft Engine Emissions Databank.

Answer comment 7 and 8): AFR is now defined in this section. ICAO is now defined in the preceding section 2.3.1, where also the reference for the EEDB is stated.

Comment 9): Fig. 7: I also suggest flipping the axes so that flight level is the left axis, which makes intuitive sense. I also don't think you have defined flight level anywhere; this term might be unfamiliar to modelers who might make use of these data.

Answer comment 9): Since both figures share the same y-axis, but have a different x-axis, we would like to keep this presentation of the results. We specified the flight levels in Section 3.1..

Comment 10): Line 273: I don't understand the sentence beginning "Assuming the size distribution of nvPM measured on the ground. . . ." What ground-based measurements are you referring to?

Answer comment 10): The particle size distribution has been measured during accompanying ground tests using the same source aircraft and fuels. As of now, those results have not been published but preliminary data for consistency checks with flight tests were communicated by T. Schripp and his team.

Comment 11): Figure 9. There need to be units on the axes. What is the dashed line?

Answer comment 11): The dashed line represents the 1:1 relationship between measurement and observation. This has been added to the figure description.

Comment 12): Lines 332-333: I'm quite confused by the reference to "filled symbols" in Fig. 9. Only the left panel has any symbols that are not "filled". Are you referring to the "x" symbols in this panel? These aren't "filled". If you label the panels "a" and "b" and refer to these in the text the meaning would be clearer. It's also not directly evident that "the model prediction skill is clearly strongest for average cruise flight conditions." There are a lot of scattered data on these plots.

Answer comment 12): By filled symbols we actually mean the filled symbols in the left plot in Fig. 9. We have labeled the panels according to the recommendation to make it easier to follow.

Exact simulation of engine emissions requires precise knowledge of a large number of fuel components, complex combustion mechanisms and the heterogeneous interactions during particle formation. The use of such a detailed model, if one would be available, also requires a large amount of computing power, which makes the use of such a model to study a large fleet of aircraft and their climate impact challenging. The MEEM method and the T4/T2 method offer a publicly available way of modeling soot emissions for further use with reasonable resources. It is therefore to be expected that the comparison of measured and modeled emission indices shows some scatter. Nevertheless, the orders of magnitude and the reductions due to the different fuels are in good agreement, so that both models can be used for further application, for example in climate models or contrail predictions. For the T4/T2 model, although there are also points showing a good agreement, the modeled emission indices deviate the most from the measured EI for low and high T30. However, both models are currently the best-established public models for modeling aircraft emissions based on accessible engine and fuel data.

> **Comment 13):** Lines 345-348: The Pearson correlation coefficient is not the only relevant metric; the slope is also important to assess how well the model reproduces the measurements. Could you please provide slopes as well as r^2 values?

Answer comment 13): The quadratic Pearson correlation coefficients are already given in this paragraph. We agree that showing the slope of the linear regression will be useful and added an evaluation of the linear regression to the section.

> **Comment 14):** Line 363: Were samples "captured" or measured continuously?

Answer comment 14): Measurements were performed continuously, we changed the wording.

> **Comment 15):** Data availability: Is the model and analysis software code available for download? Where can the (proprietary) MEEM model be found?

Answer comment 15): The MEEM method is described in Ahrens et al., 2022 as cited in the manuscript. Software code is not publicly available.

> **Comment 16)** Line 398: I don't understand this "correction function" equation at all. It looks like it would have units of #/cm^3, since the "reference concentration" is on the right side of the equation. So this doesn't appear to be a correction factor. Please provide values for the coefficients m, alpha, and b. And shouldn't the counting efficiency be a function of diameter as well as pressure?

Answer comment 16): We thank the reviewer to point this out. This sentence was not phrased very clearly. We fit the parameters of this function to model the counting efficiency of the individual CPCs as a function of sample air pressure. We evaluate this function at each measurement point and apply the inverse of the function value to the raw count data. The parameter c0 is therefore not a concentration but the counting efficiency at the highest pressure measured during the calibration run (roughly the ambient pressure at the laboratory) where it is assumed that pressure does not affect the counting efficiency. We clarified the description in this paragraph.

Comment 17): Fig. A1: Is this counting efficiency curve for 35 nm particles only? I don't find this very informative. I would much rather see efficiency curves as a function of size at different pressures as in the right panel of Fig. A2 for all the CPCs used. You should also show the calibration data rather than just the fitted curves, so the reader can get a sense of the variability and uncertainty in the measurements. This might require more panels in the figure; one for each CPC.

Answer comment 17): We added the calibration data points to the plot in Fig A1 and A2 to give the reader an impression of the variability and uncertainty of the calibrations. The lines shown in the right panel Fig A2 are computed transmission curves for the diffusion screen separators based on Feldpausch et al. (2006) where the procedures have been derived and validated experimentally in detail. The D50 diameters of the bare diffusion screen counters have been measured but are not relevant because they are well below the cutoff introduced by the screens.

We are aware that the counting efficiency is likely size-dependent, however, because we do not obtain detailed size information during our measurement flights it is not possible to account for this effect rigorously. Therefore, based on previous measurements and also the accompanying measurements on the ground, we assume that 35nm is representative for the peak of the soot size distribution and therefore captures the losses of the majority of number concentration. This assumption is clearly introducing a significant uncertainty, especially for particle size distributions with smaller mode diameters, but we feel that it is still better than no correction at all. We also measured the D50 cutoff for all CPCs at different low pressures and found a small decrease of D50 compared to ambient conditions marginally significant within the measurement uncertainty of the setup. Given this variability, we feel that a plot as suggested by the reviewer would not add much information.

Comment 18): Line 428: What is the "correction factor" that is applied? Is this the F(P) from the equation?

Answer comment 18): This refers to a correction factor to account for the losses in the thermal denuder. We clarified the sentence.

Comment 19): Line 443: Isn't contamination from shattered ice crystals on the inlet the primary concern for using measurements made within contrails or cirrus?

Answer comment 19): Ice particle shattering at the aerosol inlet is mostly a concern in presence of larger ice crystals in natural cirrus cloud. Ice particles in young contrails are typically in a size range below 5nm. Those particles are sufficiently small to be ingested into the inlet without causing significant shattering artefacts to the inlet. The measurements used for the analysis were performed outside of natural cirrus cloud.

Comment 20): References: Some reference titles are capitalized; some are not. Please check all references for consistency with the Copernicus editorial guidelines.

Answer comment 20): The manuscript was generated using the the stylesheet of the journal. We also checked for consistency with other papers recently published with ACP.

RC2: 'Comment on egusphere-2024-1224', Anonymous Referee #2, 21 May 2024

We would like to thank the Referee for the feedback on our manuscript and contributing to improve the clarity of our results. We enlarged the symbols of some plots, have modified the coloring and used a colorblind safe palette to improve readability. Answers to specific comments are discussed in the following.

> **Comment 1):** One (semi-)major point: both parts of the Figure 9 (comparison of measured and modelled emission indices) are log-log plots. As the points scatter so widely (especially in the left plot) drawing conclusions seems a bit risky. Line 344 says that there is a "significant correlation" – significant in which respect? Line 385 is more cautious: "generally correlate well". Please discuss in greater detail why you are so confident that the conclusions and the argumentation in lines 349 - 359 are valid.

Answer comment 1): All axes in Figure 9 are linear. Could you specify, which log-log plot you are referring to? However, we agree that Figure 9 shows some scatter. Exact simulation of engine emissions requires precise knowledge of a large number of fuel components, complex combustion mechanisms and the heterogeneous interactions during particle formation. The use of such a detailed model, if one would be available, also requires a large amount of computing power, which makes the use of such a model to study a large fleet of aircraft and their climate impact challenging. The MEEM method and the T4/T2 method offer a publicly available way of modeling soot emissions for further use with reasonable resources. It is therefore to be expected that the comparison of measured and modeled emission indices shows some scatter. Nevertheless, the orders of magnitude and the reductions due to the different fuels are in good agreement, so that both models can be used for further application, for example in climate models or contrail predictions. Both models are currently the best-established public models for modeling aircraft emissions based on accessible engine and fuel data.

> **Comment 2):** General remark: throughout the MS, the term "number" is often used when actually it should be "number concentration" or emission index – please check the MS carefully for proper use of these terms

Answer comment 2): We have corrected the manuscript in this respect.

> **Comment 3):** Line 39/40: ambigous sentence. "positive" in which sense?

Answer comment 3): HEFA-SPK is free of aromatics and sulfuric compounds, which can have a positive effect on contrail properties leading to a reduced radiative forcing. We clarified this in the manuscript.

> **Comment 4):** Line 50: old generation engines – old in which respect?

Answer comment 4): Here, we refer to engines from previous generations. The referenced flight measurements were performed behind the DLR research aircraft ATRA, equipped with two International Aero Engines V2527-A5 engines. The certification date of these engines was 21 November 1992.

> **Comment 5):** Line 65: please give the reason for the regulatory limit for the aromatic content of fuels for non- specialists

Answer comment 5): There is a regulatory lower limit for the aromatic content of the resulting blend fuel of 8% by volume to keep the elastomer seals swollen and prevent fuel from leaking. We clarified this in the manuscript.

> **Comment 6):** Section 2.1: please add a little description of where in the engine temperature and pressure are measured to explain the "T30" etc. In section 2.3.2 (the T4/T2 model is described very briefly – T4 a bit inconsistent with T40?)

Answer comment 6): We specified that in Section 2.21: The combustor inlet temperature (commonly designated as T3 or T30) is defined as the temperature at the outlet of the high-pressure compressor and prior to the combustion chamber. The corresponding pressure at the combustor inlet is accordingly referred to as P30. We prefer to reference the combustor inlet temperature as T30 to keep consistency with the accompanying publication of Märkl et al. 2024. A detailed description of the T4/T2 model can be found in Teoh et al., 2022a.

> **Comment 7):** Section 2.1: briefly describe difference between the two missions.

Answer comment 7): Fuels and the probed engine were different for the missions. We added a sentence in Section 2.1..

> **Comment 8):** Lines 78 – 80: logically, the second sentence should be put first.

Answer comment 8): This change has been implemented.

> **Comment 9):** Line 103: switching from one fuel tank to another: please quantitatively discuss possible fuel contamination due to these switches.

Answer comment 9): Fuel tanks are completely separated, so there is no contamination from one tank to another tank. The sampling of the fuels before and after flight in the fuel tanks were confirming the absence of contamination. To switch from one fuel to another, the pilot was operating the pumps to feed the engines with the appropriate tank, and waited several minutes (~5 minutes) in order to purge all lines from previous fuel before considering the next test point.

> **Comment 10):** Line 104/105: please rephrase – unclear whether the "which" refers to the high content of iso-paraffins or to the n-paraffins.

Answer comment 10): We clarified this in the manuscript: Both HEFA-SPK show a high content of iso-paraffins in relation to n-paraffins. This property could slightly increase the formation of soot particles.

> **Comment 11):** Figure 2: hexagons not explained

Answer comment 11): The marker show the hydrogen content of the fuels used during the flight experiments. An explanation was added to the figure caption.

Comment 12): Section 2.2: add "pointer" to the Appendix.

Answer comment 12): We added a link to the Appendix in the corresponding paragraph.

Comment 13): Line 122: what is meant by "low sulfur levels".

Answer comment 13): We added the sulfur content as well as a reference to the fuel properties table to the sentence.

Comment 14) and 15): Line 125: the term "PM" usually refers to a mass concentration – here "totPM" actually denotes a number concentration – clarify.

Line 130 (and Appendix): there are two CPC models. Which CPC (model) was used for which measurement? In the Appendix, the lower D_50 are given for the different CPCs, but there is no mention which model CPC these D_50 refer to.

Answer comment 14) and 15): Both topics are now addressed in more detail in Section 2.2. CPC 3768a showed a leak during the campaign, therefore only TSI 3010 CPCs were used for the measurements.

Comment 16): Figure 3: which fuel?

Answer comment 16): From 12:00-12:30 HEFA was used, followed by Jet A-1 until 12:55. Finally, emissions of the blend fuel were probed. We added marker to the figure.

Comment 17): Line 160: emission index – units are missing in the text (and in Fig. 9).

Answer comment 17): Units have been added to text and figure.

Comment 18): Line 180: "particulate matter mass and number" –this should be either _concentrations_ or emission indices?

Answer comment 18): MEEM provides an estimation of the emission index for non-volatile particulate matter of both, mass and number of jet engine aircraft at altitude as described by Ahrens et al..

Comment 19): Line 197: acronym AFR not explained.

Answer comment 19): AFR is now defined in this section.

Comment 20): Figures 4 and 6: which "delta T" refers to which condition?

Answer comment 20): We added an explanation in Section 3.1.

Comment 21): Line 220: give reason why "the blend fuel was sampled exclusively at FL310" (by the way: not the fuel was sampled, but the exhaust….).

Answer comment 21): Test points were limited by fuel availability and measurement time at suitable atmospheric conditions. We clarified this in the text.

> **Comment 22):** Section 3.2 particle size distribution: quantitative info on measured number concentrations should be given at least somewhere in this chapter. There is only qualitative info e.g. in line 280.

Answer comment 22): Number concentrations of nvPM > 14nm, expressed as emission index, for FL280- FL310 correspond to those in Figure 4 at medium T30. The difference in the emission index with the flight altitude is small in comparison to the measurement uncertainty. A change in particle concentration, but also the change in size distribution may cause slight deviations in the emission index with flight altitude, but the significance is uncertain.

> **Comment 23):** Line 279/280: "formation of larger nvPM aggregates due to the acting pressure in the combustion chamber": give reason or add reference on the formation process of soot in aircraft engines.

Answer comment 23): We added a reference on pressure dependence of soot particle sizes.

> **Comment 24):** Line 311: it would be nice to see at least an estimate of how much larger the reductions in soot emission would be if market average HEFA-SPK is used.

Answer comment 24): Although it would certainly be interesting, our results do not allow us to predict the reductions when using fuels with marked average hydrogen content. The specific reduction depends on many parameters and has to be assessed individually with respect to fuel properties but also specific engines.

> **Comment 25):** Line 317: "part of the gaseous sulfuric acid may be *chemisorbed and* can link with the surface of available combustion aerosol" – pleonasm. When it is chemisorbed at the particles, it is already "linked" with the surface? Or did I misunderstand what is meant here? If so, please clarify.

Answer comment 25): The sentence has been adapted accordingly.

> **Comment 26):** Line 325: "higher sulfur content…. highest fraction of volatile particles" – indicate fuel type.

Answer comment 26): We added the fuel type in the corresponding sentences.

Comment for editor:

According to newest updates of the campaigns fuel analysis, some values have been adapted in Table 2.

References

Ahrens, D., Méry, Y., Guénard, A., and Miake-Lye, R. C.: A New Approach to Estimate Particulate Matter Emissions From Ground Certification Data: The nvPM Mission Emissions Estimation Methodology, Journal of Engineering for Gas Turbines and Power, 145, https://doi.org/10.1115/1.4055477, 2022.

Feldpausch, P., Fiebig, M., Fritzsche, L., and Petzold, A.: Measurement of ultrafine aerosol size distributions by a combination of diffusion screen separators and condensation particle counters, Journal of Aerosol Science, 37, 577–597, https://doi.org/10.1016/j.jaerosci.2005.04.009, 2006.

Märkl, R. S., Voigt, C., Sauer, D., Dischl, R. K., Kaufmann, S., Harlaß, T., Hahn, V., Roiger, A., Weiß-Rehm, C., Burkhardt, U., Schumann, U., Marsing, A., Scheibe, M., Dörnbrack, A., Renard, C., Gauthier, M., Swann, P., Madden, P., Luff, D., Sallinen, R., Schripp, T., and Le Clercq, P.: Powering aircraft with 100% sustainable aviation fuel reduces ice crystals in contrails, Atmospheric Chemistry and Physics, 24, 3813–3837, https://doi.org/10.5194/acp-24-3813-2024, 2024.

Teoh, R., Schumann, U., Gryspeerdt, E., Shapiro, M., Molloy, J., Koudis, G., Voigt, C., and Stettler, M.: Aviation contrail climate effects in the North Atlantic from 2016–2021, https://doi.org/10.5194/acp-2022-169, 2022.