

General comments

This manuscript describes aerosol hygroscopicity determined under subsaturated conditions in Beijing, China during June 2014. The hygroscopicity parameter (κ) is calculated from measurements throughout the study and a closure analysis, using the size-resolved chemical composition measured by an aerosol mass spectrometer, is presented. Overall, this study presents consistent results with previous work with careful attention to analysis. However, there is little effort in demonstrating the additional knowledge gained from this study. This is not the first HTDMA study in Beijing, but there is no comparison to previous campaigns or to other studies in urban environments (e.g. Meier et al. (2009), Juranyi et al. (2012) and Yeung et al. (2014)).

I recommend that the manuscript be accepted on the condition that a more in depth comparison with other studies is included and that the specific comments listed below are addressed. This manuscript also needs careful proofreading for grammar mistakes before it should be accepted for publication.

The authors greatly thank to the referee to spend lots of time to read the manuscript and provide great comments. We realized that there is a lack of comparisons with previous studies on particle hygroscopicity in various atmospheric environments. In the revised MS, this point was strengthened.

Specific comments

Page 11501, line 14

Why did you estimate a particle density of 1500 kg/m³? An average, or time-varying, particle density could be easily calculated using the chemical composition measured by the AMS in conjunction with the BC mass measured by the PAX.

Response:

According to the particle mass concentration=volume concentration * particle density, the particle density is estimated by comparing the AMS-measured mass concentration and BC and the particle volume concentration derived from the particle number size distribution. On average, the particle density of 1.5 kg/m³ produced the best fitting between AMS-measured mass concentration and BC and the particle volume concentration. This will be clarified in the MS.

Modification in the MS:

“Here, the particle density is estimated by dividing the AMS-measured PM1 and black carbon mass concentrations by the SMPS-derived particle volume concentration.”

Page 11501, line 23

Reference to the work of Lanz et al. (2007) would be appropriate.

Response:

It was modified in the MS.

HOA and COA were both anthropogenic primary organic aerosol (POA) components (Lanz et al., 2007).

Page 11503, line 17

Justification should be provided for why 0.1 was chosen for SOA. Kammermann et al. (2010) determined to be > 0:2 for aged organics from HTDMA measurements at Jungfraujoch, whereas Wex et al. (2009) determined of laboratory generated SOA to be < 0:1 and Yeung et al. (2014) found urban org to be up to 0.29.

Response:

$\kappa_{SOA}=0.1$ was adopted from Gysel et al. (2007). By taking $\kappa_{SOA}=0.1$, the best fit between κ_{HTDMA} and κ_{chem} was obtained in this study. Kammermann et al., (2010) reported the mean hygroscopicity of the Aitken and accumulation mode aerosol at the free tropospheric site Jungfraujoch is $\kappa=0.24$, not the kappa value of organic compounds. In their study, $\kappa_{SOA}=0.1$ was also cited from Gysel et al.'s study. Yeung et al. (2014) showed that "over 90% of measured data and predictions based on Extended Aerosol Inorganics Model with a constant GF of the organic fraction (GF_{org}) of 1.18 are within 10% closure". $GF_{org}=1.18$ corresponds to $\kappa_{SOA}\approx 0.1$. In Yeung's study, GF_{org} ranged from 1 to 1.5 (κ_{org} up to 0.29) was obtained from the best fit GF_{org} values from GF^* based on the HTDMA measurements and the predicted GF_{inorg} based on the HR-ToF-AMS composition. This method may result in the GF_{org} values include all uncertainty in the closure between particle hygroscopicity and chemical composition. The SOA hygroscopicity varies with its oxidation state (Jimenez et al., 2009). The usage of a constant value in closure study may introduce uncertainty. More discussion will be given in the MS.

Modification in the MS:

"thereby more hygroscopic and has a κ_{SOA} of 0.1, which was calculated from the hygroscopic growth factor of organics at 90% RH as given in Gysel et al. (2007) using Eq. (4) in section 3.1. By taking $\kappa_{SOA}=0.1$, the best fit between κ_{HTDMA} and κ_{chem} was obtained in this study. One should note that kappa of SOA may varied with its oxidation state (Jimenez et al., 2009). The usage of a constant kappa value may introduce uncertainty in the closure of particle hygroscopicity and chemical composition."

Page 11503, line 18

There is no equation 4 in the cited paper. Please update the reference.

Response:

The equation [4] is not cited from the reference and is from the section 3.1 in the manuscript.

Modification in the MS:

"thereby more hygroscopic and has a κ_{SOA} of 0.1, which was calculated from the hygroscopic growth factor of organics at 90% RH as given in Gysel et al. (2007) using Eq. (4) in section 3.1."

Page 11503, line 25 and Table 1

What are the references for the shown in Table 1? According to Petters and Kreidenweis (2007), κ for ammonium sulphate is 0.53. Was the for ammonium

bisulphate measured by the authors?

Response:

The values listed in the Table 1 are adopted from Gysel et al. (2007). In their study, the particle hygroscopic growths of different chemical species were given. According to:

$$\kappa_{HTDMA} = (HGF^3 - 1) \left(\frac{\exp\left(\frac{A}{D_{Pdry} \cdot HGF}\right)}{RH} - 1 \right)$$

The HGFs were converted into the kappa values. It will be clarified in the MS.

Modification in the MS:

“The κ_{HTDMA} values for the individual compounds listed in the Table 2 were calculated from the hygroscopic growth factor at 90% RH as given in Gysel et al. (2007) using equation [4] in the section 3.1.”

Page 11504, line 15

The hygroscopic growth factor was measured, from which the hygroscopicity parameter was determined. The statement should be revised to reflect this.

Response:

The sentence was rearranged.

Modification in the MS:

“The hygroscopic growth factors of 50, 100, 150, 250, and 350 nm particles were measured at RH=90%. Hygroscopicity parameters (κ) were calculated using equation [4] in section 3.1.”

Page 11506, line 15

Why do you limit the size range for detectable mass to be ± 5 nm when you integrate over ± 50 nm? It seems that these should be consistent.

Response:

“ ± 5 nm” should be “ ± 50 nm”. It was corrected in the MS.

Modifications in the MS:

“ ± 5 nm” was changed to be “ ± 50 nm”.

Page 11506, lines 22–24

You should include a sensitivity analysis to estimate the uncertainties associated with the assumption that BC is uniformly distributed across particle size.

Response:

The following figure showed the BC mass size distribution in August in the atmosphere of Beijing measured by Sun et al. (2012a). The average mass size distribution of BC had one mode peaking at a volume-equivalent diameter of 207 nm. In this study, the closure between particle hygroscopicity with diameters of 150, 250, and 350 nm and chemical composition was performed. The size range from 150-350 nm covered the peak of BC mass size distribution. As a result, the BC mass concentration for particles in diameter of 150, 250, and 350 nm should be higher than

that estimated with the assumption of uniformly distributed BC across the whole particle size range. This will be mentioned in the MS. However, due to the lack of BC size distribution measurements in our study, a sensitivity analysis cannot be given here.

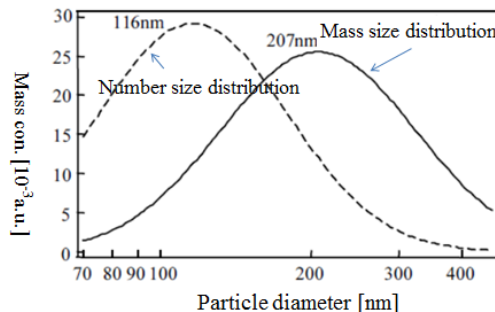


Figure: Average number size distribution and mass size distribution of BC in volume equivalent diameter (adopted from Sun et al. 2012b)

Modifications in the MS:

In section 4.3,

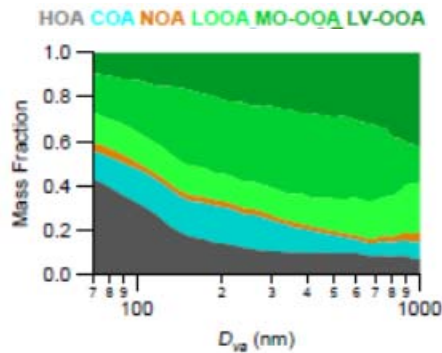
“Sun et al. (2012a) reported that the average mass size distribution of BC had one mode peaking at a volume-equivalent diameter of 207 nm in Beijing. The sizes of 150, 250, 350 nm covered the peak of BC mass size distribution. As a result, the BC mass concentration for particles in diameter of 150, 250, 350 nm should be higher than that estimated with the assumption of uniformly distributed BC across the whole particle size range.”

Page 11507, lines 1–2

Sun et al. (2012) have shown that this assumption is wrong since POA tends to be emitted at smaller sizes than the aerosol upon which SOA condense. Again, a sensitivity analysis could be used to estimate the uncertainties associated with this assumption if actual estimations cannot be made.

Response:

Sun et al.’s (2012b) study showed a size-dependency of OA factors, as displayed in the following figure. The relative contribution of POA to OA decreased with increasing particle sizes, especially, for particles larger than 150 nm in vacuum aerodynamic diameter. In this study, the closure studies were performed for particles with mobility diameters of 150, 250, and 350 nm (larger than 200 nm in vacuum aerodynamic diameter). Using the relative contribution of POA to OA in the whole size range tends to overestimate percentage of POA for the size range focused in this study, thus, underestimate the κ . In our case, the POA/OA and SOA/OA are respectively 0.39 and 0.61. According to $k_{org} = f_{POA} * k_{POA} + f_{SOA} * k_{SOA}$, the κ_{org} can be calculated as 0.06 assuming $\kappa_{SOA}=0.1$. On the basis of Sun et al.’s study, the POA/OAs for 150, 250, and 350 nm particles are 0.30, 0.23, and 0.19, respectively. Using these ratios, the κ_{org} s are 0.07, 0.08, and 0.08, respectively, which are slightly higher than the one in our case. While, the differences can be ignored. These discussions will be added into the MS.



The mass fraction of OA factors as a function of size (adopted from Sun et al., 2012b).

Modifications in the MS:

One should note that Sun et al. (2012b) found that the contributions of POA and SOA to OA showed a size-dependency. The relative contribution of POA to OA significantly increased with decreasing particle sizes. In this study, the closure studies were performed for particles with mobility diameters of 150, 250, and 350 nm (larger than 200 nm in vacuum aerodynamic diameter). Using the relative contribution of POA to OA in PM₁ tends to overestimate percentage of POA for the size range focused in this study, thus underestimate the κ . In our case, the POA/OA and SOA/OA are respectively 0.39 and 0.61. According to equation [7], the κ_{org} can be calculated as 0.06 assuming $\kappa_{SOA}=0.1$. On the basis of Sun et al.’s study, the POA/OAs for 150, 250, and 350 nm particles are 0.30, 0.23, and 0.19, respectively. Using these ratios and equation [7], the calculated κ_{org} s are 0.07, 0.08, and 0.08, respectively, which are slightly higher than the one ($\kappa_{org}=0.06$) in our case. This minor difference can be negligible.

Page 11507, lines 15–20

Have the authors ensured that the results are not unduly biased by restricting data to times when ammonium nitrate is below 20% and organics are greater than 50%? For example, this is likely reducing the number of night time data points included in the analysis, making the fit only representative for day time data points. A statement should be included in the text pointing this out.

Response:

We made a statistics on the data points included in the analysis. The result showed that the data points for 6:00-18:00 and 18:00-6:00 (next day) are 160 and 204. The night time data points were more than daytime points. This is because that the organic mass fraction during the nighttime is higher than that during daytime. This will be mentioned in the MS.

Modifications in the MS:

“Restricting data to times when ammonium nitrate is below 20% and organics are greater than 50% may lead to a bias in data points between daytime and nighttime. The statistics showed that night time data points (204 data points) were more than those during daytime (160 data points). This is because that the organic mass fraction

during nighttime is higher than that during daytime. This bias could make the fit between κ_{org} and O:C ratio more representative for the nighttime situation than daytime.”

Page 11507, lines 21–23

Although it is mentioned later, it should be emphasized here that these studies were all performed under supersaturated conditions for CCN measurements.

Response:

The studies base on the CCNc measurements were replaced by those on the basis of H-TDMA measurements.

Modifications in the MS:

Some empirical functions reported by other previous studies are also shown in Fig.8. In these previous studies (Wu et al., 2013;Jimenez et al., 2009;Rickards et al., 2013;Duplissy et al., 2011), the κ_{org} were derived from the measurements performed in the sub-saturation regime. In Massoli et al.’ study (2010) (not shown in the Fig. 8 due to the linear fitting based on HGF, not κ_{org}), they reported a linear relationship ($\text{HGF}_{90\%} = (0.58 \pm 0.15) \cdot \text{O:C} + (0.85 \pm 0.08)$) between $\text{HGF}_{90\%}$ and O:C for the laboratory-generated SOA particles. Both results displayed in Fig. 8 and Massoli’s study showed a positive correlation between κ_{org} and O:C. Such positive correlation was also reported by those studies based on CCNc measurements, for examples, Chang et al. (2010) and Mei et al. (2013).

Page 11508, lines 21–23

“The water soluble fraction of 50 nm new particles is 42%.” Is this statement referring to the chemical composition, i.e. 42% of the particle is water soluble by mass? Or is it the number fraction of the hydrophilic mode? If the former, was this determined through Eq. 3? If the latter is this before the nucleation event? In any case, this should be clarified. These results are consistent with the findings of Shantz et al. (2012), and reference should be made to this work.

Response:

The water soluble fraction is derived from:

$$\epsilon_{\text{soluble}} = \frac{\text{HGF}_{\text{measured}}^3 - 1}{\text{HGF}_{(\text{NH}_4)_2\text{SO}_4}^3 - 1}$$

It is particle volume-based method. Thus, the water soluble fraction is volume fraction, not mass or number fraction. This will be clarified in the MS.

In Shantz et al’s study (2012), they found that the 36 nm particles became increasingly CCN active within 1-4 h after the nucleation. It was hypothesized that the condensation of sulphate on these small particles enhanced their CCN activity. This article will be cited in the MS.

Modifications in the MS:

In section 2.2 “Then, the water-soluble volume fraction ($\epsilon_{\text{soluble}}$) can be calculated by”
In section 4.3 “Similar phenomenon was also observed by Shantz et al. (2012), which showed that the 36 nm particles became increasingly CCN active within 1-4 h after the nucleation. They hypothesized that the condensation of sulphate on these small

particles enhanced their CCN activity.”

Page 11508, line 6

The authors should include a discussion that compares their findings with other similar studies. This should detail the additional knowledge that their study contributes on top of the numerous other urban HTDMA studies that have previously been conducted.

Response:

We cited more literature and made the comparisons with previous studies.

Modifications in the MS:

In introduction:

Currently, some studies have been performed to investigate the relationship between particle hygroscopicity and chemical composition based on both field measurements and laboratory experiments (Massoli et al., 2010;Wong et al., 2011;Lambe et al., 2011;Rickards et al., 2013;Moore et al., 2012;Suda et al., 2014;Paramonov et al., 2013;Levin et al., 2012). These works specially focused on parametrizing the empirical correlations between the atomic Oxygen:Carbon (O:C) ratio and organic hygroscopicity parameter κ derived from either hygroscopic growth factor (Wu et al., 2013;Rickards et al., 2013) or Cloud Condensation Nuclei (CCN) activity (Mei et al., 2013;Wong et al., 2011;Lambe et al., 2011;Chang et al., 2010). Typically, a linear parametrization of the correlation between κ and O:C was presented. Rickards et al. (2013) recently summarized the literature data and pointed out the systematic variability in parametrizations between organic κ and the O:C ratio determined from the different studies remains large. A recent work done by Suda et al. (2014) tested the influence of the number and location of molecular functional groups on the hygroscopicity of organic aerosols and may help us to find out the mechanisms of organics hygroscopicity from the chemistry point of view.

Over the past several decades, particle hygroscopicity measurements have been carried out world-wide, using the HTDMA technique. Atmospheric environments, in which those measurements were performed includes marine, Antarctic, boreal forest, rural, and urban areas. Swietlicki et al. (2008) and Kreidenweis and Asa-Awu (2014) compiled the existing observations on particle hygroscopic growth in the literature. Throughout these compilations, measurements of particle hygroscopicity have been rarely performed in China, which experiences frequently severe haze pollution episodes. These few particle hygroscopicity measurements using the HTDMA technique were deployed in Yangtze River Delta (Shanghai (Ye et al., 2013) and Hangzhou (Zhang et al., 2011)), Pearl River Delta (Xinken (Cheng et al., 2008) and Hong Kong (Lopez-Yglesias et al., 2014)) and North China Plain (Beijing (Massling et al., 2009;Meier et al., 2009), Yufa (Achtert et al., 2009), and Wuqing (Liu et al., 2011)). Unfortunately, these measurements lack a linkage between particle hygroscopicity and chemical composition based on a high time resolution.

In section 4.2:

Fig. 5 (left) shows the size-dependent particle hygroscopicity parameters and

inorganic mass fraction of NR-PM₁ derived from averaging over the entire measuring period. The particle hygroscopicity increased with increasing particle size, displaying the same size-dependency with the mass fraction of inorganic composition in NR-PM₁. This is because inorganics including ammonium sulfate and ammonium nitrate are major water-soluble chemical compounds in the atmospheric particles. Compared to inorganic components, the hygroscopicity parameter of organic aerosols were typically lower than 0.1 (Varutbangkul et al., 2006; Virkkula et al., 1999). The similar size-dependency of particle hygroscopicity was observed in various environments. For examples, Levin et al. (2012;2014) and Paramonov et al. (2013) reported that particle hygroscopicity increased with particle size at a forested site in Colorado and a boreal environment of southern Finland at the SMEAR station, respectively. Jurányi et al (2013) observed that particle hygroscopic growth increased with increasing dry diameter in the urban areas of Paris. Swietlicki et al. (2008) compiled worldwide H-TDMA data and found that the particle hgyroscopicity showed a pronounced size-dependency, with hygroscopicity increasing with particle diameter.

Over the entire study, the mean κ s of 50, 100, 150, 250, and 350 nm particles were 0.16 ± 0.07 , 0.19 ± 0.06 , 0.22 ± 0.06 , 0.26 ± 0.07 , and 0.28 ± 0.10 , respectively over the entire sampling period. These values were similar to the hygroscopicity parameter $\kappa = 0.12\text{--}0.27$ (measured at RH=90%) for 35–265 nm determined in the urban atmosphere of Paris (Jurányi et al., 2013). Yeung et al. (2014) observed that hygroscopicity κ s of particles with sizes of 75, 100, 150, and 200 nm were respectively 0.28, 0.29, 0.26, and 0.27 when Hong Kong experienced a continental airstream. In their study, the particle hygroscopicity showed no obvious size-dependency and was higher than our observation in Beijing. In contrast, κ s measured were relatively low at a forested site in Colorado ($\kappa = 0.16\pm 0.08$ detected by CCNc), a boreal forest in Finland ($\kappa = 0.18$ at RH=90%) (Sihto et al., 2011), and a tropical forest site in the Amazon ($\kappa = 0.16\pm 0.06$ detected by CCNc) (Gunthe et al., 2009). At these forested locations, organic species were predominance in particles. Differently, in the atmosphere of Beijing, the inorganic fraction was relatively dominated, as shown in the Fig.3 (c).

In section 4.3

“Fig. 8 shows κ_{org} as a function of O:C ratio. From the degree of scatter point of view, κ_{org} is not correlated to the O:C ratio. Several previous studies reported the similar plots of κ_{org} values as a function of O:C ratios (Chang et al., 2010;Bhattu and Tripathi, 2015;Rickards et al., 2013). In order to derive an empirical relationship between κ_{org} and O:C ratios, κ_{org} values are usually binned by O:C in increments of 0.1. As displayed in Fig. 8, a linear fitting function ($\kappa_{\text{org}}=(0.08\pm 0.02)*\text{O:C}+(0.02\pm 0.01)$) was obtained. Some empirical functions reported by other previous studies are also shown in Fig.8. In these previous studies (Wu et al., 2013;Jimenez et al., 2009;Rickards et al., 2013;Duplissy et al., 2011), the κ_{org} were derived from the measurements performed in the sub-saturation regime. In Massoli et al.’ study (2010) (not shown in the Fig. 8 due to the linear fitting based on HGF, not κ_{org}), they reported a linear relationship ($\text{HGF}_{90\%} = (0.58\pm 0.15)*\text{O:C} + (0.85$

± 0.08)) between HGF90% and O:C for the laboratory-generated SOA particles. Both results displayed in Fig. 8 and Massoli's study showed a positive correlation between κ_{org} and O:C. Such positive correlation was also reported by those studies based on CCNc measurements, for examples, Chang et al. (2010) and Mei et al. (2013). We note that the slopes of the linear fitting varied with different studies, indicating there is no a simple parametrization to describe the relationship between organic hygroscopic and its oxidation state though the various atmospheric environments. Recently, Richards et al. (2013) had undertaken an extensive review of κ values published in the literature and showed that κ_{org} vs. O:C plot has a large degree of scatter. This indicates that other factors, such as phase state (Pajunoja et al., 2015) and molecular structures (Suda et al., 2014) of organic aerosols (OA) other than oxidation state may also play a role in the determination of the OA hygroscopicity. ”

Page 11508, lines 26

It is also possible that these are less hygroscopic primary aerosol that have coagulated to form larger particles.

Response:

Typically, the primary particles are larger than newly formed particles. If the primary particles are pre-existing during the new particle formation events, the newly formed tiny particles will coagulate with these pre-existing particles. It is very clear that the newly formed particle continuously grew to around 50 nm before around 8:30p.m., as shown in Fig. 6. The GF-PDF shows an internal mixed state of new particles. This indicates that 50 nm particles mainly came from the new particle formation via growth, not primary particles before 8:30 p.m.. However, due to the measurements performed in urban atmosphere, the influence of primary particles cannot be excluded. Around 8:30pm, the fraction of the hydrophilic mode particles dropped to 0.6, and the hydrophobic mode appeared again. This is attributed to the intensive traffic emissions at the time of rush hour, which can clearly be seen from the particle number size distribution. More discussions will be added into the MS.

Modifications in the MS:

As displayed in Fig. 9 (b), the GF-PDF of 50 nm particles showed an internal mixed state, as marked by the black dashed lines. This indicates that 50 nm particles mainly came from the new particle formation via growth, not primary particles before 8:30 p.m.. Around 8:30 p.m., the fraction of the hydrophilic mode particles dropped to 0.6, and the hydrophobic mode appeared again. This is attributed to the intensive traffic emissions at the time of rush hour, which can clearly be seen from the particle number size distribution. We also note that the growth factor of hydrophilic mode particles decreased during nighttime.

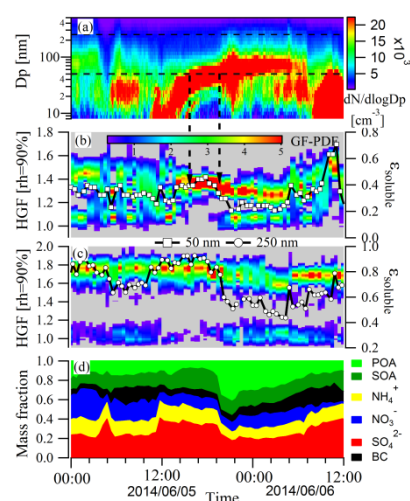


Figure 9: The variation in particle hygroscopicity, water soluble volume fraction, and

chemical composition of PM1 during the NPF event.

Page 11509, lines 7–9

This statement is confusing since 250 nm particles have a surface area that is larger than 100 nm particles but smaller than 350 nm particles. Are the authors referring to the surface area concentration?

Response:

This statement is confusing and was removed from the MS.

Page 11509, lines 11–15

These results are very interesting although hard to interpret. It would be difficult to believe that the particles suddenly converted from externally- to completely internally mixed. Even if the hydrophobic-mode grew through condensation of soluble chemical compounds such as sulphate and ammonium, there would still be a contribution from the hydrophobic core. This would show up as an increase in HGF from 1.1 up to 1.8 until the soluble components eventually dominate the hygroscopic growth. Do the authors have any insight?

Response:

The water soluble materials condensed onto the pre-existing particles. As a result, the particles converted to from external to internal state. As indicated in the following figure, this process took around 4 hours after the new particle formation started, not an instant change.

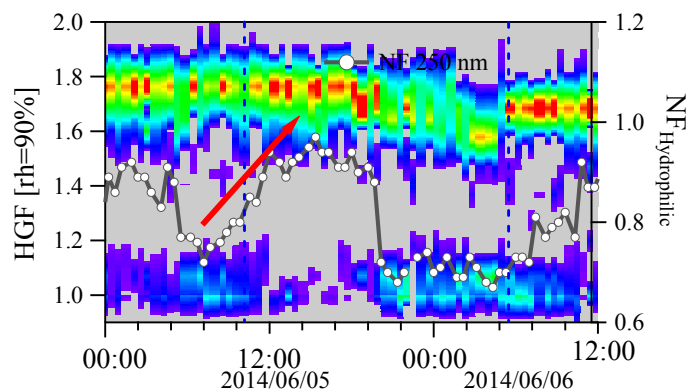


Figure: The variation in particle hygroscopicity during new particle formation event. HGF: hygroscopic growth factor, $NF_{Hydrophilic}$: number fraction of hydrophilic mode particle, $VF_{soluble}$: water soluble volume fraction.

Page 11509, line 23

Please include uncertainties associated with the listed _ and the statistic used to represent these. This could be the 25th and 75th percentile or 1.5 times the interquartile distance or 1.95 times the standard deviation, etc.

Response:

The mean values and standard deviation were given in the MS.

Modification in the MS:

“the mean hygroscopicity parameters (κ s) of 50, 100, 150, 200, and 250 nm particles

are respectively 0.16 ± 0.07 , 0.19 ± 0.06 , 0.22 ± 0.06 , 0.26 ± 0.07 , and 0.28 ± 0.10 ”

Figure 2

Volume fraction would be a more appropriate right axis for the left figure.

Clarify the meaning of the whiskers on the right figure.

Response:

This plot displays the size-dependency of particle hygroscopicity and its chemical composition. It is not used to make closure between κ and chemical compositions. Usage of mass fraction does not need to assume the densities of organic and inorganic fraction.

Figure 4

What is the RMSE of these fit?

Response:

We calculated the RMSE of the linear fit in Fig.7. All the RMSEs are 0.04. The values were added into the caption of Fig. 7.

Modification in the MS:

“Figure 7: κ_{HTDMA} vs. κ_{chem} using size-resolved chemical composition data. All the root mean square errors (RMSE) of the linear fits are 0.04.”

Figure 5

All the data from the literature are from studies conducted at supersaturated conditions. HTDMA data, such as those included in Jimenez et al. (2009), would be more appropriate.

Response:

This part was revised in the MS.

Modification in the MS:

Fig. 8 shows κ_{org} as a function of O:C ratio. From the degree of scatter point of view, κ_{org} is not correlated to the O:C ratio. Several previous studies reported the similar plots of κ_{org} values as a function of O:C ratios (Chang et al., 2010;Bhattu and Tripathi, 2015;Rickards et al., 2013). In order to derive an empirical relationship between κ_{org} and O:C ratios, κ_{org} values are usually binned by O:C in increments of 0.1. As displayed in Fig. 8, a linear fitting function ($\kappa_{\text{org}}=(0.08\pm 0.02)*\text{O:C}+(0.02\pm 0.01)$) was obtained. Some empirical functions reported by other previous studies are also shown in Fig.8. In these previous studies (Wu et al., 2013;Jimenez et al., 2009;Rickards et al., 2013;Duplissy et al., 2011), the κ_{org} were derived from the measurements performed in the sub-saturation regime. In Massoli et al.’ study (2010) (not shown in the Fig. 8 due to the linear fitting based on HGF, not κ_{org}), they reported a linear relationship ($\text{HGF}_{90\%} = (0.58\pm 0.15)*\text{O:C} + (0.85 \pm 0.08)$) between $\text{HGF}_{90\%}$ and O:C for the laboratory-generated SOA particles. Both results displayed in Fig. 8 and Massoli’s study showed a positive correlation between κ_{org} and O:C. Such positive correlation was also reported by those studies based on CCNc measurements, for examples, Chang et al. (2010) and Mei et al. (2013). We note that the slopes of the linear fitting varied with different studies, indicating there

is not a simple parametrization to describe the relationship between organic hygroscopic and its oxidation state through the various atmospheric environments. Recently, Richards et al. (2013) had undertaken an extensive review of κ_{org} values published in the literature and showed that κ_{org} vs. O:C plot has a large degree of scatter. This indicates that other factors, such as phase state (Pajunoja et al., 2015) and molecular structures (Suda et al., 2014) of organic aerosols (OA) other than oxidation state may also play a role in the determination of the OA hygroscopicity.

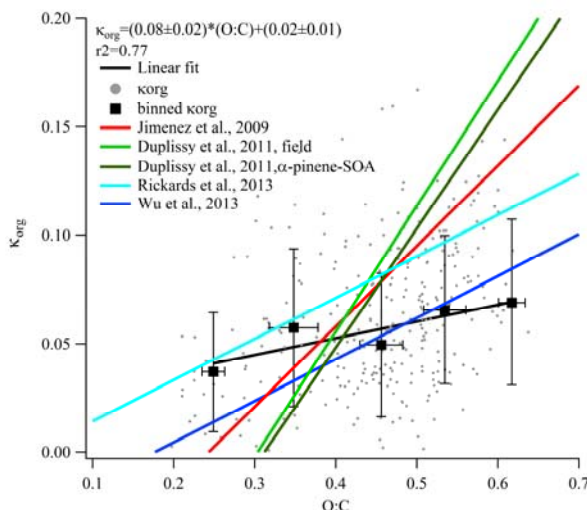


Figure 8: The relationship between organic hygroscopicity parameter (κ_{org}) and oxygen to carbon ratio (O: C).

Technical corrections

As mentioned in the general comments, there are numerous grammatical errors in the manuscript which should be corrected before the article is resubmitted.

Response:

We improved the English before resubmission.

Page 11499, line 7

This should be “mass spectrometric”.

Response:

It was corrected in the MS.

Page 11500, lines 17, 18, 20

Ammonium sulphate in the subscript is incorrect.

Response:

It was corrected in the MS.

Page 11508, line 17

The time in the figure would suggest 9.50a.m.

Response:

It was corrected in the MS.

Page 11508, line 28

This should be “the hydrophobic mode appeared again”.

Response: It was corrected in the MS.

References

- Achtert, P., Birmili, W., Nowak, A., Wehner, B., Wiedensohler, A., Takegawa, N., Kondo, Y., Miyazaki, Y., Hu, M., and Zhu, T.: Hygroscopic growth of tropospheric particle number size distributions over the North China Plain, *Journal of Geophysical Research: Atmospheres*, 114, n/a-n/a, 10.1029/2008JD010921, 2009.
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