Response to 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 3

1) My most important concern is that almost all statements in the manuscript that attempt to link EIE to aerosol sources are NOT well grounded. Some examples are Lines 25-27, 291-292, 337-339, 356-358, 364-365, 381, 391, 396-397, 446-447, 450-451, 490. The authors repeatedly attribute the EIE to high aerosol concentrations nearby.

The reviewer's assertion that our linkage of EIE to aerosol is not well-grounded is difficult to rebut without the reviewer offering a specific counter example of what would be considered a well-grounded argument. The reviewer also asserts that we "repeatedly attribute the EIE to high aerosol concentrations nearby". Below are listed, in quotations, all of the statements the reviewer lists as examples of our poorly-grounded arguments.

i)"The MERRA-2 analysis shows clear spatial correlations that link dust, black carbon (BC), organic carbon (OC) and sulfate particles with regions of EIE."
ii)"The frequency distributions do suggest that emissions from UP sources are potentially a larger source of nucleating particles in the ice clouds, in general."
iii)"In December there are EIE along the airline route between Northern Africa and South America, these would appear to be related to enhanced emissions of BB in Northern Africa and westward transport, as is discussed below."
iv)" Nevertheless, this region adjoins the area of most frequent EIE indicating high aerosol particle concentrations associated with the ice clouds."
v)" The proximity of the EIE to regions with large magnitude of AOD suggests that these clouds have likely formed on aerosol particles from relatively nearby sources"
vi)"This strongly suggests that the EIE in this region is likely related to dust, in addition to the BB that is also adjoining this region during July and whose presence is confirmed by the CO analysis."

vii)"Hence, the particles associated with the BB emissions are clearly linked with the fires, CO, OC/BC and EIE.
viii)"These high concentrations are partially reflected in the larger AOD, but the particularly striking feature are the many EIE in the region over eastern Asia.
ix)" The presence of ice clouds with extremely high crystal concentrations, clouds that in this current study have been associated with ground based emissions of anthropogenic CO and aerosol particles."
x)"The results that we have presented provide a framework for linking ice clouds in general, and EIE in particular, to surface sources of dust, BB and UP in tropical latitudes."
xi)" We conclude that the two, primary factors that are associated with the EIE encounters are the proximity to sources of dust, OC/BC or sulfate combined with strong vertical motions in deep convective clouds"

Note that of the 11 examples, only in iv) do we associate high concentrations of aerosol particle with high concentrations of ice crystal and we have now modified that text. Note, however, that the aerosol optical depth (AOD) is directly proportional to the vertical integral of the aerosol concentration and since the value of the AOD was much higher than surrounding areas, to attribute the high AOD to high aerosol concentrations was not totally incorrect.
We understand that the reviewer is not convinced by our evidence. We have now modified the text to carefully state only what the figures show. We have also included a few case studies on specific dates that provide more support for our statements. We have also toned down some of our conclusions to highlight the uncertainties involved.

2) However, according to Figure 6, the overall CO concentrations are even slightly lower in EIE as compared to the scenes with low ice concentrations

As we now explain in the revised manuscript, we use the back trajectories of the CO to identify the most likely source of the air masses in which clouds form, and do not attribute higher aerosol concentrations to higher CO anomalies. We do assert that the aerosols on which cloud particles formed are from the same source as the CO. Since the processes that remove aerosols and CO from the air masses are different, we do not use CO anomalies as proxy for aerosol concentrations.

3) Besides, Figure 11 shows that, while some EIEs do occur in the vicinity of high AOD, even a larger number of EIEs occur in regions with quite low AOD.

The revised text clarifies that the AOD is used to identify the source regions of those aerosols on which water droplets and ice crystals for, and not the regions of EIE. The reason for this, as the reviewer points out, is that regions of EIE are also regions of frequent clouds but low AOD. This is because in order to derive AOD measurements the algorithm removes data points where clouds have been

identified. Hence, the low AOD is not because there are no aerosols in these regions but because the frequent presence of clouds prevents the estimate of AOD. By comparing the maps of outgoing longwave radiation (OLR) and lightning in Fig. 12 (which indicate the presence of clouds and deep convection, respectively), it is evident that the regions of high EIE but low AOD, are regions where, on average over the 9 years of data, that there is extensive cloud activity. The new case studies discussing specific dates, show the absence of AOD data in the presence of high lightning activity.

4) After reading the manuscript, my impression is that the current results can hardly support any causal relationship between the occurrence of EIE and the occurrence of high aerosol concentrations. Please carefully reevaluate all related statements throughout the manuscript and either remove them or provide convincing supporting evidence.

We have followed the reviewer's recommendation starting with the abstract and continuing through the summary and conclusions.

5) Also, in view of the above comments, the last two objectives stated in Line 136-139 are not appropriate

The revised text now states: The four objectives of the study are: 1) to document the frequency of EIE by geographic region within the latitude band most impacted by BB and UP emissions, 2) to evaluate the seasonal variations of EIE as related to dry and rainy periods, 3) to identify regional sources of INP most closely associated with the EIE and 4) to show that there is sufficient convection to transport these INP, and the cloud particles that form on them into the UTLS.

6) Line 20: not only anthropogenic sources but also biomass burning

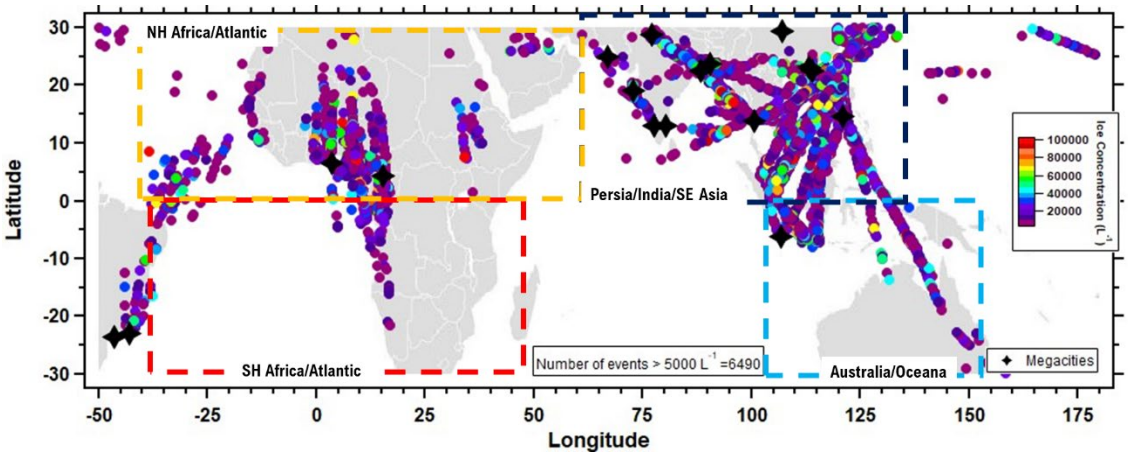
Line 20 is now modified to read "Evaluation of in situ measurements of carbon monoxide in these UT clouds, combined with back-trajectories and carbon monoxide emission inventories, identified regions of potential, anthropogenic sources of ice crystal forming particles." We consider biomass burning to be anthropogenic at low latitudes, for the most part associated with land clearing for agricultural purposes and with burning of refuse after harvest. In other parts of the world, such as temperate forests, BB can be accidental or lightning induced, but not typically in tropical regions of South America, Asia or Africa.

7) Line 168-171: Please provide more details about the SOFT-IO tool since most readers are probably not familiar with it. How does this tool link in situ detected CO to emission sources? What are the main inputs to the tool?

The revised text has been expanded to clarify the way SOFT-IO links the in situ Coat flight level to the emission sources and what the main inputs are to this back-trajectory analysis.

8) Line 205: Please show the spatial extents of these four regions in at least one figure in the main text.

Figure 1 now has the four regions outlined with dashed, colored lines, as shown here below.



9) Line 240: This paragraph can be moved to the Method section.

As the reviewer notes, this is repetitive as it has already been introduced in the methodology section so we just removed it but added a brief introduction of the results subsections.

10) Line 386: Correct the typo here.

Corrected, i.e., reference has been added.

11) I suggest that the error bars be added to Figures 4 and 8.

Added as suggested.

12) Tables 1-2 can be moved to the Supplementary Information.

Moved as recommended.