Dear Editor,

We would like to thank the two referees for their comments and suggestions. Point-by-point responses to all the comments and a revised manuscript were uploaded.

Best Regards

Chunsheng Zhao

Responses to comments of referee #2

General comments:

Although overall their revision is somewhat satisfactory, there are still several places that further improvements are necessary.

Still, I have doubt on their "assumption that the aerosol hygroscopicity parameter is constant during a specific humidifying cycle", since such a cycle under atmospheric conditions often is accompanied by composition changes of aerosols (see, for example, Ma et al., GRL, 40, 6293, 2013).

Also, the authors need to define "a specific humidifying cycle".

Furthermore, their usage of English can be further improved. For example, the last sentence "This shows the proposed κ retrieval algorithm with the *f*(RH) measurements is reasonable and robust". A conjunction needs to be added here.

Also, the use of pronoun should always be minimized.

Response: Thanks for the comments. Point-by-point responses are as follows:

1. Still, I have doubt on their "assumption that the aerosol hygroscopicity parameter is constant during a specific humidifying cycle", since such a cycle under atmospheric conditions often is accompanied by composition changes of aerosols (see, for example, Ma et al., GRL, 40, 6293, 2013).

Response: As demonstrated by most previous studies, the aerosol hygroscopicity parameter κ for subsaturated conditions does differ from that for supersaturated conditions, and the differences between the two kinds of κ results vary significantly with different aerosol sources, chemical composition changes and particle morphology transformation (Dusek et al., 2011; Kristensen, T. B., et al., 2012; Whitehead et al., 2014). However, to the *f*(RH) measurements at subsaturated

conditions of 30-90% RH in this work, the solution that we assumed a constant κ for a complete 2.5-hour humidifying cycle should be suitable to the situations of no sudden and severe air mass changes during the observation periods. This is generally in accordance with the treatment method proposed by Ervens et al. (2007).

In the study of Ma et al. (2013), they investigated the aging process of generated pure soot aerosols in a fluoropolymer environmental chamber in the laboratory. This is greatly different from our experiments conducted in the ambient environment. The measured ambient aerosols during our field campaign are of complex chemical compositions, and they have experienced different extent of aging processes in most cases. Under the condition of no significant air mass changes, the aerosol hygroscopicity during a 2.5-hour *f*(RH) measurement can be considered to be constant. In addition, Ma et al. (2012) have pointed out that approximately 90% of the total light scattering is contributed by the aerosols in the accumulation mode within the size range of 300nm − 1μm. The measured *f*(RH) is supposed to be more sensitive to the composition changes of aerosols in the accumulation mode, rather than to that of smaller particles. Therefore, the coating of freshly emitted soot aerosols (with the corresponding particle sizes much smaller than 300 nm) might be of minor influence on the overall hygroscopicity variation of the aerosol population in the ambient atmosphere.

Overall, the assumption that the aerosol hygroscopicity parameter κ is constant during a complete 2.5-hour humidifying cycle is confirmed to be applicable for our κ retrieval algorithm. Nevertheless, uncertainties inevitably lie in the assumption and consequently exist in the calculated κ results. We have discussed about the possible influence of this assumption on the uncertainties of the derived κ in the corresponding places.

References:

[1]. Dusek U., G. P. Frank, A. Massling, K. Zeromskiene, Y. Iinuma, O. Schmid, G. Helas, T. Hennig, A. Wiedensohler, and M. O. Andreae.: Water uptake by biomass

burning aerosol at sub- and supersaturated conditions: closure studies and implications for the role of organics. *Atmos. Chem. Phys.*, 11, 9519–9532, doi:10.5194/acp-11-9519-2011, 2011.

[2]. Ervens, B., M. Cubison, E. Andrews, G. Feingold, J. A. Ogren, J. L. Jimenez, P. DeCarlo, and A. Nenes.: Prediction of cloud condensation nucleus number concentration using measurements of aerosol size distributions and composition and light scattering enhancement due to humidity, *J. Geophys. Res.*, 112, D10S32, doi:10.1029/2006JD007426, 2007.

[3]. Kristensen, T. B., et al. (2012), Hygroscopic growth and CCN activity of HULIS from different environments, *J. Geophys. Res.*, 117, D22203, doi:10.1029/2012JD018249.

[4]. Ma, N., Zhao, C. S., Müller, T., Cheng, Y. F., Liu, P. F., Deng, Z. Z., Xu, W. Y., Ran, L., Nekat, B., van Pinxteren, D., Gnauk, T., Müller, K., Herrmann, H., Yan, P., Zhou, X. J., and Wiedensohler, A.: A new method to determine the mixing state of light absorbing carbonaceous using the measured aerosol optical properties and number size distributions, *Atmos. Chem. Phys.*, 12, 2381-2397, doi:10.5194/acp-12-2381-2012, 2012.

[5]. Whitehead, J. D., M. Irwin, J. D. Allan, N. Good, and G. McFiggans.: A meta-analysis of particle water uptake reconciliation studies. *Atmos. Chem. Phys. Discuss.*, 14, 9783–9800, doi:10.5194/acpd-14-9783-2014, 2014.

2. Also, the authors need to define "a specific humidifying cycle".

Response: Thanks. We have rephrased it into "a complete 2.5-hour humidifying cycle".

3. Furthermore, their usage of English can be further improved. For example, the last

sentence "This shows the proposed κ retrieval algorithm with the *f*(RH) measurements is reasonable and robust". A conjunction needs to be added here.

Also, the use of pronoun should always be minimized.

Response: We appreciate the referee's helpful comments. As suggested, the last sentence is replaced with "This shows **that** the proposed κ retrieval algorithm with the *f*(RH) measurements is reasonable and robust". We have checked the whole manuscript thoroughly and made corresponding corrections where necessary.