

## Responses to Referee #2

We thank the reviewer for the careful review of our manuscript; the comments and suggestions are greatly appreciated. All the comments have been addressed. In the following please find our one-by-one responses to the comments and the corresponding changes to the manuscript. The original comments are shown in italics.

### *Anonymous Referee #2*

*Received and published: 30 October 2015*

*This manuscript explores the effect of mixed anthropogenic and biogenic SOA on CCN activity and droplet growth kinetics, compared to pure biogenic or anthropogenic SOA. The paper is well written and logical, and I would recommend publication, subject to a few comments:*

*Page 19913 section 3.1.1: It would add an interesting angle to refer to the study of Rickards et al (2013) in which a review of existing studies attempting to link kappa and O/C were combined. <http://pubs.acs.org/doi/abs/10.1021/jp407991n>. They found that systematic variability between  $\kappa$  parameterizations determined from different studies remains large, consistent with the O/C ratio providing only an approximate guide to aerosol hygroscopicity. They do however present a new parameterization based on collated laboratory/field data. Given the propensity to rely on simplified metrics, it would be useful to highlight where your results lie as compared to these parameterizations. Indeed, does this comparison confirm any of your findings?*

### **Response:**

We thank the reviewer for the supportive remarks and bringing about the reference.

In the revised manuscript, we have compared our data with the parameterization in Rickards et al (2013) (Fig. S5). Our data are generally in line with the parameterization proposed by Rickards et al (2013) while  $\kappa_{\text{CCN}}$  of BSOA is higher than those predicted by the parameterization. However, because the variations of kappa discussed by Rickards et al (2013) are large and the parameterization is based on various chemical systems and various conditions (consistent with this study), it is difficult to conclude from the comparison whether that study supports or contradicts our data. Note that in the manuscript we compared the kappa of BSOA and ASOA with previous studies, which showed consistency with our data.

In the revised manuscript, we have added the following sentence.

**“The relationship between  $\kappa_{\text{CCN}}$  and O/C was further compared to the parameterization in the study by Rickards et al. (2013) (Fig. S5), which was obtained from their experimental data and a number of literature data and showed very large variability of  $\kappa$  versus O/C.”**

Page 19914 line 3 and figure 1: “The CCN activity of the three types of aerosol is generally similar at similar OH dose. In addition, with exception of BSOA,  $k_{CCN}$  is largely invariant over a wide range of OH doses” But in the figure,  $k_{CCN}$  appears to be slightly higher for BSOA than for ASOA or ABSOA at a given OH dose (particularly around molecules  $cm^{-3}$  s). Plus, it looks like there is a slight increase in  $k_{CCN}$  with OH dose for ASOA. Are these differences not significant / important?

**Response:**

Overall, considering the variations of the  $\kappa_{CCN}$ , no distinctive difference between  $\kappa_{CCN}$  of BSOA and that of ASOA or ABSOA could be identified.

For ASOA, we guess that the reviewer referred to experiment A4.  $\kappa_{CCN}$  at the highest OH dose ( $1.6 \times 10^{11}$  molecules  $cm^{-3}$  s) is similar to  $\kappa_{CCN}$  at the lowest OH dose of that experiment ( $1.6 \times 10^{11}$  molecules  $cm^{-3}$  s). Also considering  $\kappa_{CCN}$  of all ASOA, there is no significant difference in  $\kappa_{CCN}$  over a range of OH doses.

Page 19914, second para and figure 2: In figure 2a it is hard to follow the same SS since they change very soon after the addition of BVOC. In 2b it looks to me like there is a slight increase in  $k_{CCN}$  after addition of AVOC. Do you not consider this significant? Could you also please comment on how you decided the delay time between additions of the different VOCs?

**Response:**

In Figure 2a, the change of kappa can be tracked by the blue and green markers (SS: 0.51% and 0.68%), and at these two SS, there are data before and after BVOC addition. In the revised manuscript, we have clarified this issue.

In Figure 2b, there is a very slight increase after AVOC addition. However, such a minor change can also be attributed to the concurrent aging of SOA. Such increasing trend can be tracked by the data at 0.43% SS (light green marker). Immediately after AVOC addition, there are barely any ASOA formation (ASOA fraction  $\sim 0$ ), but there was already a slight increase of kappa.

The delay time in the experiments with sequential VOC addition was chosen in a way that particle mass concentration generated from the first VOC reached the maximum and then after 1-2 h the second VOC was added. Accordingly, the time lag was longer when AVOC was added first due to its low reactivity.

In the revise manuscript, we have explained this point.

“In experiments with sequential VOC addition, the second VOC was added 1-2 h after the SOA mass concentration from the first addition reached its maximum. Accordingly, the time lag was longer when AVOC was added first due to its low reactivity.”

*Droplet growth kinetics. Section 3.1.2: Of course, this is an area gathering increased attention. With regards to the ‘threshold droplet growth analysis’, whether there is, or isn’t, any kinetic mass transfer effects from the phase state of the aerosol might depend on how the aerosol have been treated. If there is sufficient water, or any plasticizer for that matter, in the particle then water uptake in the CCN counter is likely not going to be affected. Is the aerosol exposed to rapid/slow drying for example? It would help draw out potential effects with regards to this if more detail on the method was given rather than just referring to previous studies.*

**Responses:**

We agree that the residual water in the particle may affect droplet growth kinetics. In this study, the aerosols were produced at typically 30-60% RH and were dried to ~10% RH using a silica gel diffusion drier with a residence time of around 3 s before they were measured by CCN set up. In the revised manuscript, we have added these details.

“Before entering the instruments, the particles were dried using a silica gel diffusion drier (gradually drying to ~10% RH) with a residence time of around 3s”.

*Section 3.3, line 13: ‘an approximate cubic relationship between K and surface tension’. It is much easier to simply state that sensitivity to surface tension in the Köhler equation is increased at the point of activation. There is no need to reference a numerical approximation.*

**Response:**

We have accepted the reviewer’s suggestion. In the revised manuscript, we have modified this sentence as follows.

“ $\kappa_{CCN}$  is more sensitive to surface tension at the point of activation according to the Köhler equation.”

*Please could you add error bars to the diamonds in figure 7.*

**Response:**

Accepted. In the revised manuscript, we have added the error bars to the diamonds.