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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Free troposphere as the dominant source of CCN in the Equatorial Pacific boundary layer: long-range transport and teleconnections" *by* A. D. Clarke et al.

Anonymous Referee #1

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Summary:

This work represents a distillation of the ideas that have long been suggested by the authors and it is nice to see this come to fruition. The paper represents a significant contribution to the discussion of aerosol-cloud interactions and the potential impact of transport processes in the Equatorial Pacific maritime region. That discussion is intriguingly subject to some strong debate between groups who believe MBL CCN sources dominate and vice versa and any step forward in that debate should be welcomed. However, I have some concerns about the strength of the conclusions made in this paper and the way in which those conclusions have been arrived at. I believe they need

to be tempered and put in context. This study is an important step forward but it is not the leap to the finish that is suggested.

The over-arching statement of the study is that long-range transport of aerosol in the FT and subsidence and entrainment into the MBL over the Equatorial Pacific represents the dominant source of CCN to the MBL in the region. The authors use a variety of data sources to demonstrate this, focussing in detail (I think) on data from the PASE campaign. Correlations, back-of-the-envelope calculations and even multi-step inferences are used throughout to this end in an attempt to convince the reader; and while I am suitably convinced by a lot of the arguments made, I do also feel a little duped by the frequent over-statement of these inferences as conclusive proof and statement of fact. We are taught at high school that correlation does not necessarily mean cause but the conclusions of this paper use correlations in just such a way and without suggesting the potential coincidences, alternative causes and other correlations that might not fit the authors' hypothesis and which undoubtedly arise from the necessary use of limited data. This is not to say that the inferences are completely without merit (many are intuitive and well-reasoned) but they are over-weighted and used far too often to make bold statements that are not backed up. A simple use of the word "suggests" rather than "confirms" throughout might go some way to address this.

The basic premise of the above argument - that sometimes polluted FT air is entrained into the MBL – is entirely sound however and not unexpected (descending FT air has to be entrained into the MBL in the region and can indeed be polluted or otherwise rich in aerosol). The question in this work, though, is: does FT entrainment provide the dominant source of CCN to the MBL? I'm not entirely convinced it does and I also take issue with the "general" nature of the title of the paper that implies that FT is always the dominant source of CCN in the region. It may arguably be the dominant source during PASE but is that typical across the diurnal cycle, the meteorology and the seasons etc. I suspect it isn't and the title should really reflect the necessarily limited scope of this study.

The article is well written, largely appropriate in scope, with good quality figures and discusses topics well within the remit of ACPD. However, while I have great respect for the way the datasets used here have been carefully and expertly compiled, I do not believe that appropriate scientific rigour has been applied in the discussion of those data and some sections of the paper are very confusing. It is this reviewer's recommendation that this article is considered for publication after satisfactorily and carefully addressing the important comments below with special attention to the way in which conclusions are presented.

General comments:

1/ Airmass history assumptions:

The assertion in the abstract that correlations in CCN.2 above and below the inversion "confirm" that FT entrainment dominate CCN is not correct. At best it may "suggest" it but what if the airmasses of the MBL and FT for the flights studied had similar aerosol backgrounds to begin with, e.g. that the deep convection that is later mooted as the source of FT aerosol, actually lofted BL aerosol above and over the MBL that is still carrying the same aerosol along with it at lower altitudes? Trajectories could help to inform this argument (I'll come back to the limitations of trajectories later) and trajectories from one flight are presented in Fig. 5, which all appear to show an easterly flow at all altitudes, with some trajectories passing over SA and some remaining over the MBL before being lofted. Interestingly (in this one demonstrated example), the lofted airmass which is subsequently sampled in the FT, was lofted from the MBL, whereas the air sampled in MBL was always at low height (I use the word height and not altitude here) as the air seems to have hugged the ground as it moved over the Andes and SA (height ∼3 km at ∼80 degW, 6 degS). Now, if the lofted air was simply previously MBL air (ultimately of SA origin) with the same ultimate airmass history (S. America) and background aerosol character, then the reason the sampled FT air has similar CO and

CCN concentrations etc as the MBL would be very simply because the airmasses of the FT and MBL are still historically coherent. Furthermore, gradients in background aerosol characteristics do not have to match gradients in CO– sources are not always linked. For example, the sampled MBL air may have (and seemingly has) passed over rural South America and the Andes where it may pick up aerosol and precursors but not CO (there are sources and sinks of both CO, ozone and aerosol other than biomass burning and precipitation). I don't believe that the trajectories are conclusive proof that we should expect these FT and MBL airmasses to have a different background in the absence of entrainment (the necessary prerequisite for us to be able to conclude that the MBL aerosol sampled is of FT origin), therefore it is not possible to reasonably conclude that the observed similarity of those airmasses is dominated by mixing down from the FT. If the trajectories in the FT and MBL went in entirely different directions, I might be more inclined to agree but they do not. Correlations (or anti-correlations) between very small gradients in CO (the range of which are within the reported instrumental uncertainty) and aerosol size spectra are not enough on their own (or in conjunction with these limited and inconclusive trajectories) to suggest a causal link and dominance of CCN2 from the FT.

Remaining on the subject of trajectories, one trajectory example is given for one flight (RF14) and the reader is then expected to believe that the same conclusions made from this example holds for all other flights – this is one example of where one set of inferences are used to support an argument (and for the reasons above they do not even hold in this example), which is then applied throughout the rest of the dataset. I would like to see trajectory analysis for each flight and a better discussion of the limitations of trajectory analysis. The authors do state once that the accuracy of 10-day trajectories is questionable but then go on to reason to trust them based on the fact that they are embedded in a consistent easterly flow. But this is far from a full discussion. It is well-documented that trajectories (HYSPLIT or otherwise) that hug the surface (as many of those shown do) are subject to high error as they do not capture BL turbulence and FT exchange. Furthermore, lofting in trajectory calculations is a function of the vertical velocity calculated by convective parameterisations (where triggered) of the meteorological reanalysis dataset used to initialize HYSPLIT (no mention has been made as to which dataset was used to this end?). All global models are well-renowned for their poor capture of deep convective events in general; especially in the ITCZ. In summary, lofting by deep convection in trajectory calculations is very poorly captured (works for frontal systems etc but not for isolated convection) therefore I do not trust the inferrences made from Fig. 5.

For RF14, a MODIS plot (with no timestamp) is presented in Fig. 3 (with no recognition of where the plot was taken from as this is clearly extracted from and not a plot generated by the authors from raw data). It is referred to on P.1287, line 15, to discuss CTH and outflow from a line of convection. In the text, it appears to be a MODIS image taken "the night before" the flight, which was used to select a cloud for flying the next day. But why isn't a MODIS image that corresponds to the time of the flight shown – perhaps with the position of the aircraft labelled at the corresponding time? I would expect ITCZ-type convection to wax and wane with the diurnal cycle and I simply can't take imagery from the night before as evidence of the same cloud structure that was sampled the next day. In the text, we are told that "cloud top temperatures are indicative of outflow near 5 km" – then why isn't the plot a plot of cloud top temperature? Or what was the cloud top temperature? We are asked to believe generalities about satellite data that may not have been examined properly.

However, despite the above, the use of volatility information used later in the discussion is alot more encouraging and I enjoyed the discussion of new nucleation in deep convective outflow and the reasons for contrasts in volatility. This, when taken with the above, does begin to build a convincing case for the airmass history conclusions made, but again it must be stressed that this is all still (interesting) inference but not conclusive proof.

In summary, I really don't buy the current mismatch of satellite imagery, trajectories, in situ trace gas data and aerosol spectra that are potentially cherry-picked for analysis

to fit the hypothesis. I'd be much more convinced if I could see a complete flow of analysis (trajectory to cloud imagery to in situ data or the other way around) on a flight-by-flight basis (or even for one single flight), rather than snapshots of each data type from different flights all linked together in an unclear manner to "confirm" a favoured hypothesis.

2/ MBL nucleation

In the abstract, (line 25) we are told that the "measurements confirm that nucleation in the MBL was not evident during PASE". Do they? Have I missed something? This isn't discussed in the text so far as I can tell. Is the LDMA instrument used in this study on the C130 sensitive to ultrafine particles ($\sim$10 nm) that would see MBL nucleation. Perhaps it is but would be good to say this.

3/ Use of VOCALS data? P.1283, line 25 mentions that data from the VOCALS campaign have been used in the paper but I can see no discussion of VOCALS data later on? Which flights, where and when? I can only see PASE data. If VOCALS data have been used, why haven't any of the papers that discuss VOCALS aerosol measurements published in the ACP special issue been cited and discussed? I recall reading an interesting paper in that special issue that specifically discussed long-range transport in the region which the lead author here also co-authored, which seems to contain some interesting results that would support some of the more extensive arguments made in this paper. If VOCALS data are used in this paper it would be remiss not to cite some of the works on aerosol from that campaign as they have been discussed here for PASE.

4/ Section 9: I found this section very confusing. There are abbreviations and back-of-the-envelope calculations everywhere. Masses and diameters are discussed and then fractions of total masses etc that I simply cannot follow. But these calculations seem absolutely crucial to the overall conclusion of the paper as they arrive at a number that shows the majority of CCN may come from the FT. This needs much more careful and clear presentation given its importance to the title of the paper with careful explanation at every step. There are many assumptions implicit in the calculations that are not discussed, e.g. constant density with aerosol size (which is unlikely), integration time of the DMA instrument versus horizontal gradients, CCN sensitivity to the real range super-saturation in cloud (that could have been measured in cloud perhaps?).

5/ P.1299, line 5+: This paragraph discussed inter-flight variability between the FT and MBL and “confirms” that the apparent consistent variability of FT and MBL profiles over similar time-frames was due to entrainment. This may not be true for reasons discussed above – both FT and MBL airmasses could still be historically (and therefore chemically) coherent.

6/ Discussion and conclusions: This section is very long and mostly repetitive of earlier sections. This needs to be more concise. ———————————

Specific/technical comments:

1/ P. 1286, line 3 change “with them nucleated” to “with nucleation”

2/ p. 1287, line 8: Change “(see below)” to (see Figure 5).

3/ P. 1291, line 4. Needs a period mark after “ranges”

4/ P. 1298, line 8. A mismatch in CNVol is explained away by “unresolved cloud processes” – could you expand on this?

5/ P. 1301, line 4: What’s a “V pattern”? And what are “Vs”?

6/ P.1301, line 29, Change “after they reaches”.

7/ P.1302, line 1. What is “cloud density”? Droplet number, optical thickness, cloud fraction, thickness. . ..?

8/ P.1302, line 8; rephrase “cloud free regions along the wind that may travel together for several days” – I don’t understand what this means here.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 1279, 2013.