

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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Below are some more comments and resources that may be helpful for clarifying the third point that Nenes et al. raised: “Partitioning calculations for evaluating pH predictions”.

Nenes et al. used two field studies conducted in winter Beijing, Liu et al. (2017b) and Wu et al. (2018), to support that the metastable state for the haze fine particles is favorable. I think these measurement results may be misinterpreted. Before the

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detection of particle rebound fraction, Liu et al. (2017b) used a RH conditioner to dry the ambient particles to 20% RH and then humidified these particles to the ambient RH. Thus, it is likely that the phase state (no matter what it was) of aerosol particles has been changed to a stable state during the measurements. After converting the measured values of rebound fraction to those of hygroscopic growth factor, I found that the relationship between RH and hygroscopic growth factor agreed better with the predicted relationship using the stable state assumption and the ISORROPIA-II model. On the other hand, Wu et al. (2018) seemed to increase the RH to a value above the deliquescence RH of all the inorganic salts and thus the aerosol water content was actually calculated in a metastable state by the H-TDMA approach, which may partly explain why the “ISORROPIA metastable-predicted liquid water in good agreement with H-TDMA inferred liquid water”. In short, these measurements did not provide clear evidence of particle phase state (metastable or stable) since the RH history of ambient aerosol particles has been changed during the field measurements.

I think it is unlikely to “use the RH history of the air mass to assess whether metastable aerosol is favored based on established knowledge of the efflorescence of the major salts that form in the aerosol”, due to two factors: (1) typically, the formation of winter haze events in North China is associated with the change of wind direction (from strongly northerly winds to weak southerly winds) and the accumulation of water vapor in the atmosphere (ambient RH increases from about 20% to about 80%); (2) the efflorescence RH is unknown due to the complex aerosol chemical composition (the existence of ammonium nitrate may reduce efflorescence RH while the existence of dust materials may increase the efflorescence RH). In short, it is unlikely to figure out particle phase states from theoretical calculations because of the very large variability of ambient RH and the difficulty in estimating the efflorescence RH for multicomponent salt.

Before we have enough evidence to demonstrate what the phase state of aerosol particles is, a practical approach I believe is to predict pH for both stable and metastable

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states, which can provide an estimate of its uncertainty due to the phase state assumption.

Some additional related discussions can be found in my paper under review in ACPD:

<https://www.atmos-chem-phys-discuss.net/acp-2018-6/>

<https://www.atmos-chem-phys-discuss.net/acp-2018-6/acp-2018-6-AC2-supplement.pdf>

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