

## **Response to comments of anonymous referees # 2**

### **General comments**

The manuscript is also fairly well written. I have a few comments for the authors to clarify, and recommend a Minor Revision before publication. The manuscript by Wang et al. reports particle rebound fractions (as a measure of particle phase states) measured for SOA particles produced from the photooxidation of single and binary VOC precursors in an environmental chamber. Compared to previous studies mainly focusing on organic aerosols, this study also examined the role of ammonium sulfate seed particles by varying the organic-to-inorganic mass ratio. The authors found that particle phase states to the first order depend on the RH and organic-inorganic ratio, while the VOC precursor type plays a relatively minor role. The experiments were carefully designed and executed. The paper was well written. However, I do have a few comments that need to be addressed before I can recommend publication.

*Many thanks to the reviewer for the comments and suggestions. We have improved the manuscript accordingly. Please find a point-by-point response below.*

**Specific comments:**

**Q1.** The authors claim o-cresol as a representative anthropogenic SOA precursor. The manuscript explains that the o-cresol was chosen mainly because of its OH reactivity similar to the biogenic precursors and its modest SOA yield. However, a major source of o-cresol can be biomass burning, either from manmade or natural sources. This is not discussed in the manuscript and I think it can be misleading to simply claim that o-cresol is anthropogenic.

*Good point. The description of the o-cresol has been rephrased as follows:*

*“Building on the McFiggans et al. (2019), we added a representative aromatic VOC (o-cresol) into initially designed binary mixtures of biogenic VOCs (isoprene &  $\alpha$ -pinene). o-cresol can be emitted into atmosphere both directly from biomass burning (Coggon et al., 2019; Koss et al., 2018) and indirectly from the oxidation of toluene (e.g. motor vehicles, solvent use (Fishbein, 1985)). Therefore, anthropogenic source is one of the main contributors to the o-cresol but worth noting that the natural biomass burning can also be an important contributor.”*

**Q2.** The presence of inorganic species can significantly alter the rebound curve, and the composition and O:C of organic species play a relatively minor role. I wonder if it is possible to develop a simple mixing rule to predict the liquid-to-nonliquid phase transition? For example, is the transition RH related to liquid water content or hygroscopic growth factor?

*Thanks for your suggestion. During the experiments, we conducted concurrent hygroscopic measurement at 90% RH using a custom-made Hygroscopicity Tandem Differential Mobility Analyzer (HTDMA) for the same particle size with BF measurement. By using  $\kappa$ -Köhler theory, we calculated the growth factor (GF) at the RH of the BF, and linked the BF with the GF as shown in Figure 5 below. A brief description of HTDMA was added to the method, a new section 3.3 to the results to demonstrate the BF-GF relation, and the abstract and conclusions are revised to*

include this result accordingly.

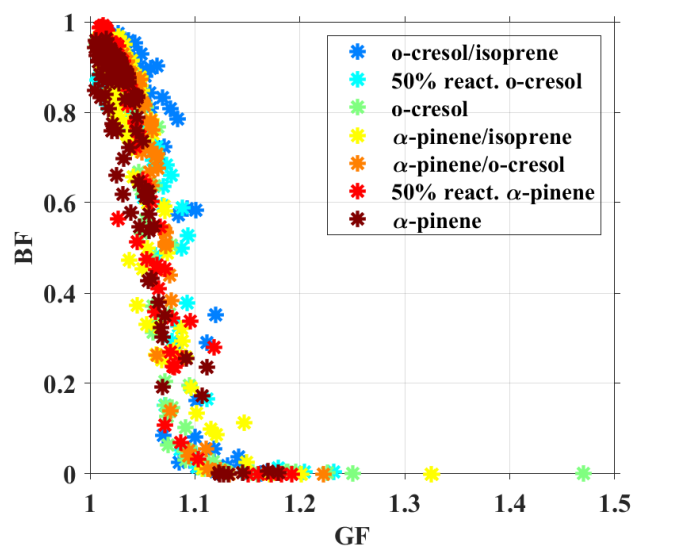


Figure 5. The relation of BF and hygroscopic GF of aerosol particles during six-hour photochemistry experiments in various VOC systems.

HTDMA description:

“Hygroscopic growth factor (GF) at 90% RH of submicron aerosol particles (75 ~ 250 nm) was recorded by a custom-made Hygroscopicity Tandem Differential Mobility Analyser (HTDMA) (Good et al., 2010), which can be used to calculate the GF at given RH using  $\kappa$ -Köhler approximation (Petters and Kreidenweis, 2007).”

### Sec. 3.3 Mixing role of chemical composition and RH (GF) on BF

“As we know that the chemical composition and RH are key factors influencing aerosol water uptake at given size (Kreidenweis and Asa-Awuku, 2014). To discuss the mixing role of chemical composition and RH on BF, the GF at the RH of BF measurement was calculated from HTDMA measurement using  $\kappa$ -Köhler equation, and the relation between the BF and the GF was plotted in Figure 5. It can be seen that the BF showed a monotonic decrease from ~ 1 to ~ 0 with the increasing GF from ~ 1 to ~ 1.15 in all VOC systems. When the GF is larger than 1.15, the aerosol particles kept in liquid

*phase ( $BF \sim 0$ ). This evidence indicated the key role of aerosol water on the phase transition from the non-liquid to the liquid. The  $BF$ - $GF$  relation of the varying multi-component aerosol particles including SOA and inorganic compounds is comparable with the previous study (Bateman et al., 2015). They measured the  $BF$  of the SOA from  $\alpha$ -pinene, toluene and isoprene and found that the  $BF$  is almost 0 when the  $GF$  is larger than 1.15 (Bateman et al., 2015)."*

*One sentence has been added in the abstract to clarify this point:*

*" $BF$  decreased monotonically with increasing hygroscopic growth factor ( $GF$ ) and the  $BF$  was  $\sim 0$  when  $GF$  was larger than 1.15."*

*Added sentences in the conclusion to clarify this point:*

*"Additionally, by combining the chemical composition and  $RH$ , we calculated the hygroscopic growth factor ( $GF$ ) and found its key role in aerosol phase behaviour. The multicomponent aerosol particles were liquid in all VOC systems when the  $GF$  is higher than 1.15 at room temperature and transmitted from liquid to non-liquid when the  $GF$  decreased to  $\sim 1$ ."*

**Q3.** Line 104-105: some studies do find that alpha-pinene SOA coating can influence the deliquescence of ammonium sulfate, although the effect is relatively smaller than isoprene SOA:

<https://www.tandfonline.com/doi/full/10.1080/02786826.2010.532178>

<https://acp.copernicus.org/articles/12/9613/2012/acp-12-9613-2012.pdf>

*Thanks for your helpful comment. The discussion of  $\alpha$ -pinene SOA condensation on deliquescence of ammonium sulfate has been added:*

“These results are partly consistent with similar studies using a different instrument (Hygroscopicity Tandem Differential Mobility Analyzer) (Smith et al., 2011; Smith et al., 2012). They found that the  $\alpha$ -pinene SOA on ammonium sulphate seed can slightly shift the deliquescence and efflorescence RH by a few percent (Smith et al., 2011) while the isoprene SOA can significantly decrease the deliquescence and efflorescence RH depending on the organic fraction (Smith et al., 2012).”

**Q4.** Were the rebound measurements performed for monodisperse or polydisperse aerosol particles? If polydisperse, were there particles smaller than the cutoff diameter of the impactor? This information might be provided in the literature cited in this paper, but it would be nice to briefly describe it here as well.

Thanks for your suggestion. In this study, a DMA was used to select a monodisperse aerosol particles (100 ~ 200 nm) to measure the rebound fraction. The measured 50% transmission diameter ( $D_a$ ) of the impactor is  $84.9 \pm 5.4$  nm, which means that smaller aerosol particles ( $< D_a$ ) will follow the gas stream and larger particles can hit to the impactor to effectively represent the bounce behaviour (Bateman et al., 2014). In this study, our selected sizes are larger than the  $D_a$ , so that we can derive reliable bounce behaviour. The clarification of the cutoff diameter of the impactor has been added to the method of the manuscript as follows:

“During the experiments, a Differential Mobility Analyzer (DMA) was used to select a monodisperse aerosol particles from chamber, with mobility diameter of 100 ~ 200 nm following the growth of the aerosol particles. The selected particle sizes are larger than the 50% transmission diameter of the used impactor ( $84.9 \pm 5.4$  nm) (Bateman et al., 2014) to ensure a reliable bounce fraction measurement.”

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