Comments to the Author:
While the revisions you have made to your manuscript have addressed some of the questions and concerns raised by the Referees and myself, another round of revisions is required to address some important remaining issues.

Response:

We are very grateful to the referees and editor for helping us to continue to improve our manuscript. Regarding the English editing, we have already contacted with Copernicus editorial services. The manuscript will be copy-edited during typesetting if it was accepted. Other comments were responded point by point.

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An important issue to address in your revisions is the lack of detailed AMS data analysis and spectra in your manuscript. Only the O:C ratio is used to explain the observed trends. This is not a sufficient use of the rich dataset that can be obtained by AMS and other co-located measurements. The following section of your manuscript highlights this issue, though it is not the only instance of this deficiency in your analysis: "We should also note that the different aerosol formation ways in different environments could also be an explanation for the different relationship between κorg 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 should be supported by presentation of representative AMS spectra. Furthermore, it would be useful to discuss what unique features of the Beijing aerosol chemical composition lead to the different observed hygroscopic behavior relative to other related studies.

Response:

By following the editor's suggestions, we made a deeper analysis on the average elemental composition, which is an approach to simply describe organic aerosol (OA) chemistry (Kroll et al., 2011). The O:C vs H:C ratios of OA (Van Krevelen (V-K) diagram) is used to characterize both the source profiles and the atmospheric evolution of OA (Heald et al., 2010;Ng et al., 2011). Fig.1 displays the V-K diagram for the O:C and H:C ratios during the entire sampling period in our study. The O:C vs H:C ratios can be fitted by a line with a slope of -0.63 and an intercept of 1.95 by the reduced-major-axis (RMA) regression method. V-K diagram for OA summarized by Chen et al (2015) is also given in Fig.1 in order to make comparisons between OA measured in Beijing and in other atmospheric environments. Chen et al (2015) reported ambient organic aerosols line up in the V-K space along a line with a slope of -0.6 by synthesizing a large dataset of surface field observations covering urban, rural and remote environments. The trajectory of our work agrees well to this general trend, as shown in Fig.1. It implies that the atmospheric aging of OA in Beijing has great similarities to the results measured in other locations.

The grey lines in the Fig.1 display a conceptual V-K diagram, illustrating how reactions involving the addition of functional groups fall along straight lines proposed by Heald et al. (2010). A slope of −1 is produced by the simultaneous addition of both functional groups, forming a hydroxycarbonyl or carboxylic acid. The horizontal line (slope=0) means the replacement of a hydrogen with an alcohol group (−OH) involves an increase in oxygen but no change in hydrogen (Heald et al., 2010). The slope for O:C vs H:C ratios in our study falls into the space between lines with a slope of -1 and 0. This indicates that the addition both alcohol and carboxylic functional groups could lead to the enhancement of O:C ratio. However, these reactions could result in different particle hygroscopic behaviors. This is one of reasons to explain that O:C does not encompass changes in detailed chemical composition that are responsible for changes in hygroscopicity.

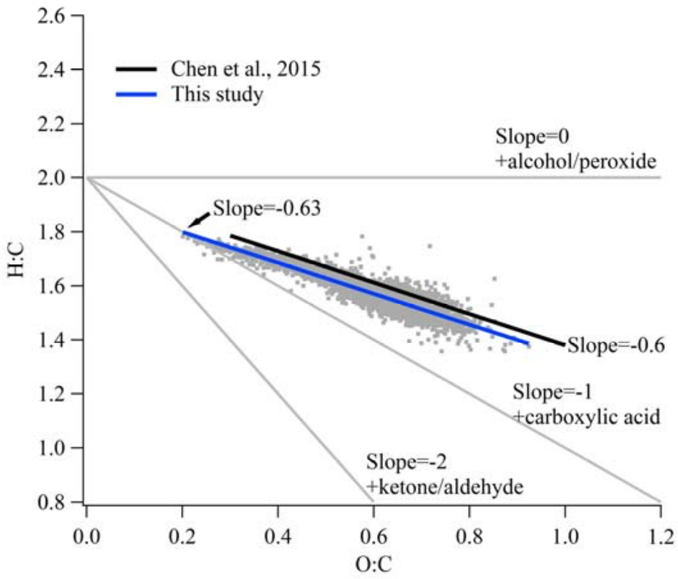

Fig. 1: Van Krevelen diagrams for the O:C and H:C ratios. The grey lines illustrating how functionalization reactions of organic species affect H:C and O:C from an arbitrary starting point (adopted from (Heald et al., 2010)).

Modifications in the MS:

In order to gain insight into the similarities of OA measured in Beijing and other environments, the average elemental composition, which is an approach to simply describe organic aerosol (OA) chemistry (Kroll et al., 2011) is analyzed, here. The O:C vs H:C ratios of OA (Van Krevelen (V-K) diagram) can be used to characterize both the source profiles and the atmospheric evolution of OA (Heald et al., 2010;Ng et al., 2011). Fig. 9 displays the V-K diagram for the O:C and H:C ratios during the entire sampling period in our study. The O:C vs H:C ratios can be fitted by a line with a slope of -0.63 and an intercept of 1.95 by the reduced-major-axis (RMA) regression method. V-K diagram for OA summarized by Chen et al (2015) is also given in Fig.9 in order to make comparisons between OA measured in Beijing and in other atmospheric environments. Chen et al (2015) found ambient organic aerosols line up in the V-K space along a line with a slope of -0.6 by synthesizing a large dataset of surface field observations covering urban, rural and remote environments. The trajectory of our work agrees well to this general trend, as shown in Fig. 9. It implies that the atmospheric aging of OA in Beijing has great similarities to the results measured in other locations.

The grey lines in the Fig.9 display a conceptual V-K diagram, illustrating how reactions involving the addition of functional groups fall along straight lines proposed by Heald et al. (2010). A slope of −1 is produced by the simultaneous addition of both functional groups, forming a hydroxycarbonyl or carboxylic acid. The horizontal line (slope=0) means the replacement of a hydrogen with an alcohol group (−OH) involves an increase in oxygen but no change in hydrogen (Heald et al., 2010). The slope for O:C vs H:C ratios in our study falls into the space between lines with a slope of -1 and 0. This indicates that the addition both alcohol and carboxylic functional groups could lead to the enhancement of O:C ratio. However, these reactions could result in different particle hygroscopic behaviors. This could be one of reasons to explain that O:C does not encompass changes in detailed chemical composition that are responsible for changes in hygroscopicity. 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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Please also address the questions raised by Referee #2:
While the authors have submitted an improved manuscript, including a number of additional references, there are still several key issues that make this manuscript not yet ready for publication:

- The grammar in the manuscript must be further improved before it can be published. It is possible that Copernicus offers editorial services.

Response:

The English editing of the revised manuscript will be made.

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- The authors use a density of POA of 1400 kg/m^3. However, the studies that they cite to back up this value were specifically conducted for secondary organic aerosol. A more realistic density for POA would be closer to 1000 kg/m3, since POA is thought to be similar to lubricating oil. This would affect the overall density of the aerosol enough that a study-long average of 1500 kg/m^3 is inappropriate.

Response:

The POA density of 1000 kg/m^3 was taken in the revised manuscript. The assumption of POA density does not affect the average particle density of 1500 kg/m^3, which is estimated from AMS and SMPS measurements directly. However, the change in POA density has an effect on the closure study. In the revised manuscript, the POA density of 1000 kg/m^3 will be taken. Correspondingly, the closure results from particle hygroscopicity and chemical composition were modified.

Modification in the MS:

"The density of SOA was taken to be 1400 kg/m^3 (Gysel et al., 2007;Alfarra et al., 2006;Dinar et al., 2006). The density for POA is chosen as 1000 kg/m^3, since POA is thought to be similar to lubricating oil."

Table 1 and Fig. 7 were modified.

Table 1: Gravimetric densities (ρ) and hygroscopicity parameters (κ) used in this study.

Species	**NH_4NO_3**	**NH_4HSO_4**	**$(NH_4)_2SO_4$**	**SOA**	**POA**	**BC**
[kg/m^3]	1720	1780	1769	1400	1000	1700
κ	0.58	0.56	0.48	0.1	0	0

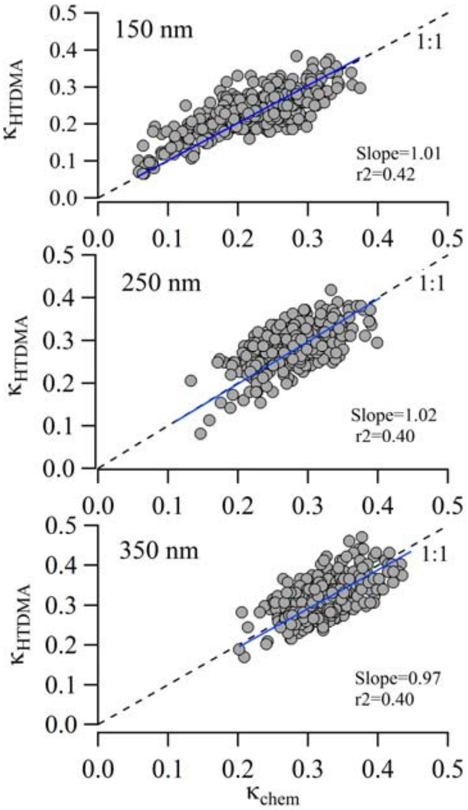

Figure 7: κ_{HTDMA} vs. κ_{chem} using size-resolved chemical composition data. All the root mean square errors (RMSE) of the linear fits were 0.04.

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- In the chemical closure part of the manuscript, a K_org of 0.1 is used. It is only mentioned in passing in the response to the reviewer that this value was chosen because it resulted in a good fit. More details should be provided on how the fitting was determined and how other values affect the final fit. In addition, this should be described clearly in Section 3.2.

Response:

First of all, κorg=0.1 was typically taken in the hygroscopicity and chemical composition closure in the previous studies (e.g. Gysel et al., 2007). Around κ =0.1, the κs, which span from 0.06 up to 0.2 were chosen for testing the closure in our study. As an example, the following figure shows the linear fitting slope vs κ values for 150 nm particles. Other particle diameters show the similar relationship. One can see that the slope is closest to the unit when κ_{SOA}=0.1. Therefore, κ_{SOA}=0.1 was chosen for final linear curve fitting. This will be clarified in the revised manuscript.

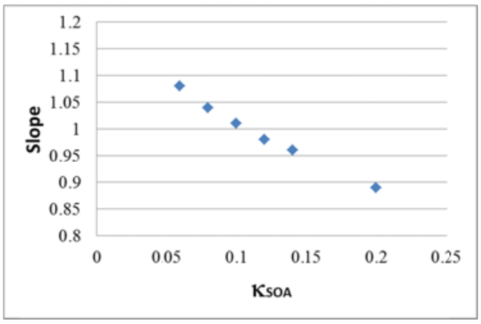

Figure 2: Linear curve fitting slope vs κ_{SOA}.

Modification in the MS:

The κs, which range from 0.06 up to 0.2, with a bin width of 0.02 were selected for testing the closure in our study. By taking κ_{SOA}=0.1, the slope of linear curve fitting is closest to 1. Therefore, κ_{SOA}=0.1 was chosen to perform the closure between κ_{HTDMA} and κ_{chem}.

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- The top panel of Figure 3, which shows the trajectory cluster based on circle size, is not meaningful since adjacent trajectories are not always related. For example Cluster 3 is from the opposite direction as Cluster 4. But it is difficult to distinguish a circle of size 3 vs 4. Perhaps you could replace it with a bar that corresponds to the colours of your clusters instead.

Response:

Figure 3 was modified.

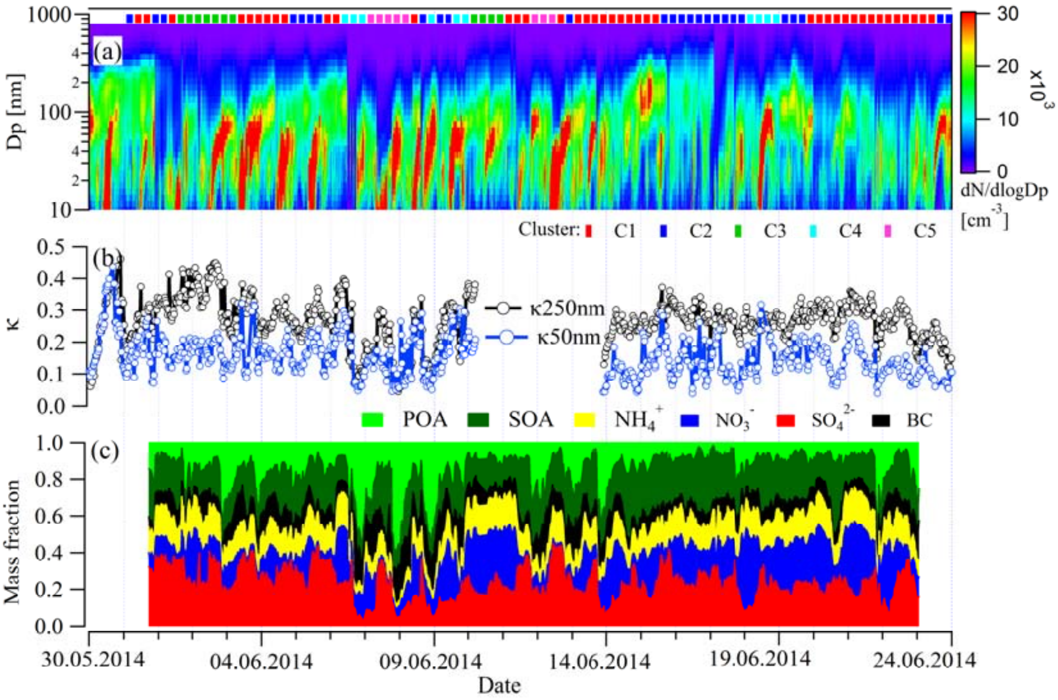

Figure 3: Time series of particle number size distribution (a), hygroscopicity

parameters (κ) (b), and chemical composition of PM1 (c) during the measuring period. The color bars in the upper panel (a) indicate the trajectory clusters. C1-C5 represents five trajectory clusters.

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- Page 9 line 29 to page 10 line 1, add ``in an urban environment" to the end of the sentence. I would argue that a 33% enhancement is not insignificant, although in absolute terms it is not as much.

Response:

"in an urban environment" was added into the manuscript. "One should note that Sun et al. (2012b) found that the contributions of POA and SOA to OA showed a size-dependency in an urban environment."

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- Page 13 line 5, the authors say that the hygroscopicity parameter of organic aerosols are typically lower than 0.1 but a quick look at Petters and Kreidenweiss (2007) shows that several organic compounds have a kappa greater than 0.1. Perhaps the authors were referring to ambient organic aerosol? The sentence should be clarified.

Response:

The hygroscopicity parameters for several pure organic acids, for examples, pinic acid and levoglucosan are greater than 0.1 (Petters and Kreidenweis, 2007). Differently, the hygroscopic growth factor derived κ for α−pinene/O_3/dark and β−pinene/O_3/dark secondary organic aerosol secondary organic aerosol is much lower than 0.1 (Petters and Kreidenweis, 2007). In the ambient atmosphere, the κs of OA are typically lower than 0.1 (Gysel et al., 2007). This will be clarified in the revised manuscript.

Modification in the MS:

"Compared to inorganic components, the hygroscopicity parameter of organic aerosols are typically lower than 0.1 in the ambient atmosphere"

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- Page 15 line 5, it should be stated at the beginning of this section that this analysis is only for 150, 250 and 350 nm aerosol.

Response:

It was stated in the MS.

"The AMS-derived particle mass concentrations for different chemical compounds were used to perform a closure study with hygroscopicity of 150, 250 and 350 nm particles."

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And be sure to also address my prior questions and comments:

Much of the manuscript is still difficult to follow, and seems to rely on a great number of assumptions, such as the SOA factor having a constant kappa of 0.1 to estimate kappa_org. The analysis of how hygroscopicity varies with aerosol composition is rather weak, especially considering that only changes in O:C were considered for the organic component. The AMS data can and should be analyzed in much greater detail to better understand the causes for the measured variations in hygroscopic growth factor, and why this does not seem to depend on O:C ratio here. The new science findings that were obtained from this study are still not presented very clearly.

Response:

It was responded in the beginning.

A few additional comments to consider:

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It appears that no size-resolved AMS measurements were made? This would have provided a lot of valuable insight into how hygroscopicity varies with particle size. If no PToF measurements were made the authors should state this and explain why. [Based on Sect. 4.3 it appears that PToF measurements were made, so these should be discussed.]

Response:

The AMS was operated in “mass spectrum” and “particle-time-of-flight” submodes for equal time periods. The size-resolved AMS data were used in the closure between particle hygroscopicity and chemical composition and the analysis on the particle hygroscopic growth size-dependency. It will be clarified in the revised manuscript.

Modification in the MS:

The Aerodyne HR-ToF-AMS (here simply referred to as AMS) (DeCarlo et al., 2006) was operated in “mass spectrum” and “particle-time-of-flight” submodes for equal time periods.

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In a reply to one of the referee’s questions the authors refer to measurements of black carbon size distributions in Beijing from summer 2012. The measurements presented here are from 2014. I do not see how measurements from a completely different time period can be reliably used to indicate what the size distribution of black carbon was during these measurements. The authors must be much more careful in the assumptions they apply to their analysis and interpretation.

Response:

We tried our best to find more published results about the black carbon size distribution, especially those measured in the summer of 2014 in the atmosphere of Beijing. However, no more articles were found. Unfortunately, the soot particle size distributions were not measured in this study. It is true that the size distribution of BC in the atmosphere of Beijing between 2012 and 2014 could be different. Considering that the weather condition, source emissions, and transformation during summertime are similar in different years in Beijing, the BC size distributions measured in 2012 are considered as be representative. This will be pointed out in the revised MS.

Modifications in the MS:

“We should note that Sun et al. (2012a)’s measurements were performed in the summer of 2012. The soot size distribution in 2012 might be different from our measurements. Considering that the weather condition and source emissions during summertime are typically similar in different years in Beijing, BC size distributions measured in 2012 are considered as be representative.”

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To estimate particle density the authors compared the AMS's mass concentration measurement with the SMPS volume concentration measurement. It is possible to estimate density this way, see for example: Kostenidou, E.; Pathak, R. K.; Pandis, S. N. An Algorithm for the Calculation of Secondary Organic Aerosol Density Combining AMS and SMPS Data. Aerosol Sci. Technol. 2007, 41, 1002–1010, doi:10.1080/02786820701666270.But it must be done carefully. First, the collection efficiency of the AMS during this study has to be determined or otherwise estimated. Only a fraction of particles sampled by the AMS are vaporized in the instrument, and this collection efficiency is known to change with changes in particle size and composition. What was done regarding the AMS's collection efficiency here? Also, the particle size transmission efficiency of the AMS must be considered, as the len inlet's efficiency decreases quickly above ~500 nm, and there can be significant variations between aerodynamic lenses on different instruments. The method used here of just dividing the AMS mass concentration by the SMPS volume concentration is too simplistic.

Response:

The previous researches showed that collect efficiency is highly sensitive to relative humidity, aerosol acidity and ammonium nitrate content etc. (Matthew et al., 2008;Middlebrook et al., 2012). A new algorithm has been developed by Middlebrooks (2012), which takes into account the aerosol composition and relative humidity in the sampling line. This algorithm has been compiled into AMS standard data analysis software (SQUIRREL 1.56A), which is used here to calculated composition-dependent CE. Since the sampled air was dried down to RH<30% before entering into the AMS, most of the aerosols were neutralized, and also the mass fraction of ammonium nitrate is expected to play minor roles in affecting CE values, particle collection efficiency could be approximated as 0.5 during the whole campaign.

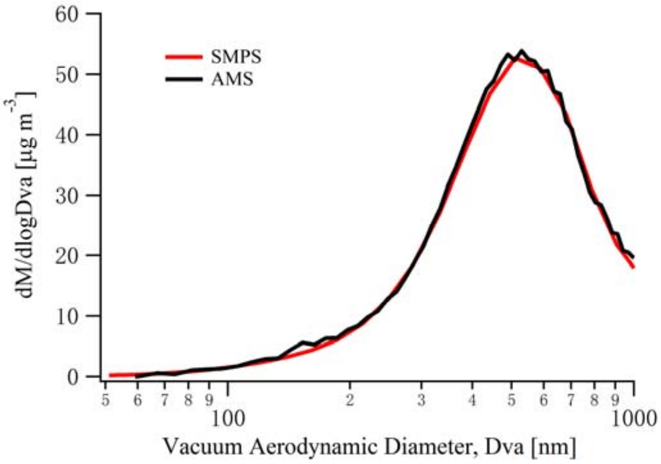

Figure 3: An example of the mass-diameter distributions derived from the AMS measurements compared to the distributions calculated from the SMPS measurements.

Particle density was estimated using the method proposed by Kostenidou (2007). Figure 3 shows an example of the mass-diameter distributions derived from the AMS measurements compared to the distributions calculated from the SMPS measurements. Here, we assume that the particles with a shape factor of 1 are spherical. The particle volume size distribution is calculated from particle number size distribution. The optimized particle effective density is 1520 kg/m^3.

Modifications in the MS:

Here, Particle density was estimated by comparing mass-diameter distributions derived from the AMS measurements and the particle volume distributions calculated from the SMPS measurements, , which is used in Kostenidou (2007) and Chen (2012). Here, we assume that the particles being detected with a shape factor of 1 are spherical. The particle volume size distribution is calculated from particle number size distribution. The optimized particle effective density is 1520 kg/m^3.

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The Referee's question does not appear to have been answered here: "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."

Response:

The aerosols properties during heavy hazy days are different from those during clean days. Our long-term measurements showed that the high number concentrations of the accumulation mode particles were observed during heavy pollution episodes (Wu et al., 2008). It is also true in this study. The analysis on particle number size distribution is hard to be involved into the discussions about the relationship between particle hygroscopicity and $PM_{2.5}$ mass concentration. Therefore, it will not be given in the revised MS.

Particle chemical composition is more relevant to particle hygroscopicity. As an example, the number fraction of the hydrophilic mode for 350 nm was colored by the inorganic (SO_4^{2-}+NO_3^-+NH_4^+) mass fraction in PM1 in the Fig. 6. Clearly, the inorganic mass fraction spanned from 0.1 up to 0.8 associating with an increasing number fraction of hydrophilic mode. When $PM_{2.5}$ mass concentration is larger than 50 μg/m^3, the inorganic mass fraction ranged from 0.5 to 0.7. This indicates that both inorganic and organic species played key roles in leading to the high aerosol loading.

Modifications in the MS:

"This means that secondary aerosol particles were dominant during severe particulate pollution episodes, occurring frequently in Beijing. As an example, the number fraction of the hydrophilic mode was colored by the inorganic (SO_4^{2-}+NO_3^-+NH_4^+) mass fraction in PM1 in the Fig. 6. Clearly, the inorganic mass fraction spanned from 0.1 up to 0.8 associating with an increasing number fraction of hydrophilic mode. When $PM_{2.5}$ mass concentration is larger than 50 μg/m^3, the inorganic mass fraction ranged from 0.5 to 0.7. This indicates that both inorganic and organic species played key roles in leading to the particle aging. The contribution of inorganic material is slightly higher than that of organic compounds."

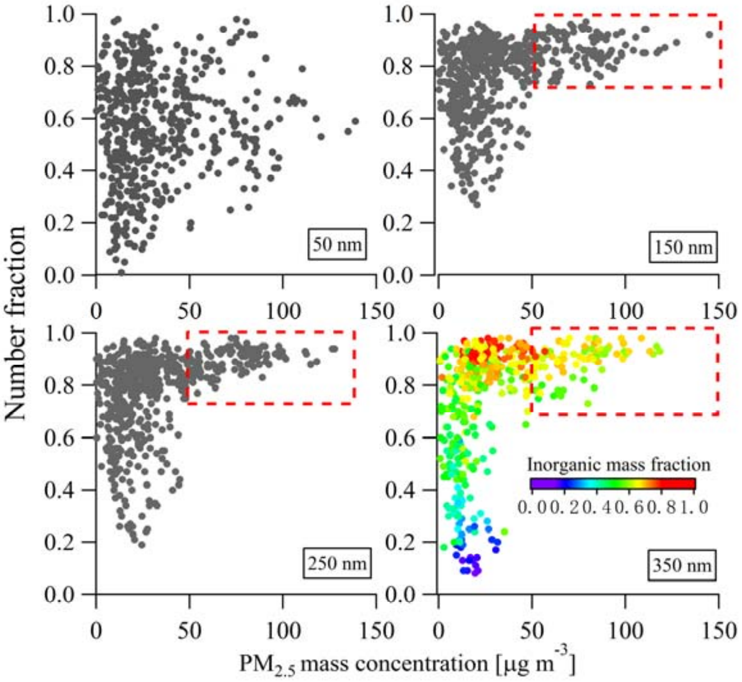

Figure 6: Number fraction of hydrophilic mode vs $PM_{2.5}$ mass concentration. For 350 nm particles, the number fraction is colored by inorganic mass fraction in PM1.

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How was the PAX instrument calibrated, using what black carbon standard?

Response:

By following PAX instruction manual and suggestions given by engineers from DMT Company, the calibration of PAX instrument was done as follows:

Mono-disperse PSL particles are used in PAX scattering calibration. 300 nm PSL particles are produced using an aerosol atomizer. After drying, the mono-disperse PSL particles selected using a DMA are introduced into PAX to detect the scattering coefficient (b_{scat}) and absorption coefficient (b_{abs}). Blank readings are obtained by measuring clean air without particles. The laser power during blank readings is set as I_0. The laser power is set as I when dry PSL samples are measured. The extinction coefficient (b_{ext}) can be calculated from Beer-Lambert law:

$$b_{ext} = \left(\frac{1}{L}\right)\ln\left(\frac{I_0}{I}\right)10^6$$

Where L is the path length of the laser beam through the cell in meters, and L =0.354 is taken in the PAX. Then, the correlation between calculated b_{ext} and b_{scat} measured by PAX is made. Since absorption coefficient is negligible for PSL, the extinction coefficient is considered as scattering coefficient. The slope, which is considered as scattering calibration factor, is used to correct the measured scattering coefficient by PAX. Regarding absorption coefficient calibration, poly-disperse propane soot particles are used. The calculation procedure is similar to the scattering calibration. Due to the light scattering of propane soot particles cannot be negligible, the correlation between the values of (b_{ext}-b_{scat}) and b_{abs} is made. The slope is considered as the absorption calibration factor to correct the measured absorption data.

Modifications in the MS:

Before carrying out the PAX measurements, the device was calibrated carefully using lab-generated monodisperse PSL particles and poly-disperse propane soot particles. The calibration curves were used to correct the measured scattering and absorption coefficients.

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Please give the flow rates used for the two SMPS instruments. Measuring up to 800 nm with an SMPS requires a very low sheath flow rate. This significantly broadens the width of the mobility selected aerosol.

Response:

TSI-SMPS (Long-DMA3081+CPC3775 and Nano-DMA3085+UCPC3776) was used to measure the particle number size distributions. According to the instruction manual, the maximum particle size can be detected is ~1000 nm. During our measurements, the sheath flow rates for Nano-SMPS were 15 L/min and 3 L/min for SMPS. The ratio of sample flowrate to the sheath air flowrate is 1:10 for both DMAs. The flowrates were calibrated using an air flow Calibrator (Gilian Gilibrator, SENSIDYNE, USA). By taking 3 L/min in the Long-DMA, the maximum size detected is 736 nm. It is true that 800 nm particles would require sheath flow rate even less than 3L/min. We will clarify this in the revised manuscript.

Modifications in the MS:

The particle number size distribution, particle hygroscopicity, and aerosol mass spectrometric measurements were concurrently made. Particle number size distributions were measured by SMPS (Long-DMA3081+CPC3775) and Nano-SMPS (Nano-DMA3085+UCPC3776). The sheath flow rates for Nano-SMPS and SMPS were 15 L/min and 3 L/min, respectively. The ratio of the sample flowrate to the sheath air flowrate is 1:10 for both DMAs. By taking 3 L/min in the Long-DMA, the maximum size detected is 736 nm. The multiple charge correction, CPC counting efficiency, and particle loss correction were carried out. Other core instruments will be briefly described below.

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The grammar and language use is still not correct in many places. Careful proofreading and editing by a person fluent in English is necessary. A few examples: “degree of scatter point of view”, “relatively dominated”.

Response:

The English writing will be improved by an English editor.

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The authors state that dust and sea salt are only present in supermicron sizes but this is a gross oversimplification. Mineral dust and sea spray aerosol can and often do extend into the submicron range, even below 350 nm. The authors cannot just assume these components were not present, though they may offer some justification for why assuming they made a small contribution is reasonable.

Response:

We agree that mineral dust particles and sea spray aerosols can extend into the submicron range. The previous studies found that a considerable number fraction of dust particles is smaller than 1 μm during the severe dust storm events in Beijing (e.g. Shao et al., 2007) . Regarding the sea salt particles, size distributions of particles produced by sea bubbles were found to have sizes as small as 0.01 μm (e.g. Clarke et al., 2006). The statistical analyses on the basis of a long-term observation showed that the dust storm events typically take place in the spring in Beijing (Wu et al., 2009). In this study, the measurements were carried out from May 31 to June 24, 2014. Meanwhile, no dust storm events were observed. The sea salt is not a major source of atmospheric particles (e.g. Song et al., 2007) because Beijing is not a coastal city. During the sampling period, the air masses spending much more time over the sea occasionally arrived in Beijing and may bring sea salt particles to the sampling site. In case of atmospheric particles being measured consist of a larger fraction of sea salt, the H-TDMA measurements should show an obvious increase in hygroscopic growth factor. However, no such phenomenon was observed.

In the revised manuscript, we will point out that the dust and sea salt can extend into the submicron range. Due to sea salt and dust particles cannot be detected by AMS, neglecting dust and sea salt may introduce bias in the closure between particle hygroscopicity and chemical composition. Neglecting sea salt particles with a high hygroscopicity growth (κ_{NaCl}=1.12 at 90% (Petters and Kreidenweis, 2007)) may cause an underestimation of particle hygroscopicity. Inversely, neglecting mineral dust with less hygroscopicity ($\kappa_{mineral\ dust}$=0.01~0.08 (Koehler et al., 2009)) may cause an overestimation of particle hygroscopicity.

Modification in the MS:

One should note that mineral dust (e.g. Shao et al., 2007) and sea salt (Clarke et al., 2006) can extend into the submicron range. Due to sea salt and dust particles cannot be detected by AMS, neglecting dust and sea salt may introduce bias in the closure between particle hygroscopicity and chemical composition. Neglecting Sea salt particles with a high hygroscopicity growth (κ_{NaCl}=1.12 at 90% (Petters and Kreidenweis, 2007)) may cause an underestimation of particle hygroscopicity. Inversely, neglecting mineral dust with less hygroscopicity (mineral dust κ= 0.01~0.08 (Koehler et al., 2009)) may cause an overestimation of particle hygroscopicity. The statistical analyses on the basis of a long-term observation showed that the dust storm events typically take place in the spring in Beijing (Wu et al., 2009). In this study, the measurements were carried out from May 31 to June 24, 2014. Meanwhile, no dust events were observed. The sea salt is not a major source of atmospheric particles (e.g. Song et al., 2007) because Beijing is not a coastal city. Therefore, the uncertainties in hygroscopicity-chemical composition closure caused by neglecting mineral dust and sea salt might be ignored.

References:

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