Response to egusphere-2024-151 reviews for RC1

Referee comments are marked in black bold and are numbered as R1Cx with x the comment number. Author (AR) 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. The former line numbers are given in "()" followed by the new line numbers.

This reviewed paper is an accompanying paper of "Soot aerosol from commercial aviation engines are poor ice nucleating particles at cirrus cloud temperatures" (doi: 10.5194/egusphere-2023-2441), which is still under review at the time of submission of these comments. Testa et al., in this follow-up study of aircraft-engine emitted soot aerosol, describe the reactivation ability of contrail-processed soot. The experiments were conducted on the ground with 2 HINC ice nucleation chambers, which simulated two consecutive steps of cloud formation with a sublimation step between them, at a range of cirrus-relevant temperatures and RHi. This study claims to advance the current knowledge in the indirect climate effects of soot emitted from jet engines, which are responsible for anthropogenic direct injection of fresh soot into higher altitude. Using real engines, the authors have demonstrated an improvement of the jet combustion soot simulation, in comparison to previous studies conducted with soot surrogates. The authors conclude that jet-engine cloud-processed combustion-soot in its compact form is poor INP, similar to activation of the unprocessed soot, despite the expected increase in activation via the PCF mechanism, so heavily reported in previous studies. The authors underline the importance of sulfuric acid and organic volatiles presence in the activation process and their inhibiting impact. The paper is well written and structured in a logical way.

I don't have much comments on the writing or presentation quality but I do have major concerns about the scientific significance and the implied generalized representation of real contrail processes in this and the accompanying paper, both fail to recognize the limitations of such ground experiments even though some of the major uncertainties are mentioned. While such steady flow studies might be relevant for natural cirrus formation, I find there are still major gaps for proper simulation of contrail formation process on the ground.

This accompanying paper focuses on the reactivation however, the evaluation of this sequential study can't be done in isolation ?and it is impossible to avoid the review of the initial soot generation and sampling methodology. It must be done in order to properly evaluate the conclusions and the usefulness of the presented results for future contrail research.

Essentially this study dismisses the relevance of PCF ice to contrails (many of the earlier studies cited in the manuscript), implying that contrail formation processes shouldn't be discussed anymore in the context of PCF. This is a strong claim, which requires a deeper substantiation and discussion of the limitations of this study and further clarification on the applicability of this study to actual contrails. To demonstrate the relevance of this study to high altitude contrails, the authors should have a more detailed description of timescales and rates of processes in their experiment with a comparison to rates known from real contrails.

We thank the reviewer for their comments and address each comment individually below. Here we would like to emphasize that indeed PCF is not relevant for contrail formation. The reason being that PCF is relevant for conditions below water saturation in the atmosphere. Contrails form in the wake of aircraft because of an abundant supply of water vapour that results in condensation and droplet formation

followed by homogeneous freezing. PCF has no role to play in this process, it is entirely driven by the availability of excessive amounts of water vapour that readily condense as bulk droplets onto the available soot particles (Kärcher, 1998) and the low temperatures that promote freezing of these droplets. In this regard, we do not make any changes to the manuscript.

Below I listed some major comments, unanswered questions, and some minor comments. Thus, after a major revision is complete, I'd recommend this manuscript for publication. Please see my comments below:

Major comments:

R1C1.1: The current design involves a stirred tank for offline characterization with isokinetic flow instruments and filter sample collection. I think the experiments could have been designed better to have a real-time sampling, similar to what was done by Korhonen et al. 2022 however, this would not solve all the problems either.

AR1.1: Real-time sampling was conducted in our study for aerosol size distribution (with SMPS) and aggregate mass (with CPMA) analysis. Offline sampling in this study and in Testa et al. (2024) was conducted for STXM/NEXAFS and electron microscopy analysis. The former gives information on the fine chemical structure of the soot aggregates, which cannot be easily determined with mass spectrometer (real-time sampling method used in Korhonen et al., 2022). Electron microscopy analysis of the soot aggregates gives shape information that cannot be obtained other than with microscopy techniques that required sampling and offline analysis of the samples.

R1C1.2: Additionally, I think expansion chambers (e.g. Crawford et al. 2011). might be more representative of the actual dynamic process in contrails. For the current ground setup, there should at least pictures, simulation of the exhaust plume, description of the ambient conditions- was the experiment conducted outdoors, wind/temperature/humidity information, was the heat-resistant alloy probe, located ~1 m downstream of the engine exhaust nozzle, sampling from the middle of the plume, at an angle, etc.

AR1.2: We agree that our setup does not mimic the dynamical processes and rates relevant for contrail formation, e.g., exhaust temperature dropping rapidly (< 1 s) from thousands of degrees to -60 °C and pressure from tens of bars to below 1 bar (Kärcher, 2018). Instead, the time scales are longer in our experiments, the pressure drops from tens of bars in the engine, to ~1 bar (atmospheric pressure in our measurement set up). The temperature also drops in three steps, from the engine temperature to the heated line 160 °C (see below) and then to room temperature followed by a third drop from room temperature to the cloud chamber temperature. Expansion ice nucleation chambers generate supersaturation by decreasing the pressure. While they represent better the dynamics of raising air parcels, they would also fail at mimicking contrail processes (no rapid cooling and dilution of the exhaust or wing vortices formation). Besides, cooling of the particles down to contrail temperature is faster in CFDC than in expansion chambers (Lacher et al., 2017; Möhler et al., 2021) and more representative of in situ rapid aerosol cooling. So, we do have similar temperature drops as would occur in the atmosphere, but they occur over a slightly longer time scale.

In the following, we further describe our ground setup and discuss the limitations of contrail processes in our setup and the impact on soot properties. The ground-based setup used in this study has been described in Testa et al. (2024), as mentioned in the paper (line (41) 45-46). The sampling unit, i.e., the probe sampling the engine exhaust in the test cell (indoor), is described in studies referenced in Testa et al. (2024). We now

also explain it here and add a description to the manuscript (see below). In brief, the air flow in the test cell and hence feeding the engine is at ambient temperature and humidity. Wind speed and sampling unit meet the International Civil Aviation Organization (ICAO) requirements for engine emission measurements. The engine exhaust particles are sampled at the engine exit plane, so not in the exhaust plume (as stated in Durdina et al. (2014) using the same sampling unit: "*Positions of the probes' orifices in the exhaust stream were adjusted such that air-to-fuel (AFR) ratios calculated from the gaseous emissions data agreed within 15% of the AFR from the engine data, and thus they provided a representative exhaust sample.*"). A long trace-heated line (12 m at 160 °C) transports the exhaust to the aerosol reservoir that was in a room next to the engine test cell (Testa et al., 2024). The primary objective of the study is to quantify the ice nucleation ability of contrail processed aviation soot. Thus, the setup was designed to closely mimic the contrail processing of the sampled soot particles (known to mainly affect soot morphology) rather than mimicking contrail formation.

Regarding the setup description, we propose the following edits. Lines (39-44) 40-51 now read: "The experimental setup was designed to simulate targeted atmospheric processes and to mimic the contrail processing of the sampled aviation soot particles (Fig. 1a). Limitations of the ground setup in representing atmospheric processes (e.g., aircraft exhaust evolution, contrail formation) and soot ice nucleation ability are discussed in Sect. 5. Soot particles were sampled from in-use commercial aircraft engines (multiple models from Pratt & Whitney and CFM International) running in an indoor test cell, with air intake at ambient temperature and humidity. The engines were all fueled with Jet A-1 fuel and ran from low to high power (30-100 % sea level thrust). The detailed sampling system is described in Testa et al. (2024) which we briefly describe here. The engine exhaust gas and particles were sampled by a heat-resistant alloy probe ~1 m downstream of the engine exhaust nozzle and directed by a long trace-heated line (12 m at 433 K) to a stirred tank, that was in a room next to the engine test cell and acted as an aerosol reservoir where the soot particles accumulated and coagulated (Fig. 1b). The exhaust temperature drops in three steps, from the engine temperature (thousands of Kelvin) to the heated line temperature (433 K) and then to room temperature followed by a third drop from room temperature to the cloud chamber temperature (< 228 K, see below)".

Line (49-50) 57 now reads: "[…] set to contrail cloud thermodynamic conditions (T = 228 K and RH_w = 105 %, HINC1 in Fig. 1b) allowing […]".

Gas phase chemistry and particle oxidation is thought to considerably slow down while exiting the combustor chamber due to lower temperatures in the exhaust nozzle and downstream of the engine (Dakhel et al., 2007; Wong et al., 2008). Such drop in temperature is also present in our sampling system (thousand degrees to 160°C), thus the primary particle overlap, size, crystallinity and oxidation are thought to be comparable to in situ emitted aviation soot particles. Differences in equilibrium temperatures and dilution would nonetheless impact the partitioning of volatile unburned hydrocarbons and sulfur compounds, i.e., the soot mixing state, and hence the soot ice nucleation abilities. The higher temperatures in our setup (160°C and then room temperatures) compared to the temperature at which the exhaust would be exposed in the atmosphere (- 60°C) would reduce the condensation of volatiles onto the soot due to their higher equilibrium saturation pressures. Higher temperature together with the drying of the exhaust before sampling into the aerosol tank would also prevent the formation of nucleation mode particles. On the other hand, reduced particle surface area (no nucleation mode particles) would favor condensation of volatiles onto the soot particles. Besides, the gases only get slowly diluted by synthetic air in the aerosol reservoir, compared to the strong in situ dilution of the exhaust gases that occur within the first seconds in the atmosphere (Kärcher et al., 2007). Nonetheless, modelling studies (Kärcher, 1998;

Kärcher et al., 2007; Yu et al., 1999) showed that nucleation mode particles and aviation soot are thought to interact in the young aircraft plume downstream of the engine, increasing the soot coating. This process did not take place in our measurement due to the absence of nucleation mode particles. Downstream of HINC1, the reemitted soot aggregates get compacted due to contrail processing and their sizes decrease. However, the formation of large soot aggregates due to the coagulation between ice crystals and the scavenging of interstitial soot aggregates is not possible due to too low concentration of ice crystals and soot and the flow being laminar in the chamber. Next, coagulation of interstitial volatile particles with ice crystals, and coagulation between interstitial soot aggregate with volatile particles is not possible in the chamber due to the absence of nucleation mode particles (although coagulation processes in real contrail remain unquantified today; Moore et al., 2017; Petzold et al., 1998).

Summarizing, we expect in situ particles to be coated with H_2SO_4 and organics. To which extent the coating of the aviation soot sample in our study is different from in situ aviation soot cannot be quantified, but any condensation of organics or sulfate in the atmosphere would first condense into soot pores which inhibits ice nucleation (Gao et al., 2022; Gao & Kanji, 2022), and thus our conclusions for the efficiency of aviation soot ice nucleation remain the same. Next, the soot particles are larger and less dense in our setup than in situ soot due to coagulation in our aerosol reservoir, but their compaction should be representative of real contrail compaction (see AR13, AR20). Finally, the primary particle properties and oxidation are fixed in the combustor (see above and AR4) and hence should be representative of their in-situ counterpart. This is described in part in the atmospheric section in Testa et al. (2024), but we now also add it as a discussion in the atmospheric implication section of the current paper.

The title of Sect. 5 has been modified for: "5 Atmospheric implications and limitations"

Lines (238) 242-283 now read: "[…] leaving cirrus cloud properties essentially unperturbed. <span style="color:red">Due to constraints of the measurement facility in this work, the representation of aircraft exhaust processes in the ground set up and hence of the aviation soot properties might differ from those at flight altitude. For instance, differences in equilibrium temperatures and dilution would impact the partitioning of volatile unburned hydrocarbons and sulfur compounds and hence the soot ice nucleation ability. The higher temperatures in our setup (433 K, then room temperature, Sect. 2.1) compared to upper tropospheric temperature (< 228 K; Krämer et al., 2020), would not promote the condensation of volatiles onto soot as much as would occur at flight altitude temperatures. The interaction of nucleation mode particles with aviation soot in the young aircraft plume downstream of the engine is thought to increase the soot coating (Kärcher, 1998; Yu et al., 1999; Kärcher et al., 2007), but does not occur in our ground setup due to the absence of nucleation mode particles. Thus, soot particles in this study are expected to have a lower amount of coating from this effect. On the other hand, total particle surface area was less in our ground setup due to the absence of the nucleation mode particles, and the exhaust gases experience reduced dilution in the aerosol reservoir with synthetic air, compared to the strong dilution that would occur within the first seconds at flight altitudes (Karcher et al., 2007). This effect would enhance the condensation of volatiles onto the soot particles in our ground setup. However, even if lower amounts of organics and sulfate condense onto the soot particles in the atmosphere, these would first condense into the pores of the soot, due to the capillary effect, and thus inhibit ice nucleation of the soot particles. Thus, our conclusions of the poor ice nucleating ability of contrail processed soot would remain the same. Downstream of HINC1, the reemitted soot aggregates get compacted due to contrail processing and their sizes decrease. The formation of large soot aggregates (Petzold et al., 1998, 1999) due to the coagulation between ice crystals and scavenging of interstitial soot aggregates was not possible due to the low concentration of ice crystals and soot and non-turbulent flow in HINC1. Due to the absence of nucleation

mode particles, coagulation of these with ice crystals, and coagulation with interstitial soot aggregates was not possible in HINC1. However, the absence of these processes does not change our conclusions as adding more organics onto the soot particles would only further result in poor ice nucleation activity (Testa et al. 2024, Gao and Kanji, 2022a). Gas phase chemistry and particle oxidation are thought to considerably slow down while exiting the combustor chamber due to much lower temperatures in the exhaust nozzle and downstream of the engine (Dakhel et al., 2007; Wong et al., 2008). Such a drop in temperature was also present in our sampling system (thousand degrees to 433 K and to room temperature), thus the primary particle overlap, size, crystallinity, and oxidation should be unaffected and comparable to in situ emitted aviation soot particles.

Summarizing, soot particles in our ground setup were larger and less dense than in situ soot due to coagulation in our aerosol reservoir. We believe the aggregate compaction in this work is atmospherically relevant as the parameters driving the soot compaction, i.e., RH_i experienced by the particles and bulk water condensation were represented in our ground setup. The primary particle properties and oxidation are fixed in the combustor and hence should be representative of their in-situ counterpart. Finally, we expect in situ particles to be coated with H_2SO_4 and organics but to which extent the coating of the aviation soot sample in our study is different from in situ aviation soot cannot be established from our study. For this reason, we quantified the ice nucleation ability of coating free (CS-CP-soot) and coated (unprocessed and CP-soot) soot in our study to constrain the possible effect of different soot mixing states on aviation soot ice nucleation."

R1C2: Line 42-43: The gap between the exhaust and the inlet, introducing dilution and turbulence: would it impact the coagulation in the tank? would a higher concentration sampled directly from the engine have resulted in a faster coagulation, a different size mode? if so are you properly simulating the contrail formation process?

AR2: As mentioned above, the sampling unit meets the ICAO requirements for engine emission measurements (Durdina et al., 2014), hence there is no air intrusion between the exhaust nozzle and the sampling probe. As explained in the paper, the sampled soot particles were confined in the restricted volume of the aerosol reservoir for several hours, where they coagulated causing a shift in the particle size distribution to larger sizes and less dense particles. We agree, the soot emission index of the different engines (soot number concentration measured at the engine exit plane) impacts the coagulation occurring in the aerosol reservoir with larger soot emission indices resulting in faster and more efficient coagulation, hence larger soot aggregate sizes. For this reason, the soot mode diameters were different for each engine test and ranged between 80-450 nm (as stated in line (47) 54).

The size of the soot aggregate could impact the activation properties to contrail ice crystals as larger particles might easily activate as cloud droplets. Yet, AF values measured at contrail conditions in HINC1 (20 to 100 %) are in the range of what is expected for contrail formation (Kärcher et al., 2015). The size of the soot aggregates would mainly affect their ability to nucleate ice via PCF (e.g., Mahrt et al., 2018; Marcolli et al., 2021; Zhang et al., 2020) rather than via homogeneous freezing hence affecting their ability to perturb natural cirrus rather than to form contrail (whose determining properties is number concentration; Kärcher et al., 2018). The effect of size, discussed in Testa et al. (2024) and in the present paper (lines (235-243) 294-302), strengthen the conclusion that the non-contrail processed, and contrail processed aviation soot would nucleate ice at or above RH_{hom} and only a small fraction (below 0.1 %) nucleates ice below RH_{hom}.

R1C3: The aforementioned meter gap between the engine exhaust and the sampling inlet: how injection of ambient air downstream of jet fuel combustion would impact soot emissions, concentrations, oxidation etc. I would like to see a discussion referring to findings reported by Kelesidis et al. 2023.

AR3: As mentioned above, there is no injection of air in the meter gap between the exhaust nozzle and the sampling probe. The only injection of (synthetic) air occurred in the aerosol reservoir during the ice nucleation measurements. In opposition to the study by Kelesidis et al. (2023) who injected O_2/N_2 in the combustor chamber (where the gas temperature is high), the low temperature of the air in our setup (ambient temperature) would not allow oxidation of the soot surface and condensed organics (Raj et al., 2013, 2014).

R1C4: What was the temperature of air intake by the engine? Would room temperature air intake affect the combustion products? The characteristics of soot particles formed? How different would it be in comparison to intake of -60C cold air?

AR4: The temperature of the air flow in the test cell and hence entering the engine was similar to the (outside) ambient temperature. The main parameter affecting the performance of aircraft engines is the air density, with lower density leading to lower engine thrust (Balicki et al., 2014). The air density at ground is higher than at flight altitudes resulting in higher thrust at ground (1.2 kg/m^{-3} at ground and 0.3 kg/m^{-3} at 200 hPa and - 60°C, i.e., flight altitude). The effect of engine thrust on particle properties and ice nucleation was investigated and discussed in Testa et al. (2024).

R1C5: Given the fact that the engines tested had differences between the produced soot, can the results of this study be generalized to the extent expressed in the title?

AR5: We agree that differences in soot chemical properties, such as functional groups and sulfur content, and soot morphology, were observed in between the different engines tested (reported in Testa et al., 2024 and in this study). The more ice active soot sampled in our study (CP-CS-soot sample) show very modest ice nucleation ability (compared to aviation soot proxies), with a minimum RH_i onset observed at 143 % (at -55°C). Yet, aviation soot that were contrail processed only (CP-soot sample) are thought to be more representative of flight altitude aviation soot (lines (232-234) 291-293) and their ice nucleation ability, for all different engines tested, converge to RH_{hom} (Figure 2). Lower RH_i onset at 137 % and 140 % were observed in Testa et al. (2024) but only for large (> 400 nm) and hence unrealistic aggregate sizes. As emphasized in the paper, the number and size of cavities within the soot aggregates are the primary controlling factors of soot ice nucleation via PCF. This is primarily controlled by the primary particle morphology and the aggregates size. High overlap of the primary particles has been observed on soot samples for all tested engines. Smaller aggregate size for aviation soot is expected for turbofan engines, hence we believe that the results from our study, that is, aviation soot particles are poor ice nucleating particle for cirrus formation can be generalized to soot emitted with the current aircraft fleet and fuel (jet A/A-1), as emphasized in the title because the size is of the particles is too small to nucleate ice other than by homogeneous freezing of droplets.

Lines 284-293 now read: "Aviation soot samples that were catalytically stripped and contrail processed were able to nucleate ice around 145 % RH_i at 218 K (~7 % lower than RH_{hom}). The modest ice nucleation ability for the CS-CP-soot likely arises from increased cavity number and sizes within the soot aggregates, which would be absent in the unprocessed soot samples coated with organics and sulfate. […] Thicker coatings for flight altitude aviation soot compared to our CP-soot would favor homogeneous nucleation and thinner coatings would be bound by the ice nucleation ability of our CS-CP-soot sample. Nevertheless,

as long as aviation soot is co-emitted with H_2SO_4, it is likely to acquire a coating upon emission and further in the exhaust plume (Kärcher et al., 2007), thus we expect our unprocessed and CP-soot samples to be of higher atmospheric relevance for engines and fuel currently in use."

To complete the soot size argument in lines (235-243) 294-302, we proposed to add the following (lines 303-309 in the revised manuscript): "The number and size of cavities within the soot aggregates are the primary controlling factors of soot ice nucleation via PCF (Sect. 1). The cavity formation is controlled by the primary particle morphology (being determined in the engine combustor and therefore well simulated in our ground setup) and the aggregates size. High overlap of the primary particles has been observed on soot samples for all tested engines. Smaller aggregate size for aviation soot is expected for turbofan engines, hence we believe that the results from our study, that is, aviation soot particles are poor ice nucleating particle for cirrus formation can be generalized to soot emitted with the current aircraft fleet and fuel (Jet A/A-1; > 90 % of global usage; Jing et al., 2022; Pires et al., 2018)"

R1C6: Sulfuric acid was mentioned as one of the main factors limiting the PCF. This is more obvious in a long and slow cooling process that allows for more efficient condensation. What about an instant temperature and significant instant pressure drop between the combustor and the high-altitude environment, would that impact the uniformity of sulfuric acid vapors condensation? The boiling point of sulfuric acid is 166 C, would thermodynamics and cooling rate in your experiment, dictate the condensation of sulfuric acid and impact its degree of soot surface coating, which in turn impacts the reactivation capacity due to the chemical content present in the voids?

AR6: We agree, the ground set up would mainly affect the particle mixing state, i.e., the level of soot surface coating (for instance with H_2SO_4), and size, and hence their ice nucleation ability via PCF. For this reason, we performed experiments for the extreme case of catalytically stripped particles (CS-CP-soot sample) by removing the organics and sulfur coated on the soot with a catalytic stripper. The ice nucleation ability of the sulfur free aviation soot was better than the coated soot, but still remains poor (see Fig 2). We believe that PCF for our catalytically stripped soot sample is limited by the availability and morphology of inter-aggregate cavities, primarily governed by the primary particle morphology, which is not affected by our setup and sampling method but rather fixed in the engine combustor (see AR1.2 and AR5 for proposed text edits).

R1C7.1: The dynamics, rates of processes are extremely important, dropping the particle and air temperature from thousands of degrees, to hundreds and then to subzero temperature during tens of minutes to several hours is not the same as an almost instantaneous transition from thousands of degrees to -60 C, would soot particle temperature be different than its carrying air temperature, would heat dissipation be slower at low pressure, would that impact INP activity and how would that compare to this slow ground test?

AR7.1: As mentioned in AR1.2, we acknowledge that the rates of processes are different to in situ rates in our ground-based setup and conclude that this will affect the soot size and mixing state, namely the coating of the sampled soot particles cannot be strictly compared to that of in situ emitted aviation soot. Yet, although in situ measurement of soot mixing state are lacking, we can make the fair assumption that aviation soot would likely acquire a coating during the processing of the aircraft exhaust (Kärcher, 1999) for current engine fleet and fuel. Thus, we quantified the ice nucleation abilities of coating free (CS-CP-soot) and coated (unprocessed and CP-soot) soot in our study to bound the possible effect of different aviation soot mixing state. A larger or smaller soot coating compared to our CP-soot sample would favor homogeneous nucleation or be bound by our CS-CP-soot sample, respectively. Next, as mentioned above,

the ice nucleation is controlled by the limited availability and morphology of inter-aggregate cavities, primarily governed by the primary particle morphology, which is not affected by our setup and sampling method but rather fixed in the engine combustor.

R1C7.2: Moreover, with the isokinetic flow into the cloud chamber slowly transitioning from room temperature around the inlet of CFDC into -60 C, would you expect glass transition of some of the coatings at higher cooling rates in the atmosphere, would that impact the INP activation? good to discuss the differences.

AR7.2: The reviewer is correct in raising a valid point that the cooling from room temperature to the chamber temperature could promote glassy aerosol coatings on the soot particles. This would depend on if a contrail forms in the atmosphere or not immediately after the exhaust plume forms. If a contrail forms, water condenses onto the soot particles immediately upon exhaust emission into the atmosphere and then freezes to form a contrail. In this case the glass aerosol formation is not favored because of the high humidity (Kilchhofer et al., 2021). As such this would not be relevant for contrail processing. However, in case a contrail does not form, glassy aerosol coatings could form, and may promote ice nucleation via deposition (Knopf et al., 2018) but for a small AF. Cooling of the soot particles to -60°C is instantaneous once the particles enter HINC (as for in situ emitted soot), i.e., glass transition of condensed organics can occur in the atmosphere and could have occurred in our study. If glassy transitions were occurring in our experiments, this would also mean the soot particles should be more effective INPs than what we observe in our studies, but our data do not support the formation of glassy coatings in our experiments since we observe very low to insignificant activated fractions (unprocessed and CP-soot nucleate ice above RH_{hom}).

Lines 287-289 now read: "We note that, if glassy organic coatings formed on the contrail processed (and unprocessed) soot particles, these particles may form ice by deposition nucleation (Knopf et al., 2018), which was not observed due to the high ice nucleation onset above RH_{hom}"

R1C8: The importance of surface properties is discussed e.g., hydrophilic surface oxygenated functionalities, polar groups etc. What's the impact on surface properties of soot aerosol under increased ozone and UV-b, UV-a conditions once emitted at high altitude in comparison to this ground experiment? would proper simulation with exposure to ozone and UV-b would change the results of this experiment? After contrail sublimation multiple things can happen to the soot particles, depending on the timespan, but even in the case of almost immediate reactivation, would ozone and UV change surface properties and the results of the next activation cycle?

AR8: Oxidation of aerosols by O_3 and OH radical can occur in the upper troposphere and has been investigated for several soot types in past studies (Browne et al., 2015; Dymarska et al., 2006; Friebel et al., 2019; Friebel & Mensah, 2019; Gao & Kanji, 2024; Ghio et al., 2020; Han et al., 2016). The combined results of these studies show that the exposure of O_3 or OH to soot of different sizes and with different organic fractions, impacts the soot properties differently. For highly graphitic soot with low organic fraction, exposure to ozone increases the soot surface water uptake capacity due to increased hydrophilic groups. For organic-rich soot, several processes might occur. Condensed organics might desorb due to the breaking of covalent bounds with the elemental soot fraction. The oxidized organic products can recondense onto the soot due to their lowered volatility, potentially filling the soot aggregate cavities. Furthermore, oxidized organics, e.g., short alkene and aldehyde that are soluble in acidic solution (e.g., H_2SO_4 solution; Yu et al., 1999) can participate to freezing point depression if condensed in pores or they can prevent water uptake by blocking the pores if there are hydrophobic, inhibiting PCF (Gao & Kanji, 2024). Overall if organics are present on the soot particles, ozone oxidation will not promote ice nucleation to conditions below RH_{hom}.

In addition, even if organics are absent on the soot particles, the increase in hydrophilicity from ozone adsorption is still secondary for ice nucleation by PCF, and the abundance of pores is still the primary controlling factor (Gao & Kanji, 2024). Since the aviation soot particles lack pores relevant for PCF and possess an organic coating, the aging in the presence of ozone or OH and UV will not enhance their ice nucleation activity.

We expect aviation soot to be coated with organics (PAH, smaller hydrocarbons, aliphatic; Abegglen et al., 2016; Marhaba et al., 2019; Parent et al., 2016) and thus following the above observations from the literature, we can say that the exposure of (organic-rich) aviation soot to O_3 or OH at flight altitude would decrease their ability to nucleate ice via PCF due to pore blocking and/or freezing point depression (with oxidation on the time scale of hours; Friebel & Mensah, 2019). An eventual increase in soot water uptake capacity from O_3/OH oxidation and thus an enhancement of aviation soot ice nucleation via PCF would remain limited owing to the lack of aggregate mesopores observe for aviation soot in this study and in the companion study. Nevertheless, we do not rule out that oxidation of soot surface organics can also turn into glass and promote heterogeneous ice nucleation of the particles (Tian et al., 2022).

Lines 310-318 now read: "In addition to contrail processing, several soot aging processes can occur in the atmosphere, such as interaction with background aerosols and volatile compounds, or oxidation of aerosols by O_3 and OH radicals (Bond et al., 2013). Interception of soluble aerosol onto the soot surface would increase the amount of soot coating preventing PCF. Exposure of aviation soot to O_3 or OH at flight altitude can cause condensed organics to desorb due to the breaking of covalent bonds with the elemental soot fraction. The oxidized organics could recondense onto the soot due to their lowered volatility, e.g., short alkanes and aldehyde that are soluble in acidic solution (e.g., H_2SO_4 solution; Yu et al., 1999) and could lead to a freezing point depression if condensed in pores or prevent water uptake by blocking the pores if there are hydrophobic, inhibiting PCF (Gao & Kanji, 2024). Nevertheless, we do not rule out that oxidation of soot surface organics can also turn into glassy coatings and promote deposition ice nucleation of the particles (Tian et al., 2022)."

Following this edit and edits from AR1.2, AR5 and AR7.2, we move down the comparison of aviation soot ice nucleation with mineral dust that was out of place (now at lines 319-321): "Additionally, the presence of other potent atmospheric INPs would further limit the effect of aviation soot on cirrus cloud microphysical properties. For instance, we note that the CS-CP-soot RH_i onset remains substantially above that for mineral desert dust, e.g., about 120 % (Ullrich et al., 2017), which all outcompete soot ice nucleation."

R1C9: On the same topic of surfaces charges, which seem to play a role in the ice nucleation process. Charging is often observed in expansion chambers experiments (e.g., AIDA). I haven't seen any discussion about charging and charge transfer of soot in the exhaust ejection process, would it play a role in measured INP activity in comparison to long mixing in the tank on the ground and isokinetic low flow aspiration with HINC?

AR9: Charging effects were reported to be significant for soot cluster coagulation within the combustor (high number of charges per particle due to the high temperature) but negligible once the particles exit the combustor (only slightly positively charged [+1 in average]; Dakhel et al. 2007). Charging effect would nonetheless impact the formation of nucleation mode particles for in situ jet plume (Kärcher et al., 2015) but these were curbed in our setup.

R1C10: Is there any impact of exhaust inhomogeneity and selected sampling inlet location/orientation?

AR10: The sampling unit was designed to meet the ICAO requirements for engine emission measurements, therefore we do not expect sample inhomogeneity induced by the inlet of the sampling unit.

R1C11: Does this setup represent well the impact of shear forces trapping the exhaust in vortices behind the aircraft, would this impact the results? could the vortices create multiple sharp supersaturation and sublimation cycles? How does that compare to the timespan of this experiment?

AR11: We agree with the comment, aircraft exhaust can be trapped in the vortices forming in the wake of the aircraft, where entrainment of air and vertical ascent/descent of the plume can create large supersaturation and freezing/sublimation cycle of the soot particle (Kärcher, 2018; Kärcher et al., 2015; Miake-Lye et al., 1993). We believe that coagulation of the soot particles (and hence effect on their sizes) would remain limited as measured with recent in situ study (Moore et al., 2017). Soot shape and mixing state could nonetheless be affected by the freezing/sublimation cycle and interactions with the numerous volatile particles, respectively.

In our ground setup, aircraft wing vortices were not represented as the aircraft engines are tested in isolation from the aircraft frames (and wings) in the test cell. Freezing/sublimation cycle in wing vortices would be essentially similar to contrail processing, as simulated in our study. The impact on aviation soot mixing state was not represented in our setup, but the effect on the soot ice nucleation properties is similar as discussed above (AR1.2 and AR5).

R1C12: A key achievement in this study (e.g., lines 31-33,224-225 and elsewhere) is the production of jet engine soot aerosol that unlike previously studied BC proxies is more representative of the contrail ice nucleation followed by "contrail" sublimation and reactivation. While the combusted jet fuel better represents the real content of the exhaust, anything downstream the exhaust is still significantly different on many dimensions in comparison to the physics and thermodynamics of high altitude. The author's claim of a greater relevance needs to be further substantiated, alternatively the claims should be toned down (as should be the accompanying paper) on its relevance to actual contrails or discuss in greater breadth the limitations of this study. These questions have to be addressed before the second step of contrail processing can be properly evaluated.

AR12: We agree that some processes occurring downstream of the aircraft engine are not represented or absent in our ground-based setup and that would affect the aviation soot properties. Those are now discussed in the above and text have been added as indicated in the previous comments.

Next, as mentioned in AR7.1, our CS-CP-soot sample could represent the best possible ice nucleation ability for aviation soot. The fact that in situ emitted soot aggregates have much smaller sizes further strengthen the claim that the large CS-CP-soot sampled in this study represent to maximal ice nucleation potential for aviation soot and that aviation soot would most likely not perturb cirrus cloud due to their poor ice nucleation ability.

R1C13: Was the CATZ time loscale representative of contrail sublimation rate? Do you think liquid/solid phase chemistry could play a role here, in the context of sulfuric acid dilution?

AR13: The ice crystals sublimation/growth rate depends on the relative humidity experienced by the ice crystals (Lohmann et al., 2016). RH_i in CATZ is about 60 %, i.e., representative of most upper tropospheric clear sky humidities (Krämer et al., 2020) hence likely encountered by sublimating contrails. We note that compaction of aviation soot aggregates might be hampered if coated with viscous material (e.g., organics)

but that it would nonetheless occur while the soot coating dilutes with liquid water during contrail cloud droplet activation (Corbin et al., 2023).

R1C14: The paragraphs and sentences dedicated to hypotheses about alternative jet fuel activation e.g., in the atmospheric implications section. In my opinion, these should be avoided. There is no need to hypothesize about experiments that have not been done, especially given the questions remaining about the applicability of the presented ground test results to real contrails. I'd recommend focusing on the applicability and substantiation of the results of this study with the fuels and engines actually tested.

AR14: We agree that a discussion on the limitation of the study and the implication for aviation soot ice nucleation need to be further developed in our paper. Such a discussion has now been added into the revised version of the paper as part of responses AR1.2, AR5 and AR7.2. We keep the discussion on the size effect, since there is a wealth of literature supporting the dependence of ice nucleation on soot particle size. We emphasize here that small soot size limits PCF will still hold for soot emitted from alternative aviation fuel s (AAFs) as those are smaller or as small of Jet A/A-1 soot (e.g., Moore et al., 2017; Durdina et al., 2021). Besides, the few studies (see references in Sect. 5) investigating other aviation soot properties such as primary particle size and hydrophilicity, point out to no significant changes of those properties and consequently, limited potential for ice nucleation enhancement for AAF soot compared to standard fuel soot. Such discussion is needed to set the priority in future research on the climate impact of aviation.

Minor comments:

R1C15: "Contrail processed…." – the title is misleading, the ground/laboratory experiment doesn't cover the full complexity of the contrail formation process at high altitude nor does it simulate the transition rates in a "violent" contrail formation but rather presents a cloud formation in a steady state environment, feeding well stirred combusted soot at room temperature into an ice nucleation chamber. Please come up with a title that properly describes the ground simulation that you present.

AR15: We agree with the reviewer comment and change the title to "Simulated contrail processed […]".

R1C16: Figure 1 caption: "incomplete combustion… emits" - combustion produces, or aircraft emits

AR16: Lines 1-2 in caption Figure 1 now reads: "From left to right: Due to the incomplete combustion of aviation fuel, aircraft engines emit soot particles […]"

R1C17: Line 14: "and they exhibit" – they hint on high likelihood of poor ice nucleation ability at cirrus relevant….

AR17: Lines (14) 14 now reads: "[…] for the first time, and suggest a high likelihood of poor ice nucleation ability at cirrus relevant temperatures […]"

R1C18: Line 28: the accompanying study (not published) states 2-50 nm. Which one is correct?

AR18: By definition, the mesopore range covers 2-50 nm pore diameters. Marcolli et al. (2021) show that the 2-30 nm range is relevant for PCF, while pore diameters above 30 nm are most likely too large to get filled with water below water saturation.

To clarify this, we propose to modify lines (28) 28-29: "Pore of relevant diameters for PCF are in the range of 2-30 nm (Marcolli et al., 2021) and fall into the mesopore size range of 2-50 nm (Haul, 1982)."

R1C19: Figure 1 caption: "forming a contrail cloud due to high concentration of water vapor and cold temperatures" – in the aerosol reservoir depicted, aren't you removing water vapor down to less than 10%? How does that correspond to the process in the atmosphere?

AR19: In our setup, contrail formation (or ice crystal formation onto the soot particles at contrail conditions) is mimicked in HINC1, there the RH_w is 105 % (see lines (47-59) 54-66).

R1C20: Line 151-152, similarly compacted regardless of their size: would a higher sublimation rate cause a greater compaction or was the maximum compaction reached?

AR20: The driver for soot compaction is the coating mechanism, i.e., the rate of condensational growth into the soot capillaries and the amount of condensing material, with the compaction reduced for RH_w > 120 % (less time for capillary formation; Corbin et al., 2023). In our study, contrail processed soot were exposed to water vapor at RH_w = 105 %, i.e., in the range of what required for maximal compaction according to the review study from Corbin et al. (2023). The aspect is discussed in the proposed text edits in AR1.2.

1C21: Same for line 159, was maximum compaction reached?

AR21: The convexity/size change of the contrail processed aviation soot sample from this study is very similar to what measured for other cloud/contrail processed soot types (China et al., 2015; Ma et al., 2013; Mahrt et al., 2020). This is explained by the harsh activation process occurring in HINC1 and CATZ (activation in cloud droplets and ice crystals followed by sublimation). Although large compaction was observed in our study (convexity approaching 1), we do not rule out that smaller condensation/sublimation rate in the atmosphere could lead to larger compaction of the particles.

We propose to clarify this as well in the paper. Lines 259-265 now read: "Downstream of HINC1, the reemitted soot aggregates get compacted due to contrail processing and their sizes decrease. We note that, the convexity/size change of the contrail processed aviation soot sample from this study is very similar to what measured for other cloud/contrail processed soot types (Mahrt et al., 2020; China et al., 2015, Ma et al. 2013). This is explained by the harsh activation process occurring in HINC1 and CATZ (activation in cloud droplets and ice crystals followed by sublimation; Corbin et al., 2023). Although large compaction was observed in our study (convexity approaching 1), we do not rule out that smaller condensation/sublimation rate in the atmosphere could lead to larger compaction of the particles. The formation […]".

R1C22: Figure 5: a colorbar should be included in the main plot and x axis titles on the adjacent box plots.

AR22: We agree, Figure 5 has been updated to include the color bar.

R1C23: Line 223 unlikely to promote ice nucleation…

AR23: We agree, line (223) 236 now reads: "showed that aviation soot is unlikely to promote ice nucleation"

R1C24: Line245-259, see major comment 14, this doesn't fit into atmospheric implications of the current study. It looks more like a discussion paragraph about future work that can be done. I would personally prefer to see a future work discussion that will present design insights from this study that can be modified to improve the simulation or guidelines to build a future ground facility for more accurate simulations of contrail formation. Alternatively, in this section there could be more discussion about the

impact of the results on radiative transfer (depicted in Figure 1), what is the difference in radiative transfer between what was previously assumed and with the new findings.

AR24: A detailed discussion on the radiation transfer is beyond the scope of this study. With regard to the ice nucleation ability measured for the aviation soot sampled in this study (and in the companion study), together with the limitations of our ground setup and implications for aviation soot ice nucleation properties (discussed in AR1.2-AR11), we consider that our conclusion, e.g. lines (278-279) 355-356 "aviation soot particles would likely not serve as INP for cirrus formation and that current radiative forcing estimates", is justified.

The present study focusses on aerosol properties and ice nucleation. This justifies a discussion on aerosol properties from AAF. Implications for radiative transfer are beyond the scope of the paper, but highly relevant to the topic of aviation soot-cirrus interaction and therefore will be investigated in follow up modelling study.

R1C25: Line 268 strong compaction (65% convexity increase) – is that the max compaction that could be achieved? What would be the typical difference between a strong and a weak compaction?

AR25: The measurements show that the initial size impacts the compaction level, with larger compaction for larger aggregates. Dedicated measurements, e.g., as a function of coating level, aggregate sizes and exposed humidities during condensation and sublimation of liquid water, would be needed to assess the maximal reachable compaction. Nonetheless, from the TEM images, the contrail processed aggregates become round shaped with no or few open aggregate branches, as visible in comparison for the unprocessed soot (see Fig. A2). Weak compaction would be characterized by little change of the shape and size of the aggregates, i.e., low convexity and open-branched aggregates.

R1C26: Line 271: In real contrails, would this condensation be impacted by the rate of the process, would the phase of the organic matter be impacted by the rate of cooling e.g. Zhang et al. 2019. Would that in turn influence the INP activity?

AR26: As mentioned in AR7.2, organics could have turned glassy due to the fast cooling of the particles once entering HINC1 (as for in situ emitted aviation soot while exiting the exhaust nozzle). Yet, no heterogeneous ice nucleation was observed for the coated, i.e., unprocessed and CP-soot soot samples that could be due to glassy organic phase on the soot surface.

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