Reply to RC1

The manuscript "Significant contribution of organics to aerosol liquid water content in winter in Beijing, China" present a filed study in 2017 in Beijing China, focusing the aerosol liquid water content (ALWC). ALWC was derived from using the growth factor (GF) measured at 90% RH at three particle sizes in nucleation mode, Aiken and accumulation mode. ALWC was also calculated using ISORROPIA II model with the chemical composition characterized by AMS as input. It was found that generally $ALWC_{HTDMA}$ correlates with $ALWC_{ISORROPIA}$ at high RH, but not at RH<60%. Including $ALWC_{org}$, the ALWC contributed by organics, i.e., $ALWC_{ISORROPIA}$ + $ALWC_{org}$ improved the correlation with ALWC (HTDMA), especially at lower RH. The contribution of organic to total ALWC was 30+/-22%. ALWC correlated with mass concentration of sulfate, nitrate and SOA. Accumulation mode was found to contribute the largest portion to ALWC. In case study, ALWC contributed by organics may play an important role in the initial stage of haze event. The manuscript addresses ALWC, which an important parameter in atmospheric chemistry. The manuscript fits the scope of ACP. I have several comments which need to be addressed before the manuscript is considered to be published in ACP.

General comments

1. The manuscript emphasized the role of organics in total ALWC. The value fraction 30+/-22% has a large uncertainty, suggesting a large variation of the contribution and likely a much smaller contribution of organics total in some cases. Also, the conclusion is in contrast with Z. Wu et al., 2018, showing dominant role of inorganics in ALWC, as the authors also cited. The variations and the difference from the literature needs to be discussed in the manuscript.

Re: Good suggestion. This is likely related with the variation in mass fraction and hygroscopicity parameter of organics (κ_{Org}). To demonstrate this, we calculated the variation of the fraction of ALWCOrg in total ALWC (ALWCHTDMA) with the ambient relative humidity (RH) and presented in Fig. R1. It is seen that the contribution of organics to total ALWC varies strongly. The mass concentration of inorganics increases more than that of organics as RH increases, leading to a lower mass fraction of organics in the case of high ambient RH. Figure R1 also shows the $ALWC_{Org}$ fraction increases signficantly with the increase of κ_{org}. All these help explain a large variation in the $ALWC_{Org}$ contribution to total ALWC. Considering the distinct differences in ambient RH and κ_{org} between clean and polluted periods, we calculated respectively the fraction of $ALWC_{Org}$ during the two periods. There is a higher $ALWC_{Org}$ fraction (33% ± 23%) during clean periods than that during polluted periods (26% ± 11%). Yet, there is little variability of $ALWC_{Org}$ fraction during polluted periods. The larger variability in $ALWC_{Org}$ fraction during clean periods is likely caused by the highly variable κ_{Org} when the ambient RH is low. In summary, the mass fraction of organics and κ_{Org} are important contributors to total ALWC, which was not investigated in Z. Wu et al. (2018). Our results also suggest inorganics (nitrate and sulfate) play key roles in determining ALWC, which is similar to Z. Wu et al. (2018).

We have added the Figure R1 in the manuscript (Figure 4) and the discussion

above in line 311-323.

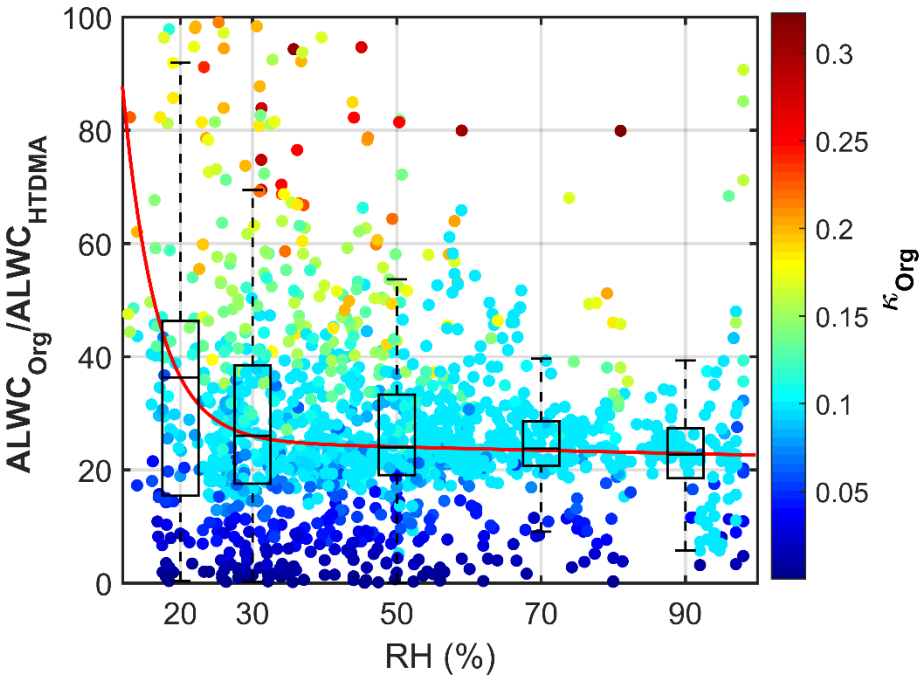

Figure R1. The variation of the fraction of $ALWC_{Org}$ in total ALWC ($ALWC_{HTDMA}$) with the ambient relative humidity (RH). The color of the dots denotes the hygroscopicity parameter of organics (κ_{Org}). The boxes show the fraction of $ALWC_{Org}$ with the 25th, 50th, and 75th percentiles. The extremities show the 5th and 95th percentiles. The red line shows the fitting curve with the function: $y = ae^{bx}$.

2. Some important details are lacking (see specific comments).

Specific comments

1. Line 77 "…its factors..." is confusing. Please specify.

Re: Good suggestion. The corresponding sentence was revised as "Overall, investigating the formation of SA and haze in North China requires an examination of ALWC and its factors including aerosol particle number size distribution (PNSD), aerosol chemical composition and ambient related humidity (RH) in this region" in line 77-78.

2. Line 156-159, at which three sizes the GF were measured?

Re: During this campaign, the H-TDMA was used to measure the GF at five particle sizes (40, 80, 110, 150, 200nm), which has been described in section 2.2. Then we follow the method of Chen et al. (2012) to calculate size-resolved GFs under different RHs, which is presented in section 3.1.

3. Line 162, the assumption of constant k in each mode may result in uncertainty in ALWC. It may be worthy discussing these lines 237-253.

Re: We assumed a constant κ in each mode based on the assumption that aerosols in a specific mode have common sources and experience similar aging processes, so κ in one mode should be the same due to the same chemical compositions (Chen et al., 2012).

This may lead to a small uncertainty in size-resolved GFs at different RHs, resulting in a deviation in ALWC calculation. The following is added: "the assumption of constant κ in each mode may lead to a small uncertainty in size-resolved GFs at different RHs, resulting in small deviations in ALWC calculation. This may be another reason for the difference between $ALWC_{HTDMA}$ and $ALWC_{ISO}$." in the line 243-245.

4. Lines 237-253, for the chemical composition measured by AMS, do the authors use bulk composition or size resolved composition? This can also contribute to the ALWC derived by ISORROPIA.

Re: The bulk chemical composition measured by HR-AMS was used to simulate ALWC from ISOPPOPIA II model. Therefore, the simulated ALWC from ISOPPOPIA II denotes the total ALWC, which can't consider the contribution of organics on ALWC. The size resolved aerosol chemical composition data are hard to use in this model. We have added the sentence as "The bulk chemical composition was used in model." in line 188.

5. Lines 204-227, in deriving ALWCorg, among inorganics, only sulfuric acid, ammonium sulfate, ammonium hydrogen sulfate, and ammonium nitrate were considered. Actually, chloride is also present, as this study found. The can result in uncertainty in ALWCorg.

Re: The chloride detected by AMS may be from organics, and it is hard to be verified. So most previous papers didn't consider 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. Figure R2 show the time series of chloride mass concentration during the sample period. It shows the average mass fraction of chloride is only 6% ± 3%. We added the sentence as in line 223-224 as "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."

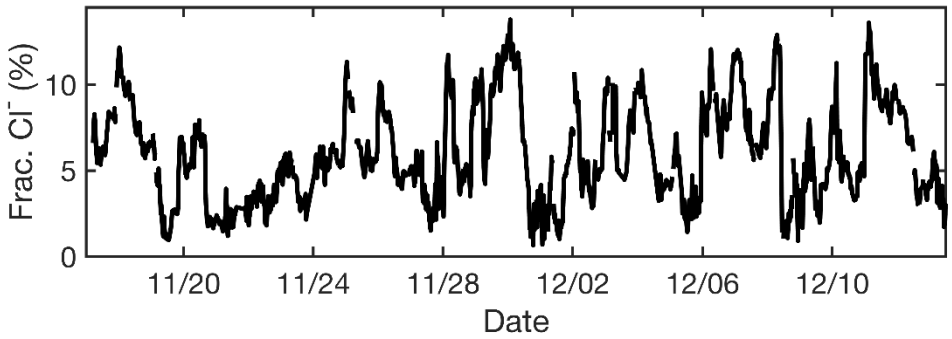

Figure R2. Time series of the mass fraction of chloride.

6. Line 281, how is b derived?

Re: b is derived by curve fitting through the data of $ALWC_{HTDMA}$ and RH using the fitting function $y = ae^{bx}$. This is clarified as "b is derived according to the relationship between $ALWC_{HTDMA}$ and RH that is fitted by the function $y = ae^{bx}$." in line 287-288.

7. Line 283, how are POA and SOA derived? Did the authors use PMF?
Re: The POA and SOA are derived using PMF. The sentence was added as "The positive matrix factorization (Paatero and Tapper, 1994) was applied on the organic aerosols (OA) spectral matrices to identify POA and SOA." in line 290-291.

8. Section 4.2.2, Fig. 4 does not provide new information to the discussion. The accumulation mode contributed dominant hygroscopic materials and ALWC to total ALWC. Naturally, the correlations are better.
Re: The analysis of the relationship between ALWC and the different modes under five different RH condition is for further evaluation of the impact of PNSD on the ALWC. Figure 4 shows the average contribution of different mode particles to $ALWC_{HTDMA}$ (f_{ALWC}) and the slope of the best linear regression. Figure 4 also shows that ALWC increases slightly as the volume concentration of accumulation mode particles increases under RH < 70 % conditions (slope < 0.001), but increases are stronger under higher RH conditions, especially under RH > 90 % conditions (slope = 0.0041). This suggests that there are more accumulation mode SA formed due to multiphase chemical reactions under high ambient RH conditions. Besides, Figure 4 also shows that the correlation between the Aitken mode particles and ALWC is enhanced significantly under RH > 90 % conditions, which suggests that the Aitken mode can also absorb a significant amount of water when RH is higher than 90%. So, we think Fig. 4 is necessary to discuss the completeness of ALWC.

9. Line 323-325, such a statement is not necessarily true because many "processed" mineral particles are highly hygroscopic, such as Ca(NO3)2, or MgSO4, as shown by many field and laboratory studies.
Re: Agree, this statement is not accurate. Some mineral particles are hygroscopic and can impact ALWC. However, these particles are mainly in the coarse mode (Hussein et al., 2004; S. Liu et al., 2008). Bian et al. (2014) and Tan et al. (2017) found the ALWC contributed by coarse mode particles exists but not substantial. Because there were no APS data in this campaign, the coarse-mode particles with diameters greater than 1 μm are not considered in this study. The statement is revised as "Particles with diameters greater than 1 μm are not considered because some particles in the coarse mode are water soluble but their contribution on the ALWC is low (e.g., Hussein et al., 2004; S. Liu et al., 2008; Bian et al., 2014; Tan et al., 2017)." In line 342-345.

10. Line 321, why upper limit of Aitken mode is set at 110 nm?
Re: In general, aerosol particles are separated into four modes in the diameter lower than 1 μm according to their different origins (Whitby, 1978). And there are some different standards to separate them. Birmili et al. (2001) defined the particle modes based on a long-term observation data (Figure R3): "fresh" nucleation mode: 3-9 nm; "aged" nucleation mode: 9-30 nm; Aitken mode: 30-110 nm; accumulation mode (110-1000 nm). Bian et al. (2014) also set the diameter of Aitken mode between 30nm and 110nm when parameterizing the particle size distribution using a multiple lognormal fitting function. Therefore, it's thus reasonable to set the upper limit of Aitken mode at

110nm.

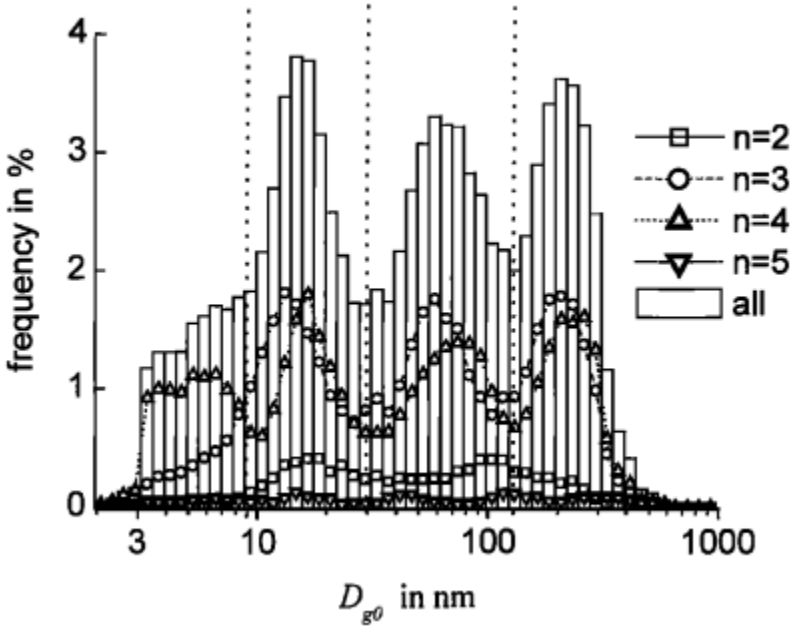

Figure R3. Frequency distribution of modal geometric mean diameters. Subsets of mode numbers 2-5 are included. The figure is from Birmili et al., (2001).

11. Line 320 (and 370), I am not sure whether using PNSD is proper here. The ALWC depends on the amount of hygroscopic materials, no matter which sizes they are present.
Re: The PNSD provides the information of the number concentration of particles at different sizes. ALWC not only depends on the number concentration of hygroscopicity particles but also depends on the particle size. Such as Aitken mode particles with small volume, because of the Kelvin effect, although the Aitken mode particles show a high hygroscopicity, they do not make a large contribution to ALWC. So, when Aitken mode particles and accumulation mode particles have the same hygroscopicity and number concentration, the particles in the accumulation mode can contribute more to ALWC than that in Aitken mode because accumulation mode particles have larger volume. Furthermore, the particles in different sizes have different hygroscopicity because of their origins, which also affects the ALWC. So PNSD needs to be considered when studying ALWC. Bian et al. (2014) and Tan et al. (2017) also found that PNSD is an important factor of ALWC.

Reference:

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