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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.

Anonymous Referee #3

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

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.

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.

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.

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. 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. 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 <100 nm in diameter. 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.

Section 4.1: While the "size-resolved mixing state" scenario was found to improve closure, it is based on 100% soluble aerosol, which is not realistic. This aspect should be discussed.

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

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 was stated in section 2.1 that "the effect of different air masses. . .is evident in. . .CCN concentrations". Please clarify.

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

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.

Minor Comments:

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

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

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.

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

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

Page 32729, line 7: Fix sentence.

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

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

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)?

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

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

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

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.

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

Page 32742, lines 8-10: Were these concentrations observed during the study?

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

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

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?

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

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