

## **Response to Referee #2:**

Thanks very much for your comments, suggestions and recommendation with respect to improve this paper. The response to all your comments are listed below.

In this study, the FTIR spectroscopy and two-D correlation analysis were used to investigate the hygroscopic behavior of typical aerosols. It demonstrated that the method is good at qualifying and quantifying the interaction between water and aerosol. In particular, the 2-D correlation analysis can provide more detail information about the hydration process. Thus, this method is helpful to understand the hygroscopicity of aerosols and it is suitable for publication as a technical note. However, the comments of referee #2 are also my concerns, and I have some other concerns as following.

**Response:** All your comments listed below have been addressed. Please check the point by point response as follows.

1. Since the samples were deposited on a ZnSe plate in stacking state, can they also be considered as nanoparticles? Further experiments using samples prepared by deposited solution with further dry process were recommended. Then the results of these two different preparation methods would be compared.

**Response:** The chemical composition of the 100nm nanoparticles is not changed during depositing onto the stacking state. And for the 100nm nanoparticles, its hydration characteristic mainly depends on its chemical composition and the kelvin effect is negligible. The enrichment for the nanoparticles is to improve the signal of FTIR because the hygroscopic signal of a single particle is too weak to be measured by the FTIR method. As a result, we believe this experiment is enough and no further experiments are needed. We have included this explanation in the revised paper.

2. Line 305, what's "surface-limited process"? Since all the initial step of particle hydration could be water adsorption on surface, surface limited process should always determine the hygroscopic behavior.

**Response:** Surface-limited process means the surface limited process could determine the hygroscopic behavior. The rate of hydrolysis depends on the water content from the surface into the substance. In our study, the decrease of  $\text{SO}_4^{2-}$  in the solid AS nanoparticles does not result in a simultaneous increase of  $\text{SO}_4^{2-}$  in the aqueous AS nanoparticles during the AS deliquescence transition. The former process occurred

predominantly before the later process. It suggests an intriguing possibility of the existence of an intermediate state between the solid and aqueous AS nanoparticles. Meanwhile, the water uptake process occurred predominantly before the decrease of  $\text{SO}_4^{2-}$  in the solid AS nanoparticles during the AS deliquescence transition. We speculate that the surface-limited process may control the transport of liquid water to the AS nanoparticle, or in other words, the surface limited process determine the hygroscopic behavior of AS nanoparticles (Leng et al., 2015).

We have added this explanation in the revised version, please check the marked up file for details.

3. In the previous study of wang et al. (2017), the formation of ammonium hydrogen oxalate ( $\text{NH}_4\text{HC}_2\text{O}_4$ ) and ammonium hydrogen sulfate ( $\text{NH}_4\text{HSO}_4$ ) from interactions between OA and AS in aerosols during the dehydration process were observed. Did you detect these species in the present study? Could the 2-D correlation analysis confirm this reaction?

**Response:** In our experiment, the peak of  $\text{HSO}_4^-$  shows in the  $1245\text{cm}^{-1}$ . Because the particles are aerosolized by an atomizer (model 255, MetOne) and dried by a diffusion dryer (model 3062, TSI). The RH would decrease to 25% under this dehydration process. But the mixture OA/AS in our experiment was in the hydration process, so we did not measure the process for the formation of ammonium hydrogen oxalate ( $\text{NH}_4\text{HC}_2\text{O}_4$ ) and ammonium hydrogen sulfate ( $\text{NH}_4\text{HSO}_4$ ) from interactions between OA and AS in aerosols. But during the dehydration process, the 2-D correlation analysis confirm this reaction due to the peak for the  $\text{HSO}_4^-$  or  $\text{HC}_2\text{O}_4^-$