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Comment on acp-2021-361

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Community comment on "Box model trajectory studies of contrail formation using a particle-based cloud microphysics scheme" by Andreas Bier et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-361-CC1, 2021

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].

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.

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.

I have a number of further points the authors may wish to clarify/re-assess/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].

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.

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?

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.

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

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

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?

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?

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.

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.

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

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.

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

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

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