Review of Dadashazar, et al.

This paper attempts to explain the seasonality of cloud drop number concentrations (Nd) off the coast of the USA and Canada with a focus on explaining why Nd is highest in the DJF season, but the aerosol optical depth (AOD) is lowest in that season. The paper presents some useful analysis and I think it should be published once the concerns have been addressed. However, there are places where the results are not fully explained, some places where there should be a more quantitative analysis and some results that are in the supplementary that should be in the main text. Some of the arguments could also be made more clearly. It seems that aerosol is more efficient at making cloud droplets in DJF, seemingly because of the prevalence of the trade cumulus and stratocumulus conditions (with high low-altitude cloud fractions). However, the paper doesn't quite get the bottom of why this should be – perhaps more discussion on this is warranted although it may be case that we don't quite know yet. Here are some suggestions that might help to get closer to an answer :-

- It would be interesting to examine what the important predictors are in the subset of data with high low altitude cloud fractions. E.g., do the aerosol parameters then become equally important between the seasons once we are in the cumulus/stratocumulus regime (particularly CCN and sulphate surface mass)? Is Nd similar in DJF and JJA for the high cloud fraction subset indicating that it is mainly the prevalence of the low cloud conditions in DJF that cause the Nd difference? You could also do a similar analysis for the subset with small low altitude cloud fractions (more typical of JJA perhaps).
- Could it be the case that the Nd retrievals don't work very well and give smaller Nd values when there are no boundary layer clouds (since they are designed to look at PBL liquid clouds)? This could then give higher Nd in the conditions with more PBL clouds (i.e., DJF). There is also likely to be more overlying higher altitude cloud in JJA, which would affect the retrieval of Nd. It would be good to look at the types of clouds and situations in which Nd is being retrieved in JJA – i.e., whether most of the retrievals come from mid-level clouds, or clouds with overlying ice cloud, etc.
- Can the composite analysis be done with the Cape Cod CCN data? Or can the CCN be used in the ML model? E.g., is it a better predictor than surface sulphate mass?
- Can you look at boundary layer decoupling index? Perhaps the high cloud fraction regime in winter is more coupled than the summer regime allowing more efficient transport of the surface aerosol to the clouds?
- It seems that the offshore flow prevalent in DJF (cf. southeasterlies from the open ocean in JJA) might also play some role in determining Nd since it may transport more CCN from the continent? Are there more measurements that can help to elucidate whether this may be the case (e.g., aerosol size distribution data or additional CCN data from the Cape Cod data)? It might be good to include the wind direction as a predictor in the ML?
- Other possible reasons for the DJF aerosols (or aerosols when there is lots of PBL cloud) being more efficient at making droplets could be discussed. E.g., could the aerosol be more hygroscopic in these conditions, could there be higher number concentrations (since here you mainly consider mass concentrations). These difference may be related to the different aerosol sources due to the different wind direction (see previous point).
- Why did you not include AOD, speciated AOD, speciated boundary layer AOD, etc., in the ML model so that their impact relative to sulphate surface mass, etc. can be quantified?

Also some more line specific comments :-

Section 3.1 – I'm not sure how well this section works where it is, or how useful the aircraft analysis is for the main conclusions of the paper. Maybe it would be better placed at the end of all the other results in order to help highlight some of the issues raised in the rest of the paper?

L318 – Is it possible to calculate an approximate activation diameter for 0.43% supersaturation given the other aircraft measurements? This would make it easier to compare to the Dp>10 and Dp>3um data.

L381 – "Consequently, humidity effects on remotely sensed aerosol parameters cannot alone explain the dissimilar seasonal cycle of Nd and AOD, but can plausibly contribute to some extent."

- You haven't proved this quantitatively. Can you do a calculation of how much impact the RH difference would have?

L411 – "but do not contribute significantly to number concentration as demonstrated clearly by airborne observations (Figure 1)."

- It's not clear which part of Fig. 1 demonstrates this? Can you explicitly point this out?

L413 – "This is supported in part by how DJF is marked by the highest fractional AOD contribution from the PBL (59 – 72%) where sea salt is concentrated. In contrast, JJA has the lowest fractional AOD contribution from the PBL (11.3 – 52.6%)."

- But this could also indicate higher CCN concentrations in the PBL in DJF perhaps due to the aerosol being more trapped there than in summer?
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Figure 3 – It looks like here the PBL AOD would be higher in DJF than for JJA for many of the regions. It would be good to quote the PBL AOD values in Table 3. You need to describe these results in more detail - the enhanced PBL values in DJF are an important point to describe even though it seems that it doesn't explain the Nd seasonality.

L447 – "We next compare MERRA-2 speciated aerosol concentrations at the surface (Figure S2) to those of speciated AOD (Figure S1)."

- I think that this information is very important and should not be in the supplementary since whether the PBL aerosol mass concentrations (or ideally CCN number concentrations, but I think they are not available from MERRA?) are lower in DJF compared to JJA is a key part of the analysis regarding why the DJF AOD is lower and yet the Nd higher compared to JJA. Indeed, Fig. S2 suggests that surface sulfate mass concentration is about the same in DJF and JJA despite the AOD difference (and sulfate AOD is also higher in JJA than DJF according to MERRA). Of course we might expect sulfate mass concentrations to be even higher in DJF than in JJA if it was to explain the Nd difference, but it does suggest that part of the issue is that AOD is vertically integrated and is not just for the PBL.

- Figs. S1 and S2 also suggest that near the coast sulfate AOD dominates over sea-salt, which argues against the higher observed (Fig. 3) PBL extinction values in DJF being due to sea-salt (as argued e.g., L413 and L707).
- Although sea-salt surface mass concentrations are higher than sulfate. Do you have speciated profiles of extinction from MERRA? These could be used to quantify the effect of sea-salt on the PBL AOD.
- What seems a bit strange given that DJF and JJA have similar surface SO4 in MERRA is that the Cape Cod observations show lower CCN concentrations in DJF. It would also be good to discuss this a bit more along with potential caveats. You mention that the 1% supersaturation at which the CCN are measured is quite high and would be counting fairly small aerosol particles – I think it would be good to show the lower supersaturations that you say are available. Or at least check whether the DJF values of these are also lower than in JJA (data permitting). Is there any other CCN data down the east coast of US since it would be very useful here. Or do you have observed aerosol size and composition measurements that might help determine whether the supersaturation has a big effect and whether there really are fewer CCN in DJF?
    - Also, how representative is the 1-year of data likely to be? Could the interannual variability be large enough to make that result uncertain?
    - It could also be that MERRA is doing a poor job of representing the sulphate mass concentration.
    - Finally, it would be good to quantify how likely it is that we can have a similar surface sulphate mass concentration, but different CCN (using observations or a more sophisticated model perhaps).
- So overall I think that the analysis discussion on the above issues can be improved quite a bit.
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Typos / grammar etc.

L252 – "The ALE value of feature S" – it's not clear here what you mean by "feature". The symbols for this equation have also not all been explained. What does subscript c refer to? What are x_s, x_c, z_s? "The value of fs,ALE(xs) can be viewed as the difference between the model's response at xs and the average prediction." – this is not very clear – I think this explanation needs to be clearer for interpreting the later figures.

L288 – the flight leg abbreviation "BCT1" is stated here without definition. It would be good to have introduced the different flight legs being described before this with reference to Fig. 1.

L305 – "ranged" -> "ranged from".

L307 – "which is a fairer comparison with the ACB1 leg" – fairer than what?

L311 – "there was a significant offshore gradient in LAS submicrometer particle number concentration and AMS non-refractory aerosol mass, ranging from 424 cm-3 and 5.60 µg m-3 (from

BCB1) to 21 cm-3 and 0.32 µg m-3 (from BCB3), respectively; these values are based on times of the maximum and minimum LAS concentrations during the BCB1 and BCB3 legs, respectively."

- This sentence was a little confusing and could be made clearer I think.

L320 – "There was a slighter gradient in particle concentrations with Dp > 3 µm (most likely sea salt) between the same two points of maximum and minimum LAS concentration in BCB1 and BCB3 legs, respectively: 0.26 cm-3 to 0.11 cm-3."

- This could be written more clearly.

Fig. 4 – "The notches in the box plots demonstrate whether medians are different with 95% confidence."

- Different to what? Or do you mean it shows the 95% confidence range of the median?

L486 – "Coefficients of determination (R2) when computing seasonal ACI values"

- What do you mean by this? Is this the correlation coefficient between Nd and the aerosol proxy?

L521 – "Subsequently, one standard deviation from both sides of the seasonal mean defined cut-off points outside of which we assign values as being low and high in each season."

- Could be written more clearly.

L683 – "will struggle for analysing".

L702 – "There were significant changes" – what were the changes?

L705 – "and surface-based aerosol mass concentrations and CCN concentrations (1% supersaturation) are generally highest in JJA and MAM and are at (or near) their lowest values in DJF"

- Surface sulfate aerosol mass concentrations were actually similar in DJF and JJA.

L725 – "by CAO type of conditions" – better as "by conditions associated with CAOs".