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23rd Dec 20

Dear Professor Kärcher,

Your manuscript titled "Aircraft soot-cirrus interactions do not matter for the climate impact of aviation" has now been seen by two reviewers, and I include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Specifically, please either tone down your main conclusions (and title) to match the strength of the evidence (taking into account the reviewers criticisms), or provide substantial additional support for your claims that aircraft soot is not important for the climate impact of aviation.

Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter) and the completed checklist:

[link redacted]

** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Heike Langenberg, PhD

Chief Editor

Communications Earth and Environment

On Twitter: @CommsEarth

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Please see my comments in the attachment.

Reviewer #3 (Remarks to the Author):

The authors seek to quantify the possible effect that aircraft soot emissions might have on cirrus formation after any initial contrail has dissipated. They do so using a sophisticated column model which incorporates an impressive state-of-the-science soot aggregate model. They investigate multiple possible situations, including the role of aircraft soot in near-term “re-formation” of a contrail, its role in longer-term formation of irregular contrail cirrus, and the anticipated effect of exhaust-like soot in base cirrus formation. They conclude that, excluding the initial effect in contrail formation, aircraft soot has a negligible effect on cirrus formation.

This work is timely and important. The question being asked is a matter of significant debate, and the methods used by the authors are novel, offering a fresh perspective on a problem which is most often addressed using global climate models (with conflicting results). The approach they have taken is mostly appropriate, but I have one serious concern regarding the scope of the study. Specifically, it was not clear to me after several readings whether the study could truly be said to capture the full scope of aviation’s impacts on cirrus formation. In particular, it seems that the possible long-term impacts of aviation soot emissions on cirrus, through the accumulation of aged combustor soot at high altitudes, may not be fully considered.

I believe that this study is of great interest to the audience of Communications Earth and Environment. It is clearly written and rigorous. However, given the importance of the question and the boldness of the claim in the title, I recommend against publication until the comments below are addressed.

Major comments

The conclusion drawn by the paper, as given in the title, is that aircraft soot-cirrus interactions do not matter. However, the paper appears to only consider the role of aircraft emissions within ~12 hours after emission, and it was not clear to me why this was the case. Given that aircraft fly in the upper troposphere and lower stratosphere, the lifetime of emitted soot is expected to significantly exceed this, with a mean lifetime on the order of a month (see e.g. Zhou and Penner 2014). On this timescale, it would be expected that soot emitted from aircraft could age significantly while also diluting, and potentially contribute significantly to formation of cirrus (presumably taking part in the “Base” scenario). However, this paper does not appear to address this possibility. The three scenarios consider either the “re-formation” of a contrail around 30 minutes (“Plume”), the formation of an irregular contrail after around 5 hours (“Background”), or formation of cirrus in air which has not been influenced by aircraft exhaust (“Base”). However, none of these scenarios appear to address a case where aged aircraft soot could take part in cirrus formation, as all three treat the soot which is present as being “unaged” (see comments below). To truly support the title of the paper, the effects of aged aircraft soot must also be discounted. If aged aircraft combustor soot is a highly efficient ice nucleus, then its role in the “Base” case has not been accounted for. I would suggest that the authors either 1) weaken the strength of their conclusions, including the title, to make it clear that their work only represents unaged soot; or they 2) perform additional sensitivity simulations to quantify the role that aged aircraft soot may have in cirrus formation, accounting for the long aging times which might be relevant (see below for more details) and the plausible range of fractional contributions that aircraft emissions might have to “baseline” soot concentrations.

A key conclusion of the paper is that many current models are overpredicting the role of aviation soot in the formation of cirrus clouds. This conclusion is based on the use of a sophisticated model, which accounts for the specific morphology of fresh and aged soot. However, I was wondering about a detail provided on lines 492-497. The authors use a contact angle of 60 degrees, consistent with Persiantseva et al (2004) which studied aircraft combustors specifically. It is stated that, to decrease the contact angle (and therefore increase the hydrophilicity of the soot), “exposure to atmospherically relevant ozone concentrations over several days is required”. It is then implied that such exposure is not relevant here, but – unless the authors reduce the scope of their paper as discussed in my previous comment - that assertion does not appear to be properly supported. The mean lifetime of aircraft soot (see previous comment) is likely to be greater than the lifetime assumed by Wei et al (2017) of “< 10 days”, which is the basis for the assumption given here. That article appears to be focused on emissions in near-surface conditions, and also states that “atmospherically relevant” concentrations of ozone would be on the order of 40 ppbv, while exposure to sulfuric acid and nitric acid would both also act to decrease the contact angle. This seems highly relevant, given that aircraft exhaust is rich in sulfur acid, while stratospheric air can contain mixing ratios of ozone which are orders of magnitude greater than 40 ppbv (in addition to significant quantities of H₂SO₄ and HNO₃). If the authors decide to retain the current scope implied by the title, I recommend that they consider performing an additional sensitivity simulation in which they further decrease the contact angle (based on a reasonable estimate of the possible effect of such aging), to simulate the potential effects of long-term aging.

Similarly, the claim that freshly emitted soot contact angles are in the range 60-110 degrees (line 492) seems misleading. Of the two studies cited (Persiantseva et al, 2004; Wei et al, 2017), only Persiantseva et al studied aircraft combustors, and they found contact angles in this situation of 63

degrees. Wei et al (2017) instead evaluated a kerosene lantern, fireplace, and bus exhaust to find the larger range of 65-110 degrees. The statement that 60 degrees corresponds to an upper limit of hydrophilicity for unaged aircraft combustor soot implies that there is data supporting aircraft exhaust having a significantly higher contact angle. If so, it should be cited here, as the two given references seem to not support it. If such evidence is not available, I would recommend removing the claim that this is an upper bound.

Minor comments

I have no minor comments. However, I would like to commend the authors on the clarity of Figure 1.

References

Persiantseva, N. M., Popovicheva, O. B. and Shonija, N. K.: Wetting and hydration of insoluble soot particles in the upper troposphere, *J. Environ. Monit.*, 6(12), 939–945, 2004.

Wei, Y., Zhang, Q., Thompson, J. E. and Others: The wetting behavior of fresh and aged soot studied through contact angle measurements, *Atmospheric and Climate Sciences*, 7(01), 11, 2017.

Zhou, C. and Penner, J. E.: Aircraft soot indirect effect on large-scale cirrus clouds: Is the indirect forcing by aircraft soot positive or negative?, *J. Geophys. Res. D: Atmos.*, 119(19), 11,303–11,320, 2014.

This study tried to suggest that there is a small impact of aircraft soot on cirrus clouds and climate. A 1-dimensional cirrus cloud model was set up to simulate the competition between heterogeneous nucleation of soot and homogeneous nucleation of soluble particles.

Based on their abstract and discussion, they attributed the small effect of aircraft soot on cirrus clouds to the small fraction ($<1\%$) of ice-active soot particles and a much smaller number concentration of aircraft soot than background soot and dust number concentration in the upper troposphere. One issue is their critique of Ref. 11. However, the model used in Ref. 11, 31, which authors commented on, only had 0.6% of aircraft soot as INP and still obtained a radiative effect of aircraft soot on cirrus clouds. So the small fraction of ice-active soot are not likely the key to explain the little effect from authors' model.

The authors set up their study using a conceptual framework (Figure 1). However, my understanding has always been that "contrail cirrus" are cirrus clouds that form from spreading contrails. Not contrails that have evaporated and then the soot from these evaporated particles forms new cirrus. Please change your description to align with this.

In the authors' model study, a single constant updraft was used to drive cirrus formation in all scenarios. This is not appropriate. Penner et al., JGR (2018) has

shown the importance to consider the range of updraft velocities needed to calculate the impact of gravity waves on cirrus clouds formation. The updraught velocity, background aerosol number concentration, temperature, and water vapor content all influence the competition between heterogeneous freezing and homogeneous freezing as well as the ability of aircraft soot to form ice crystals. I suggest to add more sensitivity experiments with different background conditions to figure out the possible effect of aircraft soot on cirrus clouds, to address the conclusion that there is an insignificant effect through the experiments with single parameter.

Their model experiment studied on the cirrus clouds with very small COD (<0.06). Are these optical depth changes (0.01 in Fig 2) limited because of the vertical depth of the clouds they consider? If so, they would only consider the near-field clouds (i.e. very close to aircraft emission) perturbations to cirrus. They therefore cannot apply their results to large-scale cirrus perturbed by soot, which global climate model always focused on.

Moreover, most of the simulation results show a significant effect of aircraft soot on the decrease in the ice crystal number concentration due to their influence on the homogeneous freezing. After those results, they noted all the assumption with aircraft soot in above simulations are unrealistic (from Line 175) and commented on the previous globe model results (from Line 199). They regard these unrealistic

assumptions as the reason for the small effect of aircraft soot on cirrus clouds. I wonder why they did not simulate using more realistic conditions they mention in their conclusion instead of so many unrealistic experiments.

I suggest authors to pay more attention to prove and discuss those assumptions used in the model are scientific and realistic. The model results should very largely depend on the assumption of the property of aircraft soot as well as background aerosol and meteorological conditions. I suggest to review studies on the property of aircraft soot (such as size distribution, Petzold et al., 1998, 1999) and discuss the uncertainty in the assumptions used in their model. The contrail process should have influence on the ability of soot acting as INP. I suggest authors to discuss more on the impact of contrail process on aircraft soot, which probably determines the effect on cirrus clouds.

This paper is novel, but there are major revisions needed to improve this paper. There are also following specific comments needed to address as well.

1. Why did you use short-wave optical depth as a measure of RF? As we know, cirrus clouds contribute little to the change in solar short-wave, but contribute to the change in radiation balance through absorption of long-wave radiation.
2. L47: Reference 8,9 suggest a big impact of aircraft soot on cirrus clouds. Why do you completely disregard references 8, 9 which did show an effect of

aircraft soot on ice (though only "correlations ").

3. L50: This statement is incorrect. Ref. 11 showed the range of forcing depends on the number of aerosol particles that may be homogeneous vs heterogeneous in the background atmosphere.
4. L54: The positive numbers quoted in ref. 3 are due to lower values of sulfate aerosols and dust aerosols, together with a small portion of aircraft soot as INP. The positive values quoted in ref. 3 result from putting limitations on the sulfate particle numbers as well as dust numbers (Ref. 13), thus they are "tuned" to give positive results and unlikely to be correct based on observations as in ref. 8, 9. The best you can say here is "may remain uncertain if you can accept small sulfate numbers (which are not supported by observations)." The gaps in knowledge are mainly from not knowing whether contrail processed soot can be an INP. However, Mahrt et al. (2020, Ref. 19) limit to only larger soot.
5. L59: The experiment in Ref. 16 start at -15 C so also include mixed phase clouds, not only cirrus.
6. L112: What effect did you mean here? It is not clear.
7. L131: I did not find any ice crystal forms from 6min in your Fig.2. If ice forms from 6min, I suggest to extent the time axis to begin at 0min instead of 20min.
8. L137: Are these deposition coeff. for soot? which case uses 0.5 and which uses 1.0?
9. L139: Please explain why turbulent diffusion lowers homogeneous ICNCs?

10. L133: Please show the quantitative evaluation of how long the homogeneous freezing is delayed because of Background and Plume scenarios.
11. L139: Please add the variation of ice supersaturation in each scenario in Fig2.
12. Fig.2: Why does the LWP increase by >50% due to the soot in included Background and Plume simulations?
13. L153: What did you mean by the effect of the onset of homogeneous freezing? Extended Fig.6 showed the result taken before homogeneous freezing. Why is the upper limit estimate of 11% higher than the highest ICNC in Extended Fig.6 for the simulation Background, but 145% higher for the simulation Plume? Can you add some explanation to this difference?
14. Line 161: I'm confused about why you did an additional simulation with updraught speed of 0.01 m/s but still dominated by homogeneous nucleation? I suggest to add some further discussion in this sensitivity experiment if you still think this is necessary for your conclusion.
15. L197: Field studies can see conditions dominated by surface sources, so it is unclear that "frequently" is correct here.
16. Line 203: In the global model study, only 0.6% of aircraft soot were INPs in Refs. 11, 13, due mainly to the deposition of sulfate ending their ability to act as INP, which agrees with the small fraction (<1%) assumed in your study. You are misquoting here.
17. Line 205: Please add the correct reference for the positive RF result here and clarify that this is a sensitivity test to examine the effect of a large increase in

INPs. This was not in ref 11, but from a sensitivity test in Ref.31.

18. Line 212: Please add: "and changes in background emissions".

19. Line 215: Please add: "due to the use of much smaller background sulfate and dust concentrations, as shown in Ref. 11".

20. Line 262: please add a consideration of the larger mode soot particles seen in Petzold et al, Atmospheric Research, 1998 and Petzold, JGR 1999.

21. Line 475: This does not say it is cumulative as stated below. please clarify.

22. Line 542: please give the parameterization in detail.

23. Fig. 1: Do you mean contrail cirrus form after dissipation? Please clarify. And what are the upper inset and lower inset?

24. Extended Data Fig. 3: I see a purple and pink curve. the use of violet and magenta is not distinguishing.

COMMSENV-20-0524

Dear editor:

We appreciate the opportunity to revise and resubmit our manuscript based on a set of constructive and thorough reviews. Below we attach a point-by-point response to all concerns raised by the reviewers. Our replies are *italicized* and the line numbers (Lnnn) and reference numbers are those from the original manuscript.

Bernd Kärcher on behalf of all co-authors

Response to reviewers**Reviewer #1**

This study tried to suggest that there is a small impact of aircraft soot on cirrus clouds and climate. A 1-dimensional cirrus cloud model was set up to simulate the competition between heterogeneous nucleation of soot and homogeneous nucleation of soluble particles.

We thank the reviewer for carefully assessing our manuscript and appreciate the detailed comments and suggestions. Below we indicate where and how we altered or extended the manuscript based on the reviewer's feedback.

Based on their abstract and discussion, they attributed the small effect of aircraft soot on cirrus clouds to the small fraction (<1%) of ice-active soot particles and a much smaller number concentration of aircraft soot than background soot and dust number concentration in the upper troposphere. One issue is their critique of Ref. 11. However, the model used in Ref. 11, 31, which authors commented on, only had 0.6% of aircraft soot as INP and still obtained a radiative effect of aircraft soot on cirrus clouds. So the small fraction of ice-active soot are not likely the key to explain the little effect from authors' model.

We believe that there is a confusion between the aircraft-emitted soot particles that are actually ice-active via the soot-PCF mechanism on the one hand and the total insoluble (soot and dust) particle number concentration on the other. The 1% AF corresponds to the fraction of soot particles that is ice-active at a supersaturation of $s = 0.5$ at 220 K, for a soot-liquid water contact angle of 60° and a PSD containing many particles at large sizes (L508-511). This translates into ice-active aviation soot number concentrations with relatively high (54/L) and low (5/L) INP concentrations in the simulations Plume and Background, respectively, compared to background upper tropospheric INP concentrations that typically amount to some 10/L (L93f). However, please note that the soot particle number concentrations of 5,448/L and 458/L used in these simulations (Extended Data Table 1) are still higher than the total number concentrations of mineral dust particles throughout the northern hemisphere, as for instance simulated in by Zhou & Penner in ref.11 (see their Fig. 2b).

We clarify this point by including the following modifications:

L95: "...derive soot particle number concentration of 5,448/L and 458/L (Extended Data Table 1) and associated upper limits ..."

L107: "... size, on the premise that real aviation soot particles have properties that are similar to the laboratory soot surrogates".

Please note that the 0.6% of the total aircraft soot assumed in refs.11,31 and the <1% of ice-active aircraft soot particles denote two distinctive quantities. Ref.11 states (in their section 2.3) that “... preactivated aircraft soot only accounts for 0.6% of the total aircraft soot in air”. This percentage refers to contrail-processed aviation soot particles **irrespective of their size** and with less than three monolayers of sulfate at their surfaces. Our study shows that at most 1% of **those** soot particles (pre-activated with less than three monolayers of sulfate) are actually able to act as INP even at high ice supersaturation (≈ 0.5). The global model in ref.11 further assumes that aviation soot activates completely already at moderate ($s \approx 0.2$ - 0.35) ice supersaturation (L200ff, L231). The assumption of complete activation at ice nucleation onsets (thresholds) as reported in laboratory studies (refs.14,15,47) overestimates ice active soot fractions, since only a minority of particles activate at the threshold value and most require higher ice supersaturation, even more so when their size is smaller than the investigated one (see e.g. Fig.2 in ref.14). Thus, the fact that ref.11 obtains a large radiative effect is primarily caused by an overestimation of the number of soot particles that are actually ice-active after contrail processing and coating with sulfate.

Thus, we believe that the reviewer’s statement that “the small fraction of ice-active soot are not likely the key to explain the little effect from authors’ model” is based on a misconception that we hope to have clarified now.

The authors set up their study using a conceptual framework (Figure 1). However, my understanding has always been that "contrail cirrus" are cirrus clouds that form from spreading contrails. Not contrails that have evaporated and then the soot from these evaporated particles forms new cirrus. Please change your description to align with this.

The reviewer is correct that contrail cirrus clouds evolve directly from spreading contrails (L29). While contrails are line-shaped, contrail cirrus are by definition irregularly-shaped and therefore much harder to observe and/or distinguish from natural cirrus. The likelihood of contrails losing their linear shape increases with time past formation depending on wind shear and relative humidity fluctuations and turbulence. Hence, if a contrail dissipates and reforms early as in scenario Plume (or late as in Background), it is more (less) likely to be line-shaped.

Please note that we used the terms contrails and contrail cirrus in an attempt to distinguish possible morphologies of the soot-perturbed ice clouds. This shape distinction is not fundamentally relevant for the results of our study, as we make no assumption about cloud morphology in the simulations. However, it may be relevant in attempts to observe the soot effect, which motivated us to include it in our conceptual framework.

Fig.1 (caption): We clarify: “... modulating ice formation in a new line-shaped cirrus cloud similar to a contrail (scenario Plume) or an irregularly-shaped cirrus cloud similar to contrail cirrus (Background).” and no longer refer to the cloud shape in the main text to avoid confusion.

In the authors’ model study, a single constant updraft was used to drive cirrus formation in all scenarios. This is not appropriate. Penner et al., JGR (2018) has shown the importance to consider the range of updraft velocities needed to calculate the impact of gravity waves on cirrus clouds formation. [...]

The reviewer is correct in pointing out the importance of dynamical forcing (updraught speeds for this matter) in cirrus formation and consequently in our simulations (cf. L384f). The single updraught speed of 15 cm/s used here is representative of widespread mesoscale gravity wave activity (L103). Regarding this choice, we stated in L167ff that “mean updraught speeds <0.15 m/s are not observed in the upper troposphere [...]. Higher mean updraught speeds strongly enhance the homogeneous freezing contribution.”

We intentionally did not consider wave-induced variability in updraught speeds, because this would broaden cirrus ice crystal number concentrations massively and make it difficult to isolate the soot effect (L171ff). Including updraught speed variability would lead to predictions with smaller or negligible perturbations (L184f), as homogeneous freezing events are triggered much more frequently. We performed a sensitivity study with a low updraught speed to demonstrate that cirrus forming at lower updraught speeds are more strongly influenced by aviation soot, but are shorter lived and optically thinner (hence contribute less to RF). A sensitivity simulation with higher updraught speeds was omitted, as higher vertical velocities lead to optically thicker, potentially longer-lived cirrus reducing any INP impact.

L171: We modify: “... contribution, leading to optically thicker cirrus clouds and reducing the impact of ice-active soot particles.

L173: We modify: „... freezing²⁷. Because this broadening would make it difficult to separate the effect of aviation soot from wave-induced variability in ICNC, we did not represent such temporal variations in our simulations. Taken together, using a higher mean updraught speed or including updraught speed variability would lead to predictions with smaller or negligible perturbations.”

Our work presents, to the best of our knowledge, the first comprehensive aerosol-cirrus interaction study that allies a detailed, spatially-resolved cloud model with a predictive model for ice activation derived from measurements. In this vein, although viewed as a simplification, the use of a single updraught speed was a deliberate choice to facilitate the analysis of the soot-PCF impact on cirrus formation (L102ff) and to assess the uppermost limit of the soot impact (L175ff).

[...] The updraught velocity, background aerosol number concentration, temperature, and water vapor content all influence the competition between heterogeneous freezing and homogeneous freezing as well as the ability of aircraft soot to form ice crystals. I suggest to add more sensitivity experiments with different background conditions to figure out the possible effect of aircraft soot on cirrus clouds, to address the conclusion that there is an insignificant effect through the experiments with single parameter.

We chose meteorological conditions representing a typical midlatitude situation with regard to air traffic volume and contrail occurrence (L398ff). Regarding the choice of values for meteorological variables other than updraught speeds (addressed above), this would basically boil down to studying less abundant high-moisture tropical and low-temperature polar contrails in areas with (currently) much lower air traffic volume. Regarding the choice of soot-PCF parameters, considering the discussion of results shown in Extended Data Figs.1+2, we used conditions ensuring high soot-PCF efficiency (L178). Regarding the choice of the particle size distribution for aviation soot emitted by jet engines at cruise, see our response below.

Instead of attempting to cover a wide range of situations in which aviation soot influences cirrus ice formation, our study provides multiple lines of evidence, demonstrating the small magnitude of the effect in typical conditions and showing that the aviation soot impact is significant only for very low updraught speeds which, when generated by mesoscale gravity waves, would only last for a few minutes (L169 and ref.25). Therefore, we believe that more sensitivity studies would not allow us to obtain important new insights or alter our conclusions.

L251: We add: „... interactions, although atmospheric conditions and soot particle properties are more variable and complex than representable in cloud models.”

L403: We add “... Atlantic³⁷, representing a typical midlatitude situation with regard to air traffic volume and contrail occurrence.”

Their model experiment studied on the cirrus clouds with very small COD (<0.06). Are these optical depth changes (0.01 in Fig 2) limited because of the vertical depth of the clouds they consider? If so,

they would only consider the near-field clouds (i.e. very close to aircraft emission) perturbations to cirrus. They therefore cannot apply their results to large-scale cirrus perturbed by soot, which global climate model always focused on.

Please note that all scenarios shown in Fig.2 develop column optical depths around 0.3. According to the ISCCP (International Satellite Cloud Climatology Project) cloud classification scheme (<https://isccp.giss.nasa.gov/cloudtypes.html>), this corresponds to a wide range of cirrus unrelated to deep convection. The optical depth changes result solely from introducing aviation soot INPs that are PCF-active. The moisture and temperature profiles are constrained by in situ measurements and do not limit the vertical depth of the developing cloud. Please recall that we simulate the cirrus formation stage, where optical depth values are evolving.

The only scenario in which the optical depth stays below 0.06 is the background scenario in the sensitivity experiment with a low updraught speed, in which soot suppresses the first (major) homogeneous freezing event but cannot prevent homogeneous freezing from occurring at the cloud top (L164 ff and Extended Data Fig.7). If aviation soot were the only ice-forming particles, it would be hard to imagine how the resulting large-scale cirrus could ever develop significant (e.g. > 0.1) column optical depth.

The soot number concentrations implicated with soot-cirrus interactions in the global model amount to 10-100/L (ref.11. their Fig.1c). These are annually and zonally averaged values at 200-400 hPa. We recall that these values correspond to a fraction of 0.6% of all aviation soot emissions when taking contrail processing and some level of sulphate scavenging into account, but are not constrained by size and ice supersaturation. Further accounting for the fact that at most 1% of those are ice-active, this range translates into 0.1-1/L. In comparison to the latter range, the total ice-active aviation soot particle number concentration is 5/L in our scenario Background; this represents a local value, not a value taken at the scale of a global model grid box.

Moreover, most of the simulation results show a significant effect of aircraft soot on the decrease in the ice crystal number concentration due to their influence on the homogeneous freezing. After those results, they noted all the assumption with aircraft soot in above simulations are unrealistic (from Line 175) and commented on the previous globe model results (from Line 199). They regard these unrealistic assumptions as the reason for the small effect of aircraft soot on cirrus clouds. I wonder why they did not simulate using more realistic conditions they mention in their conclusion instead of so many unrealistic experiments.

Our experiments are not unrealistic (we admit this term has been unfortunately chosen), but lead to upper limit estimates of the soot impact in our perturbation scenarios (L175ff). These upper limit conditions indeed decrease ice crystal number concentrations, but have no significant effect on optical depth. From this, we conclude that if upper limit conditions do not influence optical depth significantly, this is even truer for average or, as the reviewer states, more realistic conditions. With this approach, we concentrate on a few cases and avoid the calculation of many different scenarios. Our high-resolution cloud simulations with in-depth analyses of carefully selected scenarios enhance the understanding of aircraft soot-cirrus interactions at the process level, differing from the global modelling approach that aims to yield a best estimate of the aviation contribution to anthropogenic radiative forcing.

As for our suggestions in the conclusion, those apply to (future) simulations of aerosol-cirrus interactions based on observations (L235ff). In such cases, the full range of wave effects might need to be included to enable a meaningful model-data comparison.

I suggest authors to pay more attention to prove and discuss those assumptions used in the model are scientific and realistic. The model results should very largely depend on the assumption of the

property of aircraft soot as well as background aerosol and meteorological conditions. I suggest to review studies on the property of aircraft soot (such as size distribution, Petzold et al., 1998, 1999) and discuss the uncertainty in the assumptions used in their model. The contrail process should have influence on the ability of soot acting as INP. I suggest authors to discuss more on the impact of contrail process on aircraft soot, which probably determines the effect on cirrus clouds.

The reviewer is correct in pointing out that our model results depend on assumptions regarding the soot PSD, the background aerosol and meteorological conditions. We addressed the latter above responding to an earlier comment.

With regard to assumptions concerning the background aerosol, we eliminated the possibility of heterogeneous ice nucleation by INP other than aviation soot in order to maximise the soot impact. For instance, including early freezing dust (or ammonium sulphate, glassy, ...) particles in our simulations would reduce the rate of supersaturation increase and, if abundant enough, even halt the increase. This means that virtually none of the contrail-processed aviation soot particles would be able to form ice (L190f, Extended Data Fig.1). Hence, to single out the effect of aviation soot-induced ice formation, we deliberately omitted perturbations by other INPs and only let soot compete with homogeneous aerosol freezing.

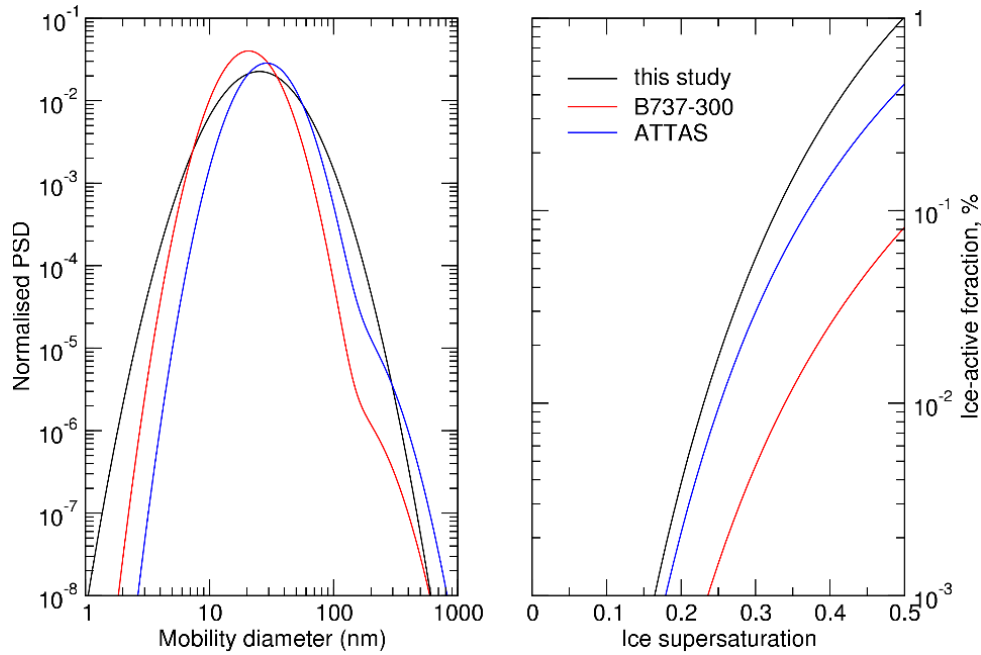
With regard to properties of soot PSDs used in our simulations, we devoted a section in the Methods to describe our parameter choices and motivate why we believe the default modal mobility diameter (35.7 nm) and g.s.d (1.8) represent upper limit estimates. We understand that this motivation needs to be expanded further.

We therefore appreciate the reviewer's suggestion to consider the 1999 study from Petzold et al., who measured bimodal soot PSDs behind various aircraft at cruise, and believe that the reviewer's comment addresses the bimodality of the PSD. We already included Petzold et al. (1998) as ref.41, because their measurements were taken in nascent contrails. Below, we analyse Petzold et al. (1999, JGR Vol.104) results using their bimodal PSDs of unprocessed soot particles (no contrail formation, plume age <1 s) taken behind a modern, large B737 and an older, smaller (hence, less representative) ATTAS aircraft. The authors conveniently provide fitted log-normal parameters for both modes and both aircraft; importantly, the ratios of total particle number concentration in the large versus the small mode stay below 3.3×10^{-3} . We point out that the authors assume that 'the larger mode is made of agglomerated particles produced by coagulation of [smaller, primary particles]' and proceed using their PSD parameters, although they caution that 'there is a potential for some unquantified bias in the size distribution measurements' (Petzold et al. (1999, p.22,174)).

We compare ice activation spectra of aviation soot calculated from soot-PCF with two sets of parameters: (i) our default setting (L421f), i.e. single log-normal mode; (ii) double mode PSDs following Petzold et al. (1999). The ice activation spectra of aviation soot in case (i) can be read off Extended Data Fig.1d as a function of ice supersaturation (s) in the limit of large mobility diameters (D); here, we calculate them directly:

$$f(s) = \int_0^{\infty} \text{PSD}(D) \text{AF}(D, s) dD$$

with size distribution, PSD, and ice-active fraction, AF (Extended Data Figs.1a,b,c). The figure below shows f alongside the underlying PSDs (for ease of comparison, normalised to unity). We confirm that the ice-active fraction in our study, which reaches a maximum of 1% at $s = 0.5$, indeed constitutes an upper limit assumption since the bimodal PSDs result in fewer ice-active particles across all supersaturations. This outcome can be rationalised considering that the abundance of particles with sizes 50-200 nm, which matter most for soot-PCF, is largest in the monomodal PSD. In particular, less than 0.1% in the B737-case soot particles are ice-active; this implies $<5/L$ ($<0.5/L$) ice-active soot particles in scenarios Plume (Background), rendering their impact on cirrus formation much smaller (negligible) compared to (i).



We could have inferred bimodal PSD parameters from ref.41 as well; given experimental uncertainty, the difference is not crucial for our argument laid out above. However, the fact that bimodal PSDs were observed by Petzold et al. (1999) in dry plumes (i.e. no contrail processing could have occurred) raises the question of the origin of the large soot mode in Petzold et al. (1998). This underscores that further experimental research is needed to resolve this issue. Thus, we recommended that “airborne measurements should characterise soot PSDs and mixing state before and after contrail processing” (L242), as little is known about how aviation soot PSDs change after compaction in sublimating ice crystals.

We hope that this extended discussion clarifies that we maximised the ice activity of aviation soot by using a monomodal PSD with large mean size and size spread, thus following our aim to establish an upper limit estimate for aviation soot-cirrus interactions. It also shows that proper representation of aviation soot PSDs in global models is crucial for improved RF predictions due to soot-cirrus interactions.

We include the above discussion in the revised Methods section. A figure summarising all newly introduced sensitivity studies is added to the Extended Data (now Supplementary Material).

In the new Methods section “Ice activity spectra of aviation soot”, we further add: “Clearly, aviation soot PSDs are crucial for accurately representing soot-cirrus interactions in models.”

This paper is novel, but there are major revisions needed to improve this paper. There are also following specific comments needed to address as well.

1. Why did you use short-wave optical depth as a measure of RF? As we know, cirrus clouds contribute little to the change in solar short-wave, but contribute to the change in radiation balance through absorption of long-wave radiation.

Solar optical depth (OD) is often used as a proxy for net RF from cirrus, which generally increases with optical depth. For optically thin cirrus ($OD < 0.3$) such as for most AIC (ref.2), the increase of RF with OD is linear, e.g. Meerkötter et al. (1999).

L116ff: We add the reference to Meerkötter et al., as our study addresses a broad audience.

2. L47: Reference 8,9 suggest a big impact of aircraft soot on cirrus clouds. Why do you completely disregard references 8, 9 which did show an effect of aircraft soot on ice (though only "correlations").

We neither share the reviewer's statement that refs.8,9 "did show an effect of aircraft soot on ice" nor completely disregard refs.8,9 (L43ff). In fact, it would be unscientific to dismiss observational evidence, but at the same time we strongly believe that it is important to put measurement data under scrutiny.

While the existence of aircraft soot-cirrus interactions cannot be ruled out in principle, our statement is based on the fact that refs.8,9 base their conclusion on inference and not on causal (i.e. direct) evidence. Let us revisit the original research works.

Ref.8 (Ström & Ohlsson) observed statistically enhanced ice crystal number concentrations in clouds influenced by aviation. We quote a paragraph from their conclusions:

*'Although the analysis strongly suggests that the enhancement in crystal concentration is linked to aircraft exhaust, we cannot arrive at a definite conclusion about the mechanism for this enhancement. **The analysis indicates that aircraft exhaust favors ice nucleation, but there is no evidence that this is caused by black carbon particles.** The simultaneous observations of enhanced crystal number densities and absorbing material could be coincidental and be a result from coagulation between black carbon particles on ice crystals.'*

Ref.9 (Urbanek et al.) measured differences in depolarisation ratios of aged cirrus cloud fields from air masses with low vs high air traffic volume. They conjecture that these differences do not result from meteorological variability or from contrails embedded in the cloud fields. We quote from their abstract:

'These results could be explained by an indirect aerosol effect where heterogeneous freezing is caused by aviation exhaust particles.'

*While we do not challenge the observational results per se, one has to be careful when interpreting the results presented. **With regard to ref.9, the assertion that neither meteorology nor contrails could have caused the observed differences by itself does not automatically imply the existence of aircraft soot-cirrus interactions.** For instance, variability in ice supersaturation affects deposition growth and ice crystal habits, and thereby depolarisation. Ref.9 offers no convincing argument directly linking heterogeneous ice nucleation by aircraft soot and lidar depolarisation ratios of associated ice crystals that have undergone significant mass growth up to the time of measurement potentially many hours after formation. We quote from their conclusions:*

'The ML-CIRRUS research flights did not focus on indirect effects; therefore, crucial information at the cirrus formation regions was not explored in detail.'

Taken together, we believe that our statement (L45ff) is justified.

3. L50: This statement is incorrect. Ref. 11 showed the range of forcing depends on the number of aerosol particles that may be homogeneous vs heterogeneous in the background atmosphere.

Thank you for pointing this out. We actually took the RF range from ref.3, which was normalised to 2018 air traffic (their Supplementary spreadsheet). We include the proper reference in L52 and note the scaling to 2018 air traffic volume to clarify this matter.

4. L54: The positive numbers quoted in ref. 3 are due to lower values of sulfate aerosols and dust aerosols, together with a small portion of aircraft soot as INP. The positive values quoted in ref. 3

result from putting limitations on the sulfate particle numbers as well as dust numbers (Ref. 13), thus they are "tuned" to give positive results and unlikely to be correct based on observations as in ref. 8, 9. The best you can say here is "may remain uncertain if you can accept small sulfate numbers (which are not supported by observations)." The gaps in knowledge are mainly from not knowing whether contrail processed soot can be an INP. However, Mahrt et al. (2020, Ref. 19) limit to only larger soot.

We appreciate the clarification. We modify the discussion of the global model results in L205-213 to include the information concerning the role of background aerosol in cirrus formation and its importance for RF due to soot-cirrus interactions.

While ref.19 mostly studied soot-PCF for particle mobility diameters >200 nm, we point out that they also tested 100 nm soot particles and found them to only yield an ice-active fraction of 0.001 at homogeneous freezing conditions (see their Fig.S4). Ref.15 measured soot IN activity for 100 nm and 800 nm particles. The results indicate ice activity in the 100 nm samples to be almost absent for ice supersaturations up to 0.45; low ice-active fractions (0.02-0.03) were detected in the range [0.45,0.47], whereby 1% AF was selected as the threshold for activation onset (see their Fig.S1).

Overall, we agree with the reviewer that the limited availability of measured size-resolved ice nucleation ability of (contrail-processed) soot particles represents a challenge in fully encompassing the effect aviation soot-cirrus interactions and should be a focus of future studies. To this end, we underscore that the previously available laboratory studies alone would have not allowed us to set up a soot-cirrus interaction case study as presented here, since it is crucial to factor in particle size distributions in order to accurately predict the ice activation spectrum of aviation soot. Importantly, the soot-PCF mechanism we develop and describe in detail in ref.23 (and outline in the Methods for the first time for aircraft soot) provides a consistent, physically-based understanding of the ice nucleation activity of soot particles that fully replicates the size- and supersaturation dependence observed in the laboratory (see Fig.6 in ref.23). In this way, we have very strong confidence in extrapolating ice-active fractions to small particle sizes. We have thereby removed the gap in knowledge whether or not contrail processed aircraft soot particles act as INP.

L106: We add: "Although size-selective ice activity measurements of particles with mobility diameters < 100 nm are lacking, the theoretical underpinning of our soot-PCF parametrization builds strong confidence to apply it to such small particle sizes."

5. L59: The experiment in Ref. 16 start at -15 C so also include mixed phase clouds, not only cirrus.

Thank you for pointing this out. We remove this reference to avoid confusion.

6. L112: What effect did you mean here? It is not clear.

L113: We add: "... determining the supersaturation quenching time and therefore the competition between the ice nucleation modes."

7. L131: I did not find any ice crystal forms from 6min in your Fig.2. If ice forms from 6min, I suggest to extent the time axis to begin at 0min instead of 20min.

*Please note that we wrote: "In scenarios Background and Plume, ice crystals already start to form on the subset of ice-active soot particles about 6 min **earlier** than in scenario Base.", i.e. not at 43 min (as in scenario Base, L117), but around 37 min. This is consistent with Figs.2f,j that show this early ice formation as a 'shoulder' preceding the steep rise due to the onset of homogeneous freezing.*

8. L137: Are these deposition coeff. for soot? which case uses 0.5 and which uses 1.0?

These are deposition coefficients for water molecules on ice, not on soot. Please note that we wrote in L140f "... growth of soot-derived ice crystals ..." and see our explanation in the Methods (L390ff). The deposition coefficients describe the uptake of water molecules on spherical ice crystal surfaces.

9. L139: Please explain why turbulent diffusion lowers homogeneous ICNCs?

Sufficiently strong turbulent diffusivity, as prescribed in case Plume, decreases homogeneously nucleated ICNCs over time due to flattening of vertical ice supersaturation gradients. This is explained in detail in ref.22 illustrating the complexity caused by the nonuniformity in nucleation and growth conditions typically present in a column of air.

L140: We add: "... in scenario Plume by flattening vertical ice supersaturation gradients.

10. L133: Please show the quantitative evaluation of how long the homogeneous freezing is delayed because of Background and Plume scenarios.

The small delays relative to scenario Base are hard to visualize and depend on the variable in question. They are best judged by inspecting the shift between the green/orange and gray curves in Fig.2b,f,j (ice crystal number) during the steep rise phase due to homogeneous freezing. The same comparison in Figs.2d,h,l (optical depths) gives slightly different delays.

11. L139: Please add the variation of ice supersaturation in each scenario in Fig2.

Extended Data Figures 3-5 show high resolution profiles of ice supersaturation vs altitude at two different simulation times for each simulation scenario. Figure 2 presents cloud properties that are averaged over the whole column as a function of time. Given the marked vertical variability in supersaturation and the strong dependence of supersaturation on ice nucleation, averaging this variable in the vertical is not helpful for the interpretation of the simulation.

12. Fig.2: Why does the LWP increase by >50% due to the soot in included Background and Plume simulations?

We believe the reviewer refers to the ice water path (IWP). Please note that IWP is not simply the product of the total ICNC and column-average mean diameter also shown in Fig.2 but results from integrating the distribution of ice water content showing significant vertical structure (please revisit Extended Data Figs.3-5). In more detail, the development of IWP is influenced by the rate of water vapour deposition; the corresponding ice crystal growth rate is influenced by ice crystal number and size and the deposition coefficient, which all vary with altitude.

The fact that ice-active soot particles modify homogeneous freezing means that they are abundant enough to remove water vapour from the gas phase and produce relatively large ice crystals. Despite the resulting decrease in homogeneously nucleated ice crystal concentrations relative to scenario Base, the surface area generated by soot-derived ice crystals leads to a net increase of IWP over time. The supersaturated layer depth is deep enough to sustain these settling ice crystals giving them time to pick up more water vapour than their homogeneously nucleated counterparts. (This may be estimated by comparing the ice water content profiles in Ext. Data Figs. 3a+4a below 10.5 km altitude at 75 min). The supersaturation dependence of deposition coefficients (and, in scenario Plume, entrainment) also play roles in how ice water is distributed vertically in the inhomogeneous supersaturation field (L390ff).

L132: We add “... because the soot-generated ice crystals settle into ice-supersaturated air and take up relatively large amounts of water vapour.”

13. L153: What did you mean by the effect of the onset of homogeneous freezing? Extended Fig.6 showed the result taken before homogeneous freezing. Why is the upper limit estimate of 11% higher than the highest ICNC in Extended Fig.6 for the simulation Background, but 145% higher for the simulation Plume? Can you add some explanation to this difference?

We suspect that the reviewer refers to the fact that the estimated peak ICNCs derived from soot (54/L and 5/L, see Ext. Data Fig.2) are larger by different relative amounts than the values actually seen in the numerical simulations right before homogeneous freezing sets in (inferred as peak values from Ext. Data Fig.6). We offered an explanation in L152: The entrainment rate is larger in the Plume scenario (cf. L98f). Faster entrainment means stronger dilution and a stronger decrease in associated ice crystal number concentrations relative to the available total soot particle number (L514ff).

14. Line 161: I'm confused about why you did an additional simulation with updraught speed of 0.01 m/s but still dominated by homogeneous nucleation? I suggest to add some further discussion in this sensitivity experiment if you still think this is necessary for your conclusion.

Please note that the low updraught scenario is not entirely “dominated by homogeneous freezing”. This idealised scenario was designed as a sensitivity experiment to study the competition between heterogeneous and homogeneous freezing. The sensitivity simulation demonstrates that for sufficiently low updraught speeds, even the few ice-active soot particles from scenario Background are capable of quenching homogeneous freezing. They deposit enough water vapour to inhibit the first homogeneous freezing event that is clearly visible in the standard scenarios. This can for instance be seen in the vertical ICNC profiles in Extended Data Fig.4b vs. Extended Data Fig.7b; see also the description in caption of Extended Data Fig.7). However, at the top of the cloud layer, soot particles are not abundant enough to suppress secondary homogeneous freezing – in Extended Data Fig.7d,e, this continuous nucleation increases ICNC and optical depth over time, albeit their magnitudes remain low owing to the smaller dynamical forcing. This is what we meant in L164-167.

L164: For clarification, we add: “We include an additional sensitivity simulation Background, performed using a much lower (synoptic) updraught speed of 0.01 m/s, to illustrate the competition between homogeneous and heterogeneous ice formation in weak forcing conditions where INP generally exert a greater impact. ...”

15. L197: Field studies can see conditions dominated by surface sources, so it is unclear that “frequently” is correct here.

L197: We remove ‘frequently’.

16. Line 203: In the global model study, only 0.6% of aircraft soot were INPs in Refs. 11, 13, due mainly to the deposition of sulfate ending their ability to act as INP, which agrees with the small fraction (<1%) assumed in your study. You are misquoting here.

This remark reiterates several previous comments. Section 2.3 of ref.11 states: “Preactivated aircraft soots remain good ice nuclei until there are three or more monolayers of sulfate coated on the surface of soot, and then it is removed from the preactivated type and put into the inactive type. [...] In section 3.3, we will show that the preactivated aircraft soot only accounts for ~0.6% of the total aircraft soot in the air.” With these assumptions, the global model study neglects the size- and supersaturation dependence of the soot INP, whereas our results imply that only 1% of the 0.6% of all emitted soot particles should have been treated as INP.

We are grateful to the reviewer for pointing to the potential confusion that may be caused by these two percentages. We add in L202: "... unless they have collected multilayer sulphate coatings."

17. Line 205: Please add the correct reference for the positive RF result here and clarify that this is a sensitivity test to examine the effect of a large increase in INPs. This was not in ref 11, but from a sensitivity test in Ref.31.

L205 now reads: "Positive RF values were obtained in sensitivity tests assuming low concentrations of ambient sulphate and dust aerosol particles."

18. Line 212: Please add: "and changes in background emissions".

L212: We add "... and changes in background aerosols."

19. Line 215: Please add: "due to the use of much smaller background sulfate and dust concentrations, as shown in Ref. 11".

It is unclear to what concentration levels the reviewer refers to by 'much smaller'.

20. Line 262: please add a consideration of the larger mode soot particles seen in Petzold et al, Atmospheric Research, 1998 and Petzold, JGR 1999.

We include a consideration and the reference to Petzold et al. (JGR, 1999) in the substantially revised Methods section. Please see changes directly therein, including a new figure in the Extended Data section.

21. Line 475: This does not say it is cumulative as stated below. please clarify.

L471: We add "... supersaturation s , the s -cumulative AF, depends on ..."

L480: We add "The fact that AF is cumulative in s reflects laboratory measurements in which the ice nucleation activity of a sample is probed at successively increasing supersaturation."

22. Line 542: please give the parameterization in detail.

Thank you for pointing out that this has been unclear. We agree that this deserves better explanation. The soot-PCF parameterization detailed in ref. 23 is deterministic and therefore provides time-independent, diagnostic ice active fractions instead of a 'nucleation rate coefficient', as in the case of stochastic (e.g. homogeneous) ice nucleation. Such a rate enters budget equations for cloud-forming and cloud particles. This has important repercussions for the numerical implementation of diagnostic AFs in models.

Ice-active fractions, $AF(D,s)$ from Eq.(2), are s -cumulative (i.e. cumulative in ice supersaturation, s), meaning that they give, for an aviation soot agglomerate with mobility diameter D , the fraction of soot particles in a given sample that is ice-active in the s -range $[0,s]$. This reflects the underlying laboratory measurements, in which samples are exposed to a pre-set supersaturation and the resulting AF is measured. When these measurements are repeated for successively increasing supersaturation, the resulting AF as a function of s can be combined in one plot. However, this is not what cloud models usually simulate. In our model, the soot particles are removed from the PSD after ice activation (equivalent to smaller and smaller samples to be used in the laboratory experiments). As a result, the true ice active fractions to be used (called differential AF), are smaller than predicted by Eq.(2), as the number of unactivated soot particles remaining in the PSD continually decreases. In other words, using the cumulative AF would artificially increase and bias the number concentration of

soot-derived ice crystals. This unphysical effect is particularly large for the large soot particles where AF approaches unity (Extended Data Fig.1b), so it must be avoided in models that budget soot and ice crystal numbers. Because the deterministic component of soot-PCF strongly dominates its stochastic component (i.e. homogeneous freezing of pore water), activation does not occur in situations where s decreases (s remaining > 0). Hence, in such situations, the stochastic component can be neglected in models and the deterministic behaviour needs to be accounted for by leaving the cloud particle number unchanged when s decreases.

We modify and extend the Methods section (“Implementation of ice-active fractions in the cirrus model”), explaining how we derived differential AF. Please see changes directly therein.

23. Fig. 1: Do you mean contrail cirrus form after dissipation? Please clarify. And what are the upper inset and lower inset?

Yes, this refers to the fact that in the Background case, the new cirrus is assumed to take an irregular shape and is therefore termed contrail cirrus (ref.2) to visually distinguish it from linear contrails (cf. our response above regarding cloud shapes).

The insets are the gray boxes in the middle top and bottom right.

24. Extended Data Fig. 3: I see a purple and pink curve. the use of violet and magenta is not distinguishing.

We change the colours to black and red.

Reviewer #3

The authors seek to quantify the possible effect that aircraft soot emissions might have on cirrus formation after any initial contrail has dissipated. They do so using a sophisticated column model which incorporates an impressive state-of-the-science soot aggregate model. They investigate multiple possible situations, including the role of aircraft soot in near-term “re-formation” of a contrail, its role in longer-term formation of irregular contrail cirrus, and the anticipated effect of exhaust-like soot in base cirrus formation. They conclude that, excluding the initial effect in contrail formation, aircraft soot has a negligible effect on cirrus formation.

This work is timely and important. The question being asked is a matter of significant debate, and the methods used by the authors are novel, offering a fresh perspective on a problem which is most often addressed using global climate models (with conflicting results). The approach they have taken is mostly appropriate, but I have one serious concern regarding the scope of the study. Specifically, it was not clear to me after several readings whether the study could truly be said to capture the full scope of aviation’s impacts on cirrus formation. In particular, it seems that the possible long-term impacts of aviation soot emissions on cirrus, through the accumulation of aged combustor soot at high altitudes, may not be fully considered.

I believe that this study is of great interest to the audience of Communications Earth and Environment. It is clearly written and rigorous. However, given the importance of the question and the boldness of the claim in the title, I recommend against publication until the comments below are addressed.

We thank the reviewer for carefully reading our manuscript and are grateful for the overall positive assessment and valuable suggestions. Below we address each comment separately.

Major comments

The conclusion drawn by the paper, as given in the title, is that aircraft soot-cirrus interactions do not matter. However, the paper appears to only consider the role of aircraft emissions within ~12 hours after emission, and it was not clear to me why this was the case. Given that aircraft fly in the upper troposphere and lower stratosphere, the lifetime of emitted soot is expected to significantly exceed this, with a mean lifetime on the order of a month (see e.g. Zhou and Penner 2014). On this timescale, it would be expected that soot emitted from aircraft could age significantly while also diluting, and potentially contribute significantly to formation of cirrus (presumably taking part in the “Base” scenario). However, this paper does not appear to address this possibility. The three scenarios consider either the “re-formation” of a contrail around 30 minutes (“Plume”), the formation of an irregular contrail after around 5 hours (“Background”), or formation of cirrus in air which has not been influenced by aircraft exhaust (“Base”). However, none of these scenarios appear to address a case where aged aircraft soot could take part in cirrus formation, as all three treat the soot which is present as being “unaged” (see comments below). To truly support the title of the paper, the effects of aged aircraft soot must also be discounted. If aged aircraft combustor soot is a highly efficient ice nucleus, then its role in the “Base” case has not been accounted for. I would suggest that the authors either 1) weaken the strength of their conclusions, including the title, to make it clear that their work only represents unaged soot; or they 2) perform additional sensitivity simulations to quantify the role that aged aircraft soot may have in cirrus formation, accounting for the long aging times which might be relevant (see below for more details) and the plausible range of fractional contributions that aircraft emissions might have to “baseline” soot concentrations.

A key conclusion of the paper is that many current models are overpredicting the role of aviation soot in the formation of cirrus clouds. This conclusion is based on the use of a sophisticated model, which accounts for the specific morphology of fresh and aged soot. However, I was wondering about a detail provided on lines 492-497. The authors use a contact angle of 60 degrees, consistent with Persiantseva et al (2004) which studied aircraft combustors specifically. It is stated that, to decrease the contact angle (and therefore increase the hydrophilicity of the soot), “exposure to atmospherically relevant ozone concentrations over several days is required”. It is then implied that such exposure is not relevant here, but – unless the authors reduce the scope of their paper as discussed in my previous comment - that assertion does not appear to be properly supported. The mean lifetime of aircraft soot (see previous comment) is likely to be greater than the lifetime assumed by Wei et al (2017) of “< 10 days”, which is the basis for the assumption given here. That article appears to be focused on emissions in near-surface conditions, and also states that “atmospherically relevant” concentrations of ozone would be on the order of 40 ppbv, while exposure to sulfuric acid and nitric acid would both also act to decrease the contact angle. This seems highly relevant, given that aircraft exhaust is rich in sulfur acid, while stratospheric air can contain mixing ratios of ozone which are orders of magnitude greater than 40 ppbv (in addition to significant quantities of H₂SO₄ and HNO₃). If the authors decide to retain the current scope implied by the title, I recommend that they consider performing an additional sensitivity simulation in which they further decrease the contact angle (based on a reasonable estimate of the possible effect of such aging), to simulate the potential effects of long-term aging.

Similarly, the claim that freshly emitted soot contact angles are in the range 60-110 degrees (line 492) seems misleading. Of the two studies cited (Persiantseva et al, 2004; Wei et al, 2017), only Persiantseva et al studied aircraft combustors, and they found contact angles in this situation of 63 degrees. Wei et al (2017) instead evaluated a kerosene lantern, fireplace, and bus exhaust to find the larger range of 65-110 degrees. The statement that 60 degrees corresponds to an upper limit of hydrophilicity for unaged aircraft combustor soot implies that there is data supporting aircraft exhaust having a significantly higher contact angle. If so, it should be cited here, as the two given

references seem to not support it. If such evidence is not available, I would recommend removing the claim that this is an upper bound.

The reviewer asks us to address the role of atmospherically processed, aged aircraft soot in cirrus ice formation in order to support the main statement of the paper as embodied in its title. We are grateful that the reviewer brings this up and admit that we could have been much clearer regarding this important issue.

In essence, the reviewer addresses two points connected to large scales. The first issue (i) states that we are considering only plumes up to 5 h past formation in our scenarios, so that the total soot particle number concentration of $458/\text{cm}^3$ in scenario Background might not represent large-scale number concentrations of aged aircraft soot particles. The second issue (ii) revolves around contact angle differences between aircraft soot in our scenarios and aged aircraft soot due to large-scale ozone oxidation.

Our reply to (i), which we reproduce verbatim in the extended Methods section “Soot particle properties” in L440, is as follows:

*“As these estimates are based on a single plume dispersion model, they do not account for multiple overlapping plumes enhancing n_{tot} -values. However, the likelihood of plumes overlapping increases with time as plume widths increase due to wind shear; therefore, ongoing dilution reduces n_{tot} -values in individual plumes. This particularly applies to potentially wide plumes in scenario Background. **In the atmosphere, both effects can offset each other, so that $n_{\text{tot}} = 458 \text{ cm}^{-3}$ in scenario Background taken at $t_b = 5 \text{ h}$ approximately represents large-scale number concentrations of aviation soot particles resulting from mixing of plumes older than 5 h.”***

With regard to issue (ii), we rearrange the discussion of contact angles and include a novel section in the Methods entitled “Role of soot particle ageing”. Our response is a verbatim quote (apart from numbering additional references) inserted after L491:

“Ice-active soot particle fractions exhibit a marked dependence on θ , which can be affected by atmospheric ageing processes. Ageing of soot particles impacts clouds and climate^{48a} via (i) condensation of semi-volatile material, (ii) coagulation with ambient aerosol particles and (iii) oxidation from the gas phase⁴⁹. While monolayer sulphuric acid (H_2SO_4) coatings have only little effect for heterogeneous ice nucleation on soot particles^{48b}, the ability of PCF is significantly reduced if soot pores are filled with H_2SO_4 or organic carbon^{47,49a} via processes (i) and (ii). If soot particles obtain thick coatings, the ice nucleation mode switches from PCF to immersion freezing, where soot has proven to be an inefficient INP^{12,48b,49a}. Therefore, we consider that only ageing process (iii) is capable of enhancing PCF efficiency by decreasing θ .

We first discuss soot-water contact angles associated with unaged soot particles. Data of experimentally determined contact angles relating to aviation soot are scarce. Ref. ^{49b} produced soot particles in a gas-turbine combustion chamber setup by burning propane-butane mixtures, mimicking aircraft cruise combustion conditions, and found $\theta = 63^\circ$. The same study reported higher contact angles (70° – 80°) when burning the aviation fuels TC1 (fuel sulphur content $> 0.11 \text{ wt-\%}$), T6 ($0.05 \text{ wt-\%}$) and kerosene in an oil lamp setup. Contact angles were found to decrease for TC1 and kerosene soot only after substantial heating and outgassing, processes that are not relevant for the upper troposphere. Using the same technique, ref. ^{49c} found $\theta = 59^\circ \pm 4^\circ$ and $69^\circ \pm 4^\circ$ when burning freshly prepared TC1 and long-time stored (oxidised) TC1 kerosene, respectively. A value of 59° was found for TS1 kerosene fuel using the same approach^{49d}. Ref. ^{49e} found the contact angle of fresh TC1 soot to be 69° . Significantly higher values ($\approx 97^\circ$ – 107°) were reported for unaged kerosene soot⁵⁰.

We now address how ageing of soot particles via gas phase oxidation (process (iii)) might affect θ . Ref. ⁵⁰ found θ to decrease by $\approx 5\text{--}10^\circ$ after exposing kerosene soot to ozone mixing ratios of 40 ppb over a period of ten days, which is at the high end of tropospheric soot particle lifetimes of $\approx 3\text{--}11$ d (ref. ⁴⁹). Following ref. ⁵⁰ and doubling the ozone mixing ratio to 80 ppbv to account for mixing of upper tropospheric and stratospheric air masses, θ might decrease by $\approx 15\text{--}20^\circ$ over 10 d. We acknowledge that lifetimes in the upper troposphere can be longer, especially for particles directly emitted into the upper troposphere, as in the case of aircraft emissions. Doubling the lifetime to 20 d with 40 ppbv of ozone would lead to the same decrease. However, ageing of aircraft-emitted soot particles already occurs in evolving exhaust plumes via condensation of H_2SO_4 formed by oxidation of SO_2 vapour from the exhaust (process (i)) and coagulation with liquid ambient and plume aerosols (process (ii))²⁸. Sulphuric acid hardly oxidises soot surfaces^{19,50a}; therefore, exposure to H_2SO_4 has next to no effect on θ . Moreover, a $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ coating shields the soot surface and prevents oxidation from e.g. ozone^{50b}. Overall, this limits the time available to reduce θ via gas phase oxidation to $\approx 1\text{--}2$ d, depending on jet fuel sulphur content and ambient aerosol load²⁸ and the above rough estimates for changes in θ reduces to a few degrees. In summary, default value of 60° for unaged aviation soot, which is already at the low end of data (see above), would decrease only slightly.”

To come full circle to ice activity for contact angles $< 60^\circ$, we include another section in the Methods entitled “Ice activity spectra”, from which we quote verbatim:

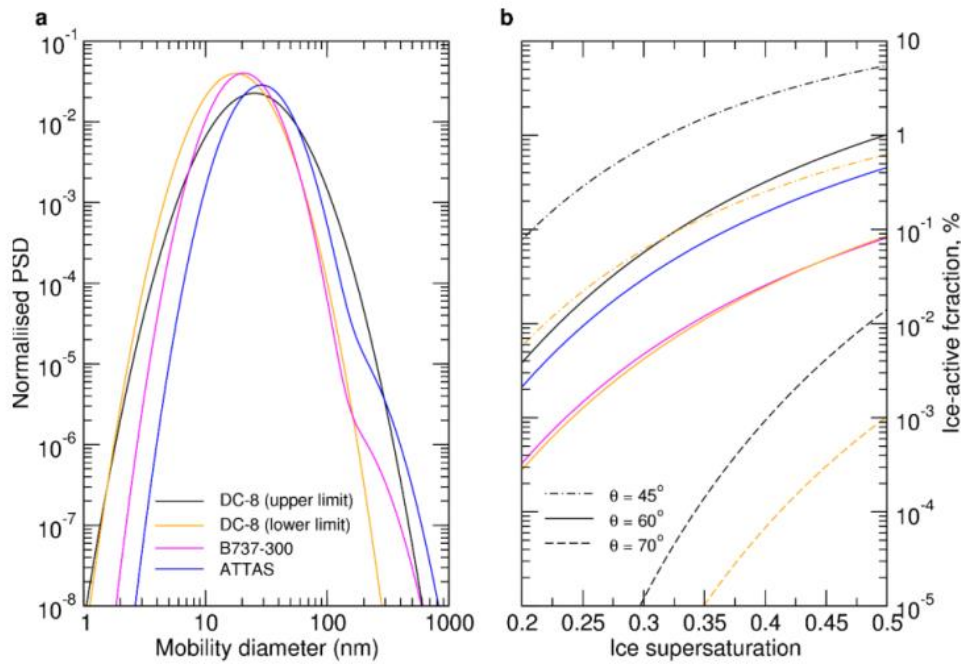
“The dependence of ice-active soot particle fractions, f , on ice supersaturation is given by integrating the ice-active PSDs shown in [Extended Data] Fig.1c over all sizes:

$$f(s) = \int_0^\infty \text{PSD}(D) \text{AF}(D, s) dD \quad (4)$$

using normalised PSDs. Results of a sensitivity study with several size distributions using our default contact angle of 60° , presented in [Extended Data] Fig.2, show that the bimodal PSDs from ref.^{41,41a} cause much fewer ice-active particle fractions across all supersaturations as compared to our default choice of an upper limit monomodal PSD. This is caused by smaller abundances of large particles. Clearly, PSDs are crucial for accurately representing soot-cirrus interactions in models.

Results of a sensitivity study with different contact angles are also shown. Increasing θ by 10° to a value of 70° renders ice activity negligible for both monomodal PSDs. We also show results of an extreme case with θ decreased by 15° to 45° , greatly overestimating the effect of large-scale soot ageing via ozone oxidation by dismissing the possibility of externally-mixed, fresh soot emissions turning into internally-mixed aerosol particles already on the plume scale. As a result, the ice activity spectra peak at 5.5 (0.6) % for our upper-limit (lower-limit) PSD. More frequently observed aviation soot PSDs, characterised by $D_m \approx 29$ nm and $\sigma \approx 1.7$ (ref. ⁴²), would yield peak ice activity fractions near 1% similar to our simulation scenarios. **In summary, we conclude that effects of atmospheric ageing on the ice activity of aviation soot particles are most likely negligible and that the conclusions drawn here hold true for both, unaged and aged soot.”**

A new figure in the Supplementary Material (formerly Extended Data) quantifies both, effects of bimodal PSDs (requested by another reviewer) and of contact angle changes:



Supplementary Figure 2. Ice activity spectra of aviation soot particles. **a**, Monomodal⁵² (black, orange) and bimodal⁵³ (blue, magenta) soot PSDs from in situ measurements in young, non-contrailing exhaust plumes of different types of aircraft. The monomodal PSDs are repeated from Supplementary Fig. 1a to facilitate comparison. **b**, Spectral distributions of ice-active fractions derived from the PSDs. Ice activity is predicted by the soot PCF-parametrization assuming a temperature of 220 K for a (solid curves) default soot-water contact angle, θ , of 60° . Results for (dashed) higher and (dot-dashed) lower θ -values are shown only based on the two monomodal PSDs. The solid black activity spectrum in panel **b** is consistent with the size-integrated results shown in Supplementary Fig. 1d in the limit of large mobility diameter.

Referring to the new material in the Methods, we add in the main text:

L178: "... effects of atmospheric soot ageing processes relevant for aviation emissions beyond t_b do not significantly increase their PCF activity."

L188: "Ozone oxidation can decrease contact angles on the surfaces of soot particles. However, gas phase oxidation becomes increasingly hampered by condensation and coagulation processes leading to thick coatings as aircraft soot ages. In the case of aircraft soot particles, contact angles decrease only slightly and therefore ice activity is not changed significantly (see Methods)."

Taken together, we hope that we have convincingly addressed that even aged aircraft combustor soot cannot be regarded as an efficient INP, hence, changed soot particle properties due to ageing do not alter our conclusions.

Minor comments

I have no minor comments. However, I would like to commend the authors on the clarity of Figure 1.

Fig.1 (caption): We clarify that the term Background (and Plume) relates to observed INP concentrations, not to large-scale average conditions by adding: "The scenarios consider number concentrations of ice-active aviation soot particles lying within the range of INP values observed in the background upper troposphere."

References

Persiantseva, N. M., Popovicheva, O. B. and Shonija, N. K.: Wetting and hydration of insoluble soot particles in the upper troposphere, J. Environ. Monit., 6(12), 939–945, 2004.

Wei, Y., Zhang, Q., Thompson, J. E. and Others: The wetting behavior of fresh and aged soot studied through contact angle measurements, *Atmospheric and Climate Sciences*, 7(01), 11, 2017.

Zhou, C. and Penner, J. E.: Aircraft soot indirect effect on large-scale cirrus clouds: Is the indirect forcing by aircraft soot positive or negative? *J. Geophys. Res. D: Atmos.*, 119(19), 11,303–11,320, 2014.

We add the following references:

48a. Lohmann, U. et al. Future warming exacerbated by aged-soot effect on cloud formation. *Nat. Geosci.* **13**, doi:10.1038/s41561-020-0631-0 (2020).

48b. DeMott, P. J., Chen, Y., Kreidenweis, S. M., Rogers, D. C. & Sherman, D. E. Ice formation by black carbon particles. *Geophys. Res. Lett.* **26**, doi: 10.1029/1999GL900580 (1999).

49. Bond, T. C. et al. Bounding the role of black carbon in the climate system: A scientific assessment. *J. Geophys. Res.* **118**, doi:10.1002/jgrd.50171 (2013).

49a. Crawford, I. et al. Studies of propane flame soot acting as heterogeneous ice nuclei in conjunction with single particle soot photometer measurements. *Atmos. Chem. Phys.* **11**, doi: 10.5194/acp-11-9549-2011 (2011).

49b. Persiantseva, N. M., Popovicheva, O. B. & Shonija, N. K. Wetting and hydration of insoluble soot particles in the upper troposphere. *J. Environ. Monit.* **6**, doi:10.1039/B407770A (2004).

49c. Popovicheva, O. et al. Effect of soot on immersion freezing of water and possible atmospheric implications. *Atmos. Res.* **90**, doi:10.1016/j.atmosres.2008.08.004 (2008).

49d. Kireeva, E. D., Popovicheva, O. B., Persiantseva, N. M., Khokhlova, T. D. & Shonija, N. K. Effect of black carbon particles on the efficiency of water droplet freezing. *Colloid J.* **71**, doi:10.1134/s1061933x09030090 (2009).

49e. Shonija, N. K., Popovicheva, O. B., Persiantseva, N. M., Savel'ev, A. M. & Starik, A. M. Hydration of aircraft engine soot particles under plume conditions: Effect of sulfuric and nitric acid processing. *J. Geophys. Res.* **112**, doi:10.1029/2006JD007217 (2007).

50a. Zhang, D. & Zhang, R. Laboratory investigation of heterogeneous interaction of sulfuric acid with soot. *Environ. Sci. Technol.* **39**, doi: 10.1021/es050372d (2005).

50b. Ray, D. et al. Hygroscopic coating of sulfuric acid shields oxidant attack on the atmospheric pollutant benzo(a)pyrene bound to model soot particles. *Sci. Rep.* **8**, doi:10.1038/s41598-017-18292-z (2018).

12th Feb 21

Dear Professor Kärcher,

Your revised manuscript titled "Aircraft soot-cirrus interactions do not matter for the climate impact of aviation" has now been seen by the original two reviewers, and I include their comments at the end of this message. Both reviewers acknowledge improvements in revision, but continue to raise apparently important points. We are still interested in the possibility of publishing your study in Communications Earth & Environment, and can see two possible ways forward:

- 1) If you are happy to tone down your paper and add caveats as requested by reviewer 1, please revise your manuscript accordingly; if appropriate, we will be able to offer to accept the paper in principle.
- 2) If you feel that you can support your current conclusion, that previous large estimates of global radiative forcing from interactions between aircraft soot and large-scale cirrus can be ruled out based on your study, we can offer adjudication from a fresh reviewer. If you opt for adjudication, please send suggestions for at least 6 potential adjudicating reviewers whom you have not collaborated with in the past 5 years, and whom you would trust to be fair. We will then send the previous reports and the new material to an adjudicator (from your list if we deem them appropriate), for an evaluation of the deadlock that has arisen between yourselves and the reviewer.

Either way, we invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter) and the completed checklist:

[link redacted]

** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to

accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Heike Langenberg, PhD

Chief Editor
Communications Earth and Environment

On Twitter: @CommsEarth

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Please refer to our data policies at <http://www.nature.com/authors/policies/availability.html>.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

I appreciate the argument that the authors make stating that they consider a “maximum” impact of aircraft soot based on their parameterization, however, I still believe they are missing conditions that would allow a better characterization of the effect of soot on cirrus clouds. In particular, they only deal with clouds having optical depth 0.3. This is smaller than many cirrus clouds and thus their calculated change in optical depth is smaller than would be calculated using their parameterization in a global model (that considers a more realistic range of optical depths). The authors also seem to argue that thicker clouds mainly form as a result of homogeneous ice nucleation. This conclusion makes me think that this model really does not simulate cases for aircraft soot effects on cirrus in the background atmosphere. It could be that there is something wrong with the 1-D representation of cirrus clouds presented here. Certainly the background conditions for liquid aerosols and aircraft soot are nothing like what is simulated in GCM models. In any case, the sweeping statements that they make in the abstract and conclusions regarding GCM studies cannot be supported. See some necessary changes below. Substantial revision of the statements has to been made and uncertainty in the 1-D model should be discussed sufficiently before consider to accept.

Line 18: add “for the conditions we evaluate” since you only evaluate a small fraction of the possible conditions

Line 19-21: delete the sentence “our results preclude...”

Line 21-22: delete “resolves a long-standing knowledge gap”. While the enclosed study applies a new parameterization for ice-active soot, it does not apply it to the situations that would apply in global atmospheres as represented in GCM’s

Line 28-9: change to “yield the largest aviation-induced RF”

Line 33: here you state that you are mainly considering contrails forming within already existing cirrus. This is not the focus of the GCM studies that you state that you “resolve” in the abstract

Line 50: These numbers are from the summary given in ref. 3, but the original reference should also be stated (Zhou and Penner, 2014), since the latter explains that the large positive numbers are associated with unrealistic assumptions about the sulfate aerosol concentrations.

Line 79: remove “and judge the magnitude of associated RF estimates” as this study does not sample enough meteorological conditions to allow a judgement.

Line 88: here you repeat that you only consider contrail cirrus at time $t - t_b > t_p$. These are now termed “irregularly shaped cirrus cloud similar to contrail cirrus” in Fig 1 caption. This term still bothers me, because it may be confused with cirrus that are close to the original contrail, but since you state in the caption that you use aircraft soot concentration active fractions “that encompass” measured INP concentrations from the background atmosphere, I suppose it applies to conditions that may be further from the contrails. I had to re-read lines 93-96 plus supplementary material, before I realized that you are referring to your parameterized soot active fraction. Hard to read, but acceptable.

Line 100. For balance, please add information on the assumed unperturbed (base) aerosol particle concentrations (and their size) used in the simulations (i.e. move from the “methods” section. The formation of ice crystals depends on both the soot amount and the sulfate amount.

Line 125-6: many cirrus clouds have optical depths > 0.3 . In fact CALIPSO typically measures cirrus with optical depths between 0.3 and 3 (Mitchell et al., ACP, 2018). You need to explain why your model is not consistent with this.

The authors will not find significant changes in optical depth due to aircraft soot with this restricted optical depth.

Line 166-7: Here is another example where your model assumptions do not fit observations or GCM simulations: not all clouds are dominated by homogeneous freezing.

176-178: Are you arguing here that the presence of cirrus with optical depth > 0.3 must mean they are formed mainly from homogeneous freezing? If so, this would be a new finding!

This makes me fear your 1-D model is not really a good representation of cirrus cloud formation!

Line 196: add: negligible perturbations in this model.

Line 207: change to “where the ice activity of aircraft soot, as measured by our parameterization” ...

Line 213-215: It is unlikely that other soot / refractory particles contribute efficiently as INP (after all, you quote 5/L as the nominal background number). Moreover, it is known from laboratory measurements that less than 100% of soot particles act as INP. Thus, you cannot conclude that aircraft soot have smaller effects in cirrus than other soot and refractory particles.

Line 2019: the statement “regardless of their size” is not true. The study you quote used a parameterization based on a detailed air parcel model that accounted for the effects of size.

Line 233-236, You have misquoted ref. 29. As I understand it, you take “ice-active fraction” as the fraction of soot particles forming ice crystals, while reference 29 refers to all particles that may form ice crystals. As a result of the size of the soot particles as well as other conditions (such as the supersaturation reached) only a fraction of these form ice.

Line 233-235: While you use reference 29 as estimating 100-300 ice-active particles in regions with high soot emission and > 10 /L in the Northern Hemisphere, not all of these particles are “ice-active” as you model here. Considerable fewer are ice active, in the model described in reference 30. (Reference 29 used the parameterization of Liu and Penner, Meteorological Zeitschrift, 2005.) Considerable fewer are ice active, in the model described in reference 30.

Line 238-239: The global model accounts for size-induced effects. This statement is wrong.

Line 247: The supersaturation for aircraft soot nucleation is not 0.2 but 0.35 in the references. This statement is wrong.

Line 249: The parcel model represents following a lagrangian parcel, and thus correctly simulates cirrus formation in a global model. It seems to me the 1-D model is inaccurate.

Line 291-3: I am not sure what is meant here. Do you treat a column of environmental air with the same updraft, turbulence, and vertical diffusivity, and entrain this air (at what rate?) into the column representing AIC? Please explain more fully or if not, please add an explanation of what is meant by “but without microphysical processes”.

Line 310-313: please explain the phrase: “each containing a number of “real” ice crystals”. Right now it reads as if you have 100,000 simulation ice particles (from liquid aerosol) and these each contain “real” crystals.

Line 327-8: Please explain what is meant by the “vertical plume extensions”

Line 322 and 366: Your liquid aerosol number is 500 cm^{-3} while the aircraft soot is 458 cm^{-3} in the background scenario. Approximately equal concentration of soot and liquid aerosol is not at all what is observed in the background atmosphere. I submit that your study is unable to comment on the conditions treated in GCM simulations.

Line 407-409: this statement requires a reference

Reviewer #3 (Remarks to the Author):

The updated manuscript is a substantial improvement, and I thank the authors for their time and effort. As this is a re-review, I have focused my efforts on the degree to which the authors have addressed my previous comments.

Broadly, I am satisfied that the authors have addressed my comments. I have only one remaining concern; once addressed, I believe this paper will be ready for publication. The arguments regarding ageing of aircraft soot are interesting and logical, but I was concerned at the focus on tropospheric air and the argument on (e.g.) line 450 that a 20 day lifetime would be "long". Roughly one third of all cruise fuel burn occurs in the stratosphere (e.g. Sheng et al 2015), meaning long lifetimes (potentially much greater than 10 days). Yu et al (2019), using a combination of observations and modeling, estimated a lifetime for wildfire black carbon entering the stratosphere at hundreds of days. Kravitz et al (2012), when simulating stratospheric emission of small black carbon particles at ~14 km as a geoengineering strategy, found an average lifetime on the order of three years. This means up to three years of ageing before re-entering the troposphere and potentially taking part in cirrus formation. This region also has high ozone concentrations, and a dry environment. While the authors do argue that (say) H₂SO₄ coatings and coagulation might limit the effect of ozone over longer lifetimes, there does not appear to be quantitative data available to test this hypothesis.

I would therefore recommend inclusion of a caveat (e.g. at line 460) to the effect that, while 60 degrees is a reasonable first estimate, and while the work presented here comprehensively covers situations involving recent (less than one month ago) aircraft emissions, additional work is needed at cruise conditions - including efforts to better constrain the lifetime and properties of aged aircraft soot - to establish the potential contribution of soot which has aged significantly in the stratosphere. Without such a caveat, the assertion in the title still appears excessive.

That having been said, I would again like to state that I find this paper both compelling and highly relevant to the audience of Communications Earth and Environment.

References

Kravitz, B., Robock, A., Shindell, D. T. and Miller, M. A.: Sensitivity of stratospheric geoengineering with black carbon to aerosol size and altitude of injection: BC GEOENGINEERING SENSITIVITY, *J. Geophys. Res.*, 117(D9), doi:10.1029/2011jd017341, 2012.

Sheng, H., Marais, K. and Landry, S.: Assessment of stratospheric fuel burn by civil commercial aviation, *Transp. Res. Part D: Trans. Environ.*, 34, 1–15, 2015.

Yu, P., Toon, O. B., Bardeen, C. G., Zhu, Y., Rosenlof, K. H., Portmann, R. W., Thornberry, T. D., Gao, R.-S., Davis, S. M., Wolf, E. T., de Gouw, J., Peterson, D. A., Fromm, M. D. and Robock, A.: Black carbon lofted wildfire smoke high into the stratosphere to form a persistent plume, *Science*, 365(6453), 587–590, 2019.

COMMSENV-20-0524A

Response to reviewers (*replies italicized*)Reviewer #1

I appreciate the argument that the authors make stating that they consider a “maximum” impact of aircraft soot based on their parameterization, however, I still believe they are missing conditions that would allow a better characterization of the effect of soot on cirrus clouds. In particular, they only deal with clouds having optical depth 0.3. This is smaller than many cirrus clouds and thus their calculated change in optical depth is smaller than would be calculated using their parameterization in a global model (that considers a more realistic range of optical depths). The authors also seem to argue that thicker clouds mainly form as a result of homogeneous ice nucleation. This conclusion makes me think that this model really does not simulate cases for aircraft soot effects on cirrus in the background atmosphere. It could be that there is something wrong with the 1-D representation of cirrus clouds presented here. Certainly the background conditions for liquid aerosols and aircraft soot are nothing like what is simulated in GCM models. In any case, the sweeping statements that they make in the abstract and conclusions regarding GCM studies cannot be supported. See some necessary changes below. Substantial revision of the statements has to been made and uncertainty in the 1-D model should be discussed sufficiently before consider to accept.

We thank the reviewer for his/her time to carefully read our revised manuscript and for providing additional comments. We provide a point-by-point response to all comments (enumerated, [nn]).

[1] Line 18: add “for the conditions we evaluate” since you only evaluate a small fraction of the possible conditions

See our reply to [7].

[2] Line 19-21: delete the sentence “our results preclude...”

L19: The modified sentence reads “Our results challenge previous large estimates of ...”.

[3] Line 21-22: delete “resolves a long-standing knowledge gap”. While the enclosed study applies a new parameterization for ice-active soot, it does not apply it to the situations that would apply in global atmospheres as represented in GCM’s

See our reply to [7].

[4] Line 28-9: change to “yield the largest aviation-induced RF”

Done.

[5] Line 33: here you state that you are mainly considering contrails forming within already existing cirrus. This is not the focus of the GCM studies that you state that you “resolve” in the abstract

Please note that we are not ‘mainly considering contrails forming within already existing cirrus’. In L30ff, the RF estimate in ref. 3 does not include the impact of contrails forming within already existing cirrus. Please also not that In L33, we refer to ref. 4, a study entitled ‘Aviation effects on already existing cirrus clouds.’ Our work deals with the soot impact on new cirrus formation.

To avoid confusion, we replace ‘This estimate’ (L32) by ‘The estimate from ref. 3’.

[6] Line 50: These numbers are from the summary given in ref. 3, but the original reference should also be stated (Zhou and Penner, 2014), since the latter explains that the large positive numbers are associated with unrealistic assumptions about the sulfate aerosol concentrations.

The sentence L49-55 in the introduction sets the stage by referring to the most recent comprehensive aviation assessment available to date (ref. 3) without detailing individual GCM studies, as we come back full circle to RF in the discussion section. In L216ff, we describe all individual references. Please note that we apply the same logic to the other two GCM studies (L52f).

To appreciate that ref. 3 was already aware of the point addressed by the reviewer, we add in L228: “... superseding large RF estimates as noted in ref. 3.” That said, please note that we already considered this concern in L225ff.

[7] Line 79: remove “and judge the magnitude of associated RF estimates” as this study does not sample enough meteorological conditions to allow a judgement.

This comment contains two issues: (i) ‘does not sample enough meteorological conditions’ and (ii) ‘judge the magnitude of associated RF estimates’.

Concerning (i), we “chose meteorological conditions representing a typical midlatitude situation with regard to air traffic volume and contrail occurrence” (L315ff). Regarding our choice of updraught speeds, “using a higher mean updraught speed or including updraught speed variability would lead to predictions with smaller or negligible perturbations” (L182f). Although not directly related to meteorological conditions, regarding our choice of baseline liquid aerosol parameters (L320f), see our reply to [9].

Concerning (ii), we agree with the reviewer that ‘judge’ may be too strong a statement regarding the largest global model-based RF estimates for aircraft soot-cirrus interactions. We remove this term, since we do not give an updated best RF estimate. Rather we showed that a maximum RF value is dramatically lower than previous large RF estimates indicate (L231f). Since this maximum value is very low, we concluded that the effect has previously been overestimated. We acknowledged that in order to arrive at a (statistically) representative global best RF estimate, climate model simulations are still needed that incorporate the latest evidence based on our study (L269f).

L78ff: The modified sentence now reads: “This allows us to unravel the microphysical mechanisms underlying aircraft soot-cirrus interactions on the process level and provides new insights and guidance to constrain the magnitude of associated RF-estimates.”

L271: We add “... enabling a more robust quantification of associated global RF.”

[8] Line 88: here you repeat that you only consider contrail cirrus at time $t_b > t_p$. These are now termed “irregularly shaped cirrus cloud similar to contrail cirrus” in Fig 1 caption. This term still bothers me, because it may be confused with cirrus that are close to the original contrail, but since you state in the caption that you use aircraft soot concentration active fractions “that encompass” measured INP concentrations from the background atmosphere, I suppose it applies to conditions that may be further from the contrails. I had to re-read lines 93-96 plus supplementary material, before I realized that you are referring to your parameterized soot active fraction. Hard to read, but acceptable.

Thank you for pointing out that this remained unclear after our initial revision. The reviewer is correct that the attribute ‘Background’ associated with the later time past emission, t_b , relates to conditions further away from the source aircraft that initiates the original contrail (visualized in Fig.1). The times t_p and $t_b > t_p$ are indeed chosen such that the parameterized ice-active soot fractions encompass

typical INP numbers observed in the upper troposphere (L92f). There should be no confusion that 'Background' refers to conditions close to the source aircraft, as those are represented by 'Plume' cases.

To better distinguish the two cases, we add in L90: "... if the exhaust plume [...] is closer to the source aircraft and therefore still line-shaped and if ...".

[9] Line 100. For balance, please add information on the assumed unperturbed (base) aerosol particle concentrations (and their size) used in the simulations (i.e. move from the "methods" section. The formation of ice crystals depends on both the soot amount and the sulfate amount.

We base the liquid aerosol particle properties (L322f) in our simulations on experiments conducted within the Cirrus Parcel Model Comparison Project (CPMC) of the GEWEX (Global Energy and Water Experiment) Cloud System Study (GCSS) Working group on Cirrus Cloud Systems (WG2) (Lin et al., 2002). As an aside, please note that in our case, these particles are not necessarily composed of sulphate; their water activity is determined more generally by a hygroscopicity parameter (L320) representing a broad range of chemical compositions.

Regardless, the sensitivity of cirrus properties resulting from homogeneous freezing rate variations due to changes in soluble aerosol particle properties is rather weak (in particular very weak when compared to changes in updraught speed), as demonstrated by cloud physical theory and numerical simulations (e.g. Kärcher & Lohmann, 2002; Liu & Penner, 2005; Cirisan et al., 2013). It has also been shown that aerosol sensitivity can be appreciable only at low temperatures and in moderate updraughts (Kay & Wood, 2008), i.e., conditions that are not relevant for our study. For these reasons, we listed characteristics of the liquid aerosol droplets in the Methods section.

To address this point, we add in L323: "The susceptibility of homogeneously nucleated ICNCs to soluble aerosol particle properties is weak for updraught speeds and temperatures relevant for the conditions in our simulations (Kay & Wood, 2008)."

[10] Line 125-6: many cirrus clouds have optical depths > 0.3 . In fact CALIPSO typically measures cirrus with optical depths between 0.3 and 3 (Mitchell et al., ACP, 2018). You need to explain why your model is not consistent with this.

The authors will not find significant changes in optical depth due to aircraft soot with this restricted optical depth.

The study the reviewer refers to estimates cirrus cloud ice-particle number concentration, effective diameter and ice water content from sensors aboard a polar orbiting satellite (CALIPSO, Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation). Mitchell and co-PIs apply their retrieval method to (quote from their abstract) 'single-layered semi-transparent clouds having a visible optical depth between about 0.3 and 3, where cloud base temperature is ≤ 235 K.'

Please note that we do not argue against the existence of cirrus with optical depths > 0.3 . Cirrus optical depth is larger at lower altitudes (higher temperatures and therefore more moisture available for deposition). Among the cirrus clouds that develop optical depths > 0.3 are cirrus influenced by convection at midlatitudes, mostly at altitudes below flight levels. Cirrus associated with tropical deep convection are mostly optically even thicker (> 3). The average air temperature along aircraft routes north of 30°N , where most aviation activity occurs, is 218 K (Fig.10 in Kärcher et al., 2009). At these temperatures, cirrus clouds are mostly optically thin, and these are most easily affected by INPs.

We add in L126: "Thin cirrus with low optical depth (< 0.3) similar to AIC² are most susceptible to INP perturbations."

[11] Line 166-7: Here is another example where your model assumptions do not fit observations or GCM simulations: not all clouds are dominated by homogeneous freezing.

We are not sure if we understand this comment. Our statement on L166f does not mean that all clouds are dominated by homogeneous freezing. It just means that aircraft soot is not ice-active enough to prevent homogeneous freezing (in the absence of INPs other than aircraft soot). If other INPs were present, aircraft soot would lose much relevance, as most other INPs are more efficient at nucleating ice than aircraft soot (please note L204ff).

[12] 176-178: Are you arguing here that the presence of cirrus with optical depth > 0.3 must mean they are formed mainly from homogeneous freezing? If so, this would be a new finding!

This makes me fear your 1-D model is not really a good representation of cirrus cloud formation!

We certainly do not mean that ‘the presence of cirrus with optical depth > 0.3 must mean they are formed mainly from homogeneous freezing’. This is not what we stated in L176ff. Please note that we have stated that if a larger updraught speed was used in our simulations, the resulting cirrus clouds would be optically thicker because homogeneous freezing is enhanced, not excluding the possible contribution of heterogeneous ice formation to optically thick clouds.

[13] Line 196: add: negligible perturbations in this model.

We add in L196: “... would lead to smaller or negligible perturbations in our simulations.”

[14] Line 207: change to “where the ice activity of aircraft soot, as measured by our parameterization”...

Thank you for spotting this. We rearrange the sentence to read: “... which become ice-active already at low ice supersaturation²⁷ where the ice activity of aircraft soot as predicted by PCF is basically absent (Supplementary Figs.1+2).”

[15] Line 213-215: It is unlikely that other soot / refractory particles contribute efficiently as INP (after all, you quote 5/L as the nominal background number). Moreover, it is known from laboratory measurements that less than 100% of soot particles act as INP. Thus, you cannot conclude that aircraft soot have smaller effects in cirrus than other soot and refractory particles.

We agree that ‘less than 100% of soot particles act as INP’ based on laboratory measurements, which is exactly what soot-PCF predicts. Even large aircraft soot particles nucleate ice well if ice supersaturation is high enough, see Supplementary Fig. 1b.

Even at a number concentration of 5/L, INPs from sources other than aviation may perturb cirrus more efficiently than aircraft soot particles if they nucleate ice at low-to-medium ice supersaturation (< 0.4). This certainly holds true for mineral dust particles (ref. 27) and could be important for widespread biomass burning smoke particles (Schill et al., 2020) or other soot particle populations containing a greater fraction of large (> 100 nm) particles compared to aircraft soot emissions.

[16] Line 2019: the statement “regardless of their size” is not true. The study you quote used a parameterization based on a detailed air parcel model that accounted for the effects of size.

In the global model (ref. 29, Zhou & Penner, 2014), the ice-activity of aircraft soot is due to processing in contrails (termed preactivation in ref. 29) without explicit mentioning a size-dependence. We quote from their section 2.3:

“Emission of aircraft soot is put into the preactivated type when the ambient temperature and relative humidity satisfy the following three criteria:

- 1. the ambient temperature is below a critical temperature, T_c (see below);*
- 2. the ambient relative humidity is higher than a critical relative humidity, RH_c ; and*
- 3. the ambient relative humidity over ice is greater than 100%.”*

Regarding the parcel model-based parameterization (Liu & Penner, 2005, hereafter LP05) mentioned by the reviewer, ref. 29 states:

“The Liu & Penner (2005) parameterization can treat the competition between homogeneous freezing on sulfate haze droplets and heterogeneous nucleation on dust (immersion freezing) as well as other potential heterogeneous IN like soot particles from both biomass burning and fossil fuel.”

*We infer from the above that ref. 29 **did not use** LP05 to calculate ice formation from aircraft soot particles. We are therefore left confused by the reviewer’s remark: ‘The study you quote [ref. 29] used a parameterization based on a detailed air parcel model that accounted for the effects of size.’ This prompts us to delete ‘parcel model-based’ in referring to the parameterization of aircraft soot-induced ice formation in the global model in L243 and correspondingly in L249.*

Let us assume for the sake of argument that ref. 29 did use LP05 to parameterize cirrus formation including aircraft soot. The parcel model described in LP05 assumes that aircraft soot nucleates ice in the immersion freezing mode. We quote from their section 2:

“... for immersion nucleation, we only consider ice nucleation on the surface of soot particles immersed in an H₂SO₄ solution.”

This would mean that the treatment of ice nucleation by aircraft soot and its competition with homogeneous freezing in the global model (ref. 29) is inconsistent with the assumptions made in LP05. Even when a size-dependence is assumed based on immersion freezing, the size-dependence will be much weaker than that observed in measurements and predicted by soot-PCF.

Nonetheless, we address the potential misunderstanding regarding size effects and replace “regardless of size” in L219 by “based solely on contrail formation and persistence criteria”. Moreover, we modify L220f to read: “Without considering the strong size-dependence of the ice nucleation ability of contrail-processed aircraft soot particles, this is in stark contrast ...”.

[17] Line 233-236, You have misquoted ref. 29. As I understand it, you take “ice-active fraction” as the fraction of soot particles forming ice crystals, while reference 29 refers to all particles that may form ice crystals. As a result of the size of the soot particles as well as other conditions (such as the supersaturation reached) only a fraction of these form ice.

*To address this comment, we quote from section 3 of ref. 29: “The annually and zonally averaged number concentrations of the **preactivated** aircraft soot in the UTLS region of the northern hemisphere (NH) is about 0.01 cm^{-3} to 0.1 cm^{-3} .” We see little room for misinterpretation here and do not find a statement in ref. 29 supporting the reviewer’s point: ‘As a result of the size of the soot particles [...] **only a fraction of these form ice**’.*

*In fact, in order to account for a size-dependence of ice nucleation of aircraft soot particles, ref. 29 needed to specify a size distribution for aircraft soot after contrail processing (called ‘preactivated’). Section 2.3 in ref. 29 does acknowledge that a soot PSD was used, but for a different purpose: “We used a lognormal size distribution with a geometric standard deviation of 1.6 and a geometric diameter of 60 nm **to calculate the initial aircraft soot number**.” (Presumably because the soot emissions were originally specified as soot particle mass per kg of fuel burnt and the size information is necessary to convert to soot particle number.)*

Please also see our reply to the related comment [18].

[18] Line 233-235: While you use reference 29 as estimating 100-300 ice-active particles in regions with high soot emission and $> 10 /L$ in the Northern Hemisphere, not all of these particles are “ice-active” as you model here. Considerable fewer are ice active, in the model described in reference 30. (Reference 29 used the parameterization of Liu and Penner, Meteorological Zeitschrift, 2005.) Considerable fewer are ice active, in the model described in reference 30.

Our statement on L233ff is directly taken from ref. 29 without further interpretation. We quote from their section 3: “The simulated preactivated aircraft soot IN number in Figure 2e ranges from 0.1 to 0.3 cm^{-3} in North America and Europe and is larger than 0.01 cm^{-3} everywhere in the middle to high latitudes in the NH.”

This comment, which is related to [17], may stem from a confusion of the parameterization of the parcel model outlined in Liu & Penner (2005) and the one used to quantify ice activity of aircraft soot in the global model described in Zhou & Penner (2014). In our study, we assume that the description given in Zhou & Penner (2014) is correct. We modify L235: “... results in 100–300 L^{-1} soot particles in regions with highest flight activities and $> 10 L^{-1}$ in the Northern Hemisphere at middle and high latitudes participating in cirrus ice formation²⁹.”

[19] Line 238-239: The global model accounts for size-induced effects. This statement is wrong.

Please refer to our responses to the comments [16], [17], and [18]. We have no compelling reason to believe that the above quoted statements taken from ref. 29 are incomplete.

[20] Line 247: The supersaturation for aircraft soot nucleation is not 0.2 but 0.35 in the references. This statement is wrong.

*We checked ref. 31 and found that 0.2 is correct, quoting from their Table 1 summarising the assumptions for aerosols to be effective INPs in the global model: “Pre-activated aircraft soot within contrails with fewer than three monolayers of sulfate **when RH_i reaches 120 %**.”*

[21] Line 249: The parcel model represents following a lagrangian parcel, and thus correctly simulates cirrus formation in a global model. It seems to me the 1-D model is inaccurate.

‘Lagrangian’ refers to the frame of reference, not to the type of model. Lagrangian air parcels can be tracked in Eulerian models.

Please note that contrary to parcel models and global models with coarse spatial resolution, cloud models with fine resolution, like the 1D model employed in our study, is capable of resolving important aspects of aerosol-cloud interactions, as key processes unfold on vertical scales of tens of meters and smaller. For instance, vertical variability in ice supersaturation affects the outcome of even a simple homogeneous freezing event (ref. 20) and numerous studies pointed out the importance of ice crystal sedimentation in simulations of cirrus (e.g., Jensen & Pfister, 2004; Lin et al., 2005; Murphy, 2014). Mainly for these reasons, we used a 1D model framework in our study.

The 1D model employed here subsumes all physical processes important for cirrus ice nucleation that are conventionally included in parcel models. In fact, process-oriented models have begun to simulate Lagrangian cirrus formation and evolution along 1D columns (e.g., Jensen et al., 2004, 2016; Kienast-Sjögren et al., 2015), mostly without compromising the amount and complexity of processes required to predict the outcome of ice nucleation events accurately. We are certainly aware of the differences between different model types, but do not want to enter this discussion in our manuscript.

We add in L.282: “Because processes controlling aerosol-cirrus interactions unfold on vertical scales of tens of meters and smaller, we use a one-dimensional model framework with a vertical grid resolution of 1 m in the computational domain (9.5–12 km).”

[22] Line 291-3: I am not sure what is meant here. Do you treat a column of environmental air with the same updraft, turbulence, and vertical diffusivity, and entrain this air (at what rate?) into the column representing AIC? Please explain more fully or if not, please add an explanation of what is meant by “but without microphysical processes”.

The reviewer is correct that the environmental air is treated with the same dynamics as the cloud column. The entrainment rate is given in L357 and corresponding values are listed in Supplementary Table 1.

We modify in L293: “without ice crystal formation”.

[23] Line 310-313: please explain the phrase: “each containing a number of “real” ice crystals”. Right now it reads as if you have 100,000 simulation ice particles (from liquid aerosol) and these each contain “real” crystals.

The reviewer is correct that each of the 100,000 simulation ice particles (SIPs) deriving from homogeneous freezing contains a number of real ice crystals. This is because representing each individual ice crystal would be computationally prohibitive.

The concept of SIPs was introduced in large-eddy simulations of cirrus (Sölch & Kärcher, 2010). “The Lagrangian particle-based approach is an emerging technique to model cloud microphysics and its coupling with dynamics, offering significant advantages over Eulerian approaches typically used in cloud models” (quoted from Grabowski et al., 2019).

[24] Line 327-8: Please explain what is meant by the “vertical plume extensions”

Depending on the time past emission, the vertical depth of the aircraft / contrail plume increases slowly by turbulent vertical diffusion. Close to the source aircraft (applicable to scenario ‘Plume’), the vertical plume extension is approximately given by the depth of the aircraft wake at the time of original contrail formation (300 m). In scenario ‘Background’, we have increased this width to 400 m using estimates of upper tropospheric vertical diffusivity. In both cases, the vertical plume extension exceeds the thickness of the evolving ice-supersaturated layer and does not notably affect our results.

L328: We add “... extensions based on typical upper tropospheric vertical diffusivities.”

[25] Line 322 and 366: Your liquid aerosol number is 500 cm⁻³ while the aircraft soot is 458 cm⁻³ in the background scenario. Approximately equal concentration of soot and liquid aerosol is not at all what is observed in the background atmosphere. I submit that your study is unable to comment on the conditions treated in GCM simulations.

Thank you for spotting this typo in the revised manuscript. The total aircraft soot number concentration in scenario ‘Background’ is of course 458 L⁻¹, as noted correctly in L93 and Supplementary Table 1.

L366: We correct the typo to read “458 L⁻¹.”

[26] Line 407-409: this statement requires a reference

The laboratory measurements used to develop the soot-PCF model framework (ref. 21) involved samples with a continuous flow diffusion chamber and provided cumulative ice-active fractions. It is generally true that activated or frozen fractions quantify the total number of INPs that form ice at a given ice supersaturation, s . Therefore, there is no primary reference for the statement on L407-409, which simply reflects the nature of the technique to measure ice activity. We are not aware of studies providing differential fractions.

L388f: We add “We apply the soot-PCF framework, developed based on ice nucleation measurements of well-characterized soot particles using a continuous flow diffusion chamber, to particles with properties characteristic for aircraft soot.

L407f: We slightly modify “The fact that AF from Eq.(2) is cumulative in s reflects the nature of laboratory measurements in which the ice nucleation activity of an aerosol sample is often probed at successively increasing supersaturation.”

Literature cited in the response to reviewer #1:

Cirisan, A. et al. Microphysical and radiative changes in cirrus clouds by geoengineering the stratosphere. *J. Geophys. Res.* **118**, doi:10.1002/jgrd.50388 (2013).

Grabowski, W. et al. Modeling of cloud microphysics: Can we do better? *Bull. Amer. Meteorol. Soc.* **100**, doi:10.1175/BAMS-D-18-0005.1 (2019).

Jensen, E. & Pfister, L. Transport and freeze-drying in the tropical tropopause layer. *J. Geophys. Res.* **109**, doi:10.1029/2003JD004022 (2004).

Jensen, E. J. et al. On the susceptibility of cold tropical cirrus to ice nuclei abundance. *J. Atmos. Sci.* **73**, doi:10.1175/JAS-D-15-0274.1 (2016).

Kärcher, B. & Lohmann, U. A Parameterization of cirrus cloud formation: Homogeneous freezing including effects of aerosol size. *J. Geophys. Res.*, **107**, doi:10.1029/2001JD001429 (2002).

Kärcher, B. et al. Aerodynamic contrails: Microphysics and optical properties. *J. Atmos. Sci.* **66**, doi:10.1175/2008JAS2768.1 (2009).

Kay, J. E. & Wood, R. Timescale analysis of aerosol sensitivity during homogeneous freezing and implications for upper tropospheric water vapor budgets. *Geophys. Res. Lett.* **35**, doi:10.1029/2007GL032628 (2008).

Kienast-Sjögren, E., Miltenberger, A. K., Luo, B. P. & Peter, T. Sensitivities of Lagrangian modelling of mid-latitude cirrus clouds to trajectory data quality. *Atmos. Chem. Phys.* **15**, 7,429–7,447 (2015).

Lin, R.-F. et al. Cirrus parcel model comparison project phase 1: The critical components to simulate cirrus initiation explicitly. *J. Atmos. Sci.* **59**, doi:10.1127/0941-2948/2005/0059 (2002).

Lin, R.-F., Starr, D. O., Reichardt, J. & DeMott, P. J. Nucleation in synoptically forced cirrostratus. *J. Geophys. Res.* **110**, doi:10.1029/2004JD005362 (2005).

Murphy, D. M. Rare temperature histories and cirrus ice number density in a parcel and a one-dimensional model. *Atmos. Chem. Phys.* **45**, doi:10.5194/acp-14-13013-2014 (2014).

Schill, G. P. et al. Widespread biomass burning smoke throughout the remote troposphere. *Nat. Geosci.* **13**, doi:10.1038/s41561-020-0586-1 (2020).

Sölch, I. & Kärcher, B. A large-eddy model for cirrus clouds with explicit aerosol and ice microphysics and Lagrangian ice particle tracking. Q. J. Roy. Meteorol. Soc. 136, doi:10.1002/qj.689 (2010).

Reviewer #2

The updated manuscript is a substantial improvement, and I thank the authors for their time and effort. As this is a re-review, I have focused my efforts on the degree to which the authors have addressed my previous comments.

Broadly, I am satisfied that the authors have addressed my comments. I have only one remaining concern; once addressed, I believe this paper will be ready for publication. The arguments regarding ageing of aircraft soot are interesting and logical, but I was concerned at the focus on tropospheric air and the argument on (e.g.) line 450 that a 20 day lifetime would be "long". Roughly one third of all cruise fuel burn occurs in the stratosphere (e.g. Sheng et al 2015), meaning long lifetimes (potentially much greater than 10 days). Yu et al (2019), using a combination of observations and modeling, estimated a lifetime for wildfire black carbon entering the stratosphere at hundreds of days. Kravitz et al (2012), when simulating stratospheric emission of small black carbon particles at ~14 km as a geoengineering strategy, found an average lifetime on the order of three years. This means up to three years of ageing before re-entering the troposphere and potentially taking part in cirrus formation. This region also has high ozone concentrations, and a dry environment. While the authors do argue that (say) H₂SO₄ coatings and coagulation might limit the effect of ozone over longer lifetimes, there does not appear to be quantitative data available to test this hypothesis.

I would therefore recommend inclusion of a caveat (e.g. at line 460) to the effect that, while 60 degrees is a reasonable first estimate, and while the work presented here comprehensively covers situations involving recent (less than one month ago) aircraft emissions, additional work is needed at cruise conditions - including efforts to better constrain the lifetime and properties of aged aircraft soot - to establish the potential contribution of soot which has aged significantly in the stratosphere. Without such a caveat, the assertion in the title still appears excessive.

That having been said, I would again like to state that I find this paper both compelling and highly relevant to the audience of Communications Earth and Environment.

We are grateful to the reviewer for carefully assessing the revised manuscript, providing further comments and endorsing our work. Below we address the last point of concern: the 'focus on tropospheric air and the argument on (e.g.) line 450 that a 20 day lifetime would be "long"'.

Long-distance, high-flying aircraft cruise in air pressure layers between 190–270 hPa; this corresponds to lowermost stratospheric altitudes along intercontinental flight routes only in the extratropics, where tropopause altitudes are low.

We quote from the Kravitz et al. (2012) study: Black carbon aerosols “were placed along the 0° longitude meridian between 10°S and 10°N” and the “injection altitude [...] was varied” (default injection altitude corresponding to air pressures 100–150 hPa). This sulphate aerosol geoengineering scenario is not applicable to aircraft emissions, since no substantial air traffic is happening, indeed no (civil) aircraft can fly safely, below (above) about 150 hPa, while injection altitude and geographical location matter.

The fate of injected material differs between particles injected in the geoengineering scenario and particles emitted by global air traffic. Emissions in the lower stratosphere in the deep tropics, as simulated by Kravitz et al. (2012), are picked up by the Brewer-Dobson circulation, leading to interhemispheric transport and associated long stratospheric residence times. H_2SO_4 condensation on and coagulation with abundant background sulphate aerosols is virtually inevitable in this scenario.

In contrast, aircraft soot particles that are emitted into the lowermost stratosphere outside the tropics are mostly subject to cross-tropopause mixing and can mix back into the troposphere through tropopause folds and cutoffs, leading on average to residence times of a few weeks, albeit with large variability (Holton et al., 1995). We acknowledge that a fraction of air parcels containing aircraft soot particles emitted into the extratropical stratosphere may have residence times exceeding 10–20 days. Even in such cases, there is no compelling reason to assume that contact angle changes due to ozone oxidation will be large, because, as we stated on L451f, “ageing of aircraft-emitted soot particles [via processes (i) and (ii)] already occurs in evolving exhaust plumes²⁶”, which holds true for both tropospheric and stratospheric aircraft emissions. Importantly, contrails form less frequently in the dry lowermost stratosphere, so that soot particles emitted there do not frequently get ice-active to begin with. (Please recall from L56ff that contrail processing is a prerequisite to render aircraft-emitted soot particles ice-active.)

Nonetheless, we appreciate the reviewer’s advice and add a paragraph after L460: “Some fraction of aircraft soot emissions is emitted above the tropopause with residence times that can be longer than the 10–20 days assumed here. Contrails form less frequently in the dry lowermost stratosphere², so that soot particles emitted there do not frequently get ice-active to begin with. The contribution of stratospherically aged soot particles to ice formation via PCF is likely negligible due to condensation of H_2SO_4 and coagulation with particles in aircraft exhaust plumes and from the $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ background aerosol. Additional work is needed to better constrain the lifetime and properties of soot particles depending on the location of emissions. This will ultimately allow to fully establish the ice-active contribution of aged soot particles from aviation to cirrus formation.”

Literature cited in the response to reviewer #2:

*Holton, J. R. et al. Stratosphere-troposphere exchange. Rev. Geophys. **33**, doi:10.1029/95RG02097 (1995).*

*Kravitz, B., Robock, A., Shindell, D. T. & Miller, M. A. Sensitivity of stratospheric geoengineering with black carbon to aerosol size and altitude of injection. J. Geophys. Res. **117**, doi:10.1029/2011JD017341 (2012).*

4th Mar 21

Dear Professor Kärcher,

Your further manuscript titled "Aircraft soot-cirrus interactions do not matter for the climate impact of aviation" has now been seen by the original two reviewers, and I include their comments at the end of this message. As you will see, reviewer 3 now recommends publication, but reviewer 1 still raises much the same concerns as before. As such, we do now feel that a deadlock has been reached, and would like re-iterate the two potential ways forward that we offered in the previous round:

- 1) tone down your manuscript according to reviewer 1's recommendations.
- or
- 2) adjudication, where a fresh reviewer evaluates the disagreement between yourselves and reviewer 1

We leave the decision to you whether you feel you can effectively counter reviewer 1's concerns.

If so, please provide a point-by-point response to the reviewer's latest comments for evaluation by an adjudicator, along with a manuscript that is revised (or not) as you deem necessary, after careful consideration of the remaining points. Thank you for sending suggestions for adjudicators already, we will look into these if you choose this option.

Alternatively, if you follow reviewer 1's recommendations in full, we will be able to accept-in-principle your manuscript.

Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter) and the completed checklist:

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** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within three weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer

than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Heike Langenberg, PhD

Chief Editor
Communications Earth and Environment

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Thanks for authors' response to my comments, but the authors have failed to take account of the fact that they do nothing to evaluate the effects of aircraft soot on large-scale cirrus as treated in GCMs. I add some further required changes to their study. It is particularly important (for the readers understanding) to at least get the abstract correct. It seems this paper is trying to disguise what they have actually done both in the abstract as well as in the discussion. They only deal with a single average updraft velocity. They do not deal with the formation of cirrus crystals in general, as they imply both in the abstract and in the discussion. Part of this may be due to their mis-reading of ref. 29. I try to correct their misunderstanding below.

Line 12-14: This study does not really deal with effects in "natural cirrus". As noted in Figure 1 they deal with "contrail cirrus", which are similar to "spreading contrail cirrus" clouds. This is stated in Figure 1, but appears here in the abstract as if they deal with "natural cirrus" which is totally misleading. At the very least natural cirrus would contain the effects of dust as well as varying amounts of sulfuric acid, organic aerosol, and soot. This manuscript only deals with the emissions behind an aircraft in the presence of a single amount of sulfate, at a single updraft velocity.

I note that this problem appears throughout the manuscript. i.e. in lines 73-75, which refers in

general to soot-cirrus interpretation and impacts on cirrus formation, and probably also appears elsewhere.

Line 18-19: This statement only applies to the conditions you evaluated. Since most conditions are NOT evaluated, you need to add this qualifier. This is absolutely needed in the abstract but also elsewhere where too broad conclusions are made.

Lines 19-21: Applying a model that does not sample the conditions for large-scale cirrus cannot possibly “challenge” these other results. This sentence should never be in an abstract. Perhaps as a speculation in the conclusions, although I would challenge even this, since a small impact on ice concentrations by no means signals a small radiative effect.

Lines 21-23: It is not true that a long-standing knowledge gap has been resolved. You have contributed to an understanding of spreading contrails, which should be stated.

Lines 78-80: add, “associated RF estimates from contrail cirrus”. This is needed to clarify that the model results do not apply to large-scale cirrus clouds in the natural atmosphere.

Line 101: I would again ask that the authors add information on how they estimated the aqueous solution droplet concentration here. This varies widely throughout the atmosphere and is important to determine how generally their study can be interpreted.

Line 128: add “are most susceptible to aircraft INP perturbations” for clarity, so that the reader does not interpret your statement as applying to mixed phase clouds, for example.

Response: [16]: You totally misread the study in ref. 29. In 29, it is correct that there is no size-dependence for treating whether aircraft soot is incorporated into an initial contrail. However, after an initial calculation of whether the soot is incorporated into a contrail, the soot particles in 29 become “preactivated”. Only the preactivated soot may interact with large-scale cirrus clouds. Perhaps your “plume” scenario is the same or similar to being incorporated into an initial contrail (or perhaps this would be even earlier and closer to the aircraft in your calculation— this is unclear to me). In any case, this calculation of whether the soot is inside contrails does not at all alter the radiation balance in the model in ref. 29. It only tells the model that subsequently (after contrail evaporation) these soot particles may act as INP (using the parameterization in LP05, as long as the sulfate deposited on the soot is <3 monolayers). The subsequent quote from 29 which you copy (regarding the use of LP05) does not specifically mention the aircraft soot that have previously participated in contrail formation, as this sentence only mentions some examples.

I note that your note that LP05 calculates ice formation via the use of the immersion mechanism for soot in is correct. However, this parameterization was a convenient way to incorporate soot acting as INP in ref. 29. I again emphasize that the current study cannot be used to comment on forcing in GCM calculations which consider a much different situation than that which is considered in this manuscript.

The addition that you make: “based solely on contrail formation and persistence criteria. Without considering the strong size-dependence of the ice nucleation ability of contrail processed aircraft soot particles” is incorrect. Ref. 29 uses LP05 for calculating ice formation of the preactivated soot.

Response: [17]: Here again you have mis-understood ref. 29. Pre-activated soot are defined in ref. 29 as those have been incorporated in contrail cirrus clouds: The abstract states: “We assume that only the aircraft soot particles that are preactivated in persistent contrail cirrus clouds are efficient IN.”

I quote from section 2 of ref. 29:

These persistent contrails are assumed to be formed “when the ambient temperature and relative humidity satisfy the following three criteria:

1. the ambient temperature is below a critical temperature, T_c (see below);
2. the ambient relative humidity is higher than a critical relative humidity, RH_c ; and
3. the ambient relative humidity over ice is greater than 100%.

The first two criteria are the Schmidt–Appleman criterion for the formation of contrails [Schmidt, 1941;

Appleman, 1953; Schumann, 1996], and the third one is required for persistent contrail cirrus to form.”

Clearly, these are conditions are those needed for contrail formation. The primary topic of ref. 29 and their calculation of radiative forcing is NOT due to contrail formation nor spreading contrails. These authors merely used these criteria (following Chen et al., 2012) to determine that fraction of soot which may further form ice crystals in large-scale cirrus (after the processing within contrails). These criteria are introduced to explain which aircraft soot particles are put into the “preactivation” category. Here even the name of the category suggests that these MAY form crystals but not necessarily. That further depends on whether they develop too much sulfate burden (≥ 3 monolayers) and whether the RH_i reaches 135% and whether the conditions for the LP05 parameterization (which accounts for size) are met.

Response [18]: I reiterate that the “preactivated” soot in ref. 29 are those that have been previously incorporated into contrails. Preactivated soot only affect large scale cirrus in the GCM if they further experience $RH_i > 135\%$ and then only the fraction that fit the LP05 criteria. The preactivated soot in ref. 29 are irrelevant for comparison to this manuscript’s PCF forming crystals in cirrus contrails.

Response [19]: Table 1 in reference 31 has a number of typos (a correction is being submitted). Reference 31 is an update to reference 30, since 31 only added a method to calculate the effects of sulfate in the model. Reference 30 specifies 135% as the critical relative humidity for preactivated aircraft soot to nucleate (see table 1 in Ref. 30).

Response [23]: Please at least add a reference to the concept of “simulation ice crystals”, if you do not want to explain what this is. Otherwise the meaning is unclear and the reader has no way to disentangle what is meant.

Response [24]: Why not just say, “the layer depths are estimated using upper tropospheric vertical diffusivities”. The term “vertical plume extensions” lacks clarity.

Reviewer #3 (Remarks to the Author):

I thank the authors for their detailed rebuttal. The new paragraph which has been added regarding long-lived soot covers my concerns adequately. As such, I have no further concerns, and recommend this paper for publication.

COMMSEV-20-0524B

Responses to reviewer comments (*italicized*)

Reviewer # 1

Thanks for authors' response to my comments, but the authors have failed to take account of the fact that they do nothing to evaluate the effects of aircraft soot on large-scale cirrus as treated in GCMs. I add some further required changes to their study. It is particularly important (for the readers understanding) to at least get the abstract correct. It seems this paper is trying to disguise what they have actually done both in the abstract as well as in the discussion. They only deal with a single average updraft velocity. They do not deal with the formation of cirrus crystals in general, as they imply both in the abstract and in the discussion. Part of this may be due to their mis-reading of ref. 29. I try to correct their misunderstanding below.

We acknowledge the additional information and suggestions. We labelled the comments by uppercase letters to allow cross-referencing. Line numbers refer to the previously revised manuscript.

We are glad that many of our previous responses were adequate and note that the past discussions leave us with two main issues. We disagree with the reviewer that our study disguises important facts and thereby potentially misguides readers: we stated clearly that we use a single updraft speed (L101ff and explaining why in L180ff) and noted the parameters of the sulphate particle size distribution (L327+328) used in our simulations. Please note that this information was already given in the originally submitted version.

The first main open issue concerns the fact that our study does not sample as many meteorological and background aerosol conditions as e.g., a global climate model does. We agree that this is the case. Regarding the presence of INP and variability in sulphate particle properties and updraught speeds, please see our responses to comments [A] and [E].

The second issue concerns the way ice nucleation is treated in, and the interpretation of, the global model study in ref.29 (Zhou & Penner, 2014), specifically the number of aircraft soot particles that participate in cirrus formation. We address this point in our responses to comments [G], [H], and [I].

[A] Line 12-14: This study does not really deal with effects in "natural cirrus". As noted in Figure 1 they deal with "contrail cirrus", which are similar to "spreading contrail cirrus" clouds. This is stated in Figure 1, but appears here in the abstract as if they deal with "natural cirrus" which is totally misleading. At the very least natural cirrus would contain the effects of dust as well as varying amounts of sulfuric acid, organic aerosol, and soot. This manuscript only deals with the emissions behind an aircraft in the presence of a single amount of sulfate, at a single updraft velocity.

I note that this problem appears throughout the manuscript. i.e. in lines 73-75, which refers in general to soot-cirrus interpretation and impacts on cirrus formation, and probably also appears elsewhere.

Please note that we intentionally did not consider more efficient INPs such as dust particles in our simulations in order to estimate the maximum possible effect of aircraft soot particles on cirrus clouds. If such INPs were included, then aircraft soot particles would play no role at all, as they nucleate ice only at the highest achievable ice supersaturations alongside homogeneous freezing (Methods and Supplementary Figs.1+2).

We already replied to the point of varying amounts of fully soluble aerosol particles in our last response and introduced modifications to clarify this point (L328ff). Please recall that the

susceptibility of homogeneously nucleated ICNCs to soluble particle properties is weak for updraught speeds (10-20 cm/s) and temperatures (215-225 K) relevant for the conditions in our simulations as demonstrated by e.g., Kay & Wood (GRL 2008), a reference (37a) we added in the last revision. Please also see our response to comment [E].

We agree with the reviewer that we capture only part of the variability in cirrus properties in homogeneous freezing events by using a single soluble droplet size distribution, but this variability is small and alters ICNCs only moderately. In addition, similar to the case of using a single updraught speed (L101), fixing the size distribution allows us to determine the characteristics of microphysical mechanisms underlying aircraft soot-cirrus interactions. We make the following changes in the revised manuscript:

We expand the sentence in L180 to read: ‘While the use of a single updraught in simulations of cirrus ice formation does not capture the full variability in ICNCs, adding wave-induced vertical wind speeds with temporal variations leads to a massive broadening of ICNC frequency distributions due to multiple homogeneous freezing events²⁵.’

We add in L330: ‘Our simulations do not capture the moderate variability in homogeneously nucleated ICNCs induced by variations in soluble aerosol properties.’

We do not simulate emissions directly behind an aircraft. This assertion probably arises from a misconception regarding (the use of) our cloud model, which we hope to clarify: our column model does not track individual aircraft exhaust plumes or contrails. Instead, we explore the role of contrail-processed soot particle populations in cirrus formation at specific times past dissipation of persistent contrails or contrail cirrus with that model. The associated properties of these soot particles enter our simulations in the form of initial conditions summarised in Supplementary Table 1.

With regard to ‘natural’ cirrus, we decide to remove ‘natural’ from the abstract, since there is no consensus in the community about how a corresponding reference state would look like. For instance, the contribution of various mineral dusts to cirrus formation is highly variable (and may be negligible) and the origin of sulphate aerosol can be anthropogenic and natural.

We have responded to the ‘single plume’ issue already in our previous reply. Please note the addition we introduced in L369ff (Methods) in the last revision. In scenario ‘Background’ 5 h past original contrail formation, it is highly unlikely that soot emissions can still be related to the source aircraft. After several hours, the associated plume area drifted hundreds of kilometres due to strong upper tropospheric winds and the processed aircraft soot emissions diluted and merged with the ambient atmosphere due to turbulent mixing. At this stage, the impact of residual entrainment-mixing on ice microphysics is very small, as the entrainment-mixing timescale, ≈ 5 h, inferred from Supplementary Table 1, is much longer than the microphysical timescales of processes governing the cirrus ice formation event.

We add in L93: “In scenario Background, soot particle number concentrations approximately quantify the aviation contribution to the large-scale atmospheric particle background (see Methods).”

We further add in L376: “In scenario Background, the impact of dilution on ice microphysics is small, as the entrainment-mixing timescale of about 5 h is much longer than the timescales of processes governing the cirrus ice formation event.”

[B1] Line 18-19: This statement only applies to the conditions you evaluated. Since most conditions are NOT evaluated, you need to add this qualifier. This is absolutely needed in the abstract but also elsewhere where too broad conclusions are made.

[B2] Lines 19-21: Applying a model that does not sample the conditions for large-scale cirrus cannot possibly “challenge” these other results. This sentence should never be in an abstract. Perhaps as a speculation in the conclusions, although I would challenge even this, since a small impact on ice concentrations by no means signals a small radiative effect.

We addressed the [B]-comments in detail in our response to [A]. Please recall that our approach intentionally overestimates the impact of aircraft soot (L187ff, L240ff), as previously acknowledged by the reviewer. We now emphasize this more explicitly earlier in the manuscript by replacing “... and provides new insights and guidance to constrain the magnitude of associated RF estimates” in L79+80 with: “We intentionally overestimate factors controlling aircraft soot-cirrus interactions to establish an upper limit impact that may serve to constrain the magnitude of associated RF estimates.”

The reviewer’s notion that a small impact [of soot] on ice concentrations by no means signals a small radiative effect astonishes us, since cirrus radiative flux changes due to ICNC perturbations scale with INP number concentrations. Moreover, please note that both ice crystal numbers and sizes are perturbed in our simulations, leaving optical depth, a proxy for RF (L125ff), nearly unchanged.

[C] Lines 21-23: It is not true that a long-standing knowledge gap has been resolved. You have contributed to an understanding of spreading contrails, which should be stated.

Thank you for pointing this out. We acknowledge that our statement ‘resolving a long-standing knowledge gap’ might be misunderstood as far as RF from aircraft-soot-cirrus interactions is concerned. While we derive, for the first time, ice nucleation properties of contrail-processed aircraft soot particles, we only constrain, not derive, a new best RF estimate, as noted in our answer to comment [B]. Therefore, we omit this statement from the abstract, but use and specify it in L78: “Clarifying the ice nucleation activity of aircraft-emitted soot particles resolves a long-standing knowledge gap and ...”

Please note that our study does not deal with spreading contrails, again as noted above. Rather, we study new cirrus formation after contrail or contrail cirrus dissipation. The simulation scenarios include soot-perturbed cirrus formation either in the volume of an aged aircraft plume (scenario ‘Plume’) or in a highly dispersed plume volume already mixed with ambient air to a large degree (scenario ‘Background’).

[D] Lines 78-80: add, “associated RF estimates from contrail cirrus”. This is needed to clarify that the model results do not apply to large-scale cirrus clouds in the natural atmosphere.

Please note our responses to [A] and [B2]. While we believe that our upper-limit estimate of the impact aircraft soot particles have on cirrus ice formation constrains previous RF estimates, we also suggest in the concluding paragraph that future global climate model simulations (with better constrained parameterizations of the ice nucleation activity of soot) are needed to quantify best global RF estimates, which was not the goal of our study.

[E] Line 101: I would again ask that the authors add information on how they estimated the aqueous solution droplet concentration here. This varies widely throughout the atmosphere and is important to determine how generally their study can be interpreted.

In choosing generic soluble aerosol particle log-normal size distribution parameters (L327f: total number 500 cm^{-3} , modal dry size 20 nm, geometric standard deviation 2.3), we were guided by a cirrus model intercomparison exercise (Lin et al. JAS, 2002), who used 200 cm^{-3} , 20 nm and 1.5. A similar haze particle size distribution with 300 cm^{-3} has been used by Jensen and Toon (GRL, 1997),

cited as ref.10. We increased the total particle number concentration roughly two-fold to represent more polluted conditions and decreased the size spread accordingly to prevent an overabundance of large (> 0.5–1 μm) solution droplets.

Aqueous solution droplet concentrations in the upper troposphere typically lie in the range 200–2,000 cm^{-3} , encompassing large-scale areas where accumulation mode aerosol had time to age by coagulation to localized regions where new particles formed via gas-to-particle conversion as observed by Schröder et al. (JGR, 2002). Changes in droplet number go along with changes in particle sizes. As detailed in our reply to [A], we point out that such variations translate only to small changes in homogeneously nucleated ICNCs, typically 10% resulting from doubling/halving the droplet number. For this reason, the choice of soluble aerosol parameters within reasonable bounds is not crucial for our upper-limit estimates of the soot impact. We quote from L328-330: “The susceptibility of homogeneously nucleated ICNCs to soluble aerosol particle properties is weak for updraught speeds and temperatures relevant for the conditions in our simulations^{37a}.” In addition to ref.37a, Hoyle et al. (JAS, 2005), Kärcher & Ström (ACP, 2003), and Kärcher & Lohmann (JGR, 2002) have studied changes in homogeneously nucleated ICNCs due to soluble aerosol variability, coming to the same conclusion.

We add Hoyle et al. (JAS, 2005) and Kärcher & Lohmann (JGR, 2002) as further references in support of our arguments.

[F] Line 128: add “are most susceptible to aircraft INP perturbations” for clarity, so that the reader does not interpret your statement as applying to mixed phase clouds, for example.

The sentence L127f reads: “Thin cirrus with low optical depth (< 0.3) similar to AIC² are most susceptible to INP perturbations.” There should be no confusion with mixed-phase clouds. We do mean INP perturbations in general, as most INPs from sources other than aviation are better ice nucleating particles than aircraft soot.

[G] Response: [16]: You totally misread the study in ref. 29. In 29, it is correct that there is no size-dependence for treating whether aircraft soot is incorporated into an initial contrail. However, after an initial calculation of whether the soot is incorporated into a contrail, the soot particles in 29 become “preactivated”. Only the preactivated soot may interact with large-scale cirrus clouds. Perhaps your “plume” scenario is the same or similar to being incorporated into an initial contrail (or perhaps this would be even earlier and closer to the aircraft in your calculation— this is unclear to me). In any case, this calculation of whether the soot is inside contrails does not at all alter the radiation balance in the model in ref. 29. It only tells the model that subsequently (after contrail evaporation) these soot particles may act as INP (using the parameterization in LP05, as long as the sulfate deposited on the soot is <3 monolayers). The subsequent quote from 29 which you copy (regarding the use of LP05) does not specifically mention the aircraft soot that have previously participated in contrail formation, as this sentence only mentions some examples.

I note that your note that LP05 calculates ice formation via the use of the immersion mechanism for soot in is correct. However, this parameterization was a convenient way to incorporate soot acting as INP in ref. 29. I again emphasize that the current study cannot be used to comment on forcing in GCM calculations which consider a much different situation than that which is considered in this manuscript.

The addition that you make: “based solely on contrail formation and persistence criteria. Without considering the strong size-dependence of the ice nucleation ability of contrail processed aircraft soot particles” is incorrect. Ref. 29 uses LP05 for calculating ice formation of the preactivated soot.

We thank the reviewer for bringing up the ref.29/LP05-issue again, because this left us somewhat puzzled after the last revision. While we argued based on the global model study in ref.29 that the “preactivated” aircraft soot particles with fewer than three monolayer sulphate coatings all form ice regardless of their size, the reviewer re-emphasises that the LP05 parameterization was used in ref.29 to calculate the fraction of “preactivated” particles that actually form ice based on immersion mode freezing. However, we find no unequivocal indication in ref.29 that LP05 was applied to aircraft soot, let alone details of its implementation and performance.

To move forward, we accept the reviewer’s standpoint and assume that ref.29 did employ LP05 based on immersion freezing of soot particles internally mixed with sulphate, implying that only a fraction of all “preactivated” particles formed ice in the global model study. In this case, as outlined in our previous response, it is inconsistent to use immersion freezing for “preactivated” aircraft soot particles, since ref.29 additionally assumes that these particles have accumulated fewer than three monolayers sulphate coatings while immersion freezing requires soot cores to be immersed in bulk droplets. Furthermore, it is not obvious how, if possible at all, to assess how small the fraction of ice stemming from “preactivated” soot might actually be. Regardless, the size dependence predicted by immersion freezing is much weaker than that observed in measurements and predicted by soot-PCF as detailed in ref.21.

We account for this information in the revision. Please see our responses to [H] and [I] for further clarification and a summary of all novel changes.

[H] Response: [17]: Here again you have mis-understood ref. 29. Pre-activated soot are defined in ref. 29 as those have been incorporated in contrail cirrus clouds: The abstract states: “We assume that only the aircraft soot particles that are preactivated in persistent contrail cirrus clouds are efficient IN.”

I quote from section 2 of ref. 29:

These persistent contrails are assumed to be formed “when the ambient temperature and relative humidity satisfy the following three criteria:

1. the ambient temperature is below a critical temperature, T_c (see below);
2. the ambient relative humidity is higher than a critical relative humidity, RH_c ; and
3. the ambient relative humidity over ice is greater than 100%.

The first two criteria are the Schmidt–Appleman criterion for the formation of contrails [Schmidt, 1941; Appleman, 1953; Schumann, 1996], and the third one is required for persistent contrail cirrus to form.”

Clearly, these are conditions are those needed for contrail formation. The primary topic of ref. 29 and their calculation of radiative forcing is NOT due to contrail formation nor spreading contrails. These authors merely used these criteria (following Chen et al., 2012) to determine that fraction of soot which may further form ice crystals in large-scale cirrus (after the processing within contrails). These criteria are introduced to explain which aircraft soot particles are put into the “preactivation” category. Here even the name of the category suggests that these MAY form crystals but not necessarily. That further depends on whether they develop too much sulfate burden (≥ 3 monolayers) and whether the RH_i reaches 135% and whether the conditions for the LP05 parameterization (which accounts for size) are met.

We fully agree with the reviewer that ref.29 does not deal with contrail formation or spreading contrails. We have never stated this. We also agree that criteria 1-3 are introduced solely to determine the fraction of all soot particles emitted by aircraft that participate in large-scale cirrus formation after being processed in persistent contrails. This is consistent with our previous reply. Therefore, we see no contradiction to our statement in L219-222: “In a global climate model study addressing RF due to soot-perturbed, large-scale cirrus²⁹, soot particles released from long-lived AIC

were assumed to act as ‘good’ INPs already at moderate ice supersaturation, unless they have collected multilayer sulphate coatings, based solely on contrail formation and persistence criteria.”

[I] Response [18]: I reiterate that the “preactivated” soot in ref. 29 are those that have been previously incorporated into contrails. Preactivated soot only affect large scale cirrus in the GCM if they further experience $RH_i > 135\%$ and then only the fraction that fit the LP05 criteria. The preactivated soot in ref. 29 are irrelevant for comparison to this manuscript’s PCF forming crystals in cirrus contrails.

We modify the text in L237ff to read: “Firstly, the size-dependence of ice activity of soot particles processed in long-lived AIC due to soot-PCF is much stronger than predicted by parameterizations based on, e.g., immersion freezing (Liu & Penner, 2005) and leads to lower soot-derived ICNCs. Scenario Background — intentionally overestimating the soot impact — yields a number concentration of ice-active soot particles of 5 L^{-1} , significantly below the values of up to $100\text{--}300 \text{ L}^{-1}$ of contrail-processed aircraft soot particles in regions with highest flight activities estimated by ref. ²⁹.”

Furthermore, we remove L222-225 (“Without considering the strong size-dependence of the ice nucleation ability of contrail-processed aircraft soot particles, this is in stark contrast to our findings that only a small fraction of uncoated soot particles ($< 1\%$) is ice-active at high supersaturation.”) as well as L242-244 (“Correcting for the size-dependence of ice activity of soot particles processed in long-lived AIC would lower soot-derived ICNCs by at least two orders of magnitude and decrease associated RF dramatically.”) and L246+247 (“... for given ice-active soot particle number concentrations, ...”).

We add Liu & Penner (Meteorol. Z., 2005) as a new reference.

We hope that these significant changes made in response to comments [G], [H], and [I] along with our other replies represent a fairer and more accurate discussion of the results of the global model study.

[J] Response [19]: Table 1 in reference 31 has a number of typos (a correction is being submitted). Reference 31 is an update to reference 30, since 31 only added a method to calculate the effects of sulfate in the model. Reference 30 specifies 135% as the critical relative humidity for preactivated aircraft soot to nucleate (see table 1 in Ref. 30).

We believe the reviewer refers to our response to the previous comment [20]. We are grateful for pointing this out. There is no way we could have possibly known that. We modify L250 to read “... processed soot particles, either bare or with minimal coatings, were assumed to act as INPs already at moderate ice supersaturation (0.35 , see ref.30).” We note, however, that this is still at odds with the ice nucleation behavior of real-world contrail-processed aircraft soot particles that only nucleate ice at ice supersaturation near 0.5 , as clearly demonstrated in our study.

[K] Response [23]: Please at least add a reference to the concept of “simulation ice crystals”, if you do not want to explain what this is. Otherwise the meaning is unclear and the reader has no way to disentangle what is meant.

We add Sölch & Kärcher (QJRM, 2010) in L318 as a new reference.

[L] Response [24]: Why not just say, “the layer depths are estimated using upper tropospheric vertical diffusivities”. The term “vertical plume extensions” lacks clarity.

We modify L335 as suggested by the reviewer.

Literature cited:

Hoyle, C. R., Luo, B. P. & Peter, T. The origin of high ice crystal number densities in cirrus clouds. *J. Atmos. Sci.* **62**, doi:10.1175/JAS3487.1 (2005).

Kärcher, B. & Lohmann, U. A parameterization of cirrus cloud formation: Homogeneous freezing of supercooled aerosols. *J. Geophys. Res.* **107**, doi:10.1029/2001JD000470 (2002).

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Kay, J. E. & Wood, R. Timescale analysis of aerosol sensitivity during homogeneous freezing and implications for upper tropospheric water vapor budgets. *Geophys. Res. Lett.* **35**, doi:10.1029/2007GL032628 (2008).

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Lin, R.-F. et al. Cirrus parcel model comparison project phase 1: The critical components to simulate cirrus initiation explicitly. *J. Atmos. Sci.* **59**, doi:10.1127/0941-2948/2005/0059 (2002).

Liu, X. & Penner, J. E. Ice nucleation parameterization for global models. *Meteorol. Z.* **14**, doi:10.1127/0941-2948/2005/0059 (2005).

Schröder, F., Kärcher, B., Fiebig, M. & Petzold, A. Aerosol states in the free troposphere at northern midlatitudes. *J. Geophys. Res.* **107**, doi:10.1029/2000JD000194 (2002).

Sölch, I. & Kärcher, B. A large-eddy model for cirrus clouds with explicit aerosol and ice microphysics and Lagrangian ice particle tracking. *Q. J. Roy. Meteorol. Soc.* **136**, doi:10.1002/qj.689 (2010).

Reviewer # 3

I thank the authors for their detailed rebuttal. The new paragraph which has been added regarding long-lived soot covers my concerns adequately. As such, I have no further concerns, and recommend this paper for publication.

We are grateful to the reviewer for the positive and very constructive evaluation that greatly helped to improve our manuscript with regard to the issue of soot ageing and thereby strengthen its conclusions.

20th Apr 21

Dear Professor Kärcher,

Your manuscript titled "Aircraft soot-cirrus interactions do not matter for the climate impact of aviation" has now been seen by our adjudicating reviewer, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewer. Please carefully address each point of the adjudicating reviewer, and document this in point-by-point responses to the comments. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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Best regards,

Heike Langenberg, PhD

Chief Editor
Communications Earth and Environment

On Twitter: @CommsEarth

REVIEWERS' COMMENTS:

Reviewer #4 (Remarks to the Author):

For clarity, my review is given on the basis of it being an adjudication review, which I understand is known by the authors.

GENERAL POINTS

There is clearly great value in the submitted manuscript since the topic of whether aviation contributes a large forcing or not, in terms of 'soot-cirrus' has been discussed for some years with little satisfactory outcome (as yet), since the estimates from one modelling group have ranged from large positive forcings to large negative forcings [m/s refs 29, 30, 33a. Two other groups have attempted to model the effect but concluded the forcings was not greatly different from zero (net) [m/s refs 32, 33]. A recent assessment and analysis [m/s ref 3] usefully pulled these differing results together (which included all of the aforementioned global modeling groups involved, so presumably had their agreement), which concluded that no best estimate of the forcing could be given, and it remained an open topic. There was a hint, however, rather tucked away in the relevant figure legend of ms ref 3 (its Fig 5) that ref 29's largely varying results had been superseded by ref 30, which at least started to constrain the sign of the forcing, but the variance within ref 30's results was still large, and even the smallest forcing was 'large' relative to other aviation forcings.

So, against this background, it seems that further study of this topic is extremely useful. However, a previous reviewer has raised serious concerns about the content of the submitted m/s, and the way in which it is presented.

I think that some of the 'heat' can be removed from this argument by simply modifying the title of the article to something less provocative and addressing some of the wording issues in the abstract.

I am not sure that the present m/s goes as far as settling the subject but rather is a useful contribution that addresses the problem from a different angle. In terms of a title, perhaps something along the lines of "Constraining the aircraft soot-cirrus climate impact of aviation"? This seems to be what the article does, and I am not convinced that the title can be concluded from the content, since the authors make recommendations as to how things may be better represented in global models. I don't think that the authors would disagree that global models are still the appropriate tools with which an assessment of the forcing should be made? The global models need improving to increase the confidence of the results, and process-based models, such as that in the present m/s, can provide useful insights. Titles are usually agreed between editors and authors in Nature Communication papers so a better one may be formulated but I do recommend changing it.

One of the other contentious parts of the abstract is the sentence (L19-31), "Our results challenge previous large estimates of global radiative forcing from interactions between aircraft soot and large-scale cirrus". Here I have some sympathy with reviewer 1, as it is difficult to argue that the authors' approach is a direct 'challenge' to global model results, since, as reviewer 1 argues, the authors do not sample all conditions in a way that a global model attempts to do, so is not an apples-with-apples comparison, which is being interpreted as a "challenge". Once, more, a linguistic softening would help this as clearly a legitimate point is being made that the authors' results are apparently inconsistent with the "previous large estimates". Perhaps something along the lines of "Our results show differences from those of global models, which imply that the large estimates of global radiative forcing from interactions between aircraft soot and large-scale cirrus may be overestimates." It could maybe be better formulated by the authors but hopefully, they get the point.

Similarly, I would ask the authors to reconsider the use of the word "advanced" in the abstract (L21). This could be interpreted either as a verb or an adjective, and the ambiguity makes it sounds somewhat prejudicial. "Improved" may be more neutral.

REFLECTIONS ON THE EXCHANGE BETWEEN REVIEWER 1 AND THE AUTHORS ('ADJUDICATION')

Points starting from reviewer "Thanks for the authors' response to my comments..."

There is a disagreement over representation of what was done in the abstract. I do not believe the authors are trying to "disguise" what they have done as suggested by Reviewer 1. However, it could be better represented. I think that this can be remedied by removal of the word "unravel". I understand the motivation for the word (short, evocative), however, a somewhat drier description would alleviate some of the concerns of Reviewer 1, if it were phrased along the lines of "A high resolution cirrus column model was used to investigate how aircraft-emitted soot particles..." This is, after all, what the authors have done, and makes it clearer that they are approaching the problem from a different angle.

[A] Line 12 - 14

The reviewer objects that the authors do not deal with natural cirrus but only aircraft-modified cirrus. My opinion is that the authors have made extensive modifications that satisfactorily clarifies what they did.

[B] Line 18 - 19", "[B] Lines 19- 21

The reviewer objects to the representation of limited conditions being modeled and the use of the word "challenge" in the abstract.

I have suggested a solution to the "challenge" problem above in 'GENERAL POINTS'. I also think that the deletion of the word "unravel" with some more descriptive words of what the authors did, would solve most of the problems, alongside the authors' own modifications

[C] Line 21 - 23

It appears that "long-standing knowledge gap" has been removed from the abstract, as requested by Reviewer 1, but persists at L79 which seems a fair and qualified use of the statement.

[D] Lines 78-80

The response from the authors seems satisfactory and they clarify the misunderstanding on the part of Reviewer 1 that they are not dealing specifically with contrail cirrus but rather new cirrus formation after dissipation.

[E] Line 101

The authors have provided an extensive response, which seems satisfactory in terms of what is being asked by Reviewer 1 – it is not clear that this is incorporated into the m/s so I would ask for the authors to ensure that it is, somewhere, even if in the SI.

[F] Line 128

The authors' response seems satisfactory that there should be no confusion with mixed-phase clouds (Reviewer 1's concern), as phrased.

[G] Response: [16]

Here there is a lengthy discussion over what was and was not done in m/s ref [29]. The authors are happy to take Reviewer 1's word over what was actually done, although they say they cannot find this documented. The authors digress onto a disagreement with Reviewer 1 over plausibility of physical mechanisms which seem to be impossible/out of scope to resolve within the confines of the submitted paper. Nonetheless, they refer forwards to responses [H] and [I], where they clarify what they have done in the present study. This seems as good an answer as is possible to this difficult discussion as I don't believe the present paper is the right place for a discussion of what was or was not done in m/s ref [29].

[H] Response: [17]

Reviewer 1 claims that the authors have misunderstood and misrepresented what was done in m/s ref [29]. From the authors' response in terms of their quote of what they say, I don't see this as a misunderstanding or misrepresentation.

[I] Response [18]

Again, Reviewer 1 takes issue with what had been previously written by the authors in their representation of what was done in m/s ref [29]. The authors have made additions and deletions that seem to answer Reviewer 1's concerns.

[J] Response [19]

Clearly, the authors could have not known of a correction in process, but they have responded to this additional information.

[K] Response [23]

Reference added as requested by Reviewer 1, so a satisfactory response.

[L] Response [24]

Authors have modified the text as suggested by Reviewer 1

MY OWN ADDITIONAL REVIEW COMMENTS

MAJOR

L49 – 55 show that the authors are acquainted with the details of m/s ref [3]. However, they may have overlooked some aspects of the relevant figure legend and/or the associated text. It is somewhat of a misrepresentation (although perhaps understandable) to place the results of m/s ref [29] alongside those of m/s ref [30], since the figure legend of m/s ref [3], in part, reads "As part of the present study, an author (JEP) re-evaluated these earlier studies and concluded that the Penner et al. (2018) results supersede the earlier Penner et al. (2009) and Zhou and Penner (2014) results because of assumptions regarding updraft velocities during cloud formation". Admittedly, this is 'tucked away information' but, correcting this or rephrasing L49 – L55 will acknowledge and alleviate some of the concerns of reviewer 1, I believe.

L279 – 284 There could be an element of 'overclaiming' here in these final three sentences. "Very likely" seems to be an overclaim, and I would prefer to see "possible" or at the least "likely" if the authors insist. I think a very short and simple reiteration at the end of the first sentence would be appropriate i.e. "...because...". Similarly, the claim "...provides the constraints..." seems very definitive? How do the authors know? There may be other constraints/factors at play, surely? Perhaps "...provides one of the major constraints.." would be more appropriate, and then link this to the final sentence. Lastly, "...which precludes the largest RF estimates..." again, this seems very definitive and needs softening in some way. I don't believe that any of these suggested

modifications take away the importance of this article. After all, the authors come at the problem from a different angle and that angle provides useful input/guidance to global modeling efforts rather than definitively solving the problem.

MINOR

Sentence ending L36 "...proves difficult to establish" – consider maybe "...proves difficult to establish with confidence."

L38 "...magnitudes that exceed..." it would be more accurate to write "...magnitudes that can exceed..."

L40-L41 The substantiation itself would not substantially alter "the role of aviation in the climate system" but rather the quantification, and/or perception.

CONCLUSION OF ADJUDICATION REVIEW

I believe that the article should be published but that the authors should respond to requests to change the title of the article, change some of the wording of the abstract (suggestions provided). Largely, I believe that the authors have provided a satisfactory response to the comments from Reviewer 1, with the aforementioned caveats on the title and abstract.

Responses to comments (*italicized*) of reviewer #4

For clarity, my review is given on the basis of it being an adjudication review, which I understand is known by the authors.

We thank the adjudicating reviewer for their advice. We are grateful for this detailed and insightful analysis and appreciate the comments on both, the manuscript and the previous exchange with reviewer #1. Below we respond to all comments.

GENERAL POINTS

There is clearly great value in the submitted manuscript since the topic of whether aviation contributes a large forcing or not, in terms of ‘soot-cirrus’ has been discussed for some years with little satisfactory outcome (as yet), since the estimates from one modelling group have ranged from large positive forcings to large negative forcings [m/s refs 29, 30, 33a. Two other groups have attempted to model the effect but concluded the forcings was not greatly different from zero (net) [m/s refs 32, 33]. A recent assessment and analysis [m/s ref 3] usefully pulled these differing results together (which included all of the aforementioned global modeling groups involved, so presumably had their agreement), which concluded that no best estimate of the forcing could be given, and it remained an open topic. There was a hint, however, rather tucked away in the relevant figure legend of ms ref 3 (its Fig 5) that ref 29’s largely varying results had been superseded by ref 30, which at least started to constrain the sign of the forcing, but the variance within ref 30’s results was still large, and even the smallest forcing was ‘large’ relative to other aviation forcings.

So, against this background, it seems that further study of this topic is extremely useful. However, a previous reviewer has raised serious concerns about the content of the submitted m/s, and the way in which it is presented.

I think that some of the ‘heat’ can be removed from this argument by simply modifying the title of the article to something less provocative and addressing some of the wording issues in the abstract.

I am not sure that the present m/s goes as far as settling the subject but rather is a useful contribution that addresses the problem from a different angle. In terms of a title, perhaps something along the lines of “Constraining the aircraft soot-cirrus climate impact of aviation”? This seems to be what the article does, and I am not convinced that the title can be concluded from the content, since the authors make recommendations as to how things may be better represented in global models. I don’t think that the authors would disagree that global models are still the appropriate tools with which an assessment of the forcing should be made? The global models need improving to increase the confidence of the results, and process-based models, such as that in the present m/s, can provide useful insights. Titles are usually agreed between editors and authors in Nature Communication papers so a better one may be formulated but I do recommend changing it.

We agree with the reviewer that improved global models are still the tool of choice to quantify the global RF of aerosol-cloud interactions, as expressed in our concluding paragraph (L270ff), and that process-based analyses using detailed cloud models such as the one employed in our study help guide such improvements. In addition, we believe that targeted field observations and laboratory studies of the ice activity of Aitken mode soot particles are needed (L110ff and L256ff).

As a new title, we choose: “Process-oriented analysis of aircraft soot-cirrus interactions constrains the climate impact of aviation”. We give three reasons in support of this alternative: it (i) better reflects what we have done; (ii) accounts the fact that any bound on thin cirrus optical depth constrains RF and ultimately the climate impact (via effective RF); (iii) hints at the different angle from which we addressed the problem (process-based vs global model).

One of the other contentious parts of the abstract is the sentence (L19-31), “Our results challenge previous large estimates of global radiative forcing from interactions between aircraft soot and large-scale cirrus”. Here I have some sympathy with reviewer 1, as it is difficult to argue that the authors’ approach is a direct ‘challenge’ to global model results, since, as reviewer 1 argues, the authors do not sample all conditions in a way that a global model attempts to do, so is not an apples-with-apples comparison, which is being interpreted as a “challenge”. Once, more, a linguistic softening would help this as clearly a legitimate point is being made that the authors’ results are apparently inconsistent with the “previous large estimates”. Perhaps something along the lines of “Our results show differences from those of global models, which imply that the large estimates of global radiative forcing from interactions between aircraft soot and large-scale cirrus may be overestimates.” It could maybe be better formulated by the authors but hopefully, they get the point.

We agree that linguistic adjustment brings the two views together. We modify L19-21 to read: “Our results imply that climate model estimates of global radiative forcing from interactions between aircraft soot and large-scale cirrus may be overestimates.”

Similarly, I would ask the authors to reconsider the use of the word “advanced” in the abstract (L21). This could be interpreted either as a verb or an adjective, and the ambiguity makes it sounds somewhat prejudicial. “Improved” may be more neutral.

Thank you for this clarification. We replace ‘advanced’ by ‘improved’ in L21.

REFLECTIONS ON THE EXCHANGE BETWEEN REVIEWER 1 AND THE AUTHORS (‘ADJUDICATION’)

Points starting from reviewer “Thanks for the authors’ response to my comments...”

There is a disagreement over representation of what was done in the abstract. I do not believe the authors are trying to “disguise” what they have done as suggested by Reviewer 1. However, it could be better represented. I think that this can be remedied by removal of the word “unravel”. I understand the motivation for the word (short, evocative), however, a somewhat drier description would alleviate some of the concerns of Reviewer 1, if it were phrased along the lines of “A high resolution cirrus column model was used to investigate how aircraft-emitted soot particles...” This is, after all, what the authors have done, and makes it clearer that they are approaching the problem from a different angle.

Thank you for this suggestion. We replace “Here we unravel ...” by “Here we use a high-resolution cirrus column model to investigate ...” in L12, and skip ‘high resolution’ in L15.

[A] Line 12 - 14

The reviewer objects that the authors do not deal with natural cirrus but only aircraft-modified cirrus. My opinion is that the authors have made extensive modifications that satisfactorily clarifies what they did.

Thank you. No further changes made to the manuscript.

[B] Line 18 - 19", "[B] Lines 19- 21

The reviewer objects to the representation of limited conditions being modeled and the use of the word "challenge" in the abstract.

I have suggested a solution to the "challenge" problem above in 'GENERAL POINTS'. I also think that the deletion of the word "unravel" with some more descriptive words of what the authors did, would solve most of the problems, alongside the authors' own modifications.

Done as suggested and noted above.

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It appears that "long-standing knowledge gap" has been removed from the abstract, as requested by Reviewer 1, but persists at L79 which seems a fair and qualified use of the statement.

Thank you. No further changes made to the manuscript.

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The authors have provided an extensive response, which seems satisfactory in terms of what is being asked by Reviewer 1 – it is not clear that this is incorporated into the m/s so I would ask for the authors to ensure that it is, somewhere, even if in the SI.

Previous changes in our manuscript related to this issue include: "The susceptibility of homogeneously nucleated ICNCs to soluble aerosol particle properties is weak for updraught speeds and temperatures relevant for the conditions in our simulations^{37b,37c,37d}." (L330ff). "Our simulations do not capture the moderate variability in homogeneously nucleated ICNCs induced by variations in soluble aerosol properties." (L335ff). We enhance clarity by combining these sentences (and reversing the order of appearance), as they appear somewhat disconnected in the earlier manuscript version.

In addition, we add the following along with a new reference.

In L327: "Midlatitude upper tropospheric aqueous solution droplet concentrations typically lie in the range 200–2,000 cm⁻³, encompassing large-scale areas where accumulation mode aerosol age by coagulation and localised regions where new particles form via gas-to-particle conversion^{37a}."

37a. Schröder, F., Kärcher, B., Fiebig, M. & Petzold, A. Aerosol states in the free troposphere at northern midlatitudes. *J. Geophys. Res.* 107, doi:10.1029/2000JD000194 (2002).

[F] Line 128

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Thank you. No further changes made to the manuscript.

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Thank you. No further changes made to the manuscript.

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Thank you. No further changes made to the manuscript.

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Thank you. No further changes made to the manuscript.

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Clearly, the authors could have not known of a correction in process, but they have responded to this additional information.

Thank you. No further changes made to the manuscript.

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Reference added as requested by Reviewer 1, so a satisfactory response.

Thank you. No further changes made to the manuscript.

[L] Response [24]

Authors have modified the text as suggested by Reviewer 1

MY OWN ADDITIONAL REVIEW COMMENTS

MAJOR

L49 – 55 show that the authors are acquainted with the details of m/s ref [3]. However, they may have overlooked some aspects of the relevant figure legend and/or the associated text. It is somewhat of a misrepresentation (although perhaps understandable) to place the results of m/s ref [29] alongside those of m/s ref [30], since the figure legend of m/s ref [3], in part, reads “As part of the present study, an author (JEP) re-evaluated these earlier studies and concluded that the Penner et al. (2018) results supersede the earlier Penner et al. (2009) and Zhou and Penner (2014) results because of assumptions regarding updraft velocities during cloud formation”. Admittedly, this is ‘tucked away information’ but, correcting this or rephrasing L49 – L55 will acknowledge and alleviate some of the concerns of reviewer 1, I believe.

Thank you for clarifying this issue.

We modify L49ff as follows: “Global climate model studies have led to a wide range of RF estimates for aircraft soot-cirrus interactions, currently from approximately –330 to 287 mW m⁻² normalised to 2018 air traffic³. This range persists despite some older estimates being superseded³. Some models assume that aircraft-emitted soot particles frozen in contrails act as good INPs in large-scale cirrus formation, while others, ...”

In this way, we keep the Introduction concise and come back full circle to RF in the Discussion, where we quote individual studies. At the same time, we avoid citing the largest negative RF estimate from ref. 29, because it has been superseded. Instead, it is replaced by the negative RF estimate from the newer study (ref. 30), consistent with the caption of Fig.5 in ref. 3.

279 – 284 There could be an element of ‘overclaiming’ here in these final three sentences. “Very likely” seems to be an overclaim, and I would prefer to see “possible” or at the least “likely” if the authors insist. I think a very short and simple reiteration at the end of the first sentence would be appropriate i.e. “...because...”. Similarly, the claim “...provides the constraints...” seems very definitive? How do the authors know? There may be other constraints/factors at play, surely? Perhaps “...provides one of the major constraints..” would be more appropriate, and then link this to the final sentence. Lastly, “...which precludes the largest RF estimates...” again, this seems very definitive and needs softening in some way. I don’t believe that any of these suggested modifications take away the importance of this article. After all, the authors come at the problem from a different angle and that angle provides useful input/guidance to global modeling efforts rather than definitively solving the problem.

We make four adjustments to prevent ‘overclaiming’ in the concluding paragraph.

We replace ‘very likely’ by ‘likely’ in L279.

We replace ‘provides the constraints’ by ‘provides a major constraint’ in L282.

We add in L280: “..., because our process-oriented analysis of soot-cirrus interactions using representative upper tropospheric conditions does not reveal a significant impact on cirrus optical depth despite changes in cloud structure.”

We modify L283f to read: “... in the short-term: at most one out of hundred emitted soot particles is ice-active after processing in persistent contrails or contrail cirrus.”

MINOR

Sentence ending L36 “...proves difficult to establish” – consider maybe “...proves difficult to establish with confidence.”

Done.

L38 “...magnitudes that exceed...” it would be more accurate to write “...magnitudes that can exceed...”

Done.

L40-L41 The substantiation itself would not substantially alter “the role of aviation in the climate system” but rather the quantification, and/or perception.

We modify L41 to read: “... the perception of the role of aviation in the climate system would be substantially altered.”

CONCLUSION OF ADJUDICATION REVIEW

I believe that the article should be published but that the authors should respond to requests to change the title of the article, change some of the wording of the abstract (suggestions provided). Largely, I believe that the authors have provided a satisfactory response to the comments from Reviewer 1, with the aforementioned caveats on the title and abstract.

Thank you.