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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Particle acidity and sulfate production during severe haze events in China cannot be reliably inferred by assuming a mixture of inorganic salts" *by* Gehui Wang et al.

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Anonymous Referee #2

General comments:

(1) Comments The manuscript "Particle acidity and sulfate production during severe haze events in China cannot be reliably inferred by assuming a mixture of inorganic salts" of Gehui Wang et al. presents laboratory measurements on the hygroscopicity and particle growth of selected particles under controlled conditions. The work contributes to the current understanding of the aqueous oxidation of SO2 by NO2. The

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authors also report on field measurements of the chemical composition of the water soluble fraction of fine PM in three sites in China during winter episodes. The results suggest little sulfate production in aqueous ammonium sulfate particles due to high acidity, whereas high sulfate production is observed from oxalic acid particles due to low acidity. The manuscript is well written and the results are relevant and I recommend the publication of the manuscript after the following aspects are addressed.

Response: We thank the reviewer's comments above.

Specific comments

(1) Comments: In Section 2, add a detailed description of the sampling conditions during the field measurements for the three sites in terms of meteorology (with emphasis on RH and T), sampling size, and a qualitative description of the emission sources potentially impacting the observations. In Section 4, discuss whether any of these sampling conditions can also contribute to the observed differences.

Response: Suggestion taken. We have added the detailed descriptions on the sampling conditions at the three sites including RH, temperature, particle cutoff size and a qualitative description of the potential emissions sources. See the above information on page 6, line 135-140. We also discussed whether these sampling conditions can also contribute to the observed differences. See page 9, line 219-221.

(2) Comments: How similar are the chemical and thermodynamic conditions in the laboratory measurements to actual haze conditions during the field measurements? The authors should also explicitly address the caveats of extrapolating the laboratory results in terms of mixing state, acidity, and chemical organic composition (organic mass factions are typically large during these events).

Response: The concentrations of gases we used for the chamber simulation are about 10 times higher than those in Beijing, but their relative abundances and the relative humidity are similar to those during the field measurements. As discussed above,

the uptake coefficient of SO2 on oxalic acid particles extrapolated from our laboratory measurements is similar to that in Beijing during the observation period, which suggests that our proposed mechanism is applicable to the actual haze conditions in China. Based on the field measurement and model simulation, Cheng et al (2016) also reported a similar result, corroborating our lab work results. As reply to referee #1, we have added related discussions into the text, see page 9, line 199-202, and page 14, line 316-322.

(3) Comments: The results in figure 2 show a large increase in the hygroscopic growth factor of oxalic acidy particles when increasing RH. Can the authors discuss possible reasons why these results differ from those of Peng et al., 2001 under high RH?

Response: The difference of hygroscopic growth factors of oxalic acid particles between Peng et al 2001 and this study is mainly due to the different methods used for the hygroscopicity measurement. In the work of Peng et al (2001), they used an electrodynamic balance (EDB) system to trap and levitate a charged particle. The relative mass of a particle equilibrated at different relative humidities was determined by measuring the balancing voltage. The size of each particle studied was not measured but was estimated from visual inspection using a microscope (Peng et al., 2001). In contrast, the hygroscopicity of oxalic acid in this study was measured by using a hygroscopicity tandem differential mobility analyzer (HTDMA) system. The particle size was measured by a SMPS (scanning mobility particle sizer). The EDB system measured the size of a charged particle through determining its optical property, while the HTDMA system measured by the SMPS system through determining its mobility in the electric field. In addition, the size range of oxalic acid particles measured by Peng et al (2001) is different from that measured by this study. The size range of charged oxalic acid droplet measured by the EDB system is 10-20 microns, while that measured by the HTDMA system in this study is less than 1.0 micron. As shown in Figure 2, our result is very close to that measured by Mikhailov et al (2009) and Prenni et al (2001), who also used a HTDMA system for the hygroscopcity measurement, further suggesting that the

difference between the measured growth factors is due to the different methods. We briefly explained this difference in the revised version, see page 7-8, line 175-180.

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