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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud condensation nuclei in polluted air and biomass burning smoke near the mega-city Guangzhou, China – Part 2: Size-resolved aerosol chemical composition, diurnal cycles, and externally mixed CCN-inactive soot particles" *by* D. Rose et al.

Anonymous Referee #1

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Rose et al., provide size-resolved chemical composition and CCN properties of aerosols measured in the polluted region of Guangzhou, China. The authors use AMS, VTDMA, and CCN data to infer information about the particle hygroscopicity, volatility, and mixing state. The authors utilize Q-AMS organic and inorganic fractions to predict overall particle hygroscopicity. VTDMA data suggest that low volatility externally mixed soot particles exist and are CCN-inactive. The inclusion of the low volatility information improves the overall prediction of aerosol hygroscopicity.

The submitted work is an extension of the authors previously published work and includes VTDMA and AMS information to improve prediction capabilities. It will add to the existing body of work on Cloud Condensation Nuclei and the influences of volatility on particle hygroscopicity. The paper addresses relevant scientific questions and is within the scope of ACP. The topic matter is extremely dense. The abstract will be improved with a clear and consistent conclusion. The overall presentation is structured but mixed messages are presented. Some of the results appear insufficient to support mixing state interpretations and conclusions. The reference list is robust but additional citations of related work can be added. The following comments once addressed will strengthen the overall flow and impact of the paper.

MAJOR CONCERNS

The conclusions are nicely summarized but the following statements in the abstract seem somewhat contradictory. The authors first state that "the constant kappa value [k=0.3] CANNOT account for the observed temporal variations in particle composition and mixing state (L28 P26843)" but then say "the results confirm that an average value of k=0.3 CAN approximate CCN concentrations when size distribution without chemical composition information are available (L3 P26844)". The following sentence (L6 P26844) then supports the original statement (L28 P26843) and says that more information is necessary to improve the k=0.3 predictions. Is it simply that that on a global and climate modeling scales the value of k=0.3 may be applied but it fails to capture regional and temporal variations of CCN? Can the authors simplify and clarify the logic in the abstract? This would apply a unifying message and improve the impact and importance of the paper.

L2. P 26849. What is the uncertainty associated with the density value (1.7 g cm-3), used to convert vacuum aerodynamic diameter to mobility equivalent diameters induce? How does this influence kappa_a,p?

The viewer has several concerns regarding the conclusions drawn from AMS and CCN

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correlations. There is an evident bias towards the proposed empirical fit suggested by Gunthe et al., 2009. E.g., the authors state "kappa_a became too high compared to the expected hygroscopicity (P26353 L14)". Is it not that the predicted value could not fully describe the measured behaviour? The authors of this paper then reconcile this discrepancy and subsequent flaws in fits to AMS data at low mass concentrations. Should we not trust the measurement more than the prediction? The reviewer has concerns that the observed discrepancies between measurements and prediction may be real. Gunthe et al., 2009 proposed the mechanism from similarly low total mass concentrations in the Amazon. The Amazon is a somewhat pristine environment however low concentrations in the Guangzhou Region maybe indicative of very aged complex organic background aerosol. What is the composition of aerosol below 1 ug m-3? What is the fraction of organic constituents? Organic composition if soluble or surface active can contribute up to 40% variability in kappa-CCN activity (Juranyi et al., 2009). In regions where inorganics may dominate such as the range between "kappa_a =0.25 to 0.55, the predicted values deviate from the observed ones on average by less than 20% (L12 P26854)" thus closure between measured and predicted CCN properties is more likely to be in better agreement.

"Non-volatile is considered to be mostly soot but contains non-refractory material" (L26849. L28). The authors should address the implications of this statement for kappa-hygroscopicity. This suggests that using assuming k_org as fully soluble will induce discrepancies in closure, especially since it is supported in this paper that the lower volatility stuff is non-hygroscopic and accounting for it improves closure.

L24. P26854. Non-constant kappa values for varying S maybe indicative of either solubility or surface tension effects in addition to error caused by steep slope of aerosol size distribution.

Fig 3. Are the slopes different for BBE and non-BBE?

L5. P26856. This correlations supports previous work that the volatile organic compo-

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nents are hygroscopic and CCN-active.

Why are the soot particles assumed to be externally mixed? It is not clear to the reader how the authors are inferring mixing state from this data set. In addition, the utility of the kappa-hygroscopcity CCN model is that predictions are independent of mixing state, hence internal and external mixture considerations are not required for kappa based predictions.

How significantly does kappa_a differ from kappa_t in this data set? Can you provide a plot of one value against the other (in addition to Fig. 11) ? If they are similar in value, then kappa_t_p is essentially a function of the observed kappa_t. Inotherwords, at larger S equations (3) and (4) are only modifications of measured values and the closure between predictions and measurements may never be perfect.

MINOR CONCERNS

P26844 L 17. What is meant by “source processes”? This phrase is somewhat ambiguous.

P26844 L25. Please include Wang et al., 20010

P26845 L26. Please include Antilla et al. 2010.

P26846 L 1. Remove “how”

P26848. L11. CDF?

P26855, L12. Why is it an external mixture?

P26858. L23. Is it (kt,p) is as follows?

P26859. L1. Replace, “the predicted k_t values” with “the predicted k_t,p values”

Does Table 3, include or exclude BBE?

Fig 4. What is the closure fit? What is the slope of the data points? How much uncertainty do the points lie between?

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Fig. 9. Axis titles are very small.

Fig 12. Figure is very small and difficult to read.

SUGGESTED REFERENCES

Wang, J., Cubison, M. J., Aiken, A. C., Jimenez, J. L., and Collins, D. R.: The importance of aerosol mixing state and size-resolved composition on CCN concentration and the variation of the importance with atmospheric aging of aerosols, Atmos. Chem. Phys., 10, 7267-7283, doi:10.5194/acp-10-7267-2010, 2010.

Anttila, T. (2010), Sensitivity of cloud droplet formation to the numerical treatment of the particle mixing state, J. Geophys. Res., 115, D21205, doi:10.1029/2010JD013995.

Zaveri, R. A., J. C. Barnard, R. C. Easter, N. Riemer, and M. West (2010), Particle-resolved simulation of aerosol size, composition, mixing state, and the associated optical and cloud condensation nuclei activation properties in an evolving urban plume, J. Geophys. Res., 115, D17210, doi:10.1029/2009JD013616.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 26841, 2010.