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Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #1

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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 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.

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

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.

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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)

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.

Minor comments

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

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.

p. 3865, l. 25: This list should also include mixing state already. It is only discussed

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later even though it has been shown by detailed studies that it might be one of the determining factors in CCN closure studies.

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.

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

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?

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

p. 3875, l. 13ff. (i) This paragraph should be a separate section. (ii) Add to the numbers in parentheses 'kappa = '

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?

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

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 3863, 2015.