Dear Bernd,

please find our answers to your comments below (your comments are repeated in italics).

Aircraft exhaust plume turbulence affects the formation and properties of contrail ice crystals and how predictions of nucleated ice numbers and sublimation losses relate to aircraft measurements [Kärcher, 2018]. This manuscript draft describes a project activity relating to the question how contrail ice formation might be affected by coupling plume turbulence and ice microphysics.

Decades of research established the basic contrail ice formation pathway (activation of size-dispersed plume and ambient aerosols present in decaying jet aircraft exhaust plumes). The most important findings, used as the basis of an intentionally simplified parameterization scheme [Kärcher et al., 2015], have been confirmed by field measurements. This is particularly true, on a quantitative basis, for the number of contrail ice crystals as a function of aircraft-related parameters and ambient conditions.

While I understand the desire to ultimately include more microphysical complexity into LES in the author's quest for gaining new insights, open research issues that potentially challenge established findings should be clearly identified and formulated. Here I mean those issues that we have incomplete knowledge of or that contradict observations. In my view, the authors could improve on this, especially in the light of their concluding statement (line 620): "Hence, using a large spatially resolving trajectory ensemble does not necessarily lead to improved scientific results contrary to what we expected in the beginning." and the significant progress in coupling turbulence and microphysics in 3D-LES reported by Lewellen [2020].

Most results and findings you refer to were obtained with simple box models. With regard to model development, the paper by Lewellen (2020) is a significant progress. For instance, Lewellen (2020) found that ice crystal formation on ultrafine volatile particles is more substantial in 3D LES than in a box model updating earlier findings in Kärcher & Yu (2009). In our opinion, it is not necessary to hypothesize in advance on established findings that are potentially challenged by a new model.
Our goal is to setup a modelling system that is similarly-tailored as the model in Lewellen (2020) and which allows similar, yet independent simulation studies.
As 3D simulations of contrail simulations are costly, an alternative less demanding approach would be also desirable. In this regard, the present manuscript investigates the approach of applying a box model for a large trajectory ensemble. We conclude that independent computations without any information exchange among the various trajectories give unsatisfactory results.
This issue is discussed in the evaluation section and corresponding remarks are also given in the next-to-last paragraph of the conclusions, but statements in the abstract were still missing. Therefore, we have added
"Using an ensemble mean framework instead of a single trajectory does not necessarily lead to an improved scientific outcome. Contrail ice crystal numbers tend to be overestimated since the interaction between the different trajectories is not considered."

at the end of the abstract.

In their project, the authors opted to include an intermediate-complexity microphysical approach into their framework, which basically replicates the original parameterization approach [Kärcher et al., 2015]. For example, neglecting the liquid phase denies the opportunity for further in-depth study or sanity checks on older results. This seems particularly relevant, as the author's ultimate goal is to include the described methodology in 3D-LES (line 23f). In a more realistic setting, consideration of a kinetic description of droplet activation and ice nucleation is arguably required for proper simulations of the interaction between droplet and ice microphysics and turbulent entrainment-mixing in plume regions, where all these processes develop on similar time scales.

It is true that our microphysical pathway for contrail formation is very similar to Kärcher et al. (2015), but there are strong conceptual differences of how the microphysical processes are modelled: Kärcher et al. (2015) assume within their analytical approach that all droplets form on soot or entrained ambient particles at one particular time in the fast cooling plume. Thereby, that droplet number concentration that is needed to balance a further increase of plume supersaturation due to condensation loss is determined. This is of course a simplification since important competition effects like subsequent activation of different particle type/sizes and the interaction of exhaust particles with plume relative humidity are not considered.

In our study, the box model explicitly simulates the contrail formation process in a simple dynamical framework. In our particle-based microphysical approach, soot particles and hydrometeors (i.e., ice crystals or water droplets) are described by simulation particles (SIPs). Each SIP stores information about the phase, dry and wet radius and weighting factor.

In the 2nd sentence, you imply that we would neglect the liquid transition phase which is not true. Soot particles first activate into water droplets (if water supersaturation is sufficiently large) and subsequently freeze to ice crystals. Opposite to Kärcher et al. (2015) droplet and ice crystal formation occurs at different plume ages (depending on the particle dry core size) and the hydrometeors directly interact with plume water vapour and temperature (latent heating) at least within one trajectory. Using the diffusional growth equations, our droplet activation automatically exhibits a kinetic character. The homogeneous ice nucleation indeed occurs instantaneously assuming that the entire droplet freezes immediately once ice nucleates within its volume. But this is a reasonable approximation since ice crystal growth in contrails is slow relative to droplet freezing (Ford, 1998).

Explicitly simulating water activation of liquid or mixed-phase aerosols (here: exhaust soot coagulating with the evolving ultrafine aqueous aerosols) alongside homogeneous freezing is pretty standard in cloud physics. Numerical representations are available that are on the one hand consistent with the original LCM treatment of aerosol and ice growth [Sölch & Kärcher, 2010] and on the other hand employed, with an even greater level of complexity than needed for contrail studies, even in global climate models [Jacobson, 2002].

The intercomparison of results with other models, be it an LES or a high-complexity microphysical model, is clearly meaningful, especially when measurements are difficult to interpret. However, in this case, airborne measurements of the temperature and humidity dependence of contrail formation [Bräuer et al., 2021] are available to put the model predictions to the test. The comparison of the author's extended approach with the original parameterization [Kärcher et al., 2015] is less valuable for validation, since they base most of their methodology (Section 2.2) on this parameterization.

The study of Bräuer et al. (2021) appeared on 28th April and we submitted our manuscript on 29th April. Hence, we were not able to include this study in our work.

Clearly, a validation effort must include a comparison with observational data in order to check if the assumption on the underlying model physics are well-chosen. But this is only one aspect. The other point is that the numerical implementation of solving the underlying model physics should do what it is supposed to do (also called “verification” in software design).

As stated above, the underlying model physics in the present study and in Kärcher et al. (2015) are similar, yet the approaches to solve the model physics are quite different. Hence, we do not agree that a comparison with Kärcher et al. (2015) would be less valuable. It is interesting to see results of different methodologies that are based on similar model physics.

I have a number of further points the authors may wish to clarify/reassess/explain/check/update/expand upon.

38-41: Ultrafine aqueous plume particles have been shown to form a second contrail ice mode from uptake of nitric acid to form ternary H2O/H2SO4/HNO3 solutions, partial activation and homogeneous freezing alongside water activation [Kärcher, 1996].

This is an interesting aspect, but Fig. 1 of Kärcher (1996) clearly shows that the number concentrations of the second volatile particle mode (H2SO4/H2O) and the HNO3/H20 solution droplets mode are several orders of magnitude lower than the number concentrations of the first volatile particle and the soot particles mode. Although the larger volatile particles in the second mode are easier to activate than the smaller ones in the first mode, their contribution to contrail ice crystal formation is very low (see also argumentation in the next answer.)

40: The large size tail of number-size distributions of ultrafine aqueous plume particles is extremely steep [Brock et al., 2000], so the supersaturation needed to water-activate these particles is highly variable and includes values that barely exceed liquid water saturation.

Even though measured volume and surface area display a second mode for particles with diameter (D) larger than 10 nm, the number distribution of volatile particles is clearly dominated for $D < 5$ nm with maxima at around 3 nm (Brock et al., 2000). These small particles require, due to the Kelvin effect, high water supersaturations (e.g., 25% for 3 nm H2SO4 particles using Kappa-Köhler-Eq.) to form water droplets. Of course, the larger particles ($D > 10$ nm) at the distribution tail are much easier to activate (critical supersaturation of a few percent), but their contribution to the overall volatile particle spectrum is very low.

The box model study of Kärcher & Yu (2009) shows that ultrafine volatile particles have only a significant impact on contrail ice nucleation for soot-poor emissions and ambient temperatures of at least more than 5 K below the formation threshold while their impact is very low for soot-rich emissions (as they typically occur behind commercial aircraft). The latter is because the droplet formation on the particularly larger soot particles and the corresponding depletion of plume humidity typically suppresses the activation of ultrafine volatile particles even at lower ambient temperatures.

In contrast, the 3D LES study of Lewellen (2020) indicates that a significant fraction of contrail ice crystals originates on ultrafine particles even in combination with high soot emissions (see his Fig. 3). This means that the relevance of exhaust or entrained ambient particles on contrail formation might change significantly when switching from a 0D box model set-up to 3D LES.

41: Can you be more specific what you mean by seconds? This assertion requires evidence. In which conditions away from the formation threshold would it take longer than 0.5–1 s to form contrail ice?

We have replaced "in the first second(s)" by **"**within the first second**"**.

This is valid at ambient temperatures (Ta) far away from the formation threshold. Only close to the formation threshold (Delta_T <~ 1.5 K), ice crystal formation might take longer than 1 s as displayed in our Fig. 2d. Note that the latter results holds for the original FLUDILES trajectories that suffer from the time lag due to the neglection of the conversion of jet kinetic in thermal energy. If these trajectories are accelerated (explained in section 5 and shown in Fig. 7), contrail ice crystals indeed form within the first second even at conditions very close (Delta_T < 0.3 K) to the formation threshold.

52-54: Small soot particles will not water-activate in threshold conditions, so this argument seems to be moot. What is the sensitivity of threshold ice numbers (then originating from the largest soot particles) on the Kelvin effect and what is the uncertainty in determining the underlying surface tension? Assuming water saturation to be sufficient for soot activation has only been used as a reasonable approximation in the contrail parameterization [Kärcher et al., 2015]; the underlying numerical process model does not make this assumption.

First, it is not true that Kärcher et al. (2015) assume water saturation to be sufficient for the activation of soot particles. (This aspect is explained in more detail in the answer to the question regarding the "adapted version of the parameterization".)

Fig. 1 B (Appendix of the current manuscript) displays that the critical saturation ratio for activation of soot particles (*Sc*) increases with decreasing dry core radius r*dry*. This increase becomes significantly stronger for *rdry* below 15 nm due to the non-linear Kelvin term. The smallest soot particles (*rdry* < 10 nm) require high plume water supersaturation (> 20%) which is not achieved at near-threshold conditions. Therefore, those soot particles cannot activate into water droplets and subsequently freeze to ice crystals (excluding heterogeneous ice nucleation). We have replaced "small" by "only several nanometer sized" for clarification. In this study, we assume the surface tension of pure water droplets that includes a temperature dependency based on experimental findings (see also answer to your last point).

56-60: Why "However"? This is not (necessarily) a contradiction.

This is true. We replaced "in general leading to a hydrophobic character" by "causing a weak hygroscopicity" and removed "However" in the subsequent sentence.

100: In my opinion, such exhaust soot particle properties are less uncertain than claimed in line 133ff [Moore et al., 2017].

Experimental data of measured soot properties are quite limited both at ground level and even more at cruise altitude conditions. Hence, there is of course a significant uncertainty. This point is also discussed in section 3.1. of Kärcher et al. (2015):

"Number EIs for aircraft soot particles can be inferred from mass EIs using fixed mass-size relationships […] Predicting EI_s from equation (7) requires accurate *rs* [median dry core radius] values. Variations in *rs* and Ms [soot mass emission index] alone cause in-flight soot number emission indices to lie between 10^{14} and 10^{15} $(kg\text{-}fuel)^{-1}$. **However, experimental data sets suitable for constraining mass-size relationships for cruising conditions are very limited.**"

144: While numerical results may converge, I wonder about the spatial resolution of the ice crystal mode. I understand that contrail ice is resolved by 50-200 SIPs, yet typically tens of thousands of ice

crystals form in contrails per cubic centimeter of air. How many real ice crystals are represented by one SIP on average? Can you estimate how many SIPs will be needed in the full 3D set-up in order to obtain a reasonable spatial coverage across the entire plume cross-section?

Let us define n_{weight} as the number of real ice crystals that are represented in one SIP. The average ice crystal number $n_{weight,m}$ can be derived from the trivial relationship $n_{weight,m} = N_{IC}/N_{SIP}$, where N_{IC} and N_{SIP} are the total numbers of ice crystals and SIPs, respectively. The total ice crystal number depends on the soot number emission index and the fraction of soot particles freezing to ice crystals. In our baseline case, $n_{weight,m}$ is around 10^{10} for a single average trajectory. In the ensemble runs with 25000 trajectories sampling the expanding plume, $n_{weight,m}$ is accordingly $N_{IC}/(25000*N_{SIP})$.

Note that the quality of a particle-based microphysics simulation cannot be judged by means of the n_{weight} value. A large absolute value of n_{weight} does not imply that a specific simulation is coarse. Analogously, one would not change the number of bins in a bin model depending on the ice crystal number in a cloud. An adequate number of SIP or bins depends much more on the dispersion of the ice crystal size distribution and other factors, but clearly not on $n_{weight.}$

At the current stage, it makes no sense to estimate the number of SIPs required in 3D simulations of contrail formation. We will make corresponding convergence tests once the 3D setup is established. By the way, Unterstrasser & Sölch (2014) found that the number of required SIPs per grid box is smaller in higher-dimensional domains due to averaging effects. We expect the same benefits from averaging in the upcoming simulations. Moreover, the LCM code is equipped with SIP merging and splitting operation to interactively adapt the number of SIPs. The splitting operation will be useful in strongly expanding exhaust jets/plumes which enables us to increase the number of SIPs over time.

165: How sensitive are the critical supersaturations (and derived variables, ultimately, the contrail ice numbers) calculated based on eq 1 to uncertainties in surface tension? In understand that the method does not track the acid or water mass fractions in the soot particle coatings deviating from the high-complexity models (and therefore also keeps the parameter kappa in eq.1 constant). How then is the surface tension of the acidic solutions estimated?

In this study, we assume the surface tension of **pure water** droplets. Acids in the soot particle coating can be positively absorbed by the water droplet, where "positive absorption" means that acid molecules concentrate near the droplet surface. This causes a decrease in surface tension relative to that for pure water droplets**.** However, the volume fraction of soot coating substances (like sulfuric acid) makes up only a few percent of the overall soot particle/droplet volume (e.g., Petzold et al., 2005). Therefore, the impact of soot particles coating on surface tension and, accordingly, on critical saturation ratios are supposed to be negligible and we do not consider the variation of droplet surface tension with acidic solution concentrations.

This is now clarified in the newly created Appendix B1) (see also the answer to your last point).

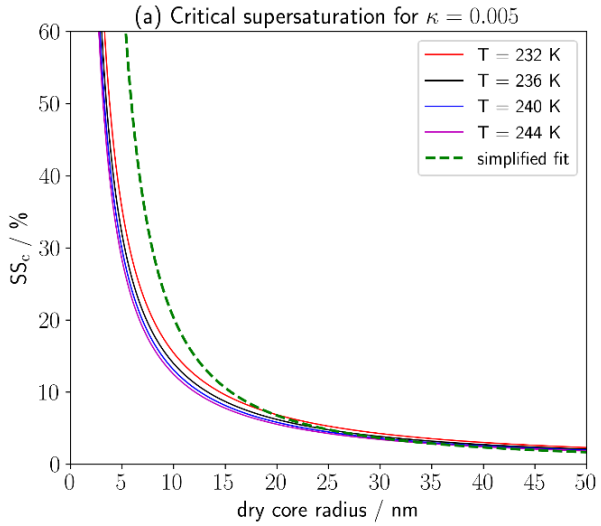
186: Why would the method to estimate the freezing threshold temperature be suitable only in "strong cooling situations" and why don't the author's refrain from basing their estimates of freezing fractions on actual freezing rates? The latter contain valuable kinetic information. In doing so identical to the original parameterization [Kärcher et al., 2015], this approach tends to maximize freezing fractions.

Concerning your first question our wording was misleading. The method is not "only" but "**also"** suitable in strong cooling situations. We have clarified the sentence by writing "This method **can handle a** strong cooling situation …".

We do not agree with the last point since instantaneous freezing of water droplets in exhaust plumes is a reasonable assumption as explained above.

584: What is an "adapted version"? if changes have been made to the original parameterization [Kärcher et al., 2015], the impact of these changes on ice crystal numbers should be documented, as the performance of the original parameterization was tested against observations.

Kärcher et al. (2015) use the Kappa-Köhler theory (Petters and Kreidenweis, 2007, abbreviated hereafter by PK2007) to calculate the so called activation dry core radius for soot ($r_{d,act,s}$) and entrained ambient particles ($r_{d,act,a}$) for given plume water saturation ratio S_p . This means that all soot (ambient) particles with dry core radii $r_{d,s} >= r_{d,act,s}$ ($r_{d,a} >= r_{d,act,a}$) will activate into water droplets if a certain water supersaturation is reached. Note that in this method, S_p is effectively considered as the critical saturation ratio S_c (maximum of the Köhler curve) and $r_{d,act}$ is calculated as the corresponding dry core radius to S_c. Kärcher et al. (2015) **prescribe the hygroscopicity (kappa) of soot with 0.005** and set the solubility of ambient particles to 0.5. For the calculation of $r_{d,act}$ Kärcher et al. (2015) use the **simplified fit Eq. (10) of PK2007** (even including a transcription error which has been corrected in Bier & Burkhardt, 2019), which is only valid for **kappa >~ 0.1**. While this equation is appropriate to treat ambient particles, it is not valid for soot particles due to their weak hygroscopicity. The Figure below displays our calculated S_c for the activation of soot particles (applying the method described in App. B2) at different plume temperatures, where typically droplet formation occurs, and that obtained from Eq. (10) of PK2007. We see that the latter significantly overestimates S_c for dry core radii < 15 nm.



Critical saturation ratio (S_c) for activation of soot particles (setting the hygroscopicity parameter to 0.005) depending on their dry core radii for different plume temperatures (T) calculated as the maximum of the Kappa-Köhler Equation (6) from Petters & Kreidenweis (2007) by applying Newtonian Iteration. The green dashed line shows the result for S_c when using the simplified Equation (10) from Petters & Kreidenweis (2007).

In the adapted version of Kärcher et al. (2015), we replace the simplified fit Eq. (10) of PK2007 for soot particles by a polynomial fit of the black line (in the Figure) for a plume temperature of 236 K. For the reasons described above, we favour to use that adapted version in our study. Testing the **original** parameterisation against observations is no reason to suppress improvements or corrections in microphysical implementations of that parameterisation.

602f: The maximum supersaturation is controlled by the contrail ice crystal number concentration.

Of course, the (maximum) plume water supersaturation is also controlled by the droplets/ice crystals that form on soot particles. In the sentence, the hypothetical supersaturation that purely results from the thermodynamic formation criterion (in absence of microphysics) was meant.

Therefore, we have added "(**in terms of pure thermodynamics)**" behind "maximum supersaturation" for clarification.

604ff: Please explain why absolute ice numbers are only sensitive to the total soot number while freezing particle fractions are only sensitive to soot particle size and solubility. At current soot emission levels, contrail ice formation is limited by the plume cooling rate. It would be interesting to know at which soot emission levels contrail ice formation will be limited by the availability of soot particles at emission for given ambient temperature.

First, there seems to be a misunderstanding. Since the apparent ice number emission index (AEI_{ice}) is the product of the freezing fraction and soot number emission index (EIs), absolute ice numbers are just as sensitive to average soot particle size and solubility like the freezing fraction (for fixed EIs). Our original concluding sentence "Absolute ice crystal numbers are, on the other hand, controlled by the soot number emission index for all ambient conditions" is simply based on the linear relationship between AEI_{ice} and EIs, which is actually trivial. Therefore, we have replaced our statement in Sect. 6 by

„[...]The freezing fraction displays a slight decrease with increasing soot number emission index, particularly for higher soot emission levels. This weakens the increase of absolute ice crystal numbers with increasing soot number emission index."

and accordingly, we have modified the wording in the abstract (lines 19-20).

Appendix A: How accurate is eq A2 at plume temperatures well in excess of ambient air temperature? How important are latent heat effects?

Thermal conductivity is only used in the diffusional growth equations and, therefore, is relevant at temperatures below 250 K when the first droplets form. Hence this physical quantity is not at all a crucial parameter in the temperature range you mention. Plume temperature changes due to latent heat have a magnitude of around 10^{-4} to 10^{-3} K per time step and, therefore, they are of low importance.

Appendix B: How is sigma in eq B1 calculated and how well is it known? See also comments above (l52ff and l165).

For the liquid phase, we assume the surface tension of pure water droplets. It is calculated according to Pruppacher & Klett (1997) using a temperature dependent fit formula based on experimental data from Hacker (1951). For the ice phase, we prescribe a fixed value (Pruppacher & Klett, 1997).

We have now documented the calculation of the surface tension in Appendix B.

References that did not appear in the manuscripts

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