F. Zhang et al., Challenges of parameterizing CCN due to…., ACPD, 2015

Overview -

The authors report measurements relevant to the problem of CCN activation. Their data set contains activated ratios (AR(D)) at six supersaturations (S ~ 0.07, 0.1, 0.2, 0.4 and 0.8%), an assessment of organic aerosol mass fraction (integrated over all particle sizes), ambient aerosol particle size distribution (PSD), and ambient CCN concentration.

Comparing measurements made at two locations (Xinzhou in 2014 and Xianghe in 2013), the authors find a difference in the shape of the AR(D) and a difference in the 50% activation dry diameter. Some of the Xinzhou AR's plateau at a value that is significantly less than unity.

What I have summarized (these results are in Figures 1 and 2) is compelling, easy to understand and worthy of publication. However, the presentation that follows (Figures 4, 5, 6, 7 and 8) is difficult to understand. Most perplexing is the use of terminology "estimated Nccn", "calculated Nccn", "predicted Nccn" and "modeled Nccn" to describe quantities derived from the measurements. As I discuss below, these things need to be described.

There another issue:

I am surprised the two methods (estimated and observed in Figure 5) do not agree better. I note that there is a bias and that the correlation coefficients (R^2) is rather small. Is it possible that coincidence in either the SMPS/CN or in the CCN contribute to this poor agreement? Related to this, Rose et al. (2008) say: "For the calibration experiments, the number concentration of monodisperse aerosol particles was kept below 3×10^3 cm^{-3} to avoid counting errors caused by coincidence." The other possibility is that using a campaign-averaged AR(D) may have contributed the discrepancy. Perhaps there are other reasons. The authors should address why the two methods compared in Figure 5 correlate so poorly.

Summary -

The manuscript needs to be reworked.

Specific Comments -

Abstract

Isn't there is a contradiction between the statements 1) variation with PSD showed a poor correlation and 2) the PSD played a dominant role?

P16146L13-16

Here you say there is little pollution from cars or industry, but in the next sentence there is mention of plumes from Xinzhou. This needs clarification.

P16147L10

These “relative deviations” are because of particle loss in the nafion dryer? Also, it is not clear what the “kinetic limitations” are.

P16147L25

It is the inner diameter, not the outer diameter, that is relevant.

P16147L29

2.5 mm ?

P16148L19

“..temperature stability was zero.” I don’t understand what you are referring to here.

P16148L21

Here you define the “aerosol number (CN) size distribution spectrum.” How is this different from the PSD mentioned on P16147L1 and on P16148L24?

P16148L27

Since Figure 1 shows efficiency curves for both sites, I don’t understand why the “campaign-averaged” efficiency was used for this. Do you mean the campaign-average for the Xianghe site? To me it is unclear what the campaign is (both Xinzhou in 2014 and Xianghe in 2013; or just Xinzhou in 2014)?

P16152L6

It is the “CCN activity”, not the “aerosol activity”, that is the focus here.

P16153L2

Transportation -> transport

P16153L26

Here is a relevant reference.

Snider, J.R., and, S.Guibert, J.-L. Brenguier, and J.-P.Putaud, Aerosol activation in marine stratocumulus clouds: Part – II Köhler and parcel theory closure studies, J. Geophy. Res., 108, doi:10.1029/2002JD002692, 2003

P16154L9-9

I don’t agree with your statement that the Ji and Shaw (1998) and/or the Twomey (1959) ($Nccn = CS^k$) parameterizations assume uniform aerosol composition. I also do not agree with your assertion that these parameterizations do not take into account variation in CCN loading. For example, you could have taken an _observed_ CCN spectrum (Nccn versus S) and fitted that as a function if S (e.g., $Nccn = CS^k$).

That spectrum accounts for the PSD and composition. The issue you mention is relevant only if you do not have a measured CCN spectrum and you proceed, in a model, with a generic "C" and a generic "k".

Figure 1

Symbols and line color (black and gray) are used to designate results obtained at the two locations (Xinzhou and Xianghe). The plot would be much more easily understood if you present with a better scheme for delineating the two stratifications (location and supersaturation).

Figure 2 – Case #1

Isn't the behavior seen here (AR < 1 at large diameter) indicative of an externally-mixed aerosol, for example one containing pure soot and hygroscopic particles? Isn't that worth mentioning/discussing? Isn't external mixing a complicating factor when it comes to converting the measurements to a CCN spectrum?

P15154L7

What are "bulk ARs"?

P16154L8

Reference to the specific section of Pruppacher and Klett is needed. Pruppacher and Klett is a huge book.

P16154L23

In Figure 4 *t*here is no obvious indication of negative correlation in the plotted data, or in the fit lines (all of these have positive slope). If you provided the "R", not R^2 (text) and if you defined the "R" (Pearson product moment), that would solve this problem. Also, the Figure legend shows $R^2 < 0$, which is mathematically impossible. All of this needs work.

P16154L25-26

I find this statement of a "CN size distribution" confusing. Isn't this the PSD define earlier. If that is the case you should use a consistent definition throughout. If a distinction is needed, you should define/distinguish these early in the manuscript.

P16155L1

This is confusing. If you multiply the CCN efficiency spectrum by the (CN) size distribution, you get a "CCN distribution", but that is only valid for the selected S. I feel more clarity is needed here.

P16155L2

What you are calling the “estimate” is the summed product of AR(D) and PSD(D)? Right? By “Observation” you are talking about the direct measurement of the ambient Nccn(S) made _without_ the DMA in front of the CCN instrument. I did not see mention of the ambient Nccn(S) measurement (without the DMA in front) in Section 2.1.

P16155L5-6

This is a big jump. Why the would the predicted CCN (you meant estimated, or is “predicted” different?) be influenced by PSD more than it is already? I note that the estimated is the summed product of AR(D) and PSD(D)?

P16155L8

Here you are using “calculated” CCN? Is this different from “estimated” CCN and “predicted” CCN? If you mean “estimated CCN”, it is not clear how the organic mass fraction is being used in these calculations.

P16155L19

I would reword this because the Figure 7 shows how the difference (estimated minus observed) varies with chi-org. The latter is the independent variable. There are other places in the manuscript where “sensitivity” is used. I would change the word order in some of these instances too. E.g., P16145L26. There are other places too.

Figure 5

These are single-parameter (slope) fits, I think. Hence, the “linear” used here is a special case of linear. This comment applies to Figures 6 and 8 too.

P16158L24

Mr. Mcribb’s. Isn’t this Maureen Cribb?