

Comment on acp-2022-95

Anonymous Referee #1

Referee comment on "Lessons from airborne VOC measurements during PAMARCMIP 2018 concerning aged biogenic emissions and ozone depletion events" by Rupert Holzinger et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-95-RC1>, 2022

Review of manuscript untitled « Possible controls on Arctic clouds by natural aerosols from long-range transport of biogenic emissions and ozone depletion events »

General comment

This study investigates the concentrations of various compounds detected from a PTR-MS onboard the POLAR 5 aircraft as part of the PAMARCMiP 2018 campaign in the Arctic. Using backtrajectories the authors link the sampled air masses to continental influences with aged biogenic emissions and anthropogenic pollution. Even though the association between the signature of oxidation products and the origins of the air masses seems to be rather convincing, the authors add a series of assumptions from which they conclude possible controls of aerosols (which are not sampled here) on clouds and longwave radiation (which are never measured, nor discussed here). Those latter results are presented in a pure qualitative style without fully investigating the underlying physical processes. The paper therefore fails to achieve the objectives given in the title, abstract and conclusions. I could go with reject, but I think some of the results are of sufficient originality and interest to merit publication. I thus encourage the authors to revise the manuscript with a more succinct discussion that focuses only on **their** observations and some supporting analyses.

Major comments

The title is totally misleading. The objective of the paper is not to evaluate the potential influence of biogenic emissions on clouds. CCN and INPs are not analyzed in this study, their role on the formation of clouds is never discussed and cannot be since no observations of clouds are mentioned in this study. Are there lidar/radar observations that support cloud formation/modification under the influence of natural aerosols ? I suggest to reformulate the title focusing on what is actually done here and of sufficient interest : evidence of the signature of biogenic compounds and their oxidation products in the Arctic atmosphere. The authors may also want to mention ODEs that have indeed been detected in their observations.

Abstract : L 38-47 : This section is rather speculative and should be removed. It is a rather a sequence of hypotheses (influence on clouds, link between ODEs and INPs) that are not supported by the analyses performed here. They shouldn't appear as key results in the abstract and conclusions. In addition, the effect of ice crystals (that could be linked to INPs, but also to secondary ice production) is much lower than that of supercooled droplets in the longwave spectrum.

L 43 : "evidence that ODEs may be". Is it a conclusion or an assumption ?

L 318-326 : This section is written in a qualitative style without any analysis to support this discussion. In addition, biogenic SOA are presented as a "major factor controlling aerosol/cloud/radiation interaction in the Arctic winter" without mentioning which physical process is concerned. Do the authors want to specify their role as CCN versus their role as INP ? Do they consider their role on SW and LW radiation ? Which minimal optical properties are required to distinguish the effects ?

Summary/conclusions : Some parts of the conclusions are not justified in the study. A large fraction of the summary (L 334-339) is taken from results from other studies, not from this study itself. None of biological INP or bromine species are measured/analysed here, but the authors nevertheless give conclusions relative to their co-occurrence with

ODEs.

The quality of the figures is very poor :

- labels are difficult to read on Fig. 2. Use larger/bold font to make this clearer.
- Fig 3, 4, 5 : It is extremely difficult (or impossible) to read as the scales for the different components are not plotted. The reader has to convert himself/herself the units to understand the variability of the different species. This is not acceptable in a published paper. Please add values on the y axis or add different sub-figures if there are too many curves. In any case, the reader should be able to read easily the values of **all** the different species that are plotted.
- Fig 6 : This figure only give mean and median values for the different components. Boxplots including the different quartiles would provide more useful information about the variability of the mixing ratios (not concentrations, as written) of the various components for each bin of the acetone mixing ratios.

Specific comments

L 50-58 : This is a strange way to start a manuscript with no reference at all. A classical introduction starts by a state-of-the-art of the scientific litterature. Please add some useful references that support the discussion.

L 58 : "anthropogenic pollution became the dominant control". Is there some evidence for that ? All INPs that are active in Arctic mixed-phase clouds are known to be of natural origin (mostly dust or biologic macromolecules) : see for examples the recent papers of Wex et al. (2019, <https://doi.org/10.5194/acp-19-5293-2019>), Kanji et al. (2020, <https://doi.org/10.1029/2019GL086764>), Welti et al. (2020, <https://doi.org/10.5194/acp-20-15191-2020>). Do the authors rather want to underline the influence of anthropogenic aerosols on liquid clouds here ? This is not clear.

L 62-76 : The two studies quoted here (Nielsen et al. , 2019 and Pernov et al., 2021) report measured at the surface. What are the connections those observations and layers containing aerosol and trace gases aloft that are sampled by an aircraft ? Are there vertical profiles available ?

L 79 : What are the scientific questions of this study ? What is the outline of the paper ?

L 152 : Which ECMWF data are precisely used in the study ? Are they reanalyses (ERA-Interim or ERA-5) ?

L 153 : How many trajectories are initialized every 10 s ? Is there only 1 trajectory initialized every 10 s along the flight track ? A series of boxes sliding along the flight track with multiple trajectories in each of them would give an idea of the uncertainties and of the dispersion.

L 157-158 : Some of the trajectories are located over the Eastern Coast of the US with potential large anthropogenic emissions ; others are over Siberia or Canada where intense fires are detected every year ; some may also originate from anthropogenic sources in

East/South Asia if they were plotted for periods larger than 5 days. It is therefore difficult to conclude something from this Fig. 2 as we don't have any information about the altitude of the backtrajectories at this stage, nor on the potential deposition during transport.

L 175-176 : How do the authors disentangle the influence of the origins of air masses from that of wet/dry deposition mechanisms along transport ?

L 190 : Do the authors mean that air masses can be influenced by pollution only when trajectories are observed in the vicinity of the surface ? What about the mixing at higher altitudes with polluted air masses (anthropogenic or biomass burning) ?

L 219-220 : Yes, halogen chemistry is an active field of research ; but the references given here are rather "old" to justify this. More recent references would be helpful here.

L 232-242 : This discussion is interesting but outcomes are from other studies and the co-emission of biological INPs is only an hypothesis here. I think it is worth mentioning this here, but not in the conclusions of the paper. Otherwise, I suggest an investigation of this effect, with at least correlations.

L 318 : "Both of these figures show" : I disagree, I don't see any evidence for that on those figures.

L 331-333 : May the role of precipitation during transport influence of result ? I don't think it has been considered here, but precipitation amounts along trajectories (drizzle vs large scale precipitation) are available in the ECMWF analyses used by the authors.

L 343 : " a natural source of cloud forming aerosol". I don't see any evidence of this in the paper. Even if biogenic emissions may produce aerosols that can serve as INPs, there is no evidence that they actually do it and that ice crystals are formed at this period in this region.

Technical comments

L 84 : Replace "abord" by "aboard".

L 108, L 115 and L 501 : Replace "(Holzinger 2015)" by "(Holzinger, 2015)"

L 118-119 : Replace "statistical uncertainty" by "relative uncertainty".

L 136-138, L 318 : What does the y in ymol mean ? Do the authors rather want to use the Greek letter "mu" (μ) for micromol ?

L 137 : "0.42 pp ymol/mol mv for CO₂" : I don't understand the units.

L 242 : Remove the "y".

L 286 : "with a lifetime".