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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of particle cloud droplet activity and composition in the free troposphere and the boundary layer during INTEX-B" *by* G. C. Roberts et al.

Anonymous Referee #1

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This paper focusses on estimating values of kappa for understanding CCN activity of particles comprised of organics and inorganics. It uses airborne measurements of CCN and hygroscopic growth factors in combination with measurements of particle size distribution and chemical composition to estimate kappa values of the aerosol in the free troposphere (FT), the marine boundary layer (MBL) and over the California Central Valley (CCV). The authors carefully calibrate the CCN and take into consideration the significant factors associated with CCN measurements at changing pressures. Despite that care, some of their kappa values in the FT are relatively high (perhaps too high); although some of the discrepancies may be based in the size distribution and composition measurements. Regardless, the authors appropriately acknowledge

this and it serves to highlight the difficulties in conducting CCN measurements from an aircraft as well as at lower supersaturations. The overall result is an interesting paper that makes a good contribution to the CCN-kappa knowledge base. Before publication, I have a few major points to deal with and some minor things for consideration.

Major points:

1. I don't doubt that the HTDMA measurements are of high quality, but there needs to be some discussion of the HTDMA measurement technique. I can't even find a reference.

2. The AMS observations are used with respect to the volume measurements from the SMPS/DMA. These are used to assess potential refractory material in the particles, and this is critical for the assessment of kappa (e.g. Figures 6a and 8). There is no discussion of the comparison of the AMS results with the SMPS/DMA. This is fundamental to this work and it needs to be clear in this work how these measurements compare.

3. Page 3525, lines 10-12 and Figure 8 - You make the statement that the results in figure 8 provide "evidence that nrOM O/C can be used to predict _org." The data in Figure 8 shows no relationship of kappa organic with m/z44/OM, and certainly provides no evidence in support of Jimenez et al. (2009) other than a vertical line crosses through a sloped line; if anything it refutes Jimenez et al. If not for this, the paper would be easily acceptable. Substantial revision of this point is required.

Minor points for consideration:

4. Page 3501, line 24 – H"TDMA".

5. Page 3502, lines 11-16 – another relevant feature of Chang et al. (2007) is that they showed that as the organic fraction of the fine particle aerosol reaches relatively high values that the composition of the organic becomes more important for CCN activity.

6. Page 3507, lines 15-16 – not important in the present context, but could not there

have been HNO3 in these particles instead of NH3NO3, especially if they contained some water and the sulphate was neutralised? The effect would be the same; this just goes to process.

7. Page 3507, line 24 – "by two research groups ON THE C-130 during INTEX-B."

8. Page 3509, line 13 – "a Single. . ."

9. Page 3512, line 17 – "with possible vertical mixing with the marine boundary layer". By this, do you mean that the trajectory dipped down near the ocean surface?

10. Page 3513, lines 6-7 - why not? This seems like an important distinction.

11. Page 3513, line 13 - the dominant mode of what?

12. Page 3515, lines 11-14 – I don't understand "which may suggest that polluted air masses enhance the rate at which particles become hygroscopic." Are you suggesting that polluted air masses contain more sulphate or more organic, or something else?

13. Page 3517, line 3 – there are also natural sources of CO, NOy and NOx.

14. Page 3517, line 14 – why wasn't MSA estimated from the AMS?

15. Page 3518, lines 21-25 – this is stretch, and I suggest revision or removal. There are a number of ways to explain this observation. It can not be inferred that high CN and low sulphate implies organics; could as easily be nucleation of H2SO4.

16. Page 3519, line 3 – "formation" and emissions.

17. Page 3519, line 7 – 0.81 is different from the value in Table 4 (1.01).

18. Page 3519, lines 7-8 – related to major comment 2 above. "the mean OM was more than 3 times the mean sulphate, yet the mean OMF is 50%. Table 4 shows that sulphate+nitrate+ammonium was about 1.9 compare with 2.9. Is the other 1.0 (needed to make the average 50%) from refractory material in the AMS size range or are there AMS collection efficiency issues?

19. Page 3519, line 15 - but it doesn't mean that they were completely externally mixed.

20. Page 3519 – Shantz et al (ACP, 2010) for example, suggests that the mass accommodation coefficient for growing droplets may be lower for anthropogenic organics compared with other organic types (e.g. biogenic). Since CCN are defined based on the ability for a particle to grow beyond some size threshold, could the lower kappa values you see in the CCV case be in part due to a kinetic effect rather than an equilibrium effect?

21. Page 3520, lines 22-23 – From Roberts et al (2006), the Wyoming chamber used in CIFEX based its CCN concentration on the peak light scattered. In that case the calibration is dependent on the chemical composition, and I believe a low value of the "true" kappa would be interpreted as a still lower value of kappa. Could this have contributed to some of the difference? Differences in growth time and detection size between the two chambers might also make a difference.

22. Page 3521, line 21 - I don't see any kappa value <0.05 in the above discussion.

23. Page 3521, line 23 – a substantial fraction? Phinney et al. (Deep Sea Research, 2006) measured the aerosol composition over the North Pacific in the summertime with an AMS. Considering the location, these measurements are somewhat relevant to your observations. For three weeks of measurements, they found for the fine mode aerosol that was sulphate was 0.74 ug/m3, sea salt was 0.6 ug/m3, organics were 0.3 ug/m3 and MSA was 0.16 ug/m3. Depending on how you classify MSA, organics were about 17-25% of the total; perhaps substantial, but likely not sufficient to significantly influence kappa unless kappa-org is relatively large.

24. Page 3525, line 4 – not coincidentally, the lower limit of your kappa values in figure 7 (for both the CCN- and htdma-derived values) looks to be close to 0.18.

25. Page 3526, line 2 - perhaps "application" rather than "demonstration". The latter does not suggest confidence in the measurements, which is not the case.