Authors' response to referee comment RC1
on manuscript
"Direct links between hygroscopicity and mixing state of ambient aerosols: Estimating particle hygroscopicity from their single particle mass spectra"

We thank Referee #1 for the comments and suggestions. We have addressed every comment and made significant changes to the paper to improve the paper. Again, the referee's comments are greatly appreciated.

Referee Comments in black bold.

Authors' Response **in** blue.

Changes in manuscript **in** *Red italic.*

Major comments

(1) The authors showed that the temporal variations of the estimated particle hygroscopicity were consistent with the back-trajectory analysis and atmospheric visibility observations. It is hard to believe that hygroscopicity could be simply explained by the back-trajectory analysis, or the observed hygroscopicity could be a major reason for the visibility. A closer look at the discussion revealed that such conclusions were not precisely summarized.

Response:

We agreed that there were some shortcomings in the original discussions. In the original manuscript, the back trajectories and visibility were included to support the estimated hygroscopicity, because of their connections to hygroscopicity. The original manuscript has been rearranged and revised considerably to address the referee's comments. In the revised manuscript, the back trajectories were just used to offer some general descriptions of the meteorological conditions. As suggested in the last comment. The correlations between PM and visibility by either considering the hygroscopicity or not have been added in the revised manuscript to extend the analysis of visibility.

Changes in manuscript:

Line 483-500:

"The temporal variation of particle estimated GF from Sep-12 to Sep-28 was illustrated in Figure 9. Four distinct periods (P1-P4) were identified based on their different hygroscopicity distributions. Generally, the P1 and P3 periods were characterized by elevated MH mode which dominated the ATOFMS particles numbers, while in P2 and P4 the MH particles decreased significantly and sea salt mode was pronounced. Back trajectories during P1-P4 were analyzed using HYSPLIT mode (Draxler, R. R. and Rolph, G. D.,2003) to inspect the airmass that influenced the sampling site (Figure S8). The 24-hour back trajectories suggests that the airmass in P1 period mainly circulated in local regions from northwest direction to Shanghai. The local circulations brought regional aerosol pollution to the sampling site, resulted in elevated concentrations of particles, especially the MH particles. During P2, the airmass originated from the ocean in northeast direction with less continental influence. The cleaner air from the ocean almost wiped out the accumulated particles observed in P1 and the concentrations of sea salt particles increased. In the majority of time during P3, the airmass

stayed over continental areas. The MH particles dominated particle numbers in this period and the sea salt mode were barely present. During Sep-18 to Sep-20 in P3, the LH particles showed increased concentrations and gradually decreased after Sep-20. Similar to P1, the origin of airmass in P4 shifted to the ocean in eastern directions and SS mode emerged again. Both the particle spectra and the back trajectories supported that the GF mode of 1.6 can be mainly attributed to sea salt particles."

Line 603-624:
'An exponential relation between visibility and PM concentrations was found by the previous study (Qu et al., 2015). After applying the exponential fitting to the visibility and particle volume concentrations, we found a moderate correlation for ATOFMS particles (R^2=0.45) and better correlations for $PM_{2.5}$ concentrations (R^2=0.64) (Figure S12). However, the fitting errors were clearly dependent on ambient RH, with larger errors in higher humidity, indicating that hygroscopicity might affect visibility degradations, which were consistent with other studies (Chen et al., 2012;Liu et al., 2012). To further examine the effect of particle hygroscopicity on visibility, we derived particle volumes in different RH using estimated κ values (Petters and Kreidenweis, 2007). The κ values were calculated using the GF of individual particles at 85% RH for ATOFMS data and the average GF of 1.36 for $PM_{2.5}$ volume data. With hygroscopicity being considered, we found notable improvements of the correlations between PM concentrations and visibility, with the improved correlation observed for $PM_{2.5}$ concentrations (R^2=0.82) after applying correction for hygroscopicity (Figure 12). However, this improvement was barely the case for NH particles, probably due to the negligible hygroscopic growth. For the ATOFMS particles in different GF modes, we found the highest R^2 (0.65) for the MH particles. The correlation between SS particles and visibility was distorted due to the visibility reached its limit (10 km) when sea salt mode was pronounced (Figure 9). The R^2 between MH particles and visibility suggests that the variation of MH particles accounted for the major part of visibility changes (65%) during this period, which coincided well with the major contribution of nitrate and sulfate to light extinction (61%) in eastern China areas (Qu et al., 2015). These results indicate the importance of discriminating particles by hygroscopicity in explaining the measured visibility.'

(2) The results indicate that particles with stronger hygroscopicities were more likely to have higher effective densities. Is it suitable for all the observed particle types? Could it be theoretically supported? This is hard to believe. For instance, aging of Dust particles lead to nitrate coatings, which would lower the effective density, but increase the hygroscopicity.
Response:
We agree with the referee's opinion, and decided to remove this part in the revised manuscript. Density is indeed a property related to particle types. We observed smaller ρ_{eff} of EC particles than dust/ash in less hygroscopic range based on our data, suggesting that it is inappropriate to discuss density in a general way without referring particle types. In the original manuscript, the effective density was included for the sake of a complete report of the measured data. As suggested by another referee, the discussions of effective density seem to be out of place, as the main objective of this study is to connect hygroscopicity to composition. We agree with the referee's opinion, since hygroscopicity is depending on

particle composition, rather than density. The presented data only suggested a statistical positive correlation between effective density and GF, but the explanation of the correlation need other information including particle shapes, which also affect ρ_{eff} (Ghazi et al., 2013;Khalizov et al., 2012). On the other hand, the effective densities were not well incorporated into the discussions on estimated hygroscopicity. We note that the hygroscopicity prediction algorithm is just depending on particle mass spectra and the density was not involved in calculation.

Changes in manuscript:

Relevant discussions are removed.

Specific comments:

1. Abstract: "These hygroscopicity estimation results with single particle mass spectra analysis can provide critical information on particulate water content, particle source apportionment, and aging processes." I wonder how hygroscopicity can provide critical information on the source of particles. The authors did not discuss this in the text but only took sea salt particles as an example in Figure 5.

Response:

We agree that the source apportionment was not clearly presented. Particle hygroscopicity can offer some aids in the source apportionment of single particles. An example is the Al-Si particle in Dust/ash type. Although most of the dust/ash particles show considerable hygroscopicity, the Al-Si particles had low hygroscopicity. Thus, we ascribe the sources of Al-Si to soil dust, as some laboratory studies also found soil dust has low hygroscopicity. Without hygroscopicity information, the identification of Al-Si particle source would be difficult, because the Si and Al peaks were also present in ash particles from coal emissions (Xu et al., 2017).

Changes in manuscript:

Line 42-46:

"Based on the combined information on particle composition, hygroscopicity, airmass back trajectories and ambient pollutants concentrations, we inferred that the NH, LH, MH, SS modes were characterized by POA/EC, SOA, SIA and salts compositions, respectively. The proposed method would provide additional information to the study of particle mixing states, source identification and visibility degradation."

Line 268-276:

"Some of the clusters showed characteristic hygroscopicity distributions which offered values in the source apportionment of these particles. As an illustration, we presented the mass spectra and hygroscopicity distribution of the Al-Si cluster in Figure 3. The mass spectra of Al-Si particles showed stronger aluminum ($27Al^+$) and silicate ($-76SiO_3^-$) peaks in their positive and negative spectra, respectively. Particle number distribution of Al-Si particles suggested that they were detected with the highest probability at GF 1.1. In the preliminary study we identified the similar Al-Si particles exclusively in NH mode (Wang et al., 2014). Based on their hygroscopicity distribution, we assumed the Al-Si particles are soil dusts according to their reported low hygroscopicity (Koehler et al., 2009)."

2. Introduction: "Herich et al. have firstly applied the HTDMA-ATOFMS system to investigate particle composition as a function of hygroscopicity." Some results related to such measurements are missing here.

Response:

We have added discussions on the findings of prior studies.

Changes in manuscript:

Line 96-102:

"Herich et al. firstly applied the tandem HTDMA-ATOFMS system to characterize particle composition of different hygroscopicity (Herich et al., 2008;Herich et al., 2009). A large portion of the less hygroscopic modes were found to be contributed by organics and combustion species in both urban and remote sites, while the sulfates and nitrates were present in almost all particles independent of hygroscopicity. These findings were similarly observed in our previous characterization using HTDMA-ATOFMS in Shanghai city (Wang et al., 2014), except higher nitrate and sulfate intensities were found in hygroscopic particles in our study."

3. Experimental section: what is the accuracy of hygroscopicity determination by the HTDMA? What is the uncertainty in GF prediction?

Response:

We have added relevant information in manuscript as follows.

Changes in manuscript:

Line 219-226:

"The uncertainties in the GF prediction in this method were estimated. The sources of uncertainties may stem from the errors in GF selection in HTDMA and the algorithm itself. After the 0.5 power treatment to peak intensities, the only parameter that can affect the estimated GF would be the matching criteria of particles. We have changed the matching criteria of 95-100% maximum dot products to 90-100% and 98-100% and the variations of GFs were inspected (Figure S5). Based on the distributions of GF variations, we estimated the overall uncertainties in the GF estimations is within ±0.15."

4. Line 148: "The DMAs were kept to select the desired diameters before significant number of particles were chemical analyzed by ATOFMS." What does a significant number refer to?

Response:

Particles with certain GFs (e.g., 1.4) typically have higher concentrations in the downstream of HTDMA, i.e. faster detection of particles in ATOFMS at these GFs. For other GFs (especially near the limits of GF distribution, e.g., 0.9, 1.7) particles had much smaller concentrations. For the sake of the statistical significance, we decided that the number of acquired spectra at each GF much be larger than 200.

Changes in manuscript:

Line 166 -167:

"The HTDMA-ATOFMS system was kept sampling until a sufficient number of particles (> 200) were analyzed by ATOFMS for each GF setting (Table 1)".

5. Line 171: "This problem was relieved by taking the 0.5 power (square root) of peaks intensities." Maybe more information is required to validate such treatments.

Response:

We took 0.5 power of mass peak intensities in the estimation of GF. Similar treatment (logarithm) of ATOFMS spectra was proposed in prior literature (Rehbein et al., 2012). This treatment was applied to both the HTDMA-ATOFMS and ambient particle data before the evaluation of particle similarity. It is known that ATOFMS mass peak intensities are affected not only by composition abundance in particles, but also by their ionization efficiencies in ATOFMS. The ATOFMS is more sensitive to metallic compositions than organics, resulting disproportionate large peaks of metals to reflect their actual concentrations in particles. We suppose the 0.5 power treatment of mass peak areas can partly solve the bias of ATOFMS toward different compositions, since it reduces larger peaks more rapidly than small peaks.

The application of 0.5 power treatment is not just based on speculation. We find it gave better results in the estimated hygroscopicity. We carried two rounds of GF estimations in which the pretreatment was either included or not. In figure S3 the two distributions of estimated GF are compared. As shown in figure, the hygroscopicity distribution without treatment suggests abnormal shape with an extra mode of GF=1.35, which was inconsistent with HTDMA observations in this area. As a comparison, with the 0.5 power treatment to peak intensities and the obtained hygroscopicity distributed regularly with smoother shapes, which agrees well with the HTDMA data of ambient particles. It is noted that in the two estimations the dataset and algorithm are identical with the only difference of the pretreatment. This fact suggests that the 0.5 power treatment could indeed be used to improve hygroscopicity estimation.

As a reply to the referee's concern, we have added relevant information in the revised manuscript.

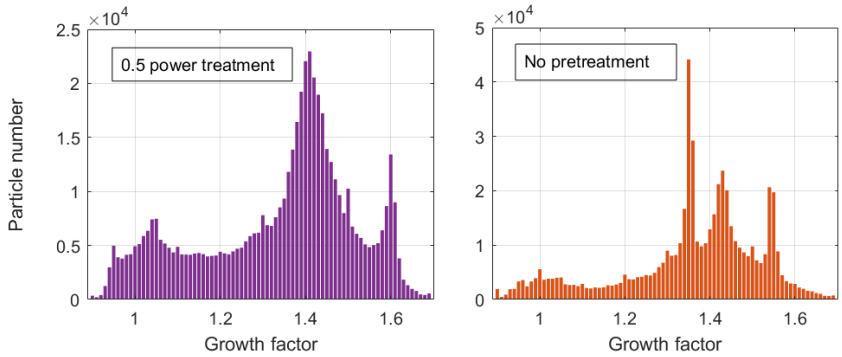

Figure S3. The estimated particle hygroscopicity distribution with (left) and without (right) taking 0.5 power of peak intensities.

Changes in manuscript:

Line 194-196:

"The 0.5 power treatment to peaks intensity was applied because it offered better results in the estimation of hygroscopicity than without it, as discussed in the supplemental information (Figure S3)."

6. Line 239: "About 20% of analyzed particles are classified as Amines-rich". Is there any explanation for such a high fraction of Amine particles?

Response:

Amine particles constitute an important type of particles and were normally detected by single particle techniques in Shanghai area. A previous single particle study suggested that the number fraction of amine particles varies considerably in different seasons (4.4% in summer vs. 23.4% in winter), which can be explained by gas-to-particle partitioning of amines at lower temperatures (Huang et al., 2012). Except temperature variations, relative humidity was also found to promote particulate amine formation through acid–base reactions of amines in high RH periods (Huang et al., 2012;Chen et al., 2019;Zhang et al., 2012). The meteorological data during the HTDMA-ATOFMS experiment indicate low temperatures (3-10 ℃, 6 ℃ in average) and higher relative humidity (50-93%, 78% in average) in this period, which both favor the formation of particulate amines in ambient aerosol. In the preliminary HTDMA-ATOFMS experiment amine particles makes up 20% of particle numbers, a comparable fraction to present study (~25%) (Wang et al., 2014).

Changes in manuscript:

Line 284-287:

"Particulate amine formation was favored in low temperatures and higher humidity conditions (Huang et al., 2012;Zhang et al., 2012). The elevated amine particle fractions may be related to the low temperature (6 ℃) and high humidity (78% RH) condition during this experiment."

7. Lines 251-252: "…CNO peaks, which are present in biomass particles, were absent very weak, suggesting that biomass burning is not their source." Do you mean CN/CNO peaks are necessarily served as markers for biomass burning?

Response:

In the ATOFMS source characterizations, biomass particles produce strong CN/CNO peak in negative spectra (Silva et al., 1999; Zauscher et al., 2013;Pratt and Prather, 2009). This mass spectral feature was also observed by studies in China (Bi et al., 2011) and our laboratory experiment on various biomass particles. We suppose the presence of CN/CNO peak is related to the ubiquity of nitrogen-containing organics in biomass, such as amino acids. Except CN/CNO, other peaks at m/z at -45/-59/-71/-73 can also be used to indicate biomass, although their intensities were typically weaker than CN/CNO. We think it is insufficient to indicate biomass particles just from the presence of CN/CNO, since CN/CNO can also be produced from other sources. The original sentence is inaccurate and we have revised it.

Changes in manuscript:

Line 298-302:

"Additionally, the -26CN and -42CNO peak which are typically present in biomass particles, were found to be weak or absent, suggesting that there are some chemical differences between Ammonium/OC and biomass particles (Silva et al., 1999;Zauscher et al., 2013;Pratt and Prather, 2009)."

8. Line 256: "…characterizations, it is possible that the ammonium/OC particles might be from coal burning sources". There are already some paper published reporting the single particle mass spectra of coal burning particles in China. The author should directly refer to these papers, rather than (Healy et al., 2010). Still, such an assignment might not be appropriate, since it is more likely produced from secondary processes, associated with high

abundance of ammonium sulfate.

Response:

We followed the referee's suggestion and revised the manuscript.

Changes in manuscript:

Line 305-310:

"A prior ATOFMS study identified the Ammonium/OC particles are from agricultural sources, and found most of them were present in higher photochemical oxidation periods (Qin et al., 2012), consistent with the prominent secondary peaks of ammonium found in this study. It is likely the organics in this type is secondary since the GF 1.2 is close to the hygroscopicity of SOA (GF=1.24 at 90% RH) (Gysel et al., 2007;Sjogren et al., 2008). "

9. Line 260: the authors further concluded that "These particles were not likely to be deeply aged particles, because their hygroscopicity was only moderate." It is a little bit confusing, since the aging should be deduced from the mass spectra, not the hygroscopicity. Hygroscopicity could be linked to the chemical compositions, but not particle age.

Response:

We agree that the original statement is inaccurate. We have modified this sentence.

Changes in manuscript:

Line 310-312:

"We inferred that the ammonium was not contributing significant mass fractions to these particles, due to the high hydrophilicity of ammonium salts while Ammonium/OC particles were only moderate hygroscopic".

10. Line 398: "…higher hygroscopicity could play more important role…" higher correlation cannot infer the higher contribution of aerosol particles with higher hygroscopicity to visibility decrease. Is it possible to estimate the relative contribution based on the combined measurements in this study?

Response:

This comment is very instructive and we attempted to analysis hygroscopicity and visibility in further detail. We acknowledge that the original statement is over simplified since PM concentrations were also contributing to visibility degradation. To evaluate the role of hygroscopicity more explicitly, we compared PM concentrations with visibility in two situations where hygroscopicity was either considered or not. In the revision we only consider particle volume concentrations since hygroscopic growth increase particle volumes rather than particle numbers. The results suggested improved correlations between visibility and particle concentrations of LH, MH, SS, all ATOFMS particles and $PM_{2.5}$ concentrations with hygroscopic growth being considered.

Changes in manuscript:

Line 584-624:

"3.3.4 Comparing the estimated hygroscopicity with visibility

Particle optical properties were closely connected to hygroscopicity (Liu et al., 2012;Qu et al., 2015;Chen et al., 2012). The hygroscopic growth increases particle volumes and cross sections and is contributing to the visibility degradation. With the estimated hygroscopicity of ATOFMS particles, we correlated atmospheric visibility with particle concentrations to study their

contributions to the visibility variation. The ATOFMS particle volume concentrations were calculated for hygroscopicity modes of NH, LH, MH and SS based on ATOFMS particle diameter and numbers. The particle volume concentrations was used because hygroscopic growth change particle sizes rather than numbers (Chen et al., 2012). The visibility data was obtained from (https://www.wunderground.com/) logged in the Hongqiao airport (31°12'N, 121°20'E) and Pudong airport (31°9.3'N, 121°49'E) during the study period (see the map in Figure S5). The temporal variations of visibility in two sites correlated strongly (Figure S5), despite the 45 kilometers distance between two airports. The Fudan site is located roughly between the two airports, and the two sets of visibility data were averaged to represent the study site. In P2 and P4 the site was under the influences from ocean, resulting visibilities larger than 10 km (Figure 9). Apart from ATOFMS particles, contemporary $PM_{2.5}$ volume concentrations were also correlated with visibility. The $PM_{2.5}$ volume concentrations were derived from $PM_{2.5}$ mass concentrations using particle density (1.4 gcm^{-3}). A strong correlation between ATOFMS particle numbers and $PM_{2.5}$ was found (R^2=0.80).

An exponential relation between visibility and PM concentrations was generally found (Qu et al., 2015). With the application of the exponential fitting to the visibility and particle volume concentrations, we found moderate correlation for ATOFMS particles (R^2=0.45) and better correlations for $PM_{2.5}$ concentrations (R^2=0.64) (Figure S12). However, the errors of fittings were clearly dependent on ambient RH conditions, with increased errors in higher humidity This result indicates that hygroscopicity have played its roles in the visibility degradations (Chen et al., 2012;Liu et al., 2012). To account for particle hygroscopicity, we derived particle volumes in different RH using κ values (Petters and Kreidenweis, 2007). The κ values were calculated using the GF of individual particles at 85% RH for ATOFMS data and the average GF of 1.36 for $PM_{2.5}$ volume data. With hygroscopicity being considered, we found notable improvements of the correlations between PM concentrations and visibility, with the best correlation observed for $PM_{2.5}$ concentrations (R^2=0.82) after correction for hygroscopicity (Figure 12). However, this improvement was barely present for NH particles due to the negligible hygroscopic growth. For the ATOFMS particles in different GF modes, we found the highest R^2 (0.65) for the MH particles and gradually loose correlations from MH to NH mode. The low correlation for SS particles is related to the saturated visibility (10 km) when sea salt mode was pronounced (Figure 9). The R^2 of MH particles suggests the MH particles were explaining the major part of visibility variations (65%) in this period, which coincided well with the major contribution of nitrate and sulfate to light extinction (61%) in eastern China areas (Qu et al., 2015). These facts indicate the importance of discriminating particles by hygroscopicity in explaining the measured visibility."

Minor:

References such as "Zelenyuk et al.(Zelenyuk et al., 2008)" is incorrectly formatted.

Response:

We have made correction to this reference.

Changes in manuscript:

Line 93-96:

"Zelenyuk et al. connected a single particle mass spectrometer SPLAT with HTDMA to demonstrate the capability of this system to derive quantitative information on aerosol

hygroscopicity, composition, and effective density (Zelenyuk et al., 2008).”

Line 98: “HTMDA-ATOFMS”

Response:

This mistyping was corrected.

Changes in manuscript:

Line 107-108:

“We conducted a comprehensive HTDMA-ATOFMS experiment with the particle GF varied in a more complete range (0.9~1.7, 85% RH)”.

Line 183: “value from 0.1 to 1.7” is it 0.9-1.7?

Response:

The mistyping was corrected.

Changes in manuscript:

Line 208-209:

“where: GF_{pred}= the estimated GF of ambient particle, GF_i= GF value from 0.9 to 1.7 interspaced by 0.1, F_i= number percentages of the matched particles in each GF bin.”

Line 314: “can be measured on by HTDMA-ATOMFS system”

Response:

The discussions of effective density were removed.

Changes in manuscript:

This sentence was removed after revision.

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