Review of “Particle hygroscopicity and its link to chemical composition in the urban atmosphere of Beijing, China during summertime” by Wu et al. This paper presents summertime measurements of aerosol chemical composition and hygroscopicity in Beijing. The authors do a good job of presenting and discussing the data and their results should be useful in increasing our understanding of aerosol chemical composition and CCN behavior in this region. I recommend that this paper be published after the following comments are addressed.

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The authors greatly appreciate that the reviewer spent a lot of time to review this manuscript and provide these constructive suggestions. We modified the manuscript according to the comments point by point.

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General Comments:

1) Some more discussion of the air masses and aerosol sources impacting the sampling site during the study would be very useful. What are the major local and regional aerosol sources? Is long range transport an important contributor to aerosol concentrations during this time? If so, what source regions are air masses coming from? More meteorological information should also be added. Was this time period characterized by stagnant conditions? Were there any rain events of frontal passages? If so, did these meteorological events lead to any changes in the measured aerosol properties? All of this information would greatly improve the utility of the findings presented here.

Response:

In the MS, one section was added to discuss the meteorological condition during the sampling period.

Modification in the MS:

2.4 Meteorological parameters

Additionally, a weather station (Met One Instruments Inc.) provided the meteorological parameters. The wind speed, wind direction, ambient temperature, and relative humidity (RH) were detected.

Air mass backward trajectories arriving at the sampling site were calculated using the NOAA “HYSPLIT-4” (Hybrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Hess, 1998). The 48 h trajectories terminated on a height of 200 m above the ground at 00:00, 06:00, 12:00 and 18:00 local time (UTC+08). In total, 100 air mass backward trajectories were grouped by assigning to five clusters using a k-means clustering algorithm. The number of clusters was identified according to the changes of total spatial variance (TSV) (cf. HYSPLIT4 user’s guide). Five was chosen as the final number of clusters considering optimum separation of trajectories (larger number of clusters) and simplicity of display (lower number of cluster).

4.1 Meteorological condition during the sampling period

Fig. 1 showed the mean air mass backward trajectories for five trajectory clusters

arriving at the sampling site from May 31 to June 24, 2014. The mean backward trajectories showed the significant differences in direction and length. The air masses from the east (45%) and the south (26%) were the dominate trajectories. The short-length air mass backward trajectories in cluster 1 and 2 indicated that air parcels moved slowly and spent much more time over the industrialized regions south and east of Beijing. As a result, the southerly and easterly air masses may be heavily polluted once they arrived at Beijing (Wehner et al., 2008). Cluster 3 spent much more time over the sea and may be associated with humid air masses. Northerly (8%) and north-westerly (10%) air masses, as represented by clusters 4–5, typically lead to the advection of dry and continental air into the Beijing area.

Fig. 2 displays the time series of wind speed, wind direction, ambient temperature, and RH during the sampling period. There was a clear diurnal cycle for all meteorological parameters. During nighttime, the wind speed was usually very low (around 1 m/s) and started to increase around noon on each day. The nighttime static wind may lead to very poor dilution with clean air and dispersion of pollutants and result in the local emissions were trapped in the urban atmosphere. The ambient temperature usually was above 30 ºC during daytime and around 20 ºC during nighttime. The average temperature and RH were respectively 24±7 °C and 45±20%. It rained several times during the measuring period, as indicated in the Fig. 2 (a). The heaviest wet deposition occurred on 17, June. The wet deposition obviously removed the atmospheric particles, as can be seen from the particle number size distribution shown in Fig. 3 (a).

In summer, the new particle formation and traffic emissions are the major sources of ultrafine particles in the atmosphere of Beijing (Wu et al., 2008;Wu et al., 2007). In addition, air masses across the industrialized regions in the south and east typically bring the high concentrations of accumulation mode particles to urban areas of Beijing (Wehner et al., 2008).

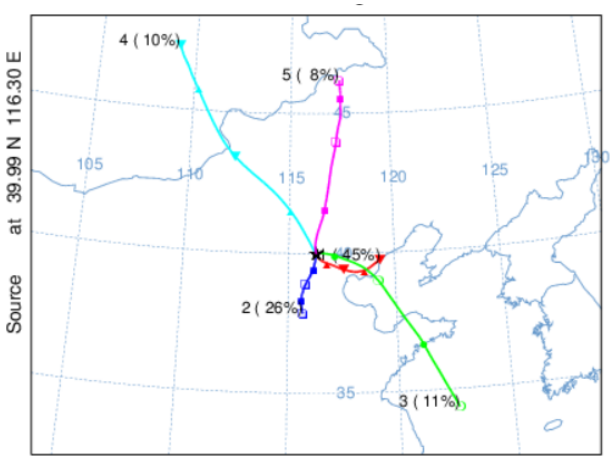

Fig.1: Mean air mass backward trajectories for five clusters arriving at the sampling site.

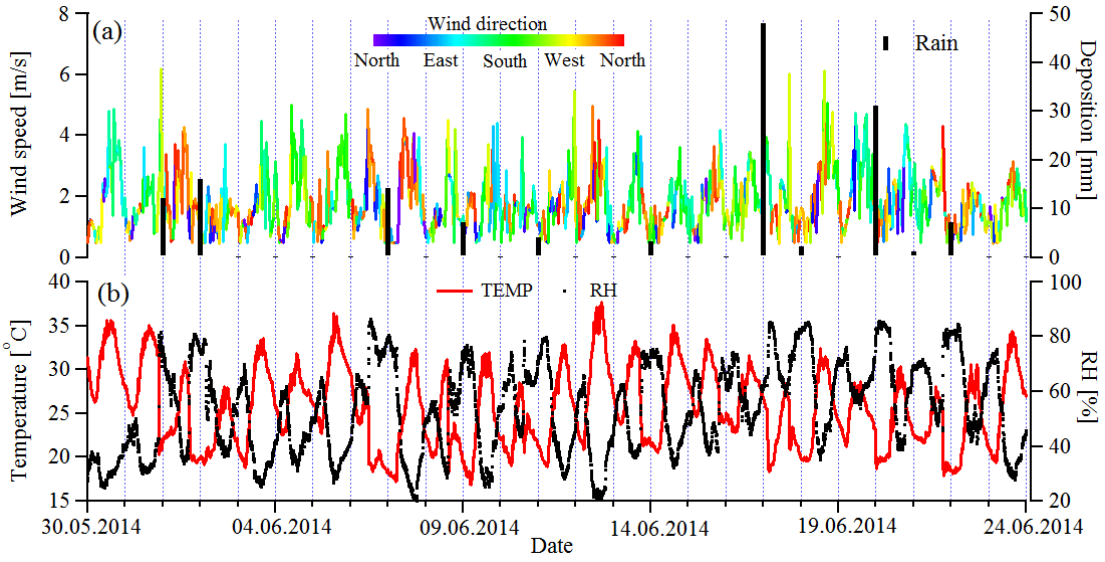

Fig.2: The time series of wind speed, wind direction, wet deposition (a) and temperature and RH (b) during the sampling period.

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2) *The authors include non-refractory aerosol components and black carbon in there analysis but no mention is made of other aerosol species. Please discus the role of dust and sea salt and any potential uncertainty or bias that may be introduced by neglecting these aerosol types*

Response:

In this study, the particles below 350 nm in mobility diameter are concerned. The dust and sea salt exist in the coarse mode which is larger than 1μm. Therefore, the potential uncertainty caused by neglecting dust and sea salt are very small.

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3) There are a number of studies that have examined aerosol hygroscopicity and chemical composition in various environments around the world. The authors allude to a few of these in the introduction but do not discuss their findings in the context of these previous measurements or compare results. For example, Paramanov et al. (2013) present size resolved κ values compiled from four different studies which agree very well with those presented here, even though they were all from remote locations. The analysis performed here is also very similar to that done by Levin et al. (2012; 2014) who compared measured aerosol hygroscopicity with that calculated from aerosol composition and also measured during new particle formation events. The results presented here for the hygroscopicity of newly formed particles is actually the opposite of that found by Levin et al. (2012) who saw a decrease in κ for newly formed particles. This difference might indicate the different roles of organic versus inorganic condensing vapors on new particle growth in remote and urban environments. Putting the results of this paper into the context of such previous measurements would again greatly help to increase the usefulness of these findings and strengthen the paper.

Response:

In revised MS, we strengthened the comparisons and citation of the previous studies.

Modifications in the MS:

In introduction:

Currently, some studies had 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-PM_1 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-PM_1. 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 κs 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 κ = 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 κs 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, κs measured were relatively low at a forested site in Colorado (κ = 0.16±0.08 detected by CCNc), a boreal forest in Finland (κ = 0.18 at RH=90%) (Sihto et al., 2011), and a tropical forest site in the Amazon (κ = 0.16±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 (κ_{org}=(0.08±0.02)*O:C+(0.02±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±0.15)*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. ”

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4) Before publication the paper should be thoroughly proof read for English grammar and word usage.

Response:

We carefully read the manuscript and improve the English writing.

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Specific Comments:

Page 11500, Line 8: What was the size range for the SMPS measurements?

Response:

The size range is 3-800 nm.

Modification in the MS:

“Particle number size distributions from 3 to 800 nm were measured by TSI-SMPS (Long-DMA3081+CPC3775 and Nano-DMA3085+UCPC3776).”

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Page 11501, Line 25: Was this a home built PAX instrument? Please provide more information. Page 11501, line 25: The reference here should be (Arnott et al. 1999). This is also wrong in the list of references.

Response:

The PAX is not home-built one. It is product of DMT Company. The reference was corrected in the MS.

Modification in the MS:

“Black carbon (BC) mass concentration in μg/m3 is derived from Photoacoustic Extinctiometer (PAX) measurements (DMT Company) (Arnott et al., 1999) equipped with PM_1 cut-off inlet.”

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Page 11503, line 14: Please add a figure showing the SOA and POA fractions during the study and discuss their relative importance during different time periods.

Response:

In the Fig.3 (a), the organics were separated into POA and SOA fractions.

Modification in the MS:

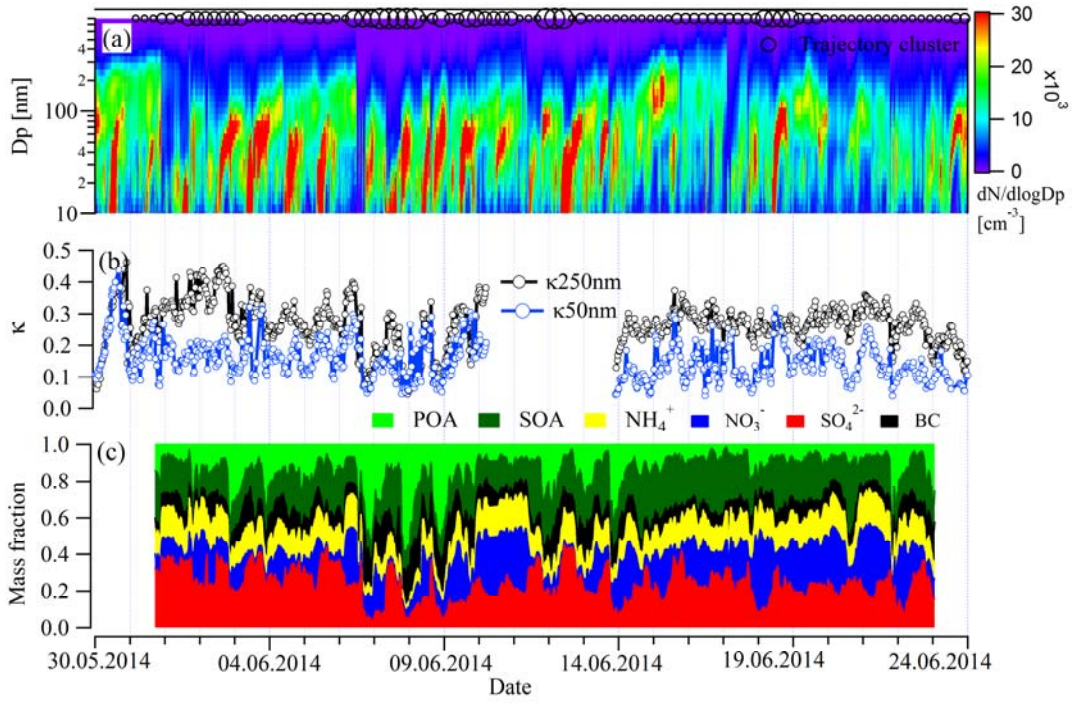

Figure 3: Time series of particle number size distribution (a), hygroscopicity parameters, κ (b), and chemical composition of PM_1 (c) during the measuring period. The black circles in the upper panel indicate the trajectory clusters. The smallest circle means cluster 1, and the biggest one is cluster 5.

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Page 11504, line 9: Were wind speed and direction also measured? If so, please add them to Figure 1 and discuss how they may have impacted the measured aerosol properties.

Response:

Yes, the wind speed and direction were measured. A new plot was added to describe the meteorological condition.

Modification in the MS:

Fig.2 displays the time series of wind speed, wind direction, ambient temperature, and RH during the sampling period. There was a clear diurnal cycle for all meteorological parameters. Typically, the wind speed was very low (around 1 m/s) during the nighttime and started to increase around noon on each day. The nighttime static wind may lead to very poor dilution with clean air and dispersion of pollutants and result in the local emissions were trapped in the urban atmosphere. The ambient temperature usually is above 30 ºC during daytime and around 20 ºC during nighttime. The average temperature and RH was respectively 24±7 °C and 45±20%. It rained several times during the measuring period. The heaviest wet deposition occurred on 17, June. The wet deposition obviously removed the atmospheric particles, as can be seen from the particle number size distribution shown in Fig. 3 (a).

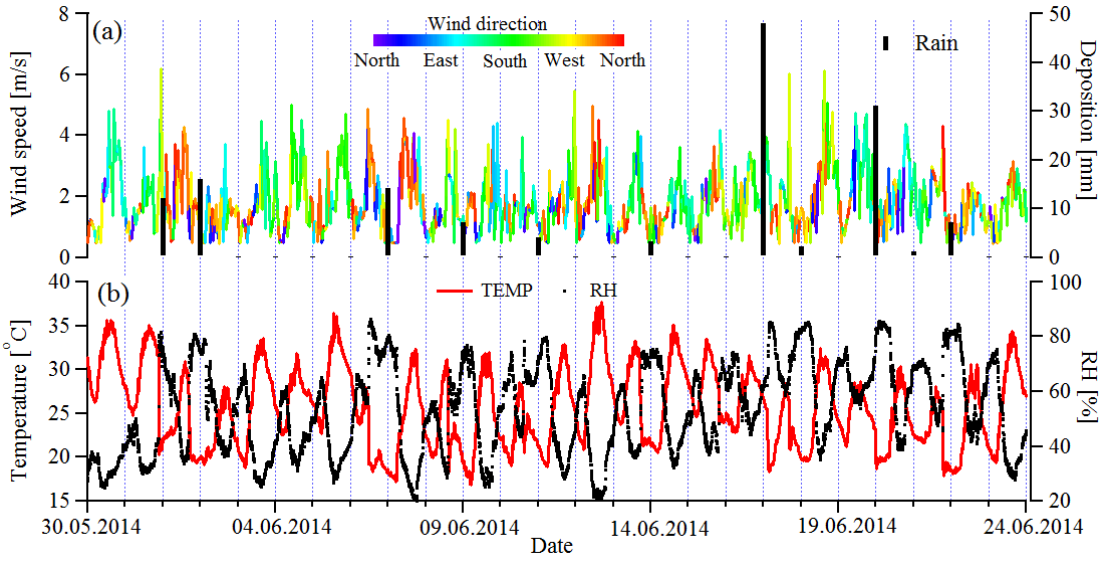

Fig.2: The time series of wind speed, wind direction, wet deposition (a) and temperature and RH (b) during the sampling period.

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Page 11505, Line 3: Please show the GF-PDF mentioned here.

Response:

One new plot was added to show the GF-PDF during the entire sampling period.

Modification in the MS:

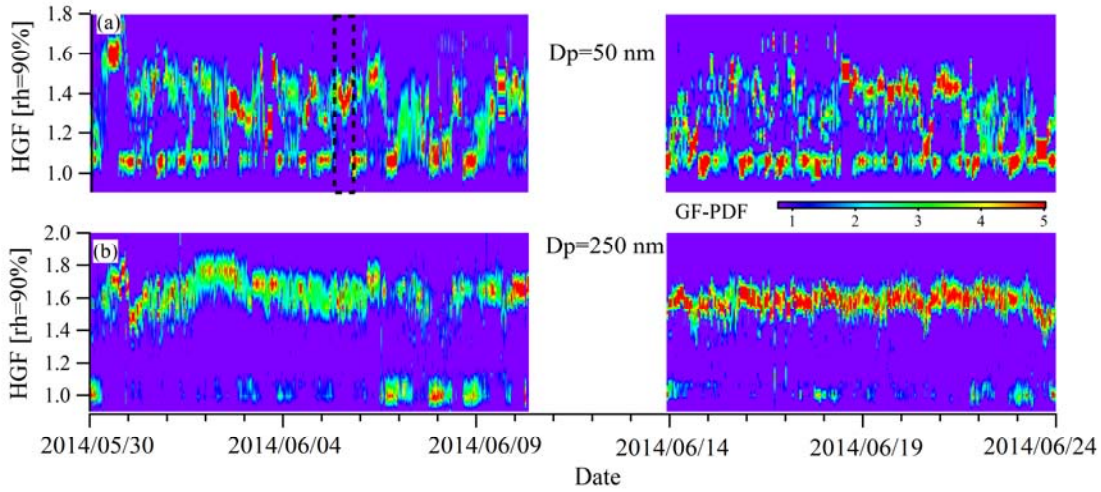

Figure 4: The time series of the GF-PDFs for 50 and 250 nm particles.

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.

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Page 11505, Line 21: Was $PM_{2.5}$ calculated from number size distributions or is this from a separate measurement?

Response:

The $PM_{2.5}$ mass concentration was measured by TEOM$^{@}$ Monitor (Series 1400ab).

Modification in the MS:

Here, the $PM_{2.5}$ mass concentration which measured by TEOM$^{@}$ Monitor (Series 1400ab), a key factor characterizing air pollution, vs. the fraction of the hydrophilic mode is plotted (Fig. 6) to analyze the relationship between particle mixing state and air pollution.

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Page 11505, Line 27: Were there other changes in aerosol characteristics (i.e. chemical composition, size distribution or κ) during high PM events?

Response:

The chemical composition and size distribution of particles should vary with PM concentration. In this study, the particle hygroscopicity is concerned. Other parameters will not be discussed in detail.

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Page 11506, Line 16: Please show a timeline of BC concentrations. This could be added to Figure 1.

Response:

The BC concentration was shown in the Fig.1 (c).

Modification in the MS:

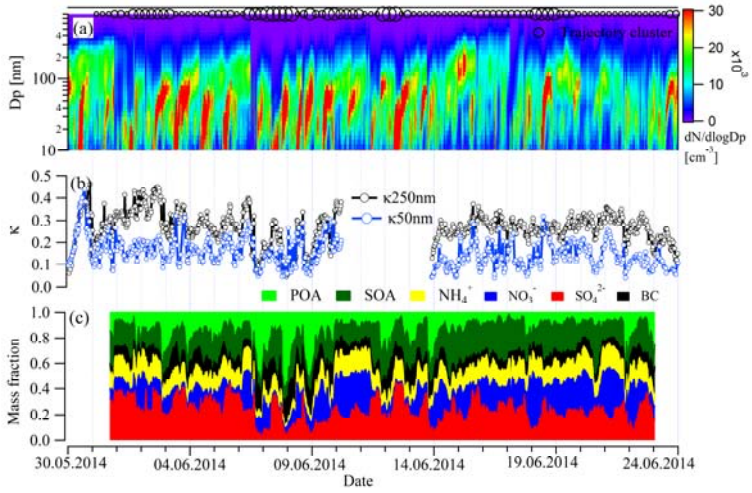

Figure 3: Time series of particle number size distribution (a), hygroscopicity parameters, κ (b), and chemical composition of PM_1 (c) during the measuring period. The black circles in the upper panel indicate the trajectory clusters. The smallest circle means cluster 1, and the biggest one is cluster 5.

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Page 11507, Line 8: While the fitted slopes are close to one, indicating no systematic

bias, there is considerable scatter around these lines in Figure 4. This means that in specific cases κ may not be well predicted, as claimed here. Please mention this and discuss what might be causing the scatter.

Response:

The assumption of BC mass size distribution and κ_{org} value in the closure as well as the measurement uncertainties for both HTDMA and AMS could introduce the biases in the closure.

Modification in the MS:

While, one should note that the assumption of BC mass size distribution and κ_{org} value in the closure as well as the measurement uncertainties for both H-TDMA and AMS could introduce the biases in the closure. This may lead to a scatter of data point around the fitting line.

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Page 11508, Line 3: The slope of the κorg vs O:C line from this work is much flatter than those of the other studies shown in Figure 5. Given the small slope and large scatter, it appears that one of the main conclusions from these measurements is that O:C is not a good predictor of κorg for this data set (as stated in the discussion). I feel this should be stated in the abstract and conclusions instead of presenting the empirical relationship between κorg and O:C, which is weak.

Response:

Yes, we agree. The abstract and conclusion were modified.

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Page 11508, Line 17: Please show, and discuss, how measured aerosol composition and κ changed during the NPF event.

Response:

The mass fraction of PM1 during the NPF events was displayed in the Fig.9 (d). More discussions were added into the MS.

Modification in the MS:

The chemical composition of PM_1 in Fig. 9 (c) showed that the inorganic species and SOA were dominated before 8:30 p.m., while mass fraction of organic compounds, especially POA increased significantly afterwards. This variation was consistent to that in particle hygroscopicity.

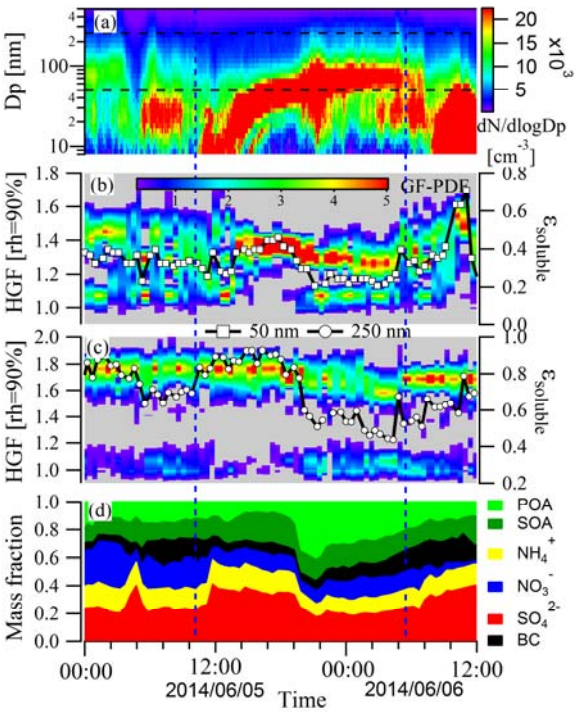

Figure 9: The variation in particle hygroscopicity, water soluble volume fraction, and chemical composition during new particle formation event.

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Page 11509, Line 7: It appears that there was an air mass change prior to the NPF, as indicated by the sharp decrease in aerosol concentrations. The change in 250 nm aerosol properties could just be due to this change and it may not be correct to directly compare these particles before and after the event.

Response:

We analyzed the local wind speed and wind direction, as shown in the Fig. 1. The wind speeds are lower than 2 m/s at the most of time. And, no significant change in wind direction was observed. This means that the decrease in aerosol concentration may not be caused by the air mass change. The possible reason may be the caused by the development of boundary layer. As a result, the pollutants were diluted by the fresh air. This analysis will be mentioned in the manuscript.

Modification in the MS:

"During the NPF events, the wind speeds were lower than 2 m/s, indicating a stable local meteorological condition. This means that the change in the hygroscopicity of 250 nm particle may not be caused by the air mass changes."

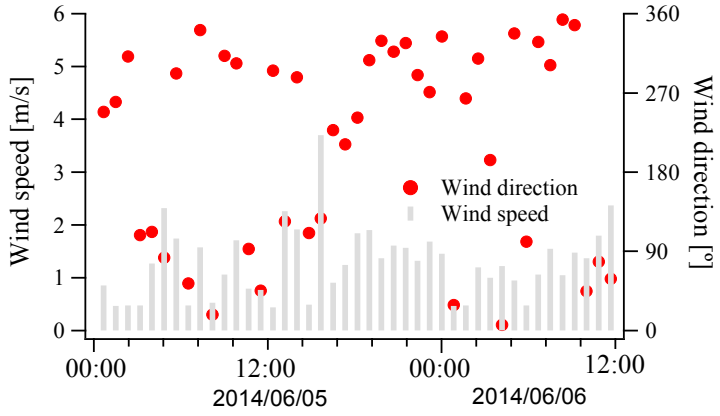

Fig 1: The wind direction and wind speed during the NPF events.

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Figure 2 Caption: This should say "Mass fraction of inorganics" since this is what is actually plotted.

Response:

Yes, it was modified in the manuscript.

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References:

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