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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Mixing state and compositional effects on CCN activity and droplet growth kinetics of size-resolved CCN in an urban environment" *by* L. T. Padró et al.

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**Reply to Reviewer # 3**

**Padro et al. discuss CCN measurements made in Atlanta, GA during a summertime study. CCN closure was examined with the knowledge of concurrent bulk water-soluble chemistry measurements. The primary aspect of the manuscript that could be improved is the description and justification of the assumed mixing state scenarios; suggestions for improvement are provided below. The conclusions could be improved by adding more discussion and clarifying what new was learned from the study (or at least how the results compare with other lo-**



**cations). Overall, the manuscript is well-written, particularly the introduction, which provides a comprehensive overview of previous CCN closure studies.**

*We thank the reviewer for the positive and thoughtful comments. We will add more discussion in conclusions and abstract on what was new and was learned from the study.*

**Major Comments:**

**Abstract: The abstract should be revised to highlight that the results of concurrent chemistry measurements were utilized for this CCN closure study; the abstract should also clearly state the new information being added to the literature by this manuscript.**

*Done.*

**Introduction: The introduction is well-written and provides a comprehensive overview of CCN closure studies and why they are needed. One thing that would be beneficial would be add a sentence or two add the end of the introduction explaining what new information this study adds to our knowledge of CCN predictions.**

*Thank you. Change is made.*

**Page 32729, lines 9-11: It is difficult to discern this trend from Figure 2a; it would be useful to add a description in the text as well.**

*Good point! A description will be added to the text.*

**Section 3.3: While it is useful to compare unrealistic chemistry scenarios for CCN closure, such as all aerosol being composed of ammonium sulfate, it should be discussed that this is simply a comparison scenario, rather than something expected of the ambient aerosol, particularly since bulk chemistry data is available.**

*This is a good point and will be explicitly stated in the revised manuscript.*

**There is some discussion of the justification of some of the scenarios, but the authors are encouraged to elaborate further. In addition, while only water-soluble material was measured, could the insoluble fraction be estimated by comparisons with number concentration or PM2.5 filter measurements? This would better inform the mixing state case scenarios.**

*We do consider the aerosol number (in the form of E) to express an "insoluble" (i.e., non-CCN active) aerosol mass. Furthermore, the PILS-based hygroscopicity when contrasted against the observed hygroscopicity suggests that the aerosol is mostly composed of soluble material. . . (see if we can estimate insoluble fraction in the CCN-active aerosol based on the hygroscopicities and mention briefly in the manuscript.).*

**In addition, there was a major Atlanta study involving several single particle mass spectrometers in August 1999 (e.g., Lee et al., 2002, Lee et al, 2003, Liu et al., 2003, Middlebrook et al., 2003, Rhoads et al., 2003, Wenzel et al., 2003); the results of this study would also help inform (and justify) realistic mixing state scenarios. In particular, Rhoads et al. (2003) examined the chemical mixing state of particles <100nm in diameter.**

*The level of CCN closure error is already very good (as discussed in section 4.1). Further improvement of closure will give a marginal improvement in predictions of CCN and droplet number. Apart from that, the aforementioned studies took place more than a decade before AMIGAS 2008, and involved the measurement primarily of much larger aerosol and do not reflect the CCN-active particles. On the other hand, the CCN data provide a direct measure of mixing state (through quantifying the fraction of CCN-active particles and their heterogeneity in hygroscopicity), and would better inform the mixing state scenarios. We will however attempt to place the mixing state information here with the studies mentioned.*

**Since the authors note in Section 4.1 that " a more precise knowledge of mixing**



**state (as a function of size) as well as chemical composition (size-resolved) is needed in order to achieve closure", the authors are encouraged to review this literature to create another scenario for evaluation. Further, the assumptions used should be considered later in evaluating "the accuracy and bias of the theoretical predictions" since unrealistic scenarios could potentially produce decent comparisons.**

*These are all good points and are addressed in the revised manuscript.*

**Section 4.1: While the "size-resolved mixing state" scenario was found to improve closure, it is based on 100% soluble aerosol, which is unrealistic. This aspect should be discussed.**

*It is not true that we assume the aerosol is 100% soluble, as the consideration of E means that only a fraction of the aerosol sampled has sufficient soluble material to act as CCN.*

**Figure 9: The day vs night aspect of this figure does not appear to be discussed.**

*This will be addressed in the text in the final manuscript.*

**Page 32741, lines 5-6: It is confusing that it is stated that there was "no clear correlation between different periods and fit parameters" when it is stated in section 2.1, that "the effect of different air masses . . . is evident in . . . CCN concentrations". Please clarify.**

*We apologize for this oversight and will clarify the text. A strong correlation of parameters with airmass was not seen, but the CCN and CN concentrations did correlate with airmass.*

**Page 32741, lines 11-14: Did the PILS WSOC data indeed show an increase in WSOC as suggested here?**

*The WSOC concentration started to decrease on August 19 and went down consis-*

*tently until August 23. The WSOC concentration started to go back up on August 25.*

**Page 32741, lines 26-28: It might also be useful to show and compare the PILS data, rather than just a derived "chemical heterogeneity" parameter.**

*The PILS provides size-averaged chemical composition and cannot be compared against the chemical dispersion (which reflects variability across size) derived from the CCN data.*

**Minor Comments:**

**Page 32724, lines 11-14: Please clarify how "the influence" . . . "varies with size" – confusing sentence.**

*We agree that this sentence is confusing and will be rewritten in the final manuscript. The sentence should read as follows: "We find that knowledge of aerosol mixing state is important for accurate predictions of CCN concentrations and that the influence of an externally-mixed, non-CCN active aerosol fraction varies with size during the day (from 31% for particle diameters less than 40nm to 93% for accumulation mode aerosol)."*

**Page 32726, line 5: "more successful" compared to what? Also, generally AMS data is not presented at $\sim$0.1 Hz time resolution.**

*In the aforementioned sentence, we state that CCN closures utilizing AMS measurements are more successful than closure studies that use other chemical composition measurement instruments (as those mentioned in the preceding sentence, page 32725, line 26). The AMS time resolution should say 1Hz instead of 0.1Hz. This will be corrected in the final manuscript.*

**Page 32726, lines 13-15: To improve this discussion even further, consider explaining why it would be beneficial to predict CCN concentrations without size-resolved chemistry information.**

*Good point! In atmospheric models, predicting size-dependent composition is done at*



*large computational cost, and often justified by the need to predict CCN concentrations accurately. The data presented here suggests that this is not always necessary.*

**Page 32726, lines 22-25: Please clarify sentence.**

*We meant to say that the calculation of droplet number is somewhat insensitive to CCN prediction error; this will be further clarified in the manuscript.*

**Page 32728, lines 12-18: These sentences seem more like discussion points that a description of measurements.**

*The Atlanta site combines a typical urban background with strong biogenic influences; we feel that mentioning this here is useful.*

**Page 32729, line 7: Fix sentence.**

*Thank you for pointing this out. The sentence will be changed in the final manuscript.*

**Page 32729, line 26: What size particles were removed by the filter?**

*The sample stream is filtered with a 0.5mm PEEK (polyetheretherketone) liquid filter.*

**Page 32731, line 17: It is stated that "plumes . . . may occasionally pass over the site"; was this observed during the study?**

*Yes we observed this in a few occasions, although we do not provide any statistics on their frequency of occurrence*

**Page 32731, lines 20-21: What was the range of ambient RH? Were the chemistry measurements done off of the same sampling line (and the particles also dried prior to measurement)?**

*The range of ambient RH measured at the site during the campaign was 26.4-100%. Our instruments were sampling from the same line as the chemistry measurements. The particles were not dried prior to the chemistry measurements, but dried prior the measurement of their size distribution and CCN activity.*

**Page 32732, line 16: Phrase in parentheses is confusing as written; please clarify.**

*The phrase will be changed from "assuming all CN are CCN active" to "assuming all CN are CCN active at a specific supersaturation"*

**Page 32733, line 12: Please clarify what "were removed".**

*We discarded data for which the integral of the fitted sigmoid deviated from the measured CCN concentration by more than 15%.*

**Page 32736: The discussion at the beginning of section 3.3 is well done.**

*Thank you.*

**Page 32736, line 6: While size-resolved chemistry could be incorporated into this method, it should be clarified that the chemistry data used were not size-resolved.**

*As the reviewer points out, this study only incorporates bulk chemistry measurements and not size-resolved chemistry. We mention this in section 2.2 and will restate it in section 3.3 too*

**Page 32737, line 23: It is confusing how PILS WSOC could provide inorganic salt information.**

*The inorganic salt information was obtained from the PILS IC measurements. To determine which salts were present in the aerosol phase, we used the sulfate molar ratio (page 32737, lines 2-7). We used the PILS WSOC along with the inorganic salt concentrations to determine the composition (fraction) of the aerosol.*

**Page 32737, lines 8-10: Were these concentrations observed during the study?**

*The concentration of CCN in the instrument were on occasion in the hundreds cm$^{-3}$. Although too low to impact the measurement of CCN, an observable (small) impact on*



*supersaturation and droplet size may occur A thorough analysis will be considered in future work.*

**Figure 2: The timeline portions of parts a) and b) could be made the same width to allow for a better comparison.**

*We will make period B match period A more closely.*

**Figure 3: This figure does not seem necessary and could be removed if desired.**

*We would like to keep the figure in the final manuscript to avoid confusion amongst the different set ups used during the study.*

**Figure 4: To avoid confusion, it should be noted that the dotted line refers to the 1:1 line Also, was the trendline intercept not found to be significant?**

*Good points. The 1:1 will be identified in the caption, and the intercepts also discussed.*

**Figure 6: For clarity, it would be useful to write out all acronyms/abbreviations and also label the top row of bins**

*Good points. Done.*

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 32723, 2011.