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Interactive Comment

Interactive comment on "Aerosol concentration and size distribution measured below, in, and above cloud from the DOE G-1 during VOCALS-REx" *by* L. I. Kleinman et al.

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This paper discusses observations and analysis of airborne measurements of aerosol particles, cloud particles and some trace gases made from the DOE G1 aircraft during the VOCALS experiment. The paper covers two main topics: 1) the vertical and horizontal distributions of CO, O3 and aerosol particles in the VOCALS project region and their relationships to dew point temperature, and 2) the impact of the aerosol particles on cloud droplet number concentrations (CDNC). The reference to the dew point as a tracer for sources and cloud processed air is an interesting concept first discussed by Kleinman and Daum. The impact of the aerosol on the CDNC is done through direct

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comparison of the CDNC with the aerosol as well as indirectly by examining the interstitial aerosol. Although many of the results have been demonstrated before in some way, from my perspective the evolution of the "Hoppel" minimum as shown in Figure 8a and the results extracted from the high frequency analysis are new. The results are important in the context of the VOCALS experiment and in a more general context they document another region of the world. The analyses are carefully done and the paper is well written and organized, but I suggest some further analyses would help: mostly related to the entrainment discussion and the fall-off in the CDNC versus Na plot. Details follow.

1) Dr. Lance posted comments on this paper that posed the question: does the drop off in the CDNC with increasing Na (Figure 10) reflect an atmospheric process or an instrument issue? A similar turnover was demonstrated by Leaitch et al. (Tellus B, 1986; prior to Martin et al. 1994 that Ramanathan et al. 2000 have attributed this to). The process vs instrument question was also an issue then even though the probes used were different (FSSP-100's). One of the things that Leaitch et al did was plot the interstitial aerosol (IA) + CDNC versus the Na where the IA and Na were both from a PMS-ASASP (precursor to the PCASP); although not conclusive, it did support the turnover. You have not done that here, in part because you are concerned about droplet shattering. You suggest that most artefact particles from the shattering of droplets are <50 nm; 50 nm is consistent with my undocumented experience of this, but the other reviewer expresses a number of valid concerns about this and as he mentions the editor of this paper is the real expert. If 50 nm is reasonable, then I don't see a good reason not to add a sum of the IA(from pcasp)+CDNC to your Figure 10 because that could help with the confidence in the fall off in the CDNC. The one case study you examine is for CDNC of about 150-200 cm-3, which is in the more linear region of your plot in Figure 10 and therefore does not provide support for the CDNC fall off. Shattering artefacts tend to be associated with larger cloud droplets and precipitation particles, and so points at higher CDNC should have reduced artefacts relative to the Oct 28 case study. Effects of shattering can be further minimized by examining data

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collected lower in the clouds where LWCs and droplets are smaller. The other 1986 paper that suggested a turnover in the CDNC, by Pueschel et al., was based on mtn-top observations and coincidence should be less of a concern. In your response to Dr. Lance, you have discussed that comparisons of the LWC suggest the turnover in the CDNC is real. This has some merit, but as the other reviewer (Prof. Snider) points out, it is not without problems.

2) There is nothing in the introduction about why this work is important.

3) Page 17292, line 8 – than rather than then?

4) Page 17297, lines 4-6 – Your assumption that the out-of-cloud segments, encountered while flying level through cloud, are representative of the precloud aerosol depends on those segments being unaffected by entrainment of air from above the MBL. Can you be certain of this? What about looking at ozone in these clear pockets versus in cloud? Given the much higher ozone above the MBL, if there is a significant influence of the FT, it should be evident in changes in ozone.

5) Page 17297, lines 18-26 – The comparisons in Table 2 suggest the CDNC measured from the G1 were biased higher than the other aircraft, but there is no reason given as to why the CDNC from the G1 should be corrected down rather than the other aircraft measurements corrected up; a comparison of three points is insignificant. Do the CDNC from the three other aircraft agree with each other in the CDNC vs longitude comparisons? I think comparisons of the pre-cloud aerosol with the Na+CDNC are another way to assess your CDNC before you simply say that they are 20% too high, and of course the LWC as suggested by the other reviewer. Raising your CDNC by 20% may alter your interpretation of Figure 10.

6) Page 17298, lines 18-20 – I don't understand how the below-cloud observations, which are the only thing shown in Fig. 3, justify the assumption discussed here?

7) Page 17299, lines 13-15 – Since you see only detection limit SO2, the implication is

that it has already being processed by the time it leaves the coast.

8) Figure 4 – the broad co-variance of the MBL and FT traces in aerosols, CO and ozone suggest a BL influence on the lower FT.

9) Page 17300, line 16 – I don't understand the "effective cold trap". I believe the other reviewer has commented on this also.

10) Page 17300, lines 21-22 – Also, in-situ production of new particles may be enhanced in this region with a reduced condensational sink.

11) Page 17301, lines 20-24 – How does this statement fit with the above discussion of dew point? Figure 7 gives no information about ozone or CO.

12) Page 17303, lines 21-22 – another test would be looking at sulphate/organics versus distance from shore.

13) Page 17303, lines 25-26 and Figure 8a – This progressive movement of the Hoppel minimum to smaller sizes is very interesting. I think it warrants some discussion of why this happens.

14) Page 17304, line 4 – period after nm.

15) CDNC is used mostly, but CD is used in Table 2 (and maybe elsewhere).

16) Page 17304, line 14 – why have you used 100 nm when you have just shown in Figure 8 that the Hoppel minimum is more like 70-90 nm? Presumably this is because the nominal lower limit of the PCASP is 100 nm. I think this is reasonable, but it should be made clear that this is the reason. How does your post-study calibration of the PCASP impact the 100 nm value?

17) Page 17304, lines 17-19 – A reference(s) should be included with this statement.

18) Page 17304, lines 19-21 – As above, this process was demonstrated much earlier than reported by Ramanathan et al. - Leaitch et al. (Tellus B, 1986); Pueschel et al.

(JCAM, 1986).

19) Page 17305, line 7 and Figure 10 - Where is Nint in Figure 10? There is only an arrow suggesting what the interstitial aerosol (IA particles >100 nm) should be.

20) Page 17305, lines 11-12 – As above, this implicitly assumes that cloud-free pockets are not due to entrainment of FT air. Was the MBL-FT division flat, such that when you flew out of these clouds you did not fly through any subsiding air? It would be helpful to have some demonstration of this, such as ozone (mentioned above).

21) Page 17309, lines 6-11 – There are also satellite observations that suggest a slight increase in LWP with increasing aerosol. You need to discuss that possibility also.

22) Page 17309, lines 16-19 – The point you are trying to make here is of course very important, but could be generalized and maybe made a little clearer. What I understand you to say is that aerosol effects happen but get masked by other aspects of cloud physics. I think it would be prudent to point out that the real aerosol indirect effect is based on changing the aerosol for the particular cloud situation you are sampling; i.e. what happens to your results in Figure 14 if you lower or increase the pre-cloud aerosol?

23) Page 17310, lines 13-14 – Not drying of droplets?

24) Page 17310 line 22 – Page 17311, line 3 – I can't find the Nenes et al. (2010) reference. See also Leaitch et al. (1996). I don't see where it is stated, but I assume that the calculated critical diameters (85-110 nm) are considered by the authors to be in agreement with their observations? The point from Guibert et al. is a good one and discussed further by Peng et al. (JGR, 2005).

25) Page 17311, lines 10-11 – A repeat question, but is this not a possible contributor to the holes you see in cloud that you suggest are equivalent to below-cloud air?

26) Page 17313, lines 1-3 - Yes, but you can also imagine situations where the air is already cold and dry (e.g. Arctic) and pollutants are emitted without significant wet

scavenging.

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