Reply to RC2

This study uses two methods (by measuring aerosol hygroscopic growth factor and particle number size distribution and by thermodynamic equilibrium modeling in combination of measured aerosol chemical species) to estimate the aerosol liquid water content ALWC from a field campaign in winter Beijing and argues that organics contribute significantly to the total ALWC and further that organics-ALWC plays a role in the formation of secondary aerosols through multiphase reactions at the initial haze stage.

Major comments

There could be inconsistency in phase states between the two methods of ALWC. Here, thermodynamic modeling assumes that the aerosol particles are in a stable thermodynamic state. But the measurements of hygroscopic growth factor increase the RH to 90% that effectively leads to a metastable thermodynamic state for the sampled aerosol particles. My question is: if the authors repeat ALWC calculations of thermodynamic models assuming a metastable state, whether the results would change? For example, whether the underestimates in ALWC by ISORROPIA would disappear? I feel that this point should be clarified in this study since it is one of the major arguments of this manuscript and since this point also affects the other argument made in this study: whether organics ALWC plays a role in the formation of secondary aerosols through multiphase reactions at the initial haze stage.

Re: Good suggestion. Bian et al. (2014) and Tan et al. (2017) both found the underestimation of ALWC simulated by ISOPPOPIA II model. Tan et al. (2017) used the ISOPPOPIA II model assuming the chemical species in the metastable state to simulate the ALWC, which also showed that the $ALWC_{ISO}$ was lower than $ALWC_{HTDMA}$ for RH below 70%. Their explanation is the lack of hygroscopicity of organic particles which was not taken into account in ISOPPOPIA II model. Here we repeat ALWC simulation using ISOPPOPIA II model assuming the chemical species in metastable state. As shown in Fig. R1a, the simulated ALWC in metastable state ($ALWC_{ISO_{metastable}}$) is similar with that in stable state ($R^2 = 0.99$), which is similar with the results in Song et al. (2018). They also showed that the simulated ALWC in metastable state is similar with that in stable state at low RH in Beijing. Figure R1b further compares the simulated ALWC in metastable state and calculated ALWC, showing $ALWC_{ISO_{metastable}}$ is still lower than $ALWC_{HTDMA}$. Moreover, regardless of the result of ISOPPOPIA II model, the average organic ALWC we inferred accounts for 30%. And for low RH and high organics hygroscopicity, the average organic ALWC accounts for up to 58%, suggesting the contribution of organics to ALWC cannot be ignored.

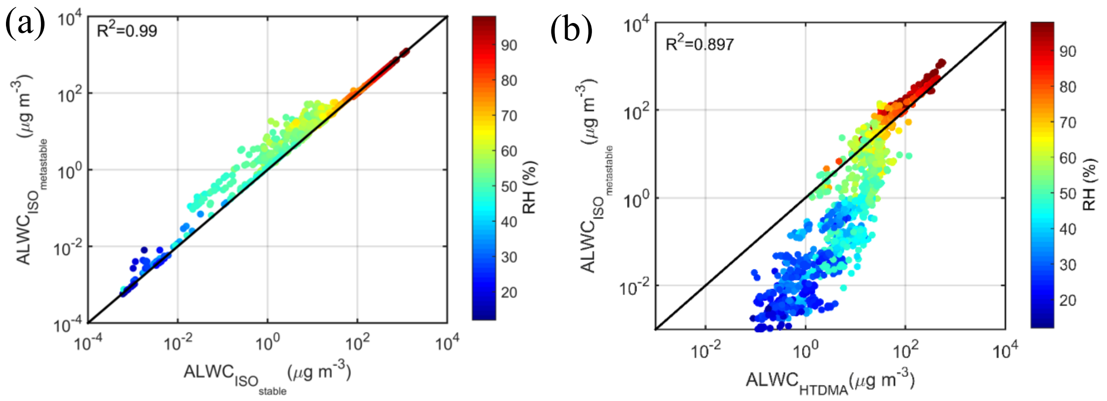

Figure R1. The correlation analysis between (a) $ALWC_{ISO_{stable}}$ and $ALWC_{ISO_{metastable}}$ and (b) $ALWC_{HTDMA}$ and $ALWC_{ISO\ metastable}$. $ALWC_{HTDMA}$ refers to calculated ALWC based on the measured growth factor and PNSDs, $ALWC_{ISO\ stable}$ refers to simulated ALWC from the ISORROPIA II model assuming chemical species in the stable state. $ALWC_{ISO\ metastable}$ refers to simulated ALWC from the ISORROPIA II model assuming chemical species in the metastable state. The coefficient of determination R^2 is given in each panel. The color of the dots denotes the ambient RH; the black solid line denotes the 1:1 line.

Minor and grammatical comments:

Line 78: can the authors elaborate on what is the direct method to measured ALWC?

Re: To our knowledge, there are no techniques for measuring the ALWC directly (Kuang et al., 2018). The sentence has been revised as: "directly measuring real-time ALWC is not feasible yet because of technical limitations (Kuang et al., 2018)." in line 79.

Line 79: why the direct measurements of ALWC is especially difficult under high RH conditions? It seems to me that such measurements under low RH conditions are more difficult.

Re: Direct measurement of ALWC is not feasible at present, for any ambient RH conditions. A possible reason is that ALWC is sensitive to RH, but the RH of aerosol sample always changes inside any instrument.

Line 94: is should be was.

Re: Thanks, we have corrected it in the manuscript.

Line 160: show should be showed.

Re: Thanks, we have corrected it in the manuscript.

Line 215: I would be surprised if n_H2SO4 were larger than 0. Would it be? Eq. 8: where is n_NH4Cl?

Re: No, the calculated n_H_2SO_4 always equals to 0 in our campaign although the ion-pairing scheme (Gysel et al., 2007) used in this paper considers the contribution of

H_2SO_4 to inorganics.

Figure R2 show the time series of the mass fraction of chloride during the campaign. The chloride detected by AMS may be from organics, and it is hard to be verified. So most previous papers didn't consider the contribution of chloride when studying aerosol hygroscopicity (e.g., Gysel et al., 2007; Sjogren et al., 2008; Guo et al., 2015; Cerully et al., 2015). In addition, the mass concentration of chloride is always low in PM_1. As shown in Figure R2, the average mass fraction of chloride is only 6% ± 3%. We added the sentence as in line 223-224 as "In this paper, the chloride was not taken into account in ion-pairing because its source is hard to determine. This may result in a minor uncertainty in κ calculation."

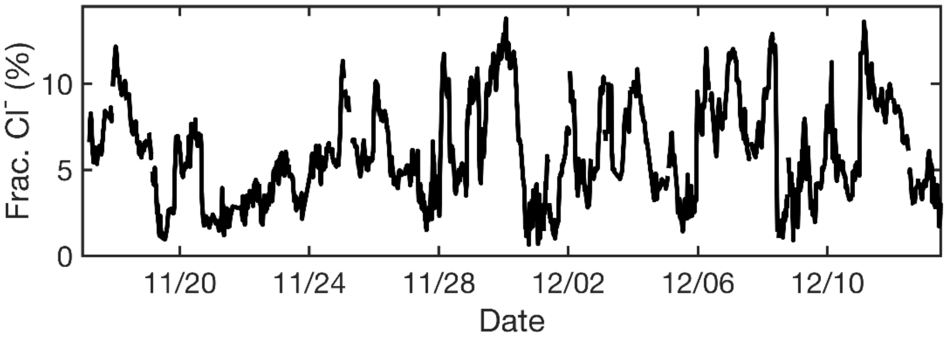

Figure R2. The time series of the mass fraction of chloride.

Line 295: ammonium is also an important component of secondary aerosols.

Re: Good suggestion, we have added the ammonium in the line 302.

Reference:

Bian, Y. X., Zhao, C.S., Ma, N., Chen, J., Xu, W.Y.: A study of aerosol liquid water content based on hygroscopicity measurements at high relative humidity in the North China Plain, Atmos. Chem. Phys., 14 (12), 6417–6426, https://doi.org/10.5194/acp-14-6417-2014, 2014.

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Gysel, M., Grosier, J., Topping, D.O., Whitehead, J.D., Bower, J.N., Cubison, M.J., Williams, P.I., Flynn, M.J., McFiggans, G.B., Coe, H.: Closure study between chemical composition and hygroscopic growth of aerosol particles during TORCH2, Atmos. Chem. Phys., 7 (24), 6131–6144, https://doi.org/10.5194/acp-7-6131-2007, 2007.

Kuang, Y., Zhao, C. S., Zhao, G., Tao, J. C., Xu, W., Ma, N., and Bian, Y. X.: A novel method for calculating ambient aerosol liquid water content based on measurements of a humidified nephelometer system, Atmos. Meas. Tech., 11, 2967–2982, https://doi.org/10.5194/amt-11-2967-2018, 2018.

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Song, S., Gao, M., Xu, W., Shao, J., Shi, G., Wang, S., Wang, Y., Sun, Y., and McElroy, M. B.: Fine-particle pH for Beijing winter haze as inferred from different thermodynamic equilibrium models, Atmos Chem Phys, 18, 7423-7438, 10.5194/acp 18-7423-2018, 2018.

Tan, H., Cai, M., Fan, Q., Liu, L., Li, F., & Chan, P. W., et al.: An analysis of aerosol liquid water content and related impact factors in pearl river delta, Science of The Total Environment, 579, 1822-1830, https://doi.org/10.1016/j.scitotenv.2016.11.167, 2017.