Authors' response to referee comment RC2
on manuscript
"Direct links between hygroscopicity and mixing state of ambient aerosols:
Estimating particle hygroscopicity from their single particle mass spectra"

We thank Referee#2 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 claim that they can predict the growth factor from the mass spectra, but they mainly just showed associations between particle types and growth factors, not that the spectra provide a degree of predictability.

Response:

We thank the reviewer for this comment. We would call it "estimate" rather than "predict". The GF of a particle can be estimated based on HTDMA-ATOFMS data for two reasons. First, different particle types had distinct GF distributions. Second, particles in different bins had different mass spectra. In the revised manuscript, we have added a section to discuss this issue. The discussions of GF estimation were inserted between the discussions of HTDMA -ATOFMS data and estimated GF ambient particles.

Changes in manuscript:

Line 391-407:

"3.2 Predictability of hygroscopicity from particle mass spectra

The GF of a particle can be estimated based on HTDMA-ATOFMS data for two reasons. First, different particle types had distinct GF distributions. Second, particles in different GF bins had different mass spectra. The GF estimation from particle spectra requires that the HTDMA-ATOFMS data is capable to represent the major particle types normally presented in atmosphere, which is evidenced in the preceding discussions. In another aspect, the GF prediction from mass spectra also demands that HTDMA-ATOFMS data are sensitive to reflect the composition differences with GF variations.

To test the sensitivity of HTDMA-ATOFMS data, we evaluated the average spectral similarities between each pair of GF groups. The average similarities were calculated from the similarities between every possible pairs of particles from the two GF groups. The self-comparing of particles within the same GF group were excluded. As shown in Figure 6, we observed a general trend that particles in the same GF bins tend to produce the highest similarities. As the GF differences increase, the mass spectra similarity between two GF bins tended to decrease. This result is an evidence that the particles with different GFs are more likely to have discriminable mass spectra, which suggests that the HTDMA-ATOFMS dataset are capable to estimate hygroscopicity just from particle mass spectra."

2. The authors also did not explore whether ion peak areas such as sulfates and nitrates, which are critical for particle water uptake, were better predictors of growth factors than the particle types themselves.

Response:

We have followed this instructive comment and revise the paper carefully. We have correlated peak intensities of nitrate and sulfate with GF for all particles in the HTDMA-ATOFMS dataset. There were indeed correlations between nitrate and sulfate signals and hygroscopicity in certain GF range but a simple trend applicable to all GF was not found. Another characteristic of peak intensities is that their trends were dependent on particle types. The discussion of peak intensities with GF were added both for HTDMA-ATOFMS data and ambient ATOFMS particles.

Changes in manuscript:

Line 344-390:

"Apart from particle number distributions, the HTDMA-ATOFMS dataset provided another aspect of information regarding peak intensities with GF. In this study, we used relative peak intensities (peak areas normalized by the total areas in spectrum) to investigate its relation to GF. Generally, the responses of peak intensity to GF variation were found to be nonlinear, since they were correlated only within specific GF ranges. A simple trend applicable to whole GF range was not observed.

We presented the statistics of peak intensity of nitrate ($-46NO_2$, $-62NO_3$, $-125H(NO_3)_2$) and sulfate ($-80SO_3$, $-97HSO_4$) which were known to be critical to particle hygroscopicity (Figure 4). As previously observed (Herich et al., 2009;Herich et al., 2008;Wang et al., 2014), the nitrate and sulfate peaks were present in the majority of particles in all GF bins. However, peak intensities of nitrate and sulfate were indeed stronger in hygroscopic particles than hydrophobic particles. In Figure 4 we observed positive correlation between nitrate and sulfate intensities and GF in GF <1.2 range, suggesting contribution of nitrate and sulfate to particle hygroscopicity in low GF range (Figure 4). However, in higher GF range (GF 1.3-1.5), nitrate and sulfate peaks seem to reach a plateau with unclear dependence on GF. Nitrate and sulfate were known to contribute large fractions of particle mass in MH particles (Swietlicki et al., 2008;Laborde et al., 2013;Liu et al., 2014). The unclear trend of nitrate and sulfate with GF seem to suggest that nitrate and sulfate were in stable ratios since nitrate and sulfate peaks were dominating peak areas in negative spectra. For particles of even higher GF, differences were observed between GF 1.3-1.5 and GF 1.5-1.7 range in that stronger nitrate and weaker sulfate peaks were detected in the GF 1.3-1.5 range. Particles classification suggests that this general characteristic is also at variance for different particle types. The same statistics for EC and Dust/ash particles were presented in Figure 4. Compared with EC particles, smaller sulfate and stronger nitrate peaks were found in Dust/ash spectra, and the observed trend in total particles were less obvious in Dust/ash. These facts highlight the nonlinearity between peak intensities and GF and that particle types should also be considered in describing peak intensities.

The analysis of peak intensities with GF can disclose some atmospheric processes happened on aerosol. We take the Sea salt as an illustration. Sea salt particles were known to react with atmospheric nitric acid, with NaCl in fresh sea salt be transformed into NaNO3 in the reacted sea salt (Gard et al., 1998). This composition transformation is indicated in corresponding

changes of NaCl and NaNO₃ peak intensities in particle spectra. The unreacted sea salt particles tend to produce larger peaks of Na_2Cl^+ and $NaCl_2^-$ in spectra (Gaston et al., 2011;Prather et al., 2013). In particle spectra of reacted sea salt, the NaCl peaks (Na_2Cl^+, $NaCl_2^-$) decrease while NaNO₃ peaks ($Na_2NO_3^+$, $Na(NO_3)_2^-$) increase. We presented peak intensities of sea salt in GF 1.5-1.7 range where sea salt particles were detected with largest numbers (Figure 5). We found that the positive correlation between NaCl peak intensity and GF, and the negative correlation for NaNO₃ peaks. Therefore, the HTDMA-ATOFMS data supported that reacted sea salt have reduced hygroscopicity (Herich et al., 2009;Gaston et al., 2018). Laboratory HTDMA study suggested that NaCl and NaNO₃ have deliquesced at 85% RH and that the NaNO₃ (GF ~1.8) is less hygroscopic than NaCl (GF ~2.2) (Hu et al., 2010). The reduced hygroscopicity of sea salt is in line with the GF of sodium salts. However, the sea salt hygroscopicity (GF 1.5-1.7) was smaller than pure NaNO₃ salt (supposing fully reacted), suggested that the chemical transformation alone is not sufficient to account for the observed hygroscopicity of sea salt. We hypothesize that other compositions as organics were mixed into sea salt and contributed to the reduction of sea salt hygroscopicity (Gaston et al., 2011;Randles et al., 2004;Facchini et al., 2008)."

Line 548-583:
"3.3.3 Peak intensity variations with estimated GF
Particle hygroscopicity and peak intensities in particle mass spectra were correlated to show their connections. The correlation was illustrated similarly to the analysis of the HTDMA-ATOFMS dataset, as shown in Figure 11. In addition to the statistics on peak intensities of different GF, the number distributions ATOFMS particles with GF and peak intensities were presented for nitrate (-46NO₂, -62NO₃, -125H(NO₃)₂) and sulfate peaks (-97HSO₄, -80SO₃) in the lower panels in Figure 11. The general trends of peak intensities with GF in HTDMA-ATOFMS dataset was preserved in estimated GF of ambient particles. The trends of nitrate and sulfate peak intensities showed increases from NH to LH range and remained constant in the MH mode. Similar to HTDMA-ATOFMS particles, stronger nitrate peaks were detected in SS particles compared with the MH particles, while an opposite trend was observed for sulfate intensities. These results highlight the nonlinearity between GF and peak intensities of ATOFMS particles.

The nonlinearity of peak intensities with GF was also suggested by the different particle types presented in ATOFMS data. As shown in the lower panels in Figure 11, the distribution of ATOFMS individual particles showed enrichment in different areas in the GF-peak intensity diagram, suggesting the presence of particle groups of different compositions. To illustrate this character, we selected two areas with clear particle enrichments in GF-peak intensity diagram and their particle composition were analyzed (denoted as A and B in lower left panel in Figure 11). Obviously, particles in area A produced much larger nitrate signals than particles in area B. Particle numbers in the two areas suggested that Dust/ash dominated particles in area A (59%) while in area B the Dust/ash only accounted for 14% of particles (Figure S10). As a contrast, particles in area B were dominated by Aged EC type (53%) followed by 25% Dust/ash. Table 2 suggests that Aged EC and Dust/ash are the major types presented in the same MH mode. However, peak intensity responses to GF were indeed different for different particle types, suggesting the importance of particle types in describing peak intensities.

The particle distribution with sulfate intensities showed similar enrichment patterns to nitrate (lower right panel in Figure 11). Sulfate peak intensities were found to correlated with hygroscopicity in GF <1.2 range but in MH range no correlated with GF was observed. We note that except the larger peaks of nitrate and sulfate, some smaller peaks were also found to correlated with GF within specific particle type. We correlated peak intensities of Na_2Cl^+ and NO_3^- with the estimated GF of sea salt particles (Figure S9). The Na_2Cl^+ peaks were positively correlated with GF while the nitrate peaks were negatively correlated with GF. The observed correlation in sea salt particles are consistent with discussed trends in the HTDMA-ATOFMS dataset. These results demonstrate that the GF estimation method have effectively reflected the minor changes in particle mass spectra into the estimated hygroscopicity."

3. This paper is missing critical information about how the ATOFMS data was treated and analyzed and what criteria were used to assign particle types and ascribe a growth factor.
Response:

We agree that the description of ATOFMS data analysis method is necessary. We have added this missing information in the revised manuscript. It is made clear that a single growth factor of particle types was not assigned, since particles in each type were distributed at more than one GF. The hygroscopicity of particle types were calculated based on their number distributions at different growth factors.

Changes in manuscript:

Line 139-146:

"The ATOFMS data was analyzed within the YAADA toolkit (http://www.yaada.org/). Particles with similar composition were grouped into clusters by an adaptive resonance algorithm (ART-2a) (Song et al., 1999). The ART-2a algorithm parameters were set to: vigilance factor = 0.85, learning rate = 0.05 and number of iterations = 20. The clusters generated by the algorithm were regrouped into major types by considering their similar composition and hygroscopicity patterns. The obtained particle types were labelled by consulting previous single particle characterization studies (Spencer et al., 2006;Silva et al., 1999;Sullivan et al., 2007a;Gaston et al., 2011;Qin et al., 2012). "

4. I don't understand the authors' claim regarding particle effective density as a predictor for particle hygroscopicity. Can the authors show a theoretical reason for such a claim?
Response:

In the original manuscript the effective density was prepared for the purpose of a complete report of the measured data, since aerodynamic diameters were measured by ATOFMS. As suggested in another item of comment, the discussions of effective density seem out of place in view of the main objective of this study is to connect hygroscopicity to composition. We accept the referee's opinion since the hygroscopicity are depending on particle composition, rather than particle density. The presented data only suggested a statistical positive correlation between effective density and GF, but a meaningful explanation of this correlation need further information including   particle physical shapes, which also affect ρ_{eff} (Ghazi et al., 2013;Khalizov et al., 2012). On the other hand, effective densities were not well incorporated into the discussions on estimated hygroscopicity. Therefore, the discussion of effective density seems somewhat redundant. In revised manuscript we decided to

concentrate on particle composition and the effective density were not discussed. We note that the hygroscopicity prediction algorithm is just based on particle composition and the density was not involved in calculation.

Changes in manuscript:

This section of information was removed.

Specific Comments

Abstract:

1. Lines 31-32: The authors need to explain why the higher effective density correlates with the hygroscopicity.

Response:

We only suppose that the lower effective densities of NH particles may be caused by the organic compositions. The higher densities of hygroscopic particles are probably related to the increasing fractions of secondary matters in particles. As responded in the major comment 4, this section of discussion was removed.

Changes in manuscript:

This part of information was removed.

2. Lines 32-34: The authors need to revise this statement or revise their work to show a degree of predictability of the GF was achieved.

Response:

As responded in the major comment 1, we re-analyzed the HTDMA-ATOFMS data and made the suggested revisions to the manuscript, in which the discussions on predictability, correlations between peak intensities and GF were added.

Changes in manuscript:

Line 31-33:

"Peak intensities in particle spectra were found nonlinearly correlated with hygroscopicity and the correlations were variant with particle types."

3. Lines 37-39: I don't understand this claim that back-trajectory analysis is consistent with particle hygroscopicity. The authors need to clarify this statement.

Response:

The original statement is problematic. We have revised the manuscript considerably according the referee's suggestions. The original statement is changed as follows.

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."

Introduction:

1. Lines 47-49: This sentence is very vague. The authors should discuss heterogeneous and

multiphase reactions that would affect particle hygroscopicity.

Response:

We have revised this sentence and relevant references were added as suggested.

Changes in manuscript:

Line 53-55:

"Aerosol particles provide surfaces for atmospheric heterogeneous reactions to occur and act as the sink for these reactions (Sullivan et al., 2007b;Gard et al., 1998;Qiu and Zhang, 2013), which are of significance to air quality, visibility and human health. "

2. While the coupling of techniques was discussed, the authors need to add discussion of growth factors observed in ambient environments and what has been learned about how composition drives hygroscopicity in tandem experiments.

Response:

As suggested, some discussions on the growth factors had been added, together with the previous findings in these studies.

Changes in manuscript:

Line 79-83:

"The HTDMA measurements suggest that several hygroscopicity modes exist simultaneously, probably due to the external mixing of atmospheric particles. HTDMA hygroscopicity were generally fell into four categories: nearly hydrophobic (NH), less-hygroscopic (LH), more-hygroscopic (MH), and sea-salt mode, with their center GFs (90% RH) lie in 1.0-1.11, 1.11-1.33, >1.33 and >1.8 ranges, respectively (Swietlicki et al., 2008)."

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 both in the urban and remote site, while the sulfates and nitrates were present in almost all particles independent of hygroscopicity. Similar findings were also observed in our preliminary 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."

Methods:

1. The data analysis methods for the ATOFMS measurements are missing yet are a critical component of this work. What clustering method was used? Art2a? What vigilance factor, learning rate, and number of iterations were used? How were particle types assigned?

Response:

We agree that the description of ATOFMS data analyzing method is necessary. We have added this missing information in manuscript as follows.

Changes in manuscript:

Line 139-146:

"The ATOFMS data was analyzed within the YAADA toolkit (http://www.yaada.org/). Particles with similar composition were grouped into clusters by an adaptive resonance algorithm (ART-2a) (Song et al., 1999). The ART-2a algorithm parameters were set to: vigilance factor = 0.85,

learning rate = 0.05 and number of iterations = 20. The clusters generated by the algorithm were regrouped into major types by considering their similar composition and hygroscopicity patterns. The obtained particle types were labelled by consulting previous single particle characterization studies (Spencer et al., 2006;Silva et al., 1999;Sullivan et al., 2007a;Gaston et al., 2011;Qin et al., 2012). "

2. What size range did the ATOFMS hit particles? Usually 250 nm is on the lower end of what the instrument can see for certain inlet configurations, what is the detection efficiency of the ATOFMS at this lower size?

Response:

According to instrument manual, the TSI ATOFMS-3800 can analyzes particles in 0.1-3 μm range (the AFL model is 3800-100). Ambient studies in Shanghai verified that most of the ATOFMS particles lie in this size range. Considering that the 250 nm is on the lower end of detection range, we expected the ATOFMS detection efficiency has decreased compared with larger particles. In HTDMA-ATOFMS experiment we selected 250 nm dry particles because the concentrations of larger particles were found to decrease further in the SMPS size distributions. We calculated ATOFMS detection efficiency at 250 nm by referring to the CPC concentration which was measured parallel to ATOFMS. Considering the different flowrates of CPC and ATOFMS, the ATOFMS and CPC data suggest that ATOFMS have detection efficiency of 1.6×10^{-2} in this experiment. The ATOFMS detection efficiency is also subjected to variation with GF as shown in Figure S2. In revised manuscript we added this part of information.

Changes in manuscript:

Line 172-181

"The ATOFMS used in this study detects particles in 100-3000 nm diameter range. The 250 nm dry diameter is near the lower end of ATOFMS detection range, therefore the detection efficiency at this size is expected to be lowered compared with larger particles. This study selected 250 nm particles because the concentrations of larger particles decreases further, as indicated by the SMPS size distributions. Therefore, the selection of D_{dry} = 250nm is a compromise between detection probability and particle concentrations (Wang et al., 2014; Herich et al., 2008). With the measured ATOFMS particle numbers and CPC concentrations, the detection efficiency of ATOFMS were calculated to be ~1.6×10^{-2} at the dry size. In Figure S2 we presented ATOFMS detection efficiencies together with the particle hit rate (hit particles/total sized particles) at different growth factors."

3. What were the hit rates for the different growth factors? This will be important for assessing how representative the observed particles are of each growth factor.

Response:

ATOFMS hit rates for different growth factors is presented in Figure S2. Higher hit rates for particles of low hygroscopicity range (GF <1.3) were observed than more hygroscopic particles (GF >1.4). This variation of hit rate is probably caused by the different particle compositions at different growth factors. In previous ATOFMS study it was verified that the coating of secondary species on particles can reduce the ionization efficiencies and lower the hit rate (Hatch et al., 2014). This is consistent with the HTDMA-ATOFMS observation that more

hygroscopic particles tend to produce stronger secondary peaks. We added this information in revised manuscript.
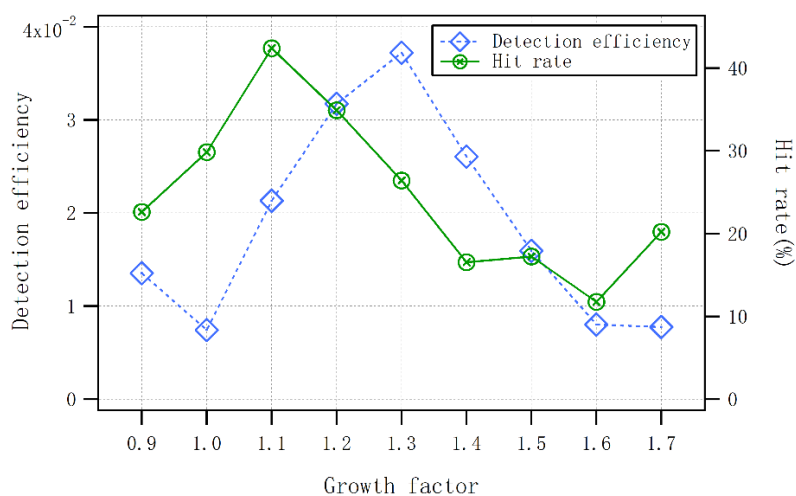
Figure S2. ATOFMS particle detection efficiency and hit rates at different growth factors.

Changes in manuscript:

Line 180-184:

"In Figure S2 we presented ATOFMS detection efficiencies together with the particle hit rate (hit particles/total sized particles) at different growth factors. Generally, the detection efficiencies suggested variations at different GFs. We found higher detection efficiencies in moderate GF range (1.2-1.4) and higher hit rates in GF<1.3 range, which is probably caused by variations of compositions with GF (Hatch et al., 2014)."

4. Lines 166-167; Lines 191-194: the authors mentioning assigning a GF for ambient particles that match particles observed during HTDMA-ATOFMS spectra. What was the criteria for matching particles and assessing their similarity? Was a threshold dot product used to assess similarity as was done for prior ATOFMS studies (e.g., [Pratt and Prather, 2009]).

Response:

We matched the HTDMA-ATOFMS particles with ambient particles by the criteria of mass spectral similarity. The similarity was calculated in the same way as that in ART-2a algorithm, that is, the dot products between the normalized mass spectra. In this preliminary study, we set a threshold dot product in matching particles. The similarities of the matched particles were recorded by the algorithm during calculation. The distribution of similarities suggests that the majority of particles (79.4%) have matching dot products in >0.8 (average dot product 0.86). We note that the 0.5 power treatment to peak intensities was applied before the calculation of similarity. To certain extend this exponentiation of peak intensities lowered the dot products between ATOFMS and HTDMA-ATOFMS mass spectra (96% of matching dot products >0.8, average dot product 0.94), because peaks of smaller intensities increased their weights in the evaluation of similarity. The figure S4 shows the distribution of the matching similarities. We have added the missing information in manuscript.
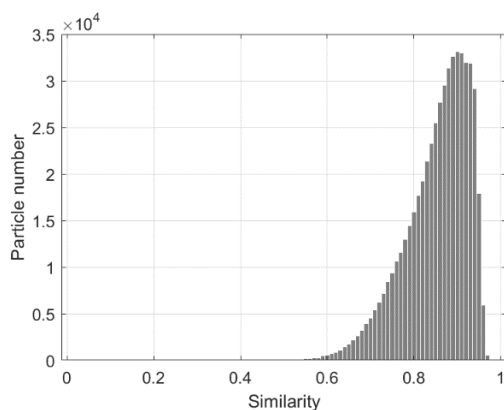
Figure S4. The distribution of matching similarities in the estimation of hygroscopicity.

Changes in manuscript:

Line 199-202:

"In this study we set a threshold similarity (0.7 dot product) in matching particles, as was required in ART-2a algorithm (Song et al., 1999). Ambient particles with matching dot products <0.7 were excluded from analysis of the estimated GF. The similarity data suggests that 96.2% of the matching similarities are > 0.7 and 79% of them are >0.8 (Figure S4)."

5. Lines 171-174: I have not heard of treating ATOFMS data this way. It is well-known that the instrument is sensitive to ionization potential energies ([Gross et al., 2000]) and usually relative intensities are used to work with such spectra. The authors' method of arbitrarily reducing the intensity of metals so that organics are relatively enhanced requires significant justification.

Response:

We took 0.5 power of mass peak intensities in the estimation of GF. This treatment was applied to both the HTDMA-ATOFMS and ambient particle data before the evaluation of particle similarity. As noted by the referee, ATOFMS mass peak intensities are affected not only by composition abundance in particles, but also by their ionization efficiencies in ATOFMS. The ATOFMS is known to be 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 this treatment reduce larger peaks more rapidly than smaller peaks. Similar treatment (take the logarithm of peak areas) of ATOFMS data was proposed in prior literature (Rehbein et al., 2012). In ATOFMS data analysis the peak relative intensities were also used. Relative intensities are the normalized intensities by the total peak area in spectrum. However, the normalization only removes the amplitude of mass spectra but the overall shape of the spectra is not changed.

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 using 0.5 power treatment or not are compared. As shown in figure, the hygroscopicity distribution without treatment shown abnormal shape with an extra mode of GF=1.35, which was inconsistent with HTDMA observations in this area. As a comparison, we applied the 0.5

power treatment to peak intensities and the obtained hygroscopicity were distributed regularly with smoother shapes, which agrees well with the HTDMA data of ambient particles. We note that in the two estimations the dataset and algorithm are identical with the only difference of pretreatment. This fact suggests that the 0.5 power treatment could indeed be used to improve hygroscopicity estimation.

To reply to the referee's concern, we have added relevant information in manuscript.
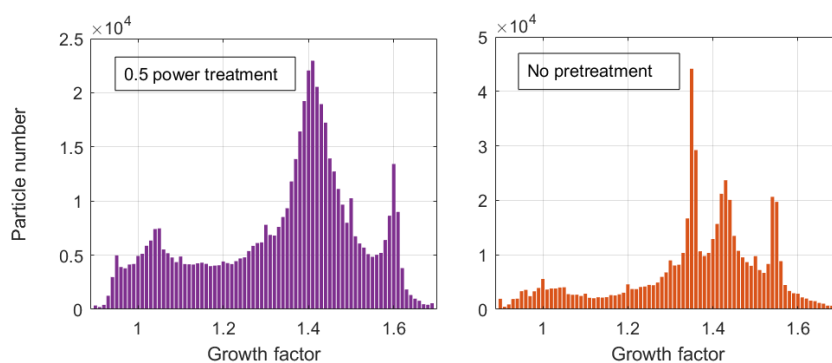
Figure S3. The estimated particle hygroscopicity distribution with (left) and without (right) taking 0.5 power of peak intensities.

Changes in manuscript:

Line 194-196:

"The 0.5 power treatment to peaks intensities was applied because it offered better results in the estimation of hygroscopicity, as discussed in the supplemental information."

6. The authors used candidate particle types to explain hygroscopicity, but did they ever simply compare sulfate and nitrate peak areas to the growth factor data to see if those soluble compounds could explain their results better than different particle types?

Response:

We have considered the referee's suggestions carefully and have added relevant discussions in revisions.

Changes in manuscript:

Line 344-390:

"3.1.2 Peak intensity variations with GF

Apart from particle number distributions, the HTDMA-ATOFMS dataset provided another aspect of information regarding peak intensities with GF. In this study we used relative peak intensities (peak areas normalized by the total areas in spectrum) to study its relations to GF. Generally, the responses of peak intensity to GF variation were found nonlinear since they were correlated within specific GF ranges but a simple trend applicable to all GF was not observed. To demonstrate this character, we presented the statistics of peak intensity of nitrate (-46NO₂, -62NO₃, -125H(NO₃)₂) and sulfate (-80SO₃, -97HSO₄) which were known to be critical to particle hygroscopicity (Figure 4). As previously observed (Herich et al., 2009;Herich et al., 2008;Wang et al., 2014), the nitrate and sulfate peaks were present in the majority of particles in all GF bins. However, peak intensities of nitrate and sulfate were indeed stronger in hygroscopic particles than hydrophobic particles. In Figure 4 we observed positive correlation between

nitrate and sulfate intensities and GF in GF <1.2 range, suggesting contribution of nitrate and sulfate to particle hygroscopicity in low GF range (Figure 4). However, in higher GF range (GF 1.3-1.5), nitrate and sulfate peaks seem to reached a plateau with unclear dependence on GF. Nitrate and sulfate were known to contribute large fractions of particle mass in MH particles (Swietlicki et al., 2008;Laborde et al., 2013;Liu et al., 2014). The unclear trend of nitrate and sulfate with GF seem to suggest that nitrate and sulfate were in stable ratios since nitrate and sulfate peaks were dominating peak areas in negative spectra. For particles of even higher GF, differences were observed between GF 1.3-1.5 and GF 1.5-1.7 range in that stronger nitrate and weaker sulfate peaks were detected in the GF 1.3-1.5 range. Particles classification suggests that this general characteristic is also at variance for different particle types. The same statistics for EC and Dust/ash particles were presented in Figure 4. Compared with EC particles, smaller sulfate and stronger nitrate peaks were found in Dust/ash spectra, and the observed trend in total particles were less obvious in Dust/ash. These facts highlight the nonlinearity between peak intensities and GF and that particle types should also be considered in describing peak intensities.

The analysis of peak intensities with GF can disclose some atmospheric processes happened on aerosol. We take the Sea salt as an illustration. Sea salt particles were known to react with atmospheric nitric acid, with NaCl in fresh sea salt be transformed into $NaNO_3$ in the reacted sea salt (Gard et al., 1998). This composition transformation is indicated in corresponding changes of NaCl and NaNO3 peak intensities in particle spectra. The unreacted sea salt particles tend to produce larger peaks of Na_2Cl^+ and $NaCl_2^-$ in spectra (Gaston et al., 2011;Prather et al., 2013). In particle spectra of reacted sea salt, the NaCl peaks (Na_2Cl^+, $NaCl_2^-$) decrease while NaNO3 peaks ($Na_2NO_3^+$, $Na(NO_3)_2^-$) increase. We presented peak intensities of sea salt in GF 1.5-1.7 range where sea salt particles were detected with largest numbers (Figure 5). We found that the positive correlation between NaCl peak intensity and GF, and the negative correlation for $NaNO_3$ peaks. Therefore, the HTDMA-ATOFMS data supported that reacted sea salt have reduced hygroscopicity (Herich et al., 2009;Gaston et al., 2018). Laboratory HTDMA study suggested that NaCl and NaNO3 have deliquesced at 85% RH and that the $NaNO_3$ (GF ~1.8) is less hygroscopic than NaCl (GF ~2.2) (Hu et al., 2010). The reduced hygroscopicity of sea salt is in line with the GF of sodium salts. However, the sea salt hygroscopicity (GF 1.5-1.7) was smaller than pure $NaNO_3$ salt (supposing fully reacted), suggested that the chemical transformation alone is not sufficient to account for the observed hygroscopicity of sea salt. We hypothesize that other compositions as organics were mixed into sea salt and contributed to the reduction of sea salt hygroscopicity (Gaston et al., 2011;Randles et al., 2004;Facchini et al., 2008;Herich et al., 2009). "

Results:

1. The particle statistics mentioned in Section 3.1.1 are really hard to follow and put into context. I suggest just keeping this section focused on particle types, then moving Section 3.2.1 up and providing better statistics for the particle types that contributed to each growth factor including a description of the percentage of each particle type observed for each growth factor and what the hit rates were on the ATOFMS for each growth factor.

Response:

We accepted the referee's suggestions and have made corresponding rearrangements of text

in manuscript. The changes include removing the statistics on particle contribution of HTDMA-ATOFMS particles and adding new statistics on particle contributions in hygroscopicity modes based on ATOFMS particles. Since the GF were estimated only for hit particles, we cannot derive the hit rates at different GF for ATOFMS particles. As an alternative, we provided the hit rates information with GF based on the HTDMA-ATOFMS data in Figure S2.

Changes in manuscript:

Line 463-481:

"In Table 2 we made the statistics on average number contributions of particle types to the NH, LH, MH and SS mode. The presented statistics were based on the temporal contributions of each particle types in daily resolution. It is noted that particle number contributions presented in Table 2 may be different from HTDMA-ATOFMS dataset (Figure 2). For example, the Cooking particles contributions to NH mode was significantly lower in the ATOFMS dataset (3%) than HTDMA-ATOFMS dataset (19%). This result is understandable because particle concentrations are variant with particle size and HTDMA-ATOFMS only analyzed a narrow size bin from the total particle size distribution. For each hygroscopicity mode, there were multiple particle types contributing significant number fractions, suggesting that even within the same hygroscopicity mode there were still some heterogeneity in particle composition. Particles in the same hygroscopicity mode may share some common features in compositions but their differences are distinguishable in single particle data. In another respect, the contributions of each type also suggest the existence of a predominant type that accounts for major fractions in respective modes compared with other types, such as Aged EC in MH mode, Ammonium/OC in LH mode. The comparison between Table 2 and Figure 2 suggests that, although their absolute contributions may be different, the hygroscopicity patterns of particle types in the two datasets are in good agreement. Based on this fact, we concluded that the composition-hygroscopicity connections contained in HTDMA-ATOFMS dataset was successfully reflected into the predicted GF."

2. Line 207: add the following references for ATOFMS detection of biomass burning aerosol: [Pratt et al., 2010; Zauscher et al., 2013].

Response:

This suggestion is accepted.

Changes in manuscript:

Line 241-243:

"The Biomass particles produced characteristic peaks of -26(CN), -42(CNO), -59($C_2H_3O_2$), -73($C_3H_5O_2$) and dominant peak at 39K and related peaks at 113(K_2Cl) or 213(K_3SO_4) (Silva et al., 1999;Zauscher et al., 2013;Pratt and Prather, 2009).

3. Lines 214-215 describing EC particles require a reference to prior work [Ault et al., 2010; Spencer et al., 2006; Toner et al., 2008].

Response:

This suggestion is accepted.

Changes in manuscript:

Line 250-251:

"The EC particles were detected by a series of elemental carbon peaks at C_n (n=1, 2, 3 …) in the

negative and positive spectra (Ault et al., 2010;Spencer et al., 2006;Toner et al., 2008). "

4. Line 217: define NH and MH.

Response:

The NH and MH were defined in the text.

Changes in manuscript:

Line 251-253:

"EC particles distributed broadly from nearly-hydrophobic (NH) mode to more-hygroscopic (MH) mode."

5. Lines 223-225: I suggest adding Figure S2 to the main text.

Response:

The peak intensity trends in Figure S2 was presented only for EC particles. To embody the referee's opinion, we transmitted the information in Figure S2 to a new figure to show the peak intensities of EC, Dust/ash and all particles, as seen in Figure 4 in the main text.

Changes in manuscript:

Line 903-906:

"
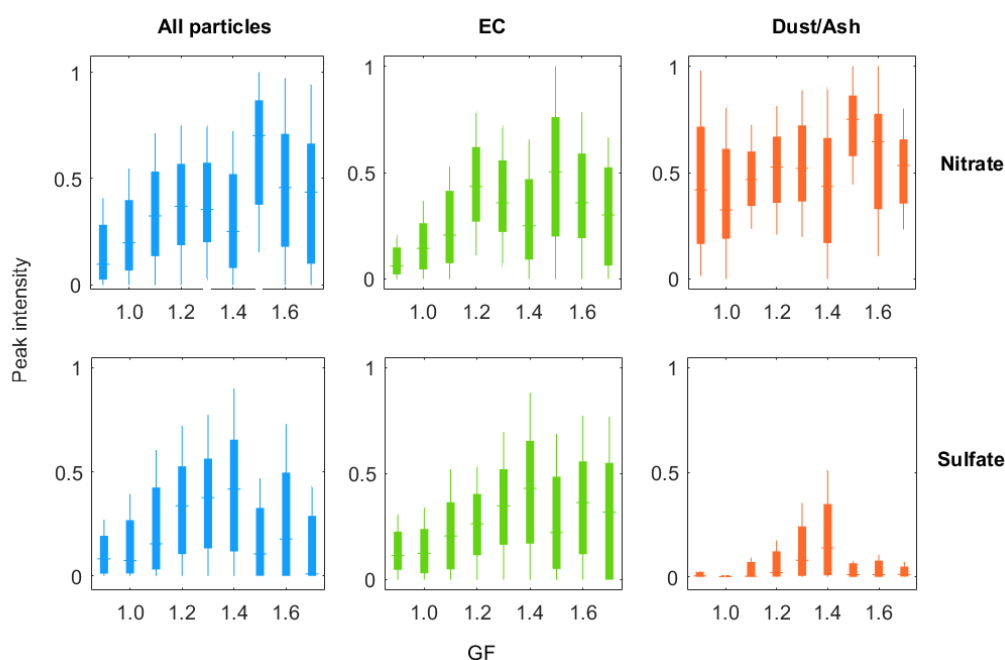
Figure 4. Statistics of nitrate and sulfate peak intensities (minimum, 25th percentile, median, 75th percentile, maximum) with GF in HTDMA-ATOFMS experiment. The intensity statistics were calculated for All particles, EC particles and Dust/Ash particles separately."

6. Lines 226-227: please add [Ault et al., 2011; Gaston et al., 2017; Sullivan et al., 2007].

Response:

This suggestion is accepted.

Changes in manuscript:

Line 264-265:

"Dust/ash type particles produced inorganic peaks of salts and metals (Gaston et al., 2017;Ault

et al., 2011;Sullivan et al., 2007a)."

7. I suggest also adding Figure S3 to the main text.

Response:

We have moved Figure S3 to the main text with relevant discussions.

Changes in manuscript:

Line 900-901:

 "
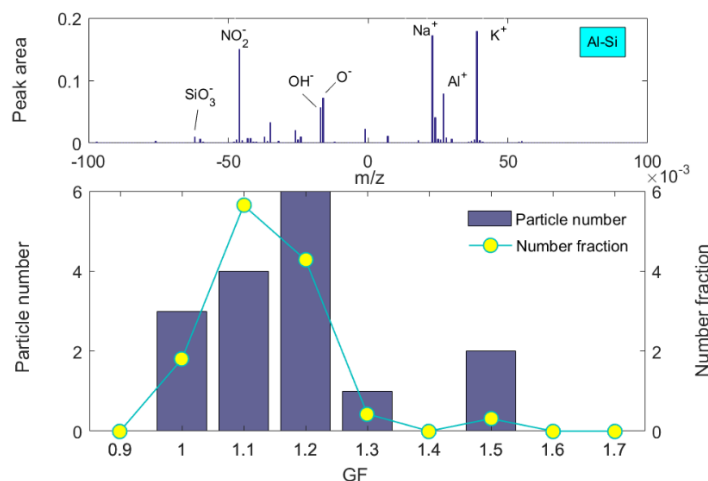
Figure 3. Average mass spectra and hygroscopicity distribution of Al-Si particles. "

8. Lines 255-257: there are other ATOFMS studies showing ammonium/OC particles associated with agricultural emissions [Qin et al., 2012]. The authors should also consider that source for their observations.

Response:

We followed the suggestions of the reviewer and made following changes.

Changes in manuscript:

Line 305-312:

"Prior ATOFMS study attribute similar Ammonium/OC particles to agricultural sources and found they were enriched in higher photochemical oxidation periods (Qin et al., 2012). The secondary nature of Ammonium/OC is also consistent with the pronounced ammonium peak. It is likely that the organics in Ammonium/OC particles are also generated from secondary processes since the GF 1.2 approximate to the hygroscopicity of SOA (GF=1.24 at 90% RH) (Gysel et al., 2007;Sjogren et al., 2008). We inferred that ammonium was not contributing major fractions to Ammonium/OC particles, since ammonium salts was very hydrophilic while Ammonium/OC demonstrate only moderate hygroscopicity."

9. Lines 260-261: aging does not always translate to high hygroscopicity.

Response:

We admit that original statement is not accurate, because aging does not necessarily promote hygroscopicity of all particles. We have revised it accordingly.

Changes in manuscript:

Line 310-312:

 "We inferred that ammonium was not contributing major fractions to Ammonium/OC particles,

since ammonium salts was very hydrophilic while Ammonium/OC demonstrate only moderate hygroscopicity.

10. Line 275: please also cite [Pratt and Prather, 2009]

Response:

This suggestion is accepted.

Changes in manuscript:

Line 323-324:

"This phenomenon was also observed for other organic particles (Shi et al., 2012;Pratt and Prather, 2009)."

11. Line 282-283: please site [Gaston et al., 2011]

Response:

This suggestion is accepted.

Changes in manuscript:

Line 332-333:

"Sea salt mass spectra contain dominant sodium peak +23Na and other sodium cluster peaks at +62Na$_2$O, +63Na$_2$OH, +81Na$_2$Cl (Gaston et al., 2017)."

12. The authors mention that only a few sea salt particles were observed, yet this was the only particle type where a thorough investigation linking the particle composition to the GF was performed. Why was this type of analysis or discussion not included for other particle types?

Response:

We analyzed sea salt particles in more detail mainly because they constitute a separated mode without interferences from other types and clearer trends was observed in sea salt. However, this treatment seems insufficient by neglecting other particles. To embody the referee's concern, in the revision we also include analysis of particles for Ammonium/OC particles in LH mode. In discussions of peak intensities, we also included discussions on Dust/ash and EC particles which made major contributions to MH mode.

Changes in manuscript:

Line 561-583:

"The nonlinearity of peak intensities with GF was also suggested by the different particle types presented in ATOFMS data. As shown in the lower panels in Figure 11, the distribution of ATOFMS individual particles showed enrichment in different areas in the GF-peak intensity diagram, suggesting the presence of particle groups of different compositions. To illustrate this character, we selected two areas with clear particle enrichments in GF-peak intensity diagram and their particle composition were analyzed (denoted as A and B in lower left panel in Figure 11). Obviously, particles in area A produced much larger nitrate signals than particles in area B. Particle numbers in the two areas suggested that Dust/ash dominated particles in area A (59%) while in area B the Dust/ash only accounted for 14% of particles (Figure S10). As a contrast, particles in area B were dominated by Aged EC type (53%) followed by 25% Dust/ash. Table 2 suggests that Aged EC and Dust/ash are the major types presented in the same MH mode. However, peak intensity responses to GF were indeed different for different

particle types, suggesting the importance of particle types in describing peak intensities.

The particle distribution with sulfate intensities showed similar enrichment patterns to nitrate (lower right panel in Figure 11). Sulfate peak intensities were found to correlated with hygroscopicity in GF <1.2 range but in MH range no correlated with GF was observed. We note that except the larger peaks of nitrate and sulfate, some smaller peaks were also found to correlated with GF within specific particle type. We correlated peak intensities of Na_2Cl^+ and NO_3^- with the estimated GF of sea salt particles (Figure S9). The Na_2Cl^+ peaks were positively correlated with GF while the nitrate peaks were negatively correlated with GF. The observed correlation in sea salt particles are consistent with discussed trends in the HTDMA-ATOFMS dataset. These results demonstrate that the GF estimation method have effectively reflected the minor changes in particle mass spectra into the estimated hygroscopicity."

13. Lines 301-302: please cite [Gaston et al., 2011; Prather et al., 2013]

Response:

This suggestion is accepted.

Changes in manuscript:

Line 375-377:

"The unreacted sea salt particles tend to produce larger peaks of Na_2Cl^+ and $NaCl_2^-$ in spectra (Gaston et al., 2011;Prather et al., 2013)."

14. Lines 306-307: please cite [Gaston et al., 2018] who found a similar result using ATOFMS data.

Response:

This suggestion is accepted.

Changes in manuscript:

Line 381-382:

"Therefore, the HTDMA-ATOFMS data supported that reacted sea salt have reduced hygroscopicity (Herich et al., 2009;Gaston et al., 2018)."

15. I had a hard time following lines 305-310, the authors need to clarify whether aged sodium nitrate contributed to the lower GF or whether other factors were responsible.

Response:

The hygroscopicity of sea salt particles was not clearly presented originally. According to HTDMA GF characterization of pure salts, the salt $NaNO_3$ (GF ~1.8) is less hygroscopic than NaCl (GF ~2.2) at 85% RH (D_{dry} 250nm) (Hu et al., 2010). The transformation of NaCl into $NaNO_3$ could reduce particle hygroscopicity, but the transformation alone could not account for the observed hygroscopicity since the observed GF of sea salt (1.5-1.7) were lower than pure $NaNO_3$ (supposing fully transformed). Therefore, we hypothesized that other factors including organics were contributing to the further reduction of sea salt hygroscopicity.

Changes in manuscript:

Line 371-390:

"The analysis of peak intensities with GF can disclose some atmospheric processes happened on aerosol. We take the Sea salt as an illustration. Sea salt particles were known to react with atmospheric nitric acid, with NaCl in fresh sea salt be transformed into $NaNO_3$ in the reacted

sea salt (Gard et al., 1998). This composition transformation is indicated in corresponding changes of NaCl and NaNO3 peak intensities in particle spectra. The unreacted sea salt particles tend to produce larger peaks of Na_2Cl^+ and $NaCl_2^-$ in spectra (Gaston et al., 2011;Prather et al., 2013). In particle spectra of reacted sea salt, the NaCl peaks (Na_2Cl^+, $NaCl_2^-$) decrease while NaNO3 peaks ($Na_2NO_3^+$, $Na(NO_3)_2^-$) increase. We presented peak intensities of sea salt in GF 1.5-1.7 range where sea salt particles were detected with largest numbers (Figure 5). We found that the positive correlation between NaCl peak intensity and GF, and the negative correlation for NaNO3 peaks. Therefore, the HTDMA-ATOFMS data supported that reacted sea salt have reduced hygroscopicity (Herich et al., 2009;Gaston et al., 2018). Laboratory HTDMA study suggested that NaCl and $NaNO_3$ have deliquesced at 85% RH and that the $NaNO_3$ (GF ~1.8) is less hygroscopic than NaCl (GF ~2.2) (Hu et al., 2010). The reduced hygroscopicity of sea salt is in line with the GF of sodium salts. However, the sea salt hygroscopicity (GF 1.5-1.7) was smaller than pure $NaNO_3$ salt (supposing fully reacted), suggested that the chemical transformation alone is not sufficient to account for the observed hygroscopicity of sea salt. We hypothesize that other compositions as organics were mixed into sea salt and contributed to the reduction of sea salt hygroscopicity (Gaston et al., 2011;Randles et al., 2004;Facchini et al., 2008). "

16. Section 3.1.2 seems out of place. While it is an interesting finding that the effective density showed a trend with the GF, the authors need to explain this finding a bit more.

Response:

In the original manuscript the effective density was included for the purpose of a complete presenting of the measured data, since aerodynamic diameters were measured by ATOFMS. The discussions of effective density seem out of place in view of the main objective of this study is to connect hygroscopicity to composition. We accept the referee's opinion since the hygroscopicity are depending on particle composition, rather than particle density. The presented data only suggested a statistical positive correlation between effective density and GF, but a meaningful explanation of this correlation need further information including particle physical shapes, which also affect ρ_{eff} . On the other hand, effective densities were not well incorporated into the discussions on estimated hygroscopicity. Therefore, in revised manuscript we decided to concentrate on particle composition and the effective density were not discussed.

Changes in manuscript:

The discussions were removed.

17. Section 3.2.1 needs some statistics to show that the authors can predict the GF from the spectra alone.

Response:

We accepted the referee's suggestion to include some particle statistics to the ambient particles. In Table 2 we have presented such information on the contributions of particle types to different hygroscopicity modes. The calculate contributions were based on ATOFMS numbers of ambient particles and offer better statistics than HTDMA-ATOFMS particles. The relevant information on HTDMA-ATOFMS particle contributions in original version were removed.

Changes in manuscript:

Line 887-890:

"Table 2. Statistics on particle number contributions of ATOFMS particle types to different GF modes. The statistics are the average contributions and variation ranges (in brackets) based on temporal data in daily resolution.

Contribution (%)	*NH (GF <1.1)*	*LH (GF 1.1-1.3)*	*MH (GF 1.3-1.5)*	*SS (GF >1.5)*
Fresh EC	*14 (7-17)*	*2 (1-4)*	*0 (0-3)*	*0 (0-1)*
Cooking	*3 (0-7)*	*1 (0-3)*	*0 (0-0)*	*0 (0-0)*
Biomass	*18 (7-35)*	*9 (2-17)*	*0 (0-1)*	*0 (0-0)*
HMOC	*40 (30-68)*	*8 (2-15)*	*0 (0-1)*	*0 (0-0)*
Ammonium/OC	*11 (3-21)*	*32 (20-45)*	*2 (1-5)*	*0 (0-0)*
Aged EC	*2 (1-5)*	*12 (5-20)*	*47 (15-72)*	*13 (1-42)*
Dust/ash	*3 (1-6)*	*13 (9-22)*	*27 (13-53)*	*26 (4-44)*
Amine-rich	*3 (1-5)*	*12 (2-41)*	*13 (4-39)*	*11 (1-44)*
Sea salt	*0 (0-1)*	*0 (0-1)*	*1 (0-4)*	*40 (7-78)*

"

18. Lines 441-442: this suggests that the authors should try comparing their GF data to peak area intensities of sulfate and nitrate.

Response:

We have added discussions on the correlations between peak intensity and estimated GF of ambient particles.

Changes in manuscript:

Line 548-583:

"3.3.3 Peak intensity variations with estimated GF

Particle hygroscopicity and peak intensities in particle mass spectra were correlated to show their connections. The correlation was illustrated similarly to the analysis of the HTDMA-ATOFMS dataset, as shown in Figure 11. In addition to the statistics on peak intensities of different GF, the number distributions ATOFMS particles with GF and peak intensities were presented for nitrate (-46NO$_2$, -62NO$_3$, -125H(NO$_3$)$_2$) and sulfate peaks (-97HSO$_4$, -80SO$_3$) in the lower panels in Figure 11. The general trends of peak intensities with GF in HTDMA-ATOFMS dataset was preserved in estimated GF of ambient particles. The trends of nitrate and sulfate peak intensities showed increases from NH to LH range and remained constant in the MH mode. Similar to HTDMA-ATOFMS particles, stronger nitrate peaks were detected in SS particles compared with the MH particles, while an opposite trend was observed for sulfate intensities. These results highlight the nonlinearity between GF and peak intensities of ATOFMS particles.

The nonlinearity of peak intensities with GF was also suggested by the different particle types presented in ATOFMS data. As shown in the lower panels in Figure 11, the distribution of ATOFMS individual particles showed enrichment in different areas in the GF-peak intensity diagram, suggesting the presence of particle groups of different compositions. To illustrate this character, we selected two areas with clear particle enrichments in GF-peak intensity diagram and their particle composition were analyzed (denoted as A and B in lower left panel in Figure 11). Obviously, particles in area A produced much stronger nitrate signals than

particles in area B. Particle numbers in the two areas suggested that Dust/ash dominated particles in area A (59%) while in area B the Dust/ash only accounted for 14% of particles (Figure S10). As a contrast, particles in area B were dominated by Aged EC type (53%) followed by 25% Dust/ash. Table 2 suggests that Aged EC and Dust/ash are the major types presented in the same MH mode. However, peak intensity responses to GF were indeed different for different particle types, suggesting the importance of particle types in describing peak intensities.

The particle distribution with sulfate intensities showed similar enrichment patterns to nitrate (lower right panel in Figure 11). Sulfate peak intensities were found to correlated with hygroscopicity in GF <1.2 range but in MH range no correlated with GF was observed. We note that except the larger peaks of nitrate and sulfate, some smaller peaks were also found to correlated with GF within specific particle type. We correlated peak intensities of Na_2Cl^+ and NO_3^- with the estimated GF of sea salt particles (Figure S9). The Na_2Cl^+ peaks were positively correlated with GF while the nitrate peaks were negatively correlated with GF. The observed correlation in sea salt particles are consistent with discussed trends in the HTDMA-ATOFMS dataset. These results demonstrate that the GF estimation method have effectively reflected the minor changes in particle mass spectra into the estimated hygroscopicity."

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