# Responses to Reviewer #1

*This is a good paper that should be published after many minor revisions. Most corrections are typical of Asian writing. Section 4 is difficult to follow and is not well supported. A figure showing the bimodal GF is in order. However, this section could be removed. Several definitions are needed. Define refractory, non-refractory, MDL, OA, PM1, downweighting, standard density, rush hours, epsilon (in Fig. 9), and SNR. Citation needed for PMF.*

A: We thank the reviewer for encouraging and helpful comments on our manuscript, and such detailed editing of our English. We believe that the quality of our manuscript is improved as we reflect the reviewer's comments. Below each of the questions/comments is written first with the Italic font and then our response is followed with the normal font.

**Comments**

*Q1: Section 4 is difficult to follow and is not well supported. A figure showing the bimodal GF is in order. However, this section could be removed.*

A: Section 4 contains crucial results that describe the relationship between the mixing state of aerosols and aerosol chemical composition based on the HTDMA – HR-ToF-AMS dataset. Therefore, we think that it would be better to add or clarify some content to make it easier to follow than to remove all of this section. In this effort we added two figures, one table and their explanations. Below is the list of figures and tables we added.

- Figure 10: Schematic plot of three aerosol Types
  (Previous Figures 10 and 11 → Figures 11 and 12)
- Figure S6: Average diurnal variation of hygroscopic growth factor distribution for (a) 30 nm, (b) 50 nm, (c) 100 nm, and (d) 150 nm dry diameters.
- Table S2: Frequency and number of data (parentheses) for each aerosol Type for four different dry diameters (30, 50, 100, and 150 nm) during the measurement period.

Q2: *Several definitions are needed. Define refractory, non-refractory, MDL, OA, PM1, downweighting, standard density, rush hours, epsilon (in Fig. 9), and SNR.*

A: As the reviewer suggested, we added the definition or full name of refractory/non-refractory (Line 148-149), MDL(Line 164), OA (Line 170), PM1 (Line 148-149), downweighting (Line 168-170 and add reference), rush hours (Line 353), epsilon in Fig.9 (Line 395) and SNR (Line 165). For standard density, we adopted the term from Decarlo et al. (2004), indicating the unity density as a reference.

- Reference for standard density

Peter F. DeCarlo , Jay G. Slowik , Douglas R. Worsnop , Paul Davidovits & Jose L. Jimenez: Particle Morphology and Density Characterization by Combined Mobility and Aerodynamic

Diameter Measurements. Part 1: Theory, Aerosol Science and Technology, 38:12, 1185-1205, DOI: 10.1080/027868290903907, 2004.

- Reference for downweighting

Paatero, P. and Hopke, P. K.: Discarding or downweighting high-noise variables in factor analytic models, Anal.Chim.Acta, 490 (1-2),277-89, 2003.

*Q3: Citation needed for PMF.*

A: Citations for the PMF method and pre-processing method used in this study were added.

References

Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, Atmos. Chem. Phys., 9, 2891–2918, https://doi.org/10.5194/acp-9-2891-2009, 2009.

Zhang, Q., Alfarra, M. R., Worsnop, D. R., Allan, J. D., Coe, H., Canagaratna, M. R., and Jimenez, J. L.: Deconvolution and quantification of hydrocarbon-like and oxygenated organic aerosols based on aerosol mass spectrometry, Environ. Sci. Technol., 39, 4938–4952, 2005.

*Q4: L11. Correspondences singular. Were to was. Delete 1st the.*

A: Changed as suggested (Line 11)

*Q5: L13. Change infer to imply.*

A: Changed as suggested (Line 13)

*Q6: L15. Delete the.*

A: Changed as suggested (Line 15)

*Q7: L18. Delete the.*

A: Changed as suggested (Line 18)

*Q8: L26. Delete from its surrounding.*

A: Changed as suggested (Line 26)

*Q9: L27. Particle plural. Delete the.*

A: Changed as suggested (Line 27)

*Q10: L30. Rogers. Delete to be.*

A: Changed as suggested (Line 30)

*Q11: L32. Add s to influence. Delete on.*

A: Changed as suggested (Line 32)

*Q12: L33. Change it to hygroscopicity.*

A: Changed as suggested (Line 33)

*Q13: L35. Delete problem. Change under a to with.*

A: Changed as suggested (Line 34-35)

*Q14: L36. Add ity to humid. Delete condition.*

A: Changed as suggested (Line 35)

*Q15: L40. Add Kim et al. 2011. Move the after of.*

A: Changed as suggested (Line 40) and added Kim et al., (2011)

Kim, J. H., Yum, S. S., Shim, S., Yoon, S.-C., Hudson, J. G., Park, J., and Lee, S.-J.: On aerosol hygroscopicity, cloud condensation nuclei (CCN) spectra and critical supersaturation measured at two remote islands of Korea between 2006 and 2009, Atmos. Chem. Phys., 11, 12627–12645, https://doi.org/10.5194/acp-11-12627-2011, 2011.

*Q16: L42. Insert the after of. Delete the.*

A: Changed as suggested (Line 42-43)

*Q17: L43. Measurement plural.*

A: Changed as suggested (Line 43)

*Q18: L44. Change mixture to mixing.*

A: Changed as suggested (Line 44)

*Q19: L45. Delete would.*

A: Changed as suggested (Line 45)

*Q20: L46. Change can to could.*

A: Changed as suggested (Line 46)

*Q21: L47. Measurement plural. Insert a after for.*

A: Changed as suggested (Line 47) but ‘a’ is omitted because this conflicts with the Q22.

*Q22: L48. Mixture plural.*

A: Changed as suggested (Line 48)

*Q23: L51. Delete values.*

A: Changed as suggested (Line 51)

*Q24: L54. Delete their. Locations singular.*
A: Changed as suggested (Line 54)

*Q25: L61. Delete the.*

A: Changed as suggested (Line 61)

*Q26: L63. Delete of aerosols.*

A: Changed as suggested (Line 63)

*Q27: L65. Estimates.*

A: Changed as suggested (Line 65)

*Q28: L67. Various.*

A: Changed as suggested (Line 67)

*Q29: L73. Delete 1st the.*

A: Changed as suggested (Line 72)

*Q30: L75. Delete 2nd the. Area plural.*

A: Changed as suggested (Line 75)

*Q31: L81. Change in to to.*

A: Changed as suggested (Line 80)

*Q32: L82. Delete last the.*

A: Changed as suggested (Line 82)

*Q33: L82-3. Move SMA in front of air.*

A: Changed as suggested (Line 82-83)

*Q34: L83. Delete of.*

A: Changed as suggested (Line 83)

*Q35: L89. Delete from a global perspective.*

A: Changed as suggested (Line 88)

*Q36: L91. Delete the. Delete territory. Korea.*

A: Changed as suggested (Line 90)

*Q37: L94. Delete last the.*

A: Changed as suggested (Line 93)

*Q38: L98. Delete 2nd the.*

A: Changed as suggested (Line 97)

*Q39: L99. Insert a after by. Insert a after and.*

A: Changed as suggested (Line 98)

*Q40: L110. Measurement plural.*

A: Changed as suggested (Line 109)

*Q41: L113. Delete the. Move road (singular) in front of traffic (singular). Delete on the.*

A: Changed as suggested (Line 112)

*Q42: L116. Change such to this. Condition plural.*

A: Changed as suggested (Line 115)

# Responses to Reviewer #2

*This paper presents aerosol chemical and hygrosopic properties from a field measurement campaign in an urban setting. The authors do a good job of presenting the data, and the analysis and conclusions are scientifically sound and relevant. I recommend that the paper be published after the minor points listed below are addressed.*

A: We thank the reviewer for encouraging and helpful comments on our manuscript. We believe that the quality of our manuscript is improved as we reflect the reviewer's comments. Below each of the questions/comments is written first with the Italic font and then our response is followed with the normal font.

*Q1: Measurements were made of non-refractory aerosol components as well as black carbon. While these likely account for the majority of sub-micron aerosol, dust and sea salt could also be present, and are not mentioned in the paper. What effect, if any, could these aerosols have on the results, especially the k-closure analysis. For sea salt, while it is likely not a major contributor to sub-micron aerosol, because of it's very high kappa even a small amount could influence the measurements. This should be discussed in the paper.*

A: This study covers the hygroscopic properties of submicron ($PM_1$) aerosols. There are two reasons why we excluded mineral dust and sea salt aerosols and included only non-refractory aerosol components and BC in our description of the chemical composition of submicron aerosols. First, as the reviewer mentioned, these components were likely to account for the majority of submicron aerosols. It is well known that even if $PM_{2.5}$ inlet system is used for BC measurement, generally BC mass is dominantly determined by submicron particles (e.g., Clarke et al., 2004; Wu et al., 2013). In addition, it was reported that sea salt aerosols measured in Seoul occupied less than 3% of $PM_{2.5}$ aerosols from a 24 hour period air sample (Heo et al., 2009). Second, the κ-closure results without mineral dust and sea salt showed very good agreement with measurement (Fig. 3), perhaps inferring that mineral dust and sea salt aerosols had little effect on the κ-closure analysis in this study. However, we agree that it would be informative to add some sentences on mineral dust and sea salt aerosols as the reviewer suggested, and therefore they were added in the revised manuscript (Line 176-182).

"In this study, other refractory and semi-refractory aerosols like mineral dust and sea salt aerosols that have their own hygroscopic properties were not considered as they were likely to account for little portion of submicron aerosols. For example, sea salt aerosol occupied less than 3% among $PM_{2.5}$ aerosols from a 24 hour period air sample collected in Seoul (Heo et al., 2009). The very good κ-closure results in Fig. 3, which did not consider mineral dust and sea salt, perhaps infers that mineral dust and sea salt aerosols had little effect on the κ-closure analysis."

Reference for answer

Clarke, A.D., Shinozuka, Y., Kapustin, V.N., Howell, S., Huebert, B., Doherty, S., Anderson, T., Covert, D., Anderson, J., Hua, X., Moore, K.G., McNaughton, C., Carmichael, G., Weber, R., 2004. Size distributions and mixtures of dust and black carbon aerosol in Asian outflow:

physiochemistry and optical properties. J. Geophys. Res. 109 (D15) http://dx.doi.org/10.1029/2003JD004378. D15S09.

Wu, Z.J., Poulain, L., Henning, S., Dieckmann, K., Birmili, W., Merkel, M., van Pinxteren, D., Spindler, G., Müller, K., Stratmann, F., Herrmann, H., Wiedensohler, A., 2013. Relating particle hygroscopicity and CCN activity to chemical composition during the HCCT-2010 field campaign. Atmos. Chem. Phys. 13, 7983e7996. http://dx.doi.org/10.5194/acp-13-7983-2013.

*Q2: Measurements were made with an HTDMA at sub-saturated conditions (RH = 85%). However, previous measurements (Petters et al., 2009, Wex et al., 2009) have shown that organic aerosol does not always behave ideally and exhibits different hygroscopic properties at sub and supersaturated conditions. This is especially true for lower RH (<90%) as used in this study. This should be mentioned in the paper, at least as a caveat, and discussion added about how the limitation of the measurements might impact the conclusions.*

A: We thank the reviewer for this legitimate comment. In the revised manuscript, we noted the difference of organic aerosol hygroscopicity for sub- and supersaturation conditions and the limitation that may arise due to the measurement condition, citing the two references the reviewer suggested (Line 561-568).

"To note is that organic aerosols do not always behave ideally and show an apparent discrepancy in hygroscopic growth between sub- and supersaturated conditions (Petters et al., 2009; Wex et al., 2009). If hygroscopic growth were measured under a supersaturated condition, the estimated hygroscopicity parameter would be significantly higher than those estimated in this study under sub-saturated condition, due to the contribution of enhanced hygroscopic growth of organic components of aerosols. This would surely affect the CCN prediction results but it is uncertain how much that would be at this point. Perhaps, however, the overestimating tendency of $\kappa_{chem}$ shown in Fig. 4 may be reduced as the measured $\kappa$ would become higher."

References:

Petters, M. D., Wex, H., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol – Part 2: Theoretical approaches, Atmos. Chem. Phys., 9, 3999–4009, 2009, http://www.atmoschem-phys.net/9/3999/2009/.

Wex, H., Petters, M. D., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards Closing the Gap between Hygroscopic Growth and Activation for Secondary Organic Aerosol: Part I – Evidence from Measurements, Atmos. Chem. Phys. Discuss., 9, 3987–3997, 2009, http://www.atmos-chemphys.net/9/3987/2009/acp-9-3987-2009.html.

*Q3; More information is needed in section 4.4 about aerosol mixing state. Specifically, how were single vs multi-modal distributions determined? Was this just subjective* *classification or was a curve fitting routine used? Also, how often, and when, were Types 1, 2 and 3 observed? Trends in the diurnal pattern are discussed in the paper but not shown. A figure showing diurnal variability would be helpful. Also, how often were more than two modes observed? The paper only says that this occurred "occasionally". Finally, this section would benefit from more editing for clarity as it was hard to follow the analysis done here.*

A: Following the reviewer's comments, Section 4.4 was supplemented with two figures, one table, and more detailed explanations in the revised manuscript. Each of the reviewer's questions were answered below:

1) We used a *peakfit* function MATLAB® to determine the single vs. multi-modal distribution. The description about this was added in Line 407 – 410 in the revised manuscript:

"For determination of mixing state, the position, height and width of each peak for HTDMA data are computed by *peakfit* function for MATLAB® that performs a least-square curve fit of a Gaussian function to the top part of the peak (O'Haver, 2016)."

2) We added a table (Table S2 in the revised manuscript) to present the frequency of occurrence of each aerosol Type for four dry diameters during the measurement period. The explanation on this was added in Line 420 – 423 in the revised manuscript:

"During the measurement period, Type 1 (externally mixed) aerosols were predominantly observed (higher than 70%) in large particles (100 and 150 nm) whereas Type 3 (internally mixed with LH mode) aerosols occupied more than 50% of all aerosols in small particles (30 and 50 nm) (Table S2)."

3) The diurnal variability of hygroscopic growth factor distribution for four dry diameters was shown in Fig. S6 in the revised manuscript. This figure was reprinted from Kim et al. (2018). Additionally, we added more explanations about the diurnal pattern of hygroscopic growth in Line 423 – 429 in the revised manuscript.

"Also found was that mixing state had a distinct diurnal pattern, as depicted in Fig. S6. Briefly, for small particles, Type 3 aerosols prevailed all day, except in the afternoon (12:00 – 18:00 LT), when a significant portion of the aerosols turned into Type 2. For large particles, externally mixed aerosols (Type 1) dominated, especially during the rush hour (07:00 – 09:00 LT) when hydrophobic particles emitted from traffics mix with preexisting large and aged particles. In the afternoon, mixing state change occurred in both small and large particles due to the photochemical processes. At night, however, no such change occurred as there was no photochemical process."

4) More than two modes (trimodal or higher modal distribution) were observed in less than 3 percent of all measurement cases. A short description was added in Line 416, "less than 3% of total measurement cases" in the revised manuscript.

5) We added a figure in the revised manuscript (Figure 10. Schematic plot of three aerosol Types) to make it easier to understand how the aerosol types were classified in this study.