Response to anonymous referee #2

General comments:

The paper "Aerosol hygroscopicity derived from size-segregated chemical composition and its parameterization in the North China Plain" by Liu et al present comprehensive study of k-parameter describing aerosol hygroscopic properties in k-Köhler theory. The paper is well structured and written in a condense manner. Maybe too condense and instead of describing what has been done and interpreting the results, authors just refer to earlier papers. It does not bring any novelty with respect to understanding the aerosol hygroscopic and cloud forming potential, but its value is in good quality analysis of size-segregated aerosol chemical composition and comprehensive and robust analysis of calculated k-values in combination with observed hygroscopic properties using HH-TDMA. It lacks on the interpretation, analysis of temporal variability and links to meteorology and variability in sources and air mass transport. This part can be certainly improved.

Response: Thanks for the comments. Our work evaluates the hygroscopicity for particles ranging from 30nm to 8μm and develops a parameterization scheme using foil sampling data to calculate particle hygroscopicity. Our further work is to dry to get a temporal variability of aerosol hygroscopicity from former foil samples based on this parameterization scheme. According to the referee's suggestions, we've added new paragraphs about the influence of meteorological conditions on aerosol hygroscopic properties and revised our manuscript.

Specific comments:

Comment:

All values given are average values for whole campaign, but analysis of temporal variability through the campaign and how it is controlled by meteorology and air mass origin (source influence) is missing. Besides deriving one general value for summer conditions, data can be certainly explored more. The range of bars in Fig.4 show great deal of variability. To what degree this is driven by uncertainty in chemical analysis and k-value calculations and by changing aerosol properties and air mass origin? Are high k-values in most abundant mode around 400 nm corresponding to high k-values for sub-100 nm particles or not? How the intermodal variability of k-values reflect changes in meteorology, aerosol sources and aerosol aging?

Response:

Thanks for the comments. We agree with the referee that besides the analysis of average values, the analysis of temporal variability should be added into the manuscript. Through analyzing the time series of κ and meteorology parameters, we found that aerosol hygroscopicity is related to wind directions. When southerly wind dominates, the aerosol hygroscopicity is fairly high. When northerly wind dominates, the aerosol hygroscopicity is relatively low. As the campaign site locates at the north edge of the North China Plain, wind direction shifting from southerly to northerly represents the aerosol origin shifts from the highly industrialized central North China

Plain to the scarce populated mountainous area. Therefore, we grouped all data into two patterns based on the wind directions and explained the relations between particle hygroscopicity and wind directions from the view of particle origin. We've added a new section (sect. 4.1) to our manuscript accordingly.

The range of bars in Fig. 4 (Fig. 5 in the latest manuscript) shows great variability, indicating particle hygroscopicity of different samples varies largely. It's mainly driven by the changes of aerosol source but not the uncertainties in chemical analysis and calculations. We've mentioned in Sect. 2.2 of our manuscript that the uncertainties of chemical components such as ions and WSOC are 5%. By the help of Monte Carlo simulation, we evaluated that the uncertainty of κ is less than 10%, which is a small value compared with the large range of bars.

The particles around 400nm and the sub-100nm particles originate from different sources. The sub-100nm particles originate from nucleation and condensation of low-volatile organic compounds and from primary emission of biomass combustion, while the particles around 400nm grow from smaller particles by coagulation and aging process. We've classified all data into two patterns. Both particles around 400nm and the sub-100nm particles behave more hygroscopic in the Southerly Pattern. And the discrepancy of hygroscopicity for particles around 400nm between two patterns is more significant than that for sub-100nm particles.

Comment:

On page 20901, L 1-11 authors present brief explanation of the observed average diurnal cycle. What about if the aged aerosol aloft transported into a growing mixed layer is more hygroscopic? It has its origin likely in previous day mixed layer and thus aged for at least a day longer than freshly emitted aerosol. On one hand photochemistry enhanced availability of condensing vapors, on the other hand the diurnal temperature cycle will likely change nitrate partitioning between aerosol and gas phase. I believe that presenting more thorough discussion on diurnal variability of aerosol composition will make the paper better.

Response:

Thanks for the referee's comments and we agree with the referee. The aged aerosols trapped aloft during nighttime are more hygroscopic. In the daytime, when the boundary layer develops, the aged aerosols are entrained downward and mixed with the newly emitted aerosols. This boundary layer mixing process results in higher hygroscopicity of particles near the ground. Besides, particle aging process can also enhance particle hygroscopicity and photochemical reactions can accelerate aging process. At night, the boundary layer collapses. We observed the enhanced black carbon emission during the nighttime (Ma et al., 2011), making particles less hygroscopic. The shallow boundary layer further amplifies the impact of black carbon on particles.

From Fig. 14 in Morino et al. (2006), we know that temperature and relative humidity influence the nitrate partitioning between NO_3^- and HNO_3. The nitrate partitioning shifts to gas state in higher temperature and lower RH conditions. Nevertheless, Morino et al. (2006) indicates that the nitrate (NO_3^- + HNO_3) shows "distinct diurnal

variations with a maximum of 12ppbv at around noon". In the daytime, ambient temperature is higher and RH is lower than at night, so the nitrate partitioning shifts to gas state. But the concentration of total nitrate is much higher than that at night. So the concentration of is controlled by both ambient conditions and concentration of total nitrate. We can't tell how the change of nitrate partitioning between NO_3^- and HNO_3 would influence particle hygroscopicity due to the lack of total nitrate measurement in our study.

Following the referee's suggestion, we have presented more thorough descriptions about the diurnal evolution of the planetary boundary layer and revised Sect. 4.4 of our manuscript.

Reference:

Ma, N., Zhao, C. S., Nowak, A., Müller, T., Pfeifer, S., Cheng, Y. F., Deng, Z. Z., Liu, P. F., Xu, W. Y., Ran, L., Yan, P., Göbel, T., Hallbauer, E., Mildenberger, K., Henning, S., Yu, J., Chen, L. L., Zhou, X. J., Stratmann, F., and Wiedensohler, A.: Aerosol optical properties in the North China Plain during HaChi campaign: an in-situ optical closure study, Atmos. Chem. Phys., 11, 5959-5973, 10.5194/acp-11-5959-2011, 2011.

Morino, Y., Kondo, Y., Takegawa, N., Miyazaki, Y., Kita, K., Komazaki, Y., Fukuda, M., Miyakawa, T., Moteki, N., and Worsnop, D. R.: Partitioning of HNO3 and particulate nitrate over Tokyo: Effect of vertical mixing, J. Geophys. Res.-Atmos., 111, 2006.

Comment:

The paper deserves publication in ACP. In addition to robust k-values calculation, additional effort on interpretation of the results with respect to other environmental parameters will increase the quality of the paper.

Response:

Thanks for the referee's support. According to the referee's comments, we've discussed the relationship between particle hygroscopicity and wind direction, which can be regarded as an indicator of synoptic patterns. We've also given more thorough interpretation about the diurnal variation of particle hygroscopicity. The referee's valuable suggestions and comments help us improve the quality of the paper.