

Atmos. Chem. Phys. Discuss., referee comment RC1
<https://doi.org/10.5194/acp-2021-288-RC1>, 2021
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Comment on acp-2021-288

Anonymous Referee #2

Referee comment on "High concentrations of ice crystals in upper-tropospheric tropical clouds: is there a link to biomass and fossil fuel combustion?" by Graciela B. Raga et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-288-RC1>, 2021

General Comments

The basic data set and analysis in this paper are very welcome additions to the literature, and are worth publishing in some final form. I enjoyed reading it, and I expect that others will. Nevertheless, I sense that the paper began as an investigation of aerosol effects on extreme ice events, as evidenced by the title, and also that it began with the notion that biomass and fossil fuel combustion particles were influencing these, but that the data do not back this up in more than a qualitative sense at best. Conflicting statements then remain in the text regarding the assured role of different aerosol types in affecting non-EIE and EIE, and the ice nucleation mechanisms are not at all resolved. Further, it only becomes evident later in the paper (obvious to the reader, and then finally stated) that there is really no distinguishing factor between EIE and non-EIE events from the aerosol standpoint, and what is most driving EIE is deep convection. Lots of strong updrafts lofting aerosols to low temperatures, potentially as liquid through homogeneous freezing conditions. Do the base aerosols even matter? The paper is still useful for framing and asking the questions at the end, but I suggest that the authors need to work a little harder at demonstrating such a conclusion, and using the provocative title that implies causal links (and will be referenced for such). The relation with aerosols is interesting but never proven to be causal. The remarkable consistency between regions and seasons in Fig. 2 does not to my mind speak to an obvious aerosol effect, and especially not a link to major cities. Hence, one wonders about whether plots of CO or delta-CO versus ice concentrations are not shown. All else being equal, would a relationship not be expected? And what would be expected if the aerosol effect were related to either heterogeneous versus homogeneous freezing nucleation? Would dust influences be distinguished from the others? The anomaly plots are not all especially revealing about what is driving things.

Hence, I suggest the need to:

- 1) Describe in the introduction how aerosols might influence cirrus concentrations via heterogeneous AND homogeneous freezing.

2) Discuss the complicating role of cloud dynamics and convection, and how the nature of the cirrus targeted (if all expected to be liquid-formed) matters. This and the first suggestion add context to the study, instead of diving immediately into aerosols as the only influence.

3) Consider if some truly quantitative analyses of relations between aerosols/gases and ice concentrations are possible, instead of only associations of fires, smoke and pollution areas with areas of EIE.

Finally, perhaps not for this paper, but would not a consideration of extratropical areas add to the understanding by taking away the immense role of tropical convection in driving EIE? I have a number of selected smaller comments, some related to these major ones.

Specific Comments

1) Abstract

Line 25: Using the term ice-forming aerosols is not exact, in that the aerosols may or may not be directly linked to the freezing mechanism. If homogeneous freezing, the factor of importance is simply that the particles carry liquid with them. If heterogeneous freezing, the nature of the particles truly matters. Perhaps, lofting aerosols that directly or indirectly lead to freezing, or better serve as seeds for heterogeneous and homogeneous freezing nucleation?

Lines 28-29: Why only heterogeneously if the cold clouds are of liquid origin? There would be a competition between heterogeneous freezing and homogeneous freezing, and what wins at cloud top will be determined by both cloud dynamics (updraft) supplying supersaturation and the propensity of particles for freezing heterogeneously and growing prior to the point where homogeneous freezing will ensue. Where would 5000 per liter INPs come from prior to -38°C ? And what concentrations would be necessary freezing prior to that temperature to defeat water persistence to -38°C that could then lead to further massive freezing? Consider the observations of Rosenfeld and Woodley (2000) in this regard. Deep convective clouds readily overcome the relatively low numbers of INP from the boundary layer.

2) Introduction

Lines 81-84 paragraph: In reference to the point above, homogeneous freezing needs mention as a potentially very important process.

Lines 90-92: "Some fraction of particles emitted from biomass and fossil fuel burning will act as CCN or INP especially as they age while lofted to the UT...". I consider the especially while they age part as not yet strongly demonstrated for the ambient atmosphere. Atmospheric measurements in this regard are not well-represented in the reference list. Recently, both Schill et al. (2020) and Barry et al. (2021) discuss ambient measurements related to biomass burning INPs, and production is mentioned in the latter study. Those measurements directly in plumes should constrain expectations on INP concentrations feasible from biomass burning, at least at temperatures in the mixed-phase regime prior to the homogeneous freezing threshold.

General comments on introduction and Figure 1: EIEs appear to occur in all regions, and quite high values occur even over oceans. Realizing that your focus is on connecting certain sources and EIEs, I wondered if the IAGOS network coverage adds any particular bias. Does the absence of occurrences between Japan and the U.S. indicate a true absence or a limitation of the network? In this regard, I felt it would be helpful to see a supplemental figure of all of the flight paths. Then it would be easier to understand where flying occurred versus where high values were seen. This point about potential bias or absence of coverage is only otherwise brought up late in the paper on lines 412-413, in regard to absence of flights over a region in Africa. Yet, large ocean regions of the Pacific in both hemispheres are missing from assessment, in a region more remote from fire and urban influences. As a second comment, some of the data are from over the maritime continent and other open ocean regions. Is there a reason not to consider sea salt as an aerosol that could affect deep convective clouds? It has been noted as a freezing nucleus at low temperatures in laboratory studies (e.g., Wagner et al., 2018), and was identified in ice residuals in deep convective anvils (Cziczo et al., 2013). When one is dealing with aerosols and ice nucleation, abundance and activation potential are both factors to consider, so I do not see a reason to exclude something in favor of something more abundant like biomass burning particles. This also arises later when the aerosols of "relevance" are mentioned on lines 373-374. It is simply what you chose to focus on.

Line 138: This is an instance of another reference to the heterogeneous ice nucleating properties of the aerosols being the only matter of interest to relate to EIE. Not so.

3) Results

Section 3.3 and Figure 6: I feel that a better explanation of the meaning of this figure is needed. What is event frequency? Is it any concentrations of ice crystals coinciding with a CO anomaly? I see nothing much distinguishing low-ice and extreme-ice, and the values of the median CO anomalies are extraordinarily low compared to say CO anomalies inside

and outside of biomass burning plumes. How does this indicate impact, if at all? Or especially, how does it show that "...frequency distributions do suggest that emissions from UP sources are potentially a larger source of nucleating particles in the ice clouds, in general.?" To me, I interpret this figure to mean that clouds and aerosols will be associated, but there is no smoking gun for any particular aerosol type or its direct involvement in creating EIEs.

4) Discussion

Lines 320-321: This inclusion of CCN here may be a nod to homogeneous freezing as a source of ice clouds, but only the INP connection is tendered earlier as a hypothesis. If the different mechanisms are made explicit in the introduction, this will all be resolved.

Line 345: AOD is an integrated measure. You do not know where in the vertical it resides, right? Most often it is in the boundary layer, although I understand that plumes can be elevated. And it seems that the full range of AOD underlies the EIE points. Like fire power and other relations, the correlation is only a spatial one as viewed from above.

Line 375: Fig. SMx? There is no AOD plot of this type in the supplement.

Line 393, paragraph: Not much is said about AOD over parts of Indonesia to Australia, which are striking for the apparent lack of any apparent influence. This is the regions that begs explanation, if it is to be contended that only certain types of particles are associated with EIEs.

Section 4.2: Not intending to beat on a point I raised already in summary, but convection so clearly shows the strongest correlation with EIE, regardless of aerosols. One has to ask for more than association with CO and other tracers in order to claim that any specific aerosol type is making a difference. Regardless, I felt that the convection link came far too late in this paper, and has to raise a question about the appropriateness of the title.

Section 4.4: A similar comment about a summary point. The discussion under this section finally acknowledges the ways that aerosols, ice formation and deep convection interplay, including mention of liquid origin ice processes and homogeneous freezing. Missing still is the fact that there must also be a relation between ice concentration and vertical velocity. And the mechanism will depend on that and on the freezing efficiency of heterogeneous INPs, as mentioned earlier. This should have been discussed up front, rather than alluding to the fact that the mechanism might be via INPs only.

Editorial notes

Line 386: Intended reference is missing.

Line 485: "temperature of EIE.."

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