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Comment

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.

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

Reviewer 1

General Comments:

The study presents aerosol measurements over two years in Tucson, a major city surrounded by weakly populated desert. The measurements include particle size distributions, aerosol composition, CCN and total number concentrations. By means of

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several assumptions and simplified models, CCN closure is attempted. The data set is clearly unique for this location and also exceeds many other data sets that are often limited to a single season or few months at a time. A new clustering method has been used to sort size distributions based on their likely origin and history. Conclusions on the sources of aerosol particles and reasons for distribution shapes are drawn based on the observed evolution of size distributions in various seasons and the skills of the CCN closure models. Therefore, the current study exceeds previous ones in terms of the measurement period and tools that are used to interpret data. However, I think previous literature should be more carefully taken into account and discussed.

I have several more comments below that should be taken into account before this manuscript can be recommended for publication.

Major comments 1) Introduction The introduction is very long and quite disorganized. It should review the current knowledge of data that are similar to those as used in the current study, e.g. data sets of size distributions and composition that are used to perform CCN closure. Details on specific organic aerosol properties such as surface tension etc. (p. 3867) distract only from the main focus of the current paper. In addition, it seems that the last paragraph on p. 3868 is redundant as it is repeated in the following.

>Response: The introduction has been shortened, removing less relevant sections relating to organic aerosol chemistry.

2) Wording At many places, quite inaccurate or misleading terminology is used. E.g. 'aerosol chemistry' or 'particle chemistry' is often used and it does not become clear whether chemical processes or composition is meant.

>Response: Cases with ambiguous terms have been amended.

Other instances include p. 3865, l. 23: 'cloud droplet distribution' – I don't think that any of the studies cited here compared their data to cloud droplet distributions.

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>Response: McFiggans et al. 2006 shows droplet distribution for an idealized salt activation case; however, we have changed “distribution” to “number” which better reflects the purpose of this statement.

p. 3866, l. 13; p. 3874, l. 18; p. 3880, l. 14; p. 3882, l. 28/9: which processes are referred to here?

>Response: These instances have either been removed or revised.

3) Data discussion In the discussion part of the paper (Sections 3 and 4) often words like 'maybe', 'likely', 'probably' etc are used. While I understand that it might be difficult to give a clear and unambiguous interpretation of the data due to the somewhat limited number of measured parameters, a somewhat more detailed discussion should be given that weights the possible processes/effects in a more quantitative way.

>Response: It is simply not possible to quantify cause-and-effect relationships definitively using purely the observations that we have reported. We have tried to use language appropriate with the level of certainty associated with interpretations made on the reported data.

Paper 4) Previous literature In the introduction, some previous CCN studies are cited together with their challenges and difficulties therein. However, in the discussion section not a single previous study is cited even though there are numerous studies that have been performed in regions where similar mixtures of fresh and aged aerosol exist. Also effects on number concentration, size distribution shapes etc due to daily, seasonal and source-dependent effects have been discussed there. One large aspect that has been highlighted in detail in many studies is the mixing state of fresh vs aged aerosol. The current study has to take into account findings from prior studies and put the current data set in their context. (see also next comment)

>Response: We thank the reviewer for pointing this out and while we are not entirely sure what specific references the reviewer had in mind, we have examined the archives

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of papers more strictly and try to make such references in the Discussion Section. A few representative papers are chosen that focused on urban environments and we added the following text:

“To put the TACO results in more context, fresh pollution aerosol in other urban areas such as Riverside, CA and Houston, TX could not be fully represented without knowledge of size-resolved composition (Cubison et al., 2008; Ervens et al., 2010). A number of other studies have shown that mixing state can help improve predictive capability of CCN behavior (Wex et al., 2010), including in Atlanta, Georgia (Padro et al., 2012) and during early morning rush hour near Mexico City (Lance et al., 2013); but studies also report that hydrophobic particles emitted in urban areas quickly (\sim few hours) become internal mixtures via condensation of secondary hygroscopic species (e.g., Wang et al., 2010; Mei et al., 2013).”

5) Mixing state In the current paper, mixing state is largely neglected and the inability of the simplified model approaches used here to predict CCN is explained by ‘probably associated with the complexity of the aerosol mixing state’ (p. 3880, l. 26). Given that the mixing state might play an important role for part of the current data set, the question arises how meaningful a single kappa is to capture the hygroscopicity of the total aerosol population. This approach should be either better justified or revised. Mixing state should be also discussed in the context of the relative role of various parameters that determine the activated fraction of an aerosol population, e.g., on p. 3881, l. 28/9 and p. 3882, l. 10.

>Response: While the reviewer raises a good point regarding the physical meaning of parameters included in a simple model, we do not believe it is unjustified to use a knowingly oversimplified parameterization as a way of identifying the occasions when it is not satisfactory. We have added an extra discussion at the beginning of this section to identify that this is our intention: “One major simplification is the limitation of the treatment of hygroscopicity to a bulk measurement, which is permitted to vary temporally but does not isolate size dependent changes in hygroscopicity nor the hygroscopicity dis-

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tribution, which may be an important component in relation to external mixing. These aspects are beyond the scope of these parameterizations and are likely to contribute to model shortfalls. Forthcoming work will separately study the degree of correspondence of hygroscopicity between the sub- and supersaturated regimes, size-dependent hygroscopicity and composition, and the closure of hygroscopicity from composition measurements.”

Minor comments p. 3864, l. 15: 'can be parameterized' should be specified here.

>Response: In handling comment 2, this should now be clearer.

p. 3865, l. 6: Cloud microphysical and optical properties are not only governed by aerosol number but also by the total amount of liquid water, which in turn is a function of cooling rates.

>Response: We have replaced “governs” to “contributes to governing”

p. 3865, l. 25: This list should also include mixing state already. It is only discussed later even though it has been shown by detailed studies that it might be one of the determining factors in CCN closure studies.

>Response: We have made this addition.

p. 3866, l. 24: In the two cited studies, CCN closure was quite satisfactory if mixing state was taken into account. This sentence should be reworded.

>Response: Agreed, the current wording is ambiguous. The phrase “under assumptions of bulk hygroscopic properties has been added” to clarify the intended meaning.

p. 3866, l. 25-27: This sentence seems out of place here.

>Response: We have revised this text.

l. 3867, l. 5: Did any of the cited studies indeed look at the effect of chemical processes and/or coagulation on size distributions and CCN properties?

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>Response: These references have been revised. Also the text was somewhat misleading in the manner it connected the statement with the references, so it has been adjusted to be more concise.

p. 3867, l. 12ff (cf. my comment 1)): This information is irrelevant for the current study. If you choose to keep it in the (already quite lengthy) introduction, a more balanced discussion should be given. E.g. sensitivity studies have shown that surface tension effects are rather negligible for CCN effects (Ervens et al., JGR, 2005).

>Response: The introduction has been revised in the process of addressing Comment 1.

p. 3874, l. 25: Is there any explanation for the higher particle concentration during weekends?

>Response: The CCN concentration is higher during the evening, of which we make mention, however, we do not have a full explanation for whether the enhancement is a result of different evening emissions patterns on weekends or some other pathway.

p. 3875, l. 13ff. (i) This paragraph should be a separate section.

>Response: We respectfully disagree with this suggestions as this section is still part of the discussion on diurnal and weekly cycles

(ii) Add to the numbers in parentheses 'kappa = '

>Response: We have made this addition.

p. 3875, l. 1-12: Have the described effects such as a shift in size-distributions due to condensation of semivolatile compounds and the switch from the importance of semivolatiles to more biomass burning been observed in previous studies?

>Response: Not specifically in Tucson. The presence of domestic wood burning emissions is unique to the winter season and is mentioned as a possible explanation for the difference seen in the diurnal cycle in winter which is not present in other sea-

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sons. There was no direct measurement of a tracer which could be used to separate quantitatively the contribution of biomass burning.

p. 3877, l. 14: Has it been observed previously that increasing partitioning of nitrate can indeed affect size distributions to an observable extent?

>Response: Indirectly this is discussed in Andreae and Rosenfeld (2008, p29) referring to the tendency for submicron nitrate to partition onto existing particles (in their discussion, they cite Kleeman et al., (1999)), which increases activation ratio but does not increase CN.

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