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 2

1) The authors have shown qualitative correlation between several independent data sources and the IAGOS incidence of Extreme Ice Events. But their suggestion that high aerosol concentrations are a cause of EIE is still circumstantial.

   As we emphasize in our opening remarks to the reviewers, and have now clarified in the revised Introduction, we did not intend to assign causality between high aerosol concentrations and EIE. Any such references to such a link have now been removed.

2) It would be interesting to somehow quantify typical values of AOD, CO anomalies, FRP and other indicators for the EIE vs. non-EIE samples, and to test the statistical significance of the differences between the two sample population.

   The case studies that we have added to the manuscript partially address the reviewer's interest in quantifying the indicators of anthropogenic emissions at the surface with the EIE. However, please note that we do not have in-situ measurements, and are only using satellite and reanalysis data sets. We concur that rigorous statistical testing is more satisfying when accepting or rejecting a hypothesis; however, as we discuss in our opening comments to the reviewers, it is the nature of the problem that makes such statistical testing impossible without Lagrangian measurements.

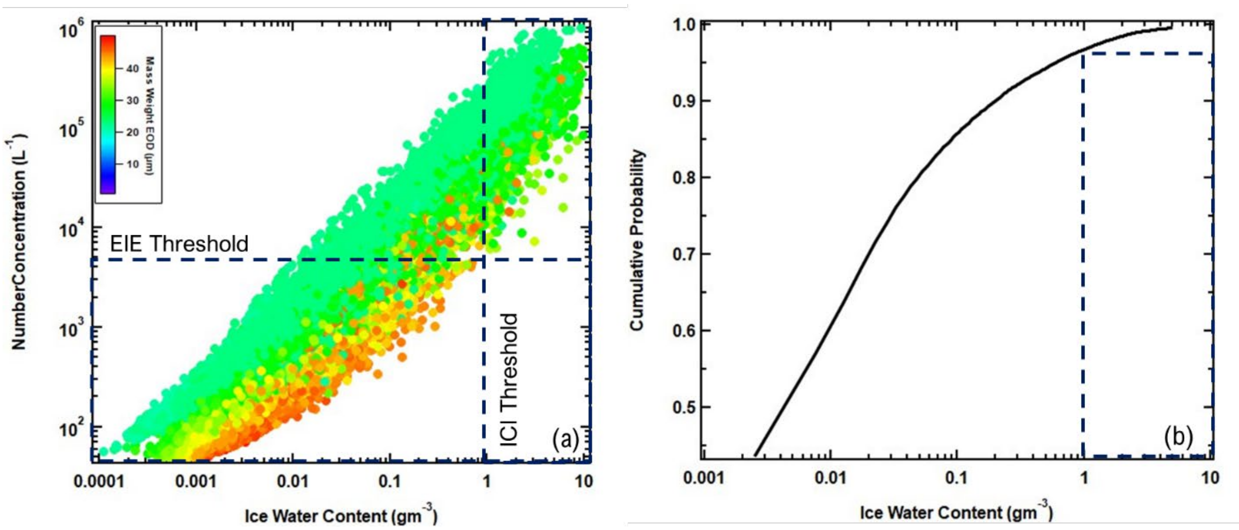
3) Introduction a. The beginning of the paper seems abrupt as the authors immediately reference prior publications by other researchers rather than introducing and motivating the topic of this paper. An introductory paragraph with some general information describing ice clouds, what is known about their interactions with

aerosols, and/or why it matters would set-up the current study more effectively before reviewing past literature.

The Introduction has been substantially modified so that now we set the stage for the remainder of the paper by discussing the two primary ways that ice clouds in the UTLS are formed, i.e. in situ and liquid origin, and how the liquid origin pathway is the most likely for the clouds we analyzed. We also discuss the importance of vertical motion along with the aerosol type and use these discussions to then explain why we are focusing on identifying the source of the aerosols on which ice crystals form in the EIE.

4) Lines 75-79. Could the authors please explain why they chose to use Nice and define a new term (Extreme Ice Events) rather than using IWC? Some of the papers cited give existing thresholds for elevated ice conditions based on IWC (i.e., HIWC), so why not be consistent? If there is a valid reason for defining an EIE threshold rather than using existing HIWC thresholds, how do the two thresholds compare?

In the revised Introduction, we clarify why we prefer to use Extreme Ice Events that use N_{ice} as the metric rather than IWC or HIWC. Research projects that investigate HIWC use thermal devices that measure IWC directly with less uncertainty than the method used in our study. We derive IWC from the BCP-measured size distribution, and estimate that the derived IWC has a >±50% uncertainty. In contrast, N_{ice}, can be determined with an accuracy of ±15% because it doesn't depend on any assumption about ice density or particle shape or size, which gives us more confidence on our results. Nevertheless, we have now added a new figure (see below) that shows the IWC vs N_{ice} in the sub-section on aircraft operations impact in the Discussion (included below). We include this to show that our derived IWC is above 1 gm^{-3} in more than 2000 of the EIE clouds, i.e. meeting the threshold of the HAIC community.



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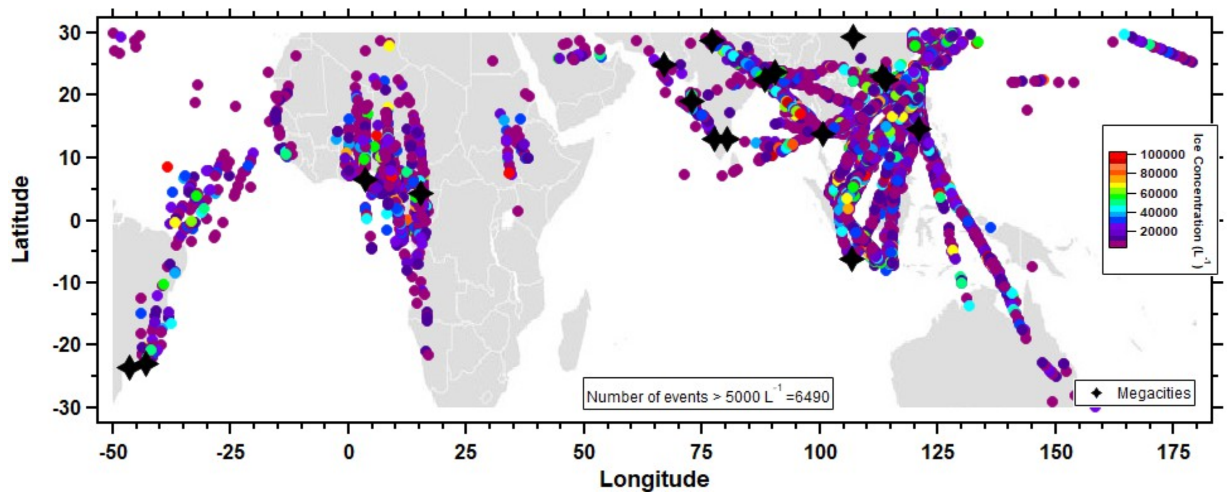
5) Line 109. Baumgardner et al. (2004) is cited here, but not listed in the Reference section.

This reference has now been added.

6) Figure 1. Some of the black stars indicating megacities are obscured by the EIE symbols.

We have redrawn the maps to bring the megacity stars to the front.



7) Measurements: Line 204 describes the geographic domain of the study as extending from -50º to +180º longitude, though Figure 1 indicates EIE outside of these boundaries and many of the subsequent figures (e.g., AOD, FRP) extend to a larger range of longitudes. It's not clear why the authors have limited their analysis to this region, or for that matter, why they didn't include events outside of the ±30º latitude range. Some explanation is in order.

The revised Introduction explains the reason that we have focused on low latitudes and Figure 1 (see figure shown in previous response) is now constrained to ±30º. In short, the highest density of EIE are in the tropics, with the largest regions of anthropogenic biomass burning and several rapidly-growing megacities.

8) Results: Figures 6 and associated discussion. Do the authors have an explanation for why the measurement-derived median CO anomalies are ~5 times larger than the modeled CO anomalies? Also, I'm confused by the assertion that, "The frequency distributions do suggest that emissions from UP sources are potentially a larger source of nucleating particles in the ice clouds, in general." Can you elaborate on how Figure 6 demonstrates this result?

We have modified the text and removed Fig. 6 to clarify our message that we are not using the CO anomaly as a proxy for either the ice crystal concentration or intensity of the combustion emissions. In our original analysis we had evaluated correlations between CO anomalies and ice crystal concentrations but concluded that no such correlation should be expected because CO and aerosol particles are removed or modified by almost completely independent processes. Hence, the CO anomaly is only a way to identify general aerosol source areas, not aerosol concentrations. Throughout the manuscript we try to return to the basic argument that the CO anomaly identifies elevated air masses under the influence of emissions larger than the background, the back trajectory analysis is used to identify from which region this air came from and if the CO is a result of BB or UP. The remaining piece of the puzzle is to identify the mechanism that transports this air to flight levels. This we have done with the best tool available, i.e. the reanalysis of the meteorological variables that provide the low-level convergence and upper-level divergence fields associated convection in reanalysis, as well as outgoing longwave radiation (OLR) to indicate the presence of clouds and lightning as proxy for deep convection.

9) Discussion: Figure 10. The density of EIE events in SE Asia obscures the values of FRP in that region.

Although we understand the reviewer's comment, we can't see an easy way to show both the important EIE and FRP so we have chosen to keep the EIE on top.

10) Lines 364-66. The following statement suggests that the evidence presented thus far proves that aerosol particles are responsible for EIE: "However, the maps suggest that these BB emissions are the source of some, but not all of the particles

that lead to EIE." While the authors have shown spatial and seasonal correlation between aerosol presence and ice crystal concentration, I think it is overstating to say that the particles cause EIE. Correlation is not causation, and as the authors discuss in later sections, other processes contribute to EIE.

The case studies in the revised manuscript provide more evidence, but we accept that the reviewer considers it an overstatement and have now modified this text.

11) Line 372-373. Please explain why only certain aerosols are considered relevant. Would sea salt not be of interest, for example.

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 water 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.

12) Lines 378-380. The authors state that Figures 11 and 12 show the high AOD over northern Africa in July is collocated with high dust concentrations and EIE in this area. Maybe I'm misreading the small map in Figure 12, but the high dust concentration and cluster of EIE points appears to coincide with a relative minimum in the AOD distribution for July (top of Figure 11). Can the authors please clarify?

As we now expand upon in the revised manuscript, AOD cannot be retrieved in the presence of generalized clouds. So what this means is that since the AOD maps are averages over nine years the very high AOD regions just north of the majority are indeed associated with the MERRA maps of dust in Fig. 12 but the region where the EIE are is also the region where there is a lot of deep convection during the July time period as is shown in Figure 13 where the region of lower AOD is where the 200 mb divergence is high, indicating strong upward motion, the OLR is a minimum, because the clouds are blocking the outgoing radiation, and lightning activity is a maximum, again associated with a lot of deep convection and ice. We have now added discussion to the text that clarifies this point.

13) Section 4.4. The authors nicely link their work to previous studies in this subsection. A key point in their argument is that the ice clouds observed in the IAGOS data set are likely liquid in origin. I'm confused about how the authors know this, and how it relates to their statement (in the abstract) that droplets are lofted and freeze heterogeneously. Please clarify.

As suggested by all the reviewers, the liquid origin assumption is discussed in the Introduction as described by Krämer et al (2016) who explain why the tropics, with *their deep convection and thicker clouds are the most likely to form by the liquid origin mechanism.*

14) Conclusions: Lines 505-506. Regarding the qualitative comparison of ICI events and EIE, wouldn't a positive correlation be expected given that both phenomena are based on high amounts of ice crystals? This again raises my earlier question about why new terminology and associated threshold for high amounts of ice crystals is introduced for this analysis.

We explain in the revised paper that ICI is used by the community that studies the impacts of high crystal concentrations on aircraft performance. The goal of our study is to contribute to improving our understanding of the processes that lead to the presence of high ice concentrations in upper-troposphere clouds analysing the IAGOS database.