

Response to All Reviewers

We are very grateful to the three reviewers, who have done an admirable job of critiquing our manuscript and raising important points related to the clarity of how we argue that there is a link between extreme ice concentrations at commercial flight altitudes and anthropogenic emissions at the surface. Many of the reviewers' comments which challenged our argument, have now been addressed. We have revised the Introduction that explains not only what the objectives of our study are, but that also emphasizes the uniqueness of this data set developed over nine years in what can be considered a totally random cloud sampling by commercial aircraft.

The reviewers criticize the study as having no quantitative evidence that supports our arguments for a link between surface sources and high ice concentrations. The only truly quantitative method to prove that such a link exists would be a Lagrangian study that measures aerosol properties at the surface and then follows these same particles as they form cloud hydrometeors. Given the near impossibility of such a study, we think that the methodology that we use, which combines quantitative aircraft measurements with multiple, independent data sets from satellite, a back-trajectory model and reanalysis, is as close to a quantitative evaluation as possible.

After reading the reviewers' comments, it became clear that we needed to state from the outset that our case was being built, by necessity, on circumstantial evidence but that the methodology that couples in situ and satellite measurements with atmospheric models makes a compelling argument for biomass burning and urban pollution as the most likely sources for the extreme ice events in the tropical regions evaluated.

We have made a number of modifications to the paper that we think will address many of the reviewers' concerns, and in particular, that we have been too aggressive in our conclusions regarding a causal link between high ice and anthropogenic emissions:

1. We have changed the title of the paper to "High Concentrations of Ice Crystals in Upper Tropospheric Tropical Clouds: Is there a Link to Biomass and Fossil Fuel Combustion?". This better represents our objectives while somewhat softening our conclusions.
2. The introduction has been rewritten to bring into sharper focus the objectives of the study, the uniqueness of the measurement platform and size of the data set, and to lay the foundation for our arguments that much of the high ice in the tropics is a result of ice-forming particles whose sources are anthropogenic emissions. The introduction now also mentions the two origins of upper troposphere ice clouds and why high ice concentration in such clouds are most likely of liquid origin, and how we connect surface sources to high ice concentrations using all the available resources at hand.
3. We have added a new subsection in the Discussion to highlight individual case studies to complement the larger data set from which our general conclusions are drawn. These case studies provide more direct evidence for the co-location of the

measured ice crystal concentrations, the surface sources and properties of potential cloud condensation nuclei (CCN) and Ice Nucleating Particles (INP) and the vertical transport mechanisms.

4. All reviewers have made comments that we have *repeatedly* asserted that high aerosol concentrations are the *cause* of high ice crystal concentrations. This was never our intent and as we look through the original manuscript, we only see a couple of times that we associate high ice with high aerosol concentrations. We have now removed those statements. In the new Introduction we explain that both biomass burning (BB) and urban pollution (UP) emissions are large area sources of particles, that the composition of many of these particles make them potential CCN or INP, and hence are a logical place to start investigating if there is a link with high ice crystal concentrations.

The point-by-point responses to reviewers are found below where we have listed each of the reviewers' comments, questions or recommendations followed by our responses highlighted in blue italics.

Response to Reviewer 1

- 1) 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.

We do not agree with this comment, and as explained in our opening comments above, we have used several, independent data sets and modeling, coupled with measurements, which support our conjecture of linking particles generated during combustion of biomass and fossil fuels to extreme ice.

- 2) 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.

We partially agree that there seems to be no distinguishing factor between the EIE and non-EIE from the aerosol standpoint, at least not from the evidence that we are able to gather at this time. Whereas the reviewer might be correct that it is deep convection that leads to the higher concentrations, we cannot rule out the possibility that anthropogenic emissions are producing higher particle concentrations which lead to higher cloud droplet concentrations and, hence, would lead to higher ice crystal concentrations from droplet freezing in the updraft.

- 3) Lots of strong updrafts lofting aerosols to low temperatures, potentially as liquid through homogeneous freezing conditions. Do the base aerosols even matter?

Base aerosols matter since CCN or IN are needed to activate as cloud hydrometeors, and this is determined by the composition and their size. While stronger updrafts mean higher supersaturations and hence, activation of smaller CCN, the right composition and size distribution of aerosols are needed to activate. Many studies have shown, from measurements and cloud models, that higher CCN concentrations lead to higher cloud droplet concentrations for the same updraft velocity.

- 4) 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.

There does not appear to be an actionable issue that we can respond to in this comment because in our discussion of Fig. 2, we make no mention of an aerosol effect or a link to major cities.

- 5) 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?

As we have now included in the introduction, and also reiterate in the discussion and conclusions, the magnitude of CO concentrations, as well as the CO anomalies, cannot be used directly as a proxy for either the magnitude of the ice crystal concentrations or precursor aerosols. The reason is because different processes remove or modify aerosols and CO concentrations. The CO is only being used as a tracer of the air masses that bring CO and aerosol particles from surface emissions to flight level. There was no physical basis for trying to correlate CO anomalies with ice concentrations

- 6) And what would be expected if the aerosol effect were related to either heterogeneous versus homogeneous freezing nucleation?

Although this question is somewhat vague as to its intent, we believe that we have now better clarified in the introduction, and reinforced in the summary, that the clouds that we are studying are for the most part form by frozen water droplets and the freezing mechanism probably has little relevance, i.e., whether the droplets freeze homogeneously at temperatures colder than -38°C or heterogeneously at warmer temperature. In the presence of the strong updrafts that loft these frozen droplets to the UTLS the only process that matters is the formation and evolution of the water droplets.

- 7) Would dust influences be distinguished from the others?

From a cloud microphysical perspective, yes, since dust tends to be better INP than CCN; however, studies have also shown that even with a little aging that can deposit hygroscopic material on their surface, dust particles can be good CCN. Hence, although we can't say unequivocally that dust would be indistinguishable from other

aerosols, without having a counterflow virtual impactor (CVI) to extract the ice crystal residuals, we are unable to distinguish dust from our analysis of the measurements. As the reviewer is aware, as reflected in the references provided at the end of his/her review, there are a very limited number of cirrus studies using a CVI on the WB-57 and many of the residuals were identified as dust particles.

- 8) The anomaly plots are not all especially revealing about what is driving things.

We agree, and have removed Fig. 6, for the reasons that we put forward above and have now put into the introduction regarding the lack of correlation between CO and ice crystal concentration magnitudes. We have also modified the text throughout to remove any discussion that would even imply that such a correlation should exist.

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

A discussion to this effect is now included in the Introduction.

- 10) 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.

We have made clear in the revised Introduction that vertical motion and boundary layer aerosols are two essential components in the clouds that were sampled.

- 11) 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.

As we explain in our opening comments before the point-wise responses, we think that the use of multiple, independent sources of data is as close to a quantitative assessment as possible without a Lagrangian measurement program, which has not been done for this type of clouds, to the best of our knowledge.

- 12) 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?

While we understand the reviewer's point, although the term we use may not be exact, neither is it incorrect and we are using it as a short form description without having to add qualifiers. Nevertheless, we do use qualifiers later in the Introduction to explain the role of aerosols in the formation of ice crystals.

- 13) 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.

We removed the word “heterogeneously” and have expanded the discussion of ice cloud origins in the Introduction.

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

We have expanded the discussion of ice cloud origins in the Introduction and added homogeneous freezing as one of the possible processes that produce ice crystals.

- 15) 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.

We agree with the reviewer and have removed “especially as they age” and added references to the Barry et al. and Schill et al. studies.

- 16) 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.

The reviewer is correct that there are large regions that unfortunately remain uncovered by the current IAGOS network. As suggested, we are adding the flight tracks to the supplemental material and in the introduction we point out the data gaps.. As we have now explained in the revised text, there are two reasons that sea salt is not included as a significant contributor to EIE: 1) sea salt aerosols, while excellent CCN, are also quite large compared to CCN from BB or UP emissions. Hence, these will be the first to form large water droplets that will very rapidly grow to precipitation-sized drops and be removed as rain before ever reaching the UTLS and 2) The MERRA results showed mass concentrations of sea salt at flight altitudes that were significantly lower than the sulfate or OC/BC at those altitude where EIE were found.

- 17) 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.

We have removed Figure 6 and modified the discussion here and throughout the manuscript, emphasizing that the anomalies are only being used to identify source regions with no intent to suggest that their magnitudes are necessarily proportional either to the intensity of the emissions or the concentration of the ice. Because the model does not specifically provide a time between when the CO was found in the clouds and when it left the surface, the magnitude of the CO anomaly may be impacted by dilution along the trajectory and should not be correlated with source intensity or particle concentration.

- 18) 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.

The Introduction now resolves this issue.

- 19) 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.

Yes, the AOD is an integrated measure and we are using monthly AODs averaged over the same nine years as the cloud data as an additional independent data set. We are showing regions that climatologically have high aerosol loading. We have now added case studies to provide a more direct connection between the emission sources at the surface, high AOD and deep convection.

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

Supplemental figure 2 (SM2) shows all 12 months of AOD. Apologies, SMx was supposed to be SM6, the MERRA maps of particle mass for the dust, sulfate and OC+BC

- 21) 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.

The reviewer raises an important omission that we have corrected in the revised manuscript. The lack of high AODs over the Indonesia is due to the generalized presence of clouds since cloud filtering is necessary to determine AOD. Looking at the OLR and lightning plots, which are a measure of cloud cover and convective activity, the reviewer will see that the region where there is no AOD data corresponds to moderate-to-high OLR and extensive lightning associated with deep convection.

- 22) 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.

With the modification of the title, the expanded Introduction, additional discussion related to lightning and upper-level divergence maps and circling back to convection

in the summary and conclusion, we think that we have adequately addressed the reviewer's concern that we don't give convection enough credit for the EIE.

- 23) 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.

We think that we have now addressed this in multiple places in the manuscript as has been highlighted in our responses above.

- 24) Line 386: Intended reference is missing.

We have added not only the Demott reference but also the Czizco reference that the reviewer provided. In addition, we would like to express our appreciation to the reviewer for all the references that were provided.

- 25) Line 485: "temperature of EIE.."

We have corrected this typo.