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“Response of cirrus clouds to idealised perturbations from aviation”

submitted by Ella Gilbert et al.

This study quantifies the impact of ice crystal number concentration (ICNC) perturbations due to aviation activity on the water budget and microphysics of cirrus clouds. For this purpose, a large eddy simulation (LES) model was set up and initialized to follow the evolution of (perturbed) cirrus over six hours. Two cirrus types were studied at temperatures in the range 224-230 K that are subject to updraft speeds in the range 1-7 cm/s. Variations in imposed updraft speed caused a range of ICNC perturbations for each type of cloud. A sensitivity parameter was inferred as the ratio of simulated relative ice water path changes over ICNC changes.

Effects of cloudiness changes due to aviation have mostly been studied based on low resolution (global) climate models, in which associated microphysical processes (e.g., ice nucleation) and underlying dynamical forcing (e.g., gravity wave activity) are only crudely parameterized. The present study uses LES in an idealized framework aiming at better understanding aviation aerosol-related changes in cirrus properties. The manuscript is well-written and the analysis of the simulations is exemplary, however, I found aspects of the methodology and approach to be unclear or questionable, leaving a central claim of the study unsupported. These issues should be resolved before the manuscript can be accepted for publication in ACP.

General comments

The authors address in their introduction two possible mechanisms that may lead to cloudiness changes due to aviation that I briefly recall and comment on in more detail below:

> A. Aircraft emitted black carbon soot (BC) may impact the ice formation process in cirrus clouds. Numerous studies have shown that aviation BC particles are unlikely to nucleate ice heterogeneously in the upper troposphere (UT) and that their ice activity can be enhanced by processing in contrails. Even after such processing, they are poor ice nucleating particles (INPs).
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> B. A significant amount of flights in the UT lead to the formation of contrails. Besides forming in outside of clouds, contrails can also form within pre-existing cirrus clouds or natural cirrus forms around contrails. Contrail ice formation in aircraft exhaust plumes involves aircraft BC particle emissions and differs from aerosol-induced cirrus ice formation in the UT.

Both mechanisms are unrelated and perturb cirrus clouds in their own characteristic ways depending on whether a new cloud is formed or one already exists.

1. Attributing aviation activity to simulated cirrus perturbations.

The authors describe the main purpose of their study (lines 20-22): “Here, we use a large eddy simulation model to quantify the impact of ice crystal number concentration (ICNC) perturbations on the water budget and microphysics of pre-existing cirrus clouds. These perturbations aim to represent the second half of the chain of effects linking aircraft aerosol emissions to changes in ICNC and ice water path.”

Thus, the authors address mechanism A, but on the basis of a pre-existing cirrus cloud. This raises the question whether it is reasonable to assume that (line 555-556) "… cirrus are then perturbed by abruptly increasing their ICNC to mimic, in an idealised way, the effect of an aircraft emitted aerosol plume reaching an existing cirrus cloud."

While I understand that aviation perturbations are idealized in the model (which I do not cast into doubt in principle), I have strong reservations about the model perturbations representing real-world aviation activity. Perturbing "instantaneously across the whole model domain and throughout the entire profile" (line 184) is similar to what is done in coarser models in which clouds are not resolved. Instead, aerosol-cloud interactions occur locally and in particular, an aviation soot layer would perturb only a small atmospheric region (e.g., in the vertical) so that the association of the model perturbation with aviation activity (or any kind of aerosol-cloud interaction for that matter) appears to be unrealistic.

Cirrus perturbations as represented in the present LES model do not mimic mechanism A, because when an aircraft exhaust plume containing contrail-processed BC particles reaches already existing cirrus, it will not lead to a significant change in cloud properties. This is because pre-existing cloud ice will prevent, via rapid quenching of ice supersaturation, new ice nucleation. This holds true especially for aircraft BC particles that only nucleate ice at high supersaturation. New ice formation may occur only in regions where ICNCs stay below a few per liter of air for the updraft speeds considered in the manuscript.

Moreover, mechanism A cannot simply be replicated by "applying a multiplication factor to their distributions at the end of the spin-up phase" (line 183). A change in cirrus ICNC due to aviation soot-cirrus interaction involves additional changes in ice crystal size distributions and in the magnitudes of other cloud variables (e.g., ice water path) that do not simply scale with ICNC according to detailed simulations (Karcher et al., 2021). The latter study also suggests that the effect depends on where the aircraft BC particles are located in the vertical relative to ice supersaturation maximum and at which after emission the perturbation occurs.

The cirrus perturbations do not mimic mechanism B as well, because when an aircraft passes through a pre-existing cloud, it may generate a pencil-shaped (contrail) volume containing ice crystals which much higher number concentrations and much smaller sizes than present in the cirrus. The resulting bimodal ice crystal size distributions change microphysical processes locally. Such changes cannot be captured by scaling pre-existing cirrus ICNC instantaneously with a constant factor.

While I agree in principle that for the sake of conceptualization (line 114): "The aerosol-cloud interaction problem can be divided into two steps.", deliberately excluding the first half of the aerosol-ICNC-IWP chain in the present study prevents a proper treatment of the cirrus perturbation due to aircraft BC particles. Arguably, in order to treat mechanism A realistically, the aerosol impact has to be modeled explicitly. As implied on line 573, the study of Karcher et al. (2021) suggests that shortly after the aircraft BC perturbation, $\Delta\ln(\mathrm{IWP})>0$ and $\Delta\ln(\mathrm{ICNC})<0$, which invalidates the statement on line 576f: "The IWP response would then follow the pathway that corresponds to our simulation where ICNC is decreased." This means that realistically simulated aviation soot-related perturbations of cirrus cloud formation are not captured by linear $\Delta\ln(\mathrm{IWP})$-$\Delta\ln(\mathrm{ICNC})$ relationships, since the prescribed ICNC-perturbation of pre-existing cirrus can only yield $\Delta\ln(\mathrm{IWP})<0$ for $\Delta\ln(\mathrm{ICNC})<0$ (Fig.9 and Table 3).

In view of the statement (line 571-573): "Therefore, more work is required to take this analysis one step further and explore the first half of the chain, from aerosols to ice crystals, and whether representing that first half has an influence on the IWP sensitivities described here.", it might be a good idea to acknowledge that the "first half of the aerosol-ICNC-IWP chain" has been studied previously and that such work suggests it does matter for the second half. Otherwise, the notion is created that this is completely uncharted territory.

In summary, this challenges the fundamental assumption made in this study that the second half of the chain of effects linking aircraft aerosol emissions to cirrus changes can be treated independently of the first half of the chain and thus raises serious doubts as to the validity of the final statement in the abstract: “These results suggest that aviation has the potential to increase the lifetime and radiative effects of pre-existing cirrus clouds.”

2. Model set-up, initialization and cirrus classification.

Both selected cases have some observational meteorological background, but appear to be too low in altitude and too warm to represent average conditions at (midlatitude) cruise altitudes. One case (Spichtinger et al., 2005) relates to ice supersaturation between 320-408 hPa, where aircraft don’t even cruise. Yang et al. (2012) is not in the reference list so I cannot comment further on it, but what is written about this case study on lines 146-147 seems counterintuitive: a lee wave cirrus should have encountered rather strong lifting and should initially contain many ice crystals with ICNCs at the upper end of observed values.

According to long-term in-situ measurements in the extratropical northern hemisphere, temperatures at cruise altitudes are in the average range 214-224 K; values >226 K are rare even at the lowest flight levels (down to 270 hPa). It is unclear how the choice of meteorological case studies affects the representativeness of the results presented in the study. At any rate, the synoptic analyses underlying both cases may not capture the full gravity wave (GW) impact, especially the high frequency components in the vertical wind field that are crucially important for ice nucleation in cirrus are likely missed.

Regardless of the meteorological set-up, the distinction between cirrus types (line 99): “Cirrus can further be classified according to vertical velocity as either slow (<10 cm/s) or fast (>10 cm/s) updraft cirrus” makes little sense to me. This is because the lifetimes of the simulated cirrus (6 h, Fig. 8) or even the times past cirrus formation after which sensitivity parameters are evaluated (30-45 min, Fig. 9) well exceed the time intervals over which updraft speeds generated by high frequency GWs are coherent, typically a fraction of a buoyancy period (Podglajen et al., 2016). During and after formation, cirrus clouds are subject to a very wide range of rapidly fluctuating vertical wind speeds superimposed on much slower variability.

Thus, the W=10 cm/s updraft speed separation appears to be an arbitrary choice, and it remains unclear why it is introduced in the first place. Updraft speed standard deviations due to GW lie in the range 10-20 cm/s. The values of W listed in Table 2 are therefore not representative for GW activity. These may rather represent a slowly variable (background) updraft onto which GW variability is to be superimposed. According to in-situ measurements, mesoscale vertical wind variability is the primary cause of broad ICNC distributions within cirrus clouds.

The method of creating pre-existing cirrus (lines 223-224) during the relaxation of the model spin up phase is obviously a numerical experiment that does not guarantee UT ice formation processes to be reliably replicated, due mainly to the lack of realistic dynamical forcing as noted above. Since ICNC distributions of the resulting cirrus are key to the present study, it would be helpful if the authors could produce from their Fig.3 probability distributions of ICNC (perhaps during the stable phase) that may be compared with field data of midlatitude cirrus representative for cruise conditions (e.g., Karcher and Strom, 2003; Hoyle et al., 2005; Jensen et al., 2013).

In that regard, (lines 141-142): “We emphasise that this method of initialising the model cloud is not necessarily reflective of the actual ice nucleation mechanisms of the two cirrus cases studied.” introduces an inconsistency that may be consequential for the simulated cloud evolution that remains to be explored.

All in all, this challenges the representativeness of the selected cirrus cloud cases for deriving the sensitivity parameter shown in Fig.9, again questioning the validity of the statement (lines 28-29): “These results suggest that aviation has the potential to increase the lifetime and radiative effects of pre-existing cirrus clouds.”

Other comments

i. “Parameterisations of updraft velocity are especially important in the kind of large-scale models that are typically used to make global estimates of the radiative effect …” (lines 68ff). I agree, but such parameterizations would also be needed in the LES framework. Please refer to general comment 2.

ii. “… whereas those assuming soot is an inefficient INP show much smaller effects (Gettelman & Chen, 2013; Kärcher et al., 2021)”. The assumption holds for the climate model. By contrast, the latter study did not just “assume” soot to be an inefficient INP, but based the microphysical simulations on a physically-based parameterization of the nucleation mechanism constrained by laboratory data. They did find a non-negligible reduction of nucleated ice crystal number concentrations due to the ability of aviation soot to weaken homogeneous freezing events. Please clarify.

iii. The introduction mentions mechanism B on lines 104-112. This confused me and only after a second reading I understood that this study does not deal with contrail formation (within cirrus). Is it necessary to elaborate on mechanism B? If so, it should be clearly distinguished from A to avoid confusion.

iv. “The perturbations induced are intended to be plausible.” (line 119). I don’t think so. Please refer to general comment 1 above.

v. Please state explicitly why / how the domain experiences an updraft (see W-entries in Table 2).

vi. How realistic is a water vapour forcing applied over 3 km depth in a subsidence region (lines 156-159)? Why would an otherwise dissipating cirrus be representative for aviation perturbation estimates?

vii. The authors chose a larger vertical than horizontal resolution (line 131). I wonder whether this choice is compatible with e SGS turbulence scheme in the LES model and which effect the rather coarse vertical resolution of 120 m has on the treatment of sedimentation?

viii. On which grounds do you decide that the “ICNC×0.5 and ×2 experiments represent large but still-plausible perturbations” (line 190)? Please refer to my comment on mechanism B above (general comment 1).

ix. “Homogenous nucleation rates are relatively higher at the cloud base, whereas heterogenous nucleation is comparatively higher at cloud top.” on lines 229-230 sounds counterintuitive. Does the nucleation parameterization scheme include competition between different nucleation modes for available water vapor during ice formation?

x. The statement on lines 575-576: “Kärcher et al. (2021) suggest using 1D LES modelling that the first half of the chain from aerosol to ICNC perturbation is not straightforward. They find that adding soot aerosol into their simulations actually forms clouds with fewer, larger particles because if soot aerosols act as efficient INP they suppress homogeneous freezing.” does not reflect results of that study, namely that aircraft BC particles are poor INPs only forming ice alongside homogeneous freezing of supercooled solution droplets, even under conditions that maximize their impact. Please correct.

Overall recommendation for revision

Besides considering and clarifying the above issues, I recommend the authors remove the inference (line 29): “These results suggest that aviation has the potential to increase the lifetime and radiative effects of pre-existing cirrus clouds.” I strongly suggest to remove (not weaken), since the way cirrus perturbations are treated have no link to aviation activity whatsoever that could be justified on a physical basis, plus the perturbed cirrus properties and dynamical regime may not be capturing cirrus types and small-scale meteorological conditions prevalent at cruise altitudes.

Importantly, the above criticism extends to the Short Summary, stating: “We show that the main effect of our experiments – which intend to mimic the effect of aircraft soot emissions reaching existing high-altitude cirrus clouds – is to extend cloud lifetime, thereby enhancing their effect on climate.” This far-reaching statement cannot be maintained and I strongly recommend to remove it as well.

The perturbation of a pre-existing cirrus cloud, in which cloud properties are abruptly changed over the whole cloud domain does not harvest the full potential of what the high-resolution LES framework employed in this study may actually accomplish. It is necessary to explicitly discuss issues with relating the idealized perturbation to aviation activity. Even an idealized treatment should be reasonably realistic. What would be its real-world equivalent?

Concerning the discussion of sources of uncertainties in the introduction, it may be worth pointing out that there is convergence in experimental studies (both lab and ground-based measurements) showing that aircraft BC particles are poor INPs when their small size (< 50-100 nm) is considered. This stems mainly from morphological considerations (BC particle nanopore geometry after cloud processing) which should apply and thus allow extrapolation to UT conditions. None of the quoted climate model studies represents the competition between aircraft BC particles and liquid solution droplets based on the latest information from the lab, let alone includes the competition with other, more potent INPs such as mineral dust, see Karcher et al. (2023).

Besides covering the many meteorological conditions in which various cirrus types form and evolve (lines 580-582), a major future challenge is the explicit treatment or parameterization of aerosol-induced cirrus ice formation in a limited-area LES framework along with realistic gravity wave forcing. Maybe this is worth pointing out in the conclusions.

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