A Note to All of the Reviewers

The authors are appreciative of the detailed attention that the reviewers have paid to the reading and review of our manuscript. In particular, their level of understanding of cloud dynamics and microphysics have provided us with a perspective that is on occasions, somewhat different than ours. Their comments have reminded us that not all of our readers may have the same level of experience measuring cloud properties as we have, nor the familiarity of how clouds in the tropics differ from those that form in other latitudes. Hence, we have tried to be responsive to the opinions put forth by the reviewers while also maintaining our own interpretation of our measurement results. All three reviewers have acknowledged the value of this data set and, to our knowledge, no other study has taken the analysis of these cloud measurement to the depth that we have, especially assembling auxiliary satellite and reanalysis products to support our case for how ice clouds form in the UTLS in tropical latitudes.

Our observational study is, nevertheless, unable to produce the "smoking gun" that unequivocally links ice crystal microphysics to particle emissions at the surface. To do so will require modeling expertise beyond the scope of the current paper and by modelers more skilled at this activity than our team, whose expertise is in measurements and data interpretation. The reviewers have offered their own explanations of how these clouds might form and we have incorporated at least a summary; however, in our opinion, based upon all the evidence that we have assembled, our explanations are more likely and the reviewers have not offered evidence to the contrary.

Response to Referee #1

1) The last round of revision has improved the manuscript since the authors have removed or at least weakened many statements regarding the causal relationships between extreme ice crystal events (EIE) and emissions from biomass burning and urban pollution. However, there remain some statements that articulate or suggest the physical links between the two. After reading the revised manuscript and the authors' response to the reviewers' comments, I insist that the results presented in this manuscript cannot prove any physical links between EIE and biomass burning/urban pollution aerosols. Instead, the results only show that CO/aerosols from biomass burning/urban pollution often coincide or coexist with EIE. As one reviewer pointed out and the authors also agreed on, there is really no distinguishing factor between EIE and non-EIE events from the aerosol standpoint. For example, it is possible that some background or natural aerosols are already enough to allow EIE events to happen and that the aerosols from biomass burning/urban pollution are not actually making a difference. I don't want to get into more details since this would largely repeat many previous comments from myself and the other two reviewers. I do not intend to kill this manuscript since the dataset reported herein is really valuable. My suggestion is that the authors remove the arguments/conclusions on the causal relationships or physical links between EIE and biomass

burning/urban pollution aerosols. Otherwise, I still feel reluctant to support the publication of this manuscript.

As the referee acknowledges, we went to great length to remove wording that would be mistaken as an attempt to unequivocally attribute not only EIE but all of the cirrus measured in this region. The revised manuscript was thoroughly screened for wording that would indicate attribution, which can only be done via modelling and not through an observational study.

2) A minor issue: Line 625-627, "Of all the cells that had either clouds or CO anomalies, 52% had CO anomalies with no clouds, 10% had clouds with no CO anomalies, and 48% had concurrent observations of clouds and CO anomalies." The three percentages do not add up to 100%.

Thank you, this has been corrected. The 48% should have been 38%.

Response to Referee #2

1) The authors have captured the essence of my comments and those of other reviewers regarding the need to frame this study mechanistically before diving in to demonstrate relations to aerosols. Thus, the paper and its general focus are much better now. I understand the utility of the added case studies in addressing other reviewer concerns, even though I did not personally find them to add greatly to the paper. The introduction regarding nucleation processes still needs a little work in my opinion, and hence I make a few suggestions for consideration in laying out the basis for thinking about how the high ice concentrations could ensue.

The authors would like to thank the reviewer for recognizing that we strove to address the comments and recommendations in the previous review. As acknowledged by this reviewer, the case studies were in response to another reviewer; however, we respectfully disagree and consider that their addition was important to show that our results and conclusions were not strictly based on a statistical association over the 9 years of measurements, but that statistics are founded on individual cases that can provide a direct connection between surface emission sources, particle composition, cloud microphysics and atmospheric dynamics. Without the detailed modeling that we mention in our comments to all reviewers (above), we believe that the statistical results supplemented by selected case studies do show compelling evidence that supports our conclusions.

The reviewer indicated making some suggestions for further consideration, but we are unable to determine from what is written below the specific suggestions referred to. Hence, in this new revision we have included additional text on homogeneous versus heterogeneous freezing and we have also strengthened the discussion of the importance of strong convection in the formation of these high ice concentration clouds.

There is no discussion at all of homogeneous freezing, how it could come about, and whether composition really matters, preferring still to focus on heterogeneous nucleation. Instead, as in the response to reviews, the authors prefer to call them all ice-forming particles. That is not accurate. Some particles trigger ice formation, while ice formation ensues in others due to an ice formation process that cares very little about the particle except that there is dilute water in it.

In the introduction we have expanded on the freezing types, and then again in the discussion we reiterate how homogeneous freezing may be equally likely as heterogeneous freezing under conditions of vigorous updrafts. We think that our use of the term "ice-forming particles" is more precise than the phrase that "particles trigger ice formation" or that some ice formation processes care little about the particle. Perhaps we are arguing semantics here but in the earth environment, with no particles on which to form, there will be neither water droplets nor ice crystals. Although in all cases of ice formation, the ice is forming on particles, we believe that our terminology of "ice forming particles" is a reasonable way to describe the particle on whose surfaces ice forms. In the revised version we have added a sentence to clarify this point.

One reason that poor INPs such as biomass burning particles and urban pollution can impact ice formation in these clouds so strongly is most easily argued to be a process that is less deferential in selecting them for freezing (i.e., not due to the special properties of the particles). Excepting the regions influenced by mineral dusts, where one could plausibly say that thousands per liter might activate before the onset of homogeneous freezing as droplets are lofted, one apparently only needs high aerosol numbers and strong updrafts in order to promote more ice formation in the presence of more particles. This could be a working hypothesis for anyone pursuing this topic further, after reading this paper.

We are in complete agreement with the reviewer that we expect, and would indeed welcome, modelling work motivated by this observational study to address some of the specific details that cannot be done with the datasets analyzed.

For example, the argument now made is that sea salt can be ignored on a number basis alone. Well, there a second point made about its preferential liquid-phase scavenging in lower cloud regions, an argument that appears to ignore the role of cloud dynamics on impacting CCN activation and scavenging by any aerosol entering deep convection.

We think that perhaps the reviewer misunderstood our point regarding the liquid-phase scavenging and upon reading that section again have edited it to make our point clearer, i.e., that marine aerosols, due to their size and hygroscopicity would be those that activate first and grow rapidly to raindrops that will remove them from the cloud before they are lofted to freezing temperatures.

In general, the paper lacked an expressed appreciation for how dynamics can overcome restrictions on CCN activation. Under strong updrafts, one could easily posit that chemistry as a player in CCN hygroscopicity is likely irrelevant, and that is why BB and urban aerosols become important for anvil microphysics over the tropics. If this had been considered, Fan et al. (Science 359, 411–418, 2018) might have been referenced as a case for even small pollution particles as likely CCN that can freeze in upper cloud regions, due to cloud dynamics and the role of coalescence in driving supersaturations in elevated cloud regions. But I am not trying to rewrite the paper, only state what is apparent to a reader.

We have added what we expect will satisfy the reviewer's interest in making the role of strong updrafts more obvious. Our use of the maps of upper air divergence and the other analysis tools that we employed to underscore this role, indicate our awareness of the role of strong updrafts.

Line 21: Acronym SOFT-IO not defined in abstract.

The paper by Sauvage et al., 2017 introduces the name "SOFT-IO version 1.0" to describe the software developed which combines the FLEXPART (Stohl et al., 2005) Lagrangian dispersion model with the inventory of the Emissions of atmospheric Compounds & Compilation of Ancillary Data (ECCAD) emission database (Granier et al., 2012). It does not indicate that SOFT-IO is an acronym, but rather the way to identify the software developed in their study, which is now available to users.

Lines 30-31: Heterogeneous and homogeneous freezing. Saying both would make it clearer that both are likely involved.

We have added "Heterogeneous and homogeneous" to the text.

Lines 81 paragraph: It is a little odd to start this paragraph this way, since you have already mentioned heterogeneous and homogeneous freezing above this point. It would be more appropriate to speak to both mechanisms then at this point. The paragraph only mentions heterogeneous INP concentrations and ice crystal concentrations, ending presently with a very nice statement about that. But why not say that homogeneous freezing would be driven by strong updrafts that send condensed water not already frozen by limited heterogeneous freezing or limited consumption by ice growth into the regime where remaining drops can freeze? Noting the role of updraft on activating particles pre- and post-coalescence could add some context as to why this mechanism might be particularly powerful in affecting upper anvil ice concentrations.

We believe that perhaps the reviewer didn't understand the purpose of this paragraph since in the discussion of the previous paragraph, we were only referring to homogeneous freezing whereas in the paragraph beginning on line 81, we are talking about depositional nucleation. Yes, when we reference Krämer et al. (2016) and cirrus of in situ origin, we

know they are talking about depositional nucleation but this paragraph further explains why that is unlikely to be the mechanism forming the ice in the tropical clouds. That being said, we realize that we have not made this very clear and have added a couple of sentences to link this type of ice formation to the in situ type of cirrus formation.

In addition, we have added to the previous paragraph words to the same effect as suggested by the reviewer, reinforcing the important role of strong updrafts to loft liquid water that has not already frozen heterogeneously into regions where it freezes homogeneously. That is a point that we failed to include in our discussion, i.e., that the two mechanisms are not mutually exclusive.

Lines 105-107: Not a nuanced discussion. Would one not expect some major fraction of all particles to be available as CCN from combustion sources, but some more limited fraction available as INPs prior to onset of homogeneous freezing conditions? I say this only because it otherwise sounds like very little is known, but that is not the case.

We agree that this single sentence by itself offered little information. We have added a bit more context as suggested by the reviewer, in particular here is where we highlight that it might not be the composition of the combustion particles as much as their sheer number that get activated due to high supersaturations generated in strong updrafts.

Line 125-127: Contrast the above discussion with the apparent need to state that rBC particles are coated. This is probably a nearly irrelevant factor since supersaturations could be high in deep convective updrafts (especially following initial coalescence). Also, is it not the case that rBC is but a small fraction of all biomass burning particles, all of which are available to act as CCN? You later rule out sea salt for similar number-based arguments.

We agree and here we have reiterated what we have now added in the previous discussion on the role of combustion particles that it is likely the number of BC and not their size or composition that could contribute to high ice in the presence of strong updrafts.

Line 230: “Concentrations” or “Mixing ratios”?

We have changed concentrations to mixing ratios here and throughout when talking about CO in order to not confuse the reader with number concentration.

Lines 285-292: The statements made on not considering sea salt remains speculative. It will “likely” be less of a factor would solve this for now.

Changed to “..it will likely be less of a factor..”

4) Results

Line 317: 0.01 cm-3 as a lower bound on ice concentrations. I thought the lower limit of detection used in this study was 50 per liter?

We have changed 0.01 to 0.05.

Lines 414-415: Dust acting as good INPs by "deposition" of water vapor to their surfaces? If all of the cirrus discussed were of liquid-origin (also stated as a conclusion on lines 584-585), then why mention a mechanism that is unrelated to liquid droplet activation?.

We have now included words to the effect that dust particles, although very good INP will also activate as CCN under the high updraft scenario.

Lines 440-441: This repeats the same point about dust, but is better-spoken here. Could say this just once, in one spot or the other.

We have added here a sentence that refers back to what we added after line 415.

Line 454: Second mention of sulfate "mixed with dust", but it is unclear why this is at all important, or if it is important (which I doubt, if CCN activation is the concern –i.e., big dust > smaller CCN for activation under strong forcing, all else being equal and no matter the sulfate).

We agree and have now reworded this statement with a reiteration that even though these mixture make them better CCN, under vigorous updrafts they would activate regardless of their composition.

Line 577: Discussion of ice crystal residuals as "ice nuclei". You could stop that phrase at their "composition", since "ice nuclei" infers a component that freezes heterogeneously. It could say ice crystal nuclei, but ice residual nuclei is already stated, and a better way to discuss it.

Agreed. We have removed "that could identify the composition of the ice nuclei"

Lines 587-588: "…cloud chamber and field studies have shown that some fraction of the BB and UP aerosol are hygroscopic and can serve as CCN." Just how hygroscopic do they really need to be in these circumstances? Would kappa of 0.1 not be sufficient? See Twohy et al. (2021; https://doi.org/10.1029/2021GL094224) on how easily smokes are activated even in modest cumuli.

Now that we have clarified in the introduction and throughout the text the importance of the high updraft, we have inserted again that the composition might not matter under those conditions.

Line 664: Indeed, any boundary layer aerosols are lofted by strong convergence, and hence, one expects pollution, smoke, dust, and even sea spray particles (primary and secondary formed ones) to influence convective cirrus that dominate in the tropics. This paper confirms that.

Agreed.