

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 #2

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This paper estimates the contributions of different aerosol sources to marine CCN concentrations measured during the central equatorial Pacific during PASE campaign in August–September, 2007. The campaign consisted of several flights carrying an impressive set of instruments, and the data obtained in these measurements is valuable for our understanding of what makes marine CCN. The paper suggests that during the campaign a major fraction of remote marine CCN originated from combustion sources some 10000 km away. This result is obtained as a combination of trajectory analysis, CO concentration measurements, volatility and size distribution measurements of aerosols, and supporting satellite data. So the long range transport itself appears to be rigorously proven. Also, the paper presents some very interesting data about the

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contributions of volatile and non-volatile aerosols to CCN concentrations during the campaign. Overall, the paper is of high scientific value and is suitable for ACP after revisions.

The marine CCN production mechanisms are still largely uncertain particularly due to lack of measurements. The understanding of marine CCN sources is vital to our understanding of past and future aerosol forcings. Because of this, the data analysis presented in this paper should be more critical and also take into account the limitations of the data set.

The data set is large enough to explore some key features of the CCN particles (volatility, size distribution, transport), but the measurements coincide with highly active biomass burning episode downwind in SA (Aug-Sep 2007) that is only briefly mentioned in the paper, and not discussed in the abstract or in the conclusions. As mentioned in Section 2, the CN and CCN concentrations during PACE campaign were unusually high compared to other clean MBL regions. It appears that the patchy pollution episodes are the ones that raise the concentrations to high levels during the time of year when biomass burning in SA is particularly active. Therefore it is unlikely that the data represents average conditions in Equatorial pacific boundary layer.

Because of the uniqueness and importance of the data presented I would gladly see this paper published in ACP after major revisions. The following points should be addressed before publication:

1. The results are based on a relatively short measurement campaign, and there is no discussion how well the results are expected to represent annual averages in the region. It is mentioned in Section 2 that PACE campaign took place during the time of active biomass burning in the Amazon region, and the abstract should clearly point this out as well. The biomass burning episodes in South America tend to be sporadic with high interannual variability. According to Giglio et al. (2010), burning in SA peaked during the measurement period and was higher than normal in 2007. This should be

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discussed in the paper. It would be a good to separate “clean, CO below 63 ppbv” and “polluted” cases in the abstract and in Table 1. Also changes in wind speed affect sea salt aerosol emissions, and their variability should be discussed in connection with winds speeds during the campaign.

2. I feel that there is some inner conflict in some of the conclusions: the growth of small particles to CCN sizes is claimed to be important (p. 1299), and this growth is linked to sulfuric acid produced from DMS (p. 1295). However, in Discussion and conclusions (p.1307) it is claimed that CLAW is not effective since all sulfate produced is accumulated on larger entrained aerosols. This claim is then stated in the abstract as well. Also I think it is wrong to claim that CLAW requires BL nucleation to be effective. While the CLAW hypothesis has been recently shot down by several other studies, I don't think that the data presented here justifies strong conclusions about the CLAW hypothesis.

3. In section 9 it is suggested that 30-40nm volatile particles entraining to BL are rapidly removed by some unidentified cloud processing, and therefore do not contribute to BL CCN. This sounds quite suspicious, but seems to be a rather important assumption when calculating the contribution of CCN originating from UT nucleated particles. A few lines earlier it is suggested that the uptake of sulfate by volatile particles entraining from UT may convert CN_vol to be detected as CN_hot. Is this not a more plausible explanation for the smaller CN_vol than unidentified cloud processing?

4. I am confused of the nature of particles that grow to CCN sizes in the boundary layer, that contribute 25% (50 particles/cc) to MBL CCN on average. Are these combustion aerosols? Or is this UT nucleated aerosol (as suggested by intercept of the linear line with y-axis in Fig 7b)? Please discuss.

5. The paper considers three sources of CCN: combustion aerosol, sea salt aerosol, and aerosol nucleated in cloud outflow. The abstract mentions that no boundary layer nucleation was observed, but this discussion is very limited in the rest of the paper. The

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authors claim that all observed volatile MBL aerosols originated from UT nucleation in cloud outflow, but I find this highly speculative. Nucleation also takes place in very large air volumes close to tropopause (for example Young et al., 2007 and references within), and the subsequent entrainment of these particles could be responsible for the majority of the volatile UT aerosol. I find it difficult to believe that cloud outflow nucleated particles with concentrations around 3000 particles/cc around noon right after nucleation could generate a stable background aerosol of some hundreds particles/cc some 3000 km away, after being distilled into such a large airmass. Please discuss other UT nucleation as well or justify why cloud outflow nucleation should be the only source.

6. Introduction should include more discussion about previous measurements and modeling studies of marine CCN. Also, discussion about combustion aerosol and its role as CCN should be included.

References:

Giglio, L., Randerson, J. T., van der Werf, G. R., Kasibhatla, P. S., Collatz, G. J., Morton, D. C., and DeFries, R. S.: Assessing variability and long-term trends in burned area by merging multiple satellite fire products, *Biogeosciences*, 7, 1171–1186, doi:10.5194/bg-7-1171-2010, 2010.

Young, L.-H., et al. (2007), Enhanced new particle formation observed in the northern midlatitude tropopause region, *J. Geophys. Res.*, 112, D10218, doi:10.1029/2006JD008109

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