Interactive comment on “CCN activity of organic aerosols observed downwind of urban emissions during CARES” by F. Mei et al.

Anonymous Referee #1
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Mei et al. consider the nucleation of droplets (using a CCNC) from a size selected fraction of the atmospheric aerosol (using a DMA) and compare this to aerosol chemistry using an AMS instrument at a remote California field site. Mei et al. derive the compositional parameter k, which is rather low compared to past continental studies, and show this is likely due to the high organic fraction of the aerosol at this location. The values, while somewhat lower than typical continental, are comparable to studies in similar areas (chemistry) and this should help the field to better understand that these low k values are common.

The paper is well written and it is an excellent addition to this special issues on CARES.

I have a few points I hope the authors will consider.

We appreciate the thoughtful comments and constructive suggestions from the referee. We have modified the manuscript accordingly. Below we address each of the comments specifying where in the manuscript we have made changes.

(1) I found the maximum value of only 0.9 (90% of particles activated) at 108 nm in Figure 3 a bit surprising at the highest supersaturations (_0.8%). Would not even particles of this size with minimal soluble material activate at this supersaturation? Can the authors include the activation RH required for insoluble particle of this size for comparison?

Yes, only particles with very low hygroscopicity ($\kappa<0.014$) remain unactivated at supersaturation of 0.8%. Wettable but insoluble (i.e., nonhygroscopic) particles of 108 nm activate at a supersaturation of 2%. In the manuscript, these unactivated particles were considered as nonhygroscopic.

(2) It is not clear what additional information is provided in panels (b) and (c) of figure 1 (don’t they essentially repeat concentration with the only difference being density)?
We believe the reviewer is referring to figure 2 (b) and (c). Those figures provide somewhat different information. The mass concentrations of both sulfate and organics in Figure 2(b) showed a relatively modest increase (~30% over otherwise nearly constant background) during this period. However, the species volume fractions (in Figure 2(c)) were essentially constant during the day. For multi-component particles, particle hygroscopicity is a volume average of the component hygroscopicity. Therefore particle hygroscopicity varies with volume fraction of the species. The organic volume fraction also influences the uncertainty in derived organic hygroscopicity as described in section 4.

(3) This manuscript requires more careful editing. There are several grammatical mistakes. E.g. 9379 “suggesting (the) vast majority” as one example.
We have corrected the mistake and the revised manuscript has been checked carefully.

(4) My major concern is that the paper, especially the Results section, is too AMS-centric and, as a result, is rather too long. What I mean by this is that there is excessive detail spent on e.g. O:C ratio and fraction of signal in mass 44 peak. While this is likely of interest to the AMS user it is not an atmospheric quantity (rather these are instrumental outputs, not results). The Results section could consequently be shortened a great deal by concentrating on the important compositional trend impact on the hygroscopicity parameter k. The amount of detail in O:C ratio and f44 can be moved to the supplementary material where an interested user can find it. Figures 7 and 8 don’t really add much information to the non-AMS user and can be moved to supplementary materials as well. Again, this detail is something that should be presented at a users meeting, not the scientific section of the paper. It detracts from the real message.

Indeed O:C and f44 are derived from instrumental outputs, but they also provide key information about sampled aerosol. O:C ratio is an important chemical property of organic compounds and the O:C of an organic aerosol indicates the average oxidation level of the molecules that compose the OA. As shown in this work and previous studies, the relationship between O:C and κ_{org} can be utilized to predict the CCN activity of an aerosol based on its composition. However, whereas f44 is available from any AMS spectra, O:C can only be determined directly from high-resolution mass spectra. Since f44 and O:C are usually found to correlate well in ambient aerosols [e.g., Aiken et al., 2008], f44 has been frequently used to estimate the O:C of an OA when only the unit-resolution mass spectrum is available. An important finding from this work is that the correlation between κ_{org} and f44 is substantially better than that between κ_{org} with O:C, suggesting that organic acids likely dominate OA water uptake and activation. However, this finding needs to be verified in future studies and at other locations. As the main objective of this study is to examine the relationship between κ_{org} and organic oxidation level, we think it is important to include the details describing how size-resolved O:C ratio was derived, and the relationships between O:C and f44. In addition, given the broad application of AMS and aerosol chemical speciation monitors (ACSM) for measurements of atmospheric aerosol composition, it is important to understand the variability of the relationships among the oxidation level (i.e., O:C), f44, and organic hygroscopicity at different locations. For these reasons, we feel it is necessary to show the correlations between f44/O:C and κ_{org} and their comparison with earlier studies in the main text, which is one of the key results of this study.

Reference:

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Duplissy, A. Metzger, U. Baltensperger, and J. L. Jimenez (2008), O/C and OM/OC Ratios of Primary, Secondary, and Ambient Organic Aerosols with a High Resolution Time-of-Flight Aerosol Mass Spectrometer, Environ. Sci. and Technol., 42(12), 4478-4485.