

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 #1

General comments:

(1) Comments: The manuscript presents field and laboratory results of sulfate formation during winter haze conditions in China. It points to the potentially important role that organics play in aerosol acidity and hygroscopic growth in China. An important finding is that SO₂ oxidation by NO₂, a new mechanism proposed for Beijing winter haze, is significant only on oxalic acid as seed particles, but not on ammonium sulfate

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due to the strong acidity. This finding was not stressed in the earlier work of the authors (Wang et al. 2016, PNAS). The paper is well written for most parts and should be published in ACP after the following comments are addressed.

Response: We thank the reviewer for the comments above.

(2) Comments: My main comment is the representativeness of the lab experiment for atmospheric conditions in winter haze in China. I would expect the organics and inorganic compounds are mixed in ambient aerosol particles, so there will not exist pure oxalic acid aerosol or pure ammonium sulfate aerosol as used in the chamber experiments. I would suggest the authors discuss this issue in the manuscript, particularly with regard to how their proposed mechanism should be adopted in models where aerosols are typically assumed internally mixed.

Response: We agree with the reviewer that organic and inorganic compounds are mixed in ambient aerosol particles, and there are no pure oxalic acid aerosols or pure ammonium sulfate aerosols in the atmosphere. In fact, in our chamber experiment the seeded particles were initially pure oxalic acid and became a mixture of ammonium oxalate and ammonium sulfate with the proceeding of the formation of sulfate on the seeded oxalic acid particles. Based on our laboratory simulation, we calculated the uptake coefficient SO₂ on oxalic acid particles in the reaction chamber, which is $8.3 \pm 5.7 \times 10^{-5}$ (Table S6, Wang et al., 2016) under the humid conditions and consistent with that $(4.5 \pm 1.1 \times 10^{-5})$ (Table S6, Wang et al., 2016) observed in Beijing during the haze period of 2015. Such a consistence indicates that our chamber results including the uptake coefficient would be applicable for model simulation. We added those related discussions into the text. See page 9, line 199-202 and page 13-14, line 313-322.

Specific comments:

(3) Comments: Figure 1 shows no particle growth with ammonium sulfate as the seed particle. It would be convenient to compare the same growth chart with oxalic acid as the seed particle.

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Response: Suggestion taken. We added the growth of the seeded oxalic acid particles into the figure for the comparison. See Figure 2 and related statements in page 7, line 167-169.

(4) Comments: Figure 2: the graph shows growth of oxalic acid seed particles in the chamber experiments at different RH. How much of the growth is due to water and how much is it due to sulfate formation?

Response: Figure 2 shows the hygroscopic growth factor of oxalic acid measured by a HTDMA system, which is not the growth of oxalic acid seed particles in the chamber experiment.

(5) Comments: Figure 3. Please add sulfate time series and discuss if sulfate correlates with WSOC and/or oxalic acid.

Response: Suggestion taken. We have revised the figure by adding sulfate time series (see Figure 3 g and h). As shown in Figure 3g and h, during the field observation periods sulfate at the three sites showed a temporal variation pattern similar to that of oxalic acid with a robust linear correlation ($r^2=0.67$, 0.84 and 0.61 in Xi'an, Beijing and Hebei Province, respectively). Such a correlation was also reported by other researchers (Wang et al, 2017, Yu et al, 2005), suggesting the cooccurrence of both compounds in atmospheric aerosols. The related discussion was added into the revised manuscript. See page 10, line 227-235.

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