

Wu et al. conducted a set of ambient measurements from which they calculated size dependent hygroscopic growth factors (HGF) of aerosols in Beijing. Two primary results are reported. First, the hygroscopicity parameter calculated from HGF (κ HGF) increases with increasing particle diameter. Second, κ HGF of the organic aerosol fraction is correlated with the corresponding oxygen-to-carbon (O/C) ratio that is used as a surrogate for the aerosol oxidation level. The authors hypothesize that the increase in κ HGF with increasing particle diameter is associated with condensational aging processes that increase the hydrophilic fraction of the aerosol. New particle formation events are found to be responsible for aerosols with higher κ HGF. Overall, this manuscript addresses an important research topic regarding the influence of particle composition on hygroscopic properties. It extends previous laboratory and field studies with related measurements in an area that is subject to severe pollution problems. I would support publication in ACP after consideration of the following comments.

We greatly thank to the referee to review this manuscript and provide many constructive suggestions. We modified the MS point by point.

COMMENTS

1. P11497, L25: Several other studies have investigated the relationship between hygroscopicity and chemical composition of laboratory and ambient organic aerosols, including but not limited to: Massoli et al. (2010), Wong et al. (2011), Lambe et al. (2011), Moore et al. (2012a), Rickards et al. (2013), and Suda et al. (2014). None are referenced or discussed in this manuscript. In particular, Massoli et al. and Rickards et al. characterize water uptake under subsaturated conditions as is done in this work, and Suda et al. characterize functional-group-dependent trends in aerosol hygroscopicity at a specific O/C ratio. The introduction should be expanded to include discussion of prior work that is relevant to the current manuscript.

Response:

Thanks for this suggestion. We strengthened the comparisons and citation of the 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 $\kappa = 0.12$ - 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 ($\kappa_{org} = (0.08 \pm 0.02) * O:C + (0.02 \pm 0.01)$) 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 ($HGF_{90\%} = (0.58 \pm 0.15) * O:C + (0.85 \pm 0.08)$) 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. ”

2. In the abstract, the authors report median κ_{HGF} values ranging from 0.15 to 0.29; these values should be cited somewhere in Section 4.1 because as far as I can tell they are not mentioned anywhere except the abstract.

Response:

The statistics of κ_s was given in the texts.

Modification in the MS:

In the section 4.2: “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 $\kappa = 0.12-0.27$ (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).”

3. I think it would be useful to compare results that are presented in Figure 2 with related measurements such as those presented in Moore et al. (2012b). This comparison could help to generalize the observations detailed in this work. Further, it would aid in the interpretation of other field measurements where chemical composition data may not be available.

Response:

Moore (2012)’s work was cited in the Introduction. Due to their study was performed at a supersaturation regime, not sub-saturated condition, the comparisons of size-dependency of particle hygroscopicity did not include Moore et al.’s study.

Modification in the MS:

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 are 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.

4. P11505, L3-5: The growth factor probability distribution function is an important result. In my opinion the campaign average GF-PDF should be shown as a figure. It might also be interesting to contrast multiple GF-PDF’s during events that are dominated by specific sources or aging processes.

Response:

The GF-PDF during the entire sampling period was plotted and added into the MS. The analysis was also added into the MS.

Modification in the MS:

Fig. 3 (b) displays the time series of hygroscopicity parameters for 50 nm ($\kappa_{50\text{nm}}$) and 250 nm ($\kappa_{250\text{nm}}$) particles. Both $\kappa_{50\text{nm}}$ and $\kappa_{250\text{nm}}$ had an obvious temporal variability. Their variations were similar to that of inorganic mass fraction in NR-PM₁ displayed in Fig. 3 (c). An in-depth analysis of the relationship between particle hygroscopicity and chemical composition will be given in section 4.3. Fig. 4 gives an overview of growth factor probability density distributions (GF-PDF) for 50 and 250 nm particles during the entire field campaign. The GF-PDFs of both 50 and 250 nm showed two distinct modes, which are identified as hydrophobic mode (GF<1.2) and hydrophilic mode (GF>1.2). This implied that the particles were usually externally mixed. The hydrophilic mode of 250 nm particles is more prominent most of the time. Differently, the hydrophobic mode was dominated in 50 nm particles. As marked in the Fig. 4 (a) by the square with dashed line, the hydrophobic mode disappeared occasionally, indicating that the vast majority of particles in this size range can be fully hygroscopic. This phenomenon took place during the NPF events. A case study of particle hygroscopic behavior during new particle formation events will be given in section 4.4.

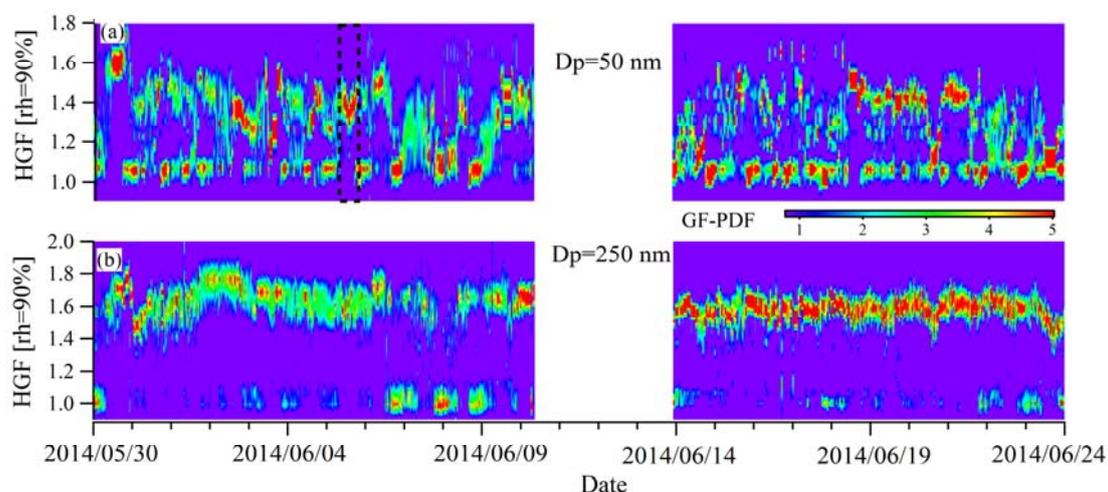


Figure 4: The time series of the GF-PDFs for 50 and 250 nm particles.

5. P11507, L27 – P11508, L5: For the reasons that are mentioned in the text, it is not appropriate to compare κHGF -versus-O/C trends observed here with κCCN -versus-O/C parameterizations from other studies that are referenced in the Figure 5 caption (Bhattu and Tripathi, 2015; Chang et al., 2010 and Mei et al., 2013). I suggest modifying Figure 5 and related discussion by removing the κCCN parameterizations and instead comparing with other κHGF measurements. Some suggested studies for comparison include Jimenez et al. (2009), Massoli et al. (2010), Rickards et al. (2013), Pajunoja et al. (2015).

Response:

The MS was revised, see below:

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 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.

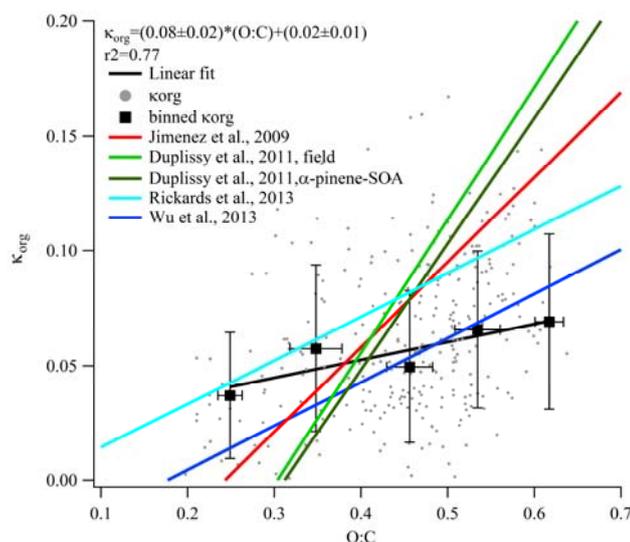


Figure 8: The relationship between organic hygroscopicity parameter (κ_{org}) and oxygen to carbon ratio (O: C).

6. P11508, L3: The authors state: “We should also note that the different aerosol formation ways in different environments could also be an explanation for the different relationship between κ_{org} and O:C.” Other studies also suggest that O:C does not encompass changes in detailed chemical composition that are responsible for changes in hygroscopicity (e.g. Mei et al., 2013; Rickards et al., 2013; Suda et al., 2014). However, AMS spectra obtained in this study are not discussed aside from the O/C ratio. This claim it should be supported by presentation of representative AMS spectra. Further, it would be useful to discuss what unique features of the Beijing aerosol chemical composition lead to different hygroscopic behavior relative to other studies.

Response:

As pointed out by the referee, several previous studies suggested that O:C does not encompass changes in detailed chemical composition that are responsible for changes in hygroscopicity (Rickards et al., 2013; Suda et al., 2014; Mei et al., 2013). Our results also showed that κ_{org} is not well correlated to the O:C ratio, as shown in the scattering plot (κ_{org} vs O:C) in the manuscript. There could be some other factors determining the organic hygroscopicity except for oxidation state. Our previous studies performed in the same sampling site with this study showed that no significant difference in the mass spectra observed between in Beijing and in other places (Huang et al., 2010). It would be difficult to find out the unique features of aerosol chemical composition in the atmosphere of Beijing. Suda et al. (2014) systematically studied the influence of the number and location of molecular functional groups on the hygroscopicity of organic aerosols. The great work done by Suda et al. (2014) may help us to find out the mechanisms of organics hygroscopicity from the chemistry point of view. In the future, we will make a deeper study using Suda et al.’s method, not in present study.

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