Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2022-529-RC1, 2022

Comment on acp-2022-529

Blaž Gasparini (Referee)

Referee comment on "A global climatology of ice-nucleating particles under cirrus conditions derived from model simulations with MADE3 in EMAC" by Christof G. Beer et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-529-RC1, 2022

The manuscript by Beer et al., 2022 presents simulated ice nucleating particles and their effects on cirrus cloud properties in a global chemistry-climate model. As a difference from other similar studies, they include two ice nucleating particle types that are rarely included in model simulations: crystalline ammonium sulfate and glassy organic particles. They find that ammonium sulfate plays a large role in nucleating ice at cirrus conditions, of larger or at least comparable magnitude compared to dust aerosols.

This is a robust scientific study describing the results of careful extensions to the model freezing scheme. However, after reading it through I think the paper reads too much as a report on the (otherwise excellent) work, missing a sharper line of thought including more discussion on the main novel outcomes of this work and sharper conclusions. Because of this, I was initially not certain what is the key novel result of the manuscript.

I would therefore suggest bringing the large importance of crystalline ammonium sulfate for cirrus freezing (which is in my opinion the main novel result of this work) more to the forefront of the manuscript. Indeed, this may be a somewhat controversial conclusion, that demands more discussion and (if possible/available) more evidence beyond your modeling work.

Secondly, the manuscript falls short of pointing at the radiative importance of the separate ice nucleating particle species. I think the manuscript would significantly benefit if the authors could add the analysis of the radiative relevance of implemented INP species for the top-of-the-atmosphere radiative budget.

The manuscript will be suitable for publication in final form, once the above-mentioned and other comments are addressed.

Additional general comments

- Is there a way to find observational evidence of ammonium sulfate playing such a large role (outside of lab studies)? This would go against the generally accepted notion of dust as the key ice nucleating particle at cirrus levels (e.g. Cziczo et al., 2013, Froyd et al., 2022, and many more).
- What is the climatic role of different ice nucleating modes? Is the addition of ammonium sulfate as separate ice nucleating species as important also from the radiative perspective?
- Could you comment on the uncertainties in ammonium sulfate ice nucleating properties, e.g. in its onset ice nucleation temperature, critical supersaturation, etc? As an example, a recent study by Bertozzi et al., 2021 found the onset of ammonium sulfate freezing at -54°C at a critical supersaturation of 1.3. Such assumptions would probably significantly reduce the role of ammonium sulfate as ice nucleating particles compared to the used assumptions.
- Detrainment of ice is one of the key sources of upper tropospheric clouds and likely accounts for most of the upper tropospheric clouds in the tropics below about 14 km and a large fraction of the extratropical summertime high clouds. How does detrainment affect in-situ cirrus and their ice nucleation? How are the ice number and mass sources of detrainment treated, and does detrainment interact with in-situ ice nucleation?

Specific comments

Page 1, line 2: the word climate modifications alludes to artificial geoengineering-type of modifications. You could consider using a synonym.

Page 1: The last sentence of the abstract describes (in my opinion) the main result of the manuscript. The reader should ideally be informed of the key result earlier than at the end of a relatively long abstract.

Page 7: It seems like you have put a lot of effort into numerically representing and due to the need for 3 additional tracers also computing ammonium sulfate aerosols. Is there a simpler way of simulating crystalline ammonium sulfate that the other modeling groups may be more likely to implement in their models?

Figure 1, panel (a): Surface or 300 hPa level?

Figure 5: panel (c) should use the same colorbar limits as all other panels, not to artificially overemphasize the role of aircraft soot.

Page 19, lines 414-117: The two sentences seem to be contradicting each other (the first one claiming the occurrence frequencies of heterogeneous freezing are largely preventing homogeneous freezing, the second one saying that heterogeneous INPs do not completely suppress homogeneous freezing)

Page 19, lines 429 and further: I agree INP measurements at cirrus conditions are practically non-existent, but you may still want to comment on the surprising result that ammonium sulfate is found to be the key INP species at cirrus conditions, contrary to the mainstream cirrus literature.

Page 21, lines 477-484: You don't comment at all about the surprising result of the large importance of ammonium sulfate! From this paragraph, it sounds like dust is clearly the main INP, which is different from your model results.

Page 22, lines 497-500: I would suggest modifying for clarity the italicized part of the sentence: "….this study demonstrates the importance of including *additional ice nucleating particle types* together with…. " to ammonium sulfate
And then just conclude with "Glassy organic particles probably have only a minor influence…"

References

Bertozzi et al., 2021, doi: 10.5194/acp-21-10779-2021

Cziczo et al., 2013, doi: 10.1126/science.1234145

Froyd et al., 2022, doi: 10.1038/s41561-022-00901-w