Atmos. Chem. Phys. Discuss., 15, C2449–C2453, 2015
www.atmos-chem-phys-discuss.net/15/C2449/2015/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the competition among aerosol number, size and composition in predicting CCN variability: a multi-annual field study in an urbanized desert" *by* E. Crosbie et al.

E. Crosbie et al.

ewan@email.arizona.edu

Received and published: 11 May 2015

>We thank both reviewers for thoughtful suggestions that have helped us improve the manuscript.

Reviewer 2

General Comments:

This paper presents a detailed climatology of CCN concentrations, characteristics, and related aerosol properties based on two years of measurements in an "urbanized desert." In addition, CCN closure analyses are conducted and the predictive skill of pa-

rameterized models of CCN concentrations are explored. This paper makes important contributions to the field by presenting long-term measurements in an under-studied environment; however, this paper would benefit from a more thorough discussion of some of the methodology and the broader implications of this work. I recommend publication after the consideration of the following comments.

Specific Comments Paper 1. Two years of CCN measurements conducted at a supersaturation of 0.2% are presented. The authors should consider noting briefly why this value was chosen. Is this somewhat arbitrary or does it reflect a "typical" updraft condition/conditions specific to the climate of interest?

>Response: It is not an especially relevant supersaturation for the Tucson climate, however, it is a value that has been used in a number of other field measurements. It is also a worthy choice from the perspective of understanding CCN variability since it results in an activation diameter in the 100-200 nm range, for typical particle hygroscopicities. This range is close to the peak in the number distribution and hence CCN at 0.2% supersaturation is well suited to making comparisons with variability in the aerosol size distribution. The latter point is not relevant to the conclusions, and so we do not feel it is beneficial to attempt to try and offer further explanation for this choice in the manuscript.

2. Details regarding the cluster analysis are provided in the supplement; however, some of this information should be provided in the main text. In addition, further information is needed to describe this portion of the methodology, either in the supplement or the main text. Specifically:

- The reasoning for the selection of 4 clusters in the K-means clustering analysis should be included in the main text, as should a brief description of the definition of cluster associations/cluster assignment weights, especially given the presentation of this latter metric in Figure 5.

>Response: We have made this addition.

- Because the “fuzzying” of the cluster associations is outside of the more traditional application of K-means clustering, in which distributions are assigned to a single cluster, a justification of this choice should be provided. Would changing the number of clusters be another method by which the authors could achieve their goal of considering the transitions between/combination of physical process/regimes?

>Response: Part of this has been covered in the additional text added in response to the comment above. K-means is most successful in isolating patterns when there are distinct low-density regions separating high-density regions that are in a small neighborhood surrounding the centroid. In this case the data are far more continuous and thus the transition “region” between two clusters can have a significant effect on the cluster centroid and respective mean properties, since there are a large number of observations in this category. The implication of this is two-fold: 1) it is difficult to objectively determine the optimal number of clusters; and 2) the decision on how the cluster boundary is defined has an important effect on the cluster. Fuzzying helps to relieve the sensitivity to the latter, which could also be achieved by increasing the number of clusters, but this carries the penalty of more degrees of freedom i.e. 100% membership of C could be adequately approximated by 60% A + 40% B.

While the objective was to retain the smallest number of clusters, was there evidence that other potentially important physical processes/aerosol regimes contributed to variability in particle size distribution properties when more clusters were considered?

>Response: The answer to the question is “no”. Retaining the 4 clusters revealed the important result of the split seen in the summer between the “nucleation” cluster and “coagulation/condensation” cluster. Adding more clusters did not yield additional nonlinear behavior (at least for moderate increases in the number of clusters) such that intermediate states could be well represented by combinations of the current clusters, which is permitted by the fuzzy boundaries.

3. Under some circumstances, cluster-derived parameters led to improvements in

model performance. How do the authors envision the application of the presented clustering methodology in future field studies in other geographic regions and climates? I am particularly interested to know if there is evidence for the potential of a more generalized approach that could be applied to a large number of sites. In other words, some of the clusters presented here are likely to be specific to the region of interest or other very similar climates/sites. In their clustering analysis, did the authors see evidence for the potential for defining the clusters such that they more broadly represent the processes influencing size distributions at a wide number of sites?

>Response: We acknowledge that there could be many ways to implement this type of clustering to a more generalized problem. The method relies on using training data to first establish the cluster shapes – but following that, any data could be classified. If there is the potential for a regime not included in the training data to be important then some minimum threshold should be established to define an “undefined” cluster association such that it could be flagged that a particular site is not well reflected in the clusters. The critical aspect of establishing generalized clusters would be to capture variability that is sufficiently “extreme” to describe the salient modes in the size distribution, while sufficiently frequent to garner enough member associations for quality statistics. We do not claim to be able to optimally determine this, however, it is an interesting problem.

4. Along similar lines, with the aim of improving the representation of cloud properties and processes in large-scale models, in what ways does this work inform future similar campaigns in which long-term measurements of CCN and aerosol properties are measured? What measurements are crucial to this effort?

>Response: The community could benefit from studies of this type (arid and non-arid) regarding the feasibility of simplistic closure assumptions to identify how consistent this is. We would suggest that, at minimum, continuous CCN and aerosol size distribution data are needed to perform sensitivity studies that assess the respective role of number, size and hygroscopicity on CCN variability. We feel no additional text is required in

the manuscript to address this comment.

5. Particle composition measurements are for PM2.5, while size distribution measurements cover a range of 13 – 748 nm. Could differences in composition and/or mixing state for particles larger and smaller than 748 nm contribute to heterogeneity in the degree of variance in CCN concentrations explained by size and composition? Could the influence of this vary temporally, on hourly and seasonal scales?

>Response: Yes, without doubt, one could consider cases where size dependent composition effects significantly affect the inferences that can be made from PM2.5 speciation on CCN relevant composition. A good example of this in Tucson would be the prevalence of dust in PM2.5 as seen in the IMPROVE fine soil mass concentration. However, where we focus on emissions from urban mobile sources and secondary aerosol, we anticipate that the majority of the mass be attributable to sizes relevant to CCN. While we acknowledge that the mixing state and composition may be function of size within the CCN relevant range, we feel that it still offers some insight into CCN variability. Future efforts in the region should consider the use of measurements of size resolved composition. We feel that no additional text is required in the manuscript to address this comment.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 3863, 2015.