

Response to anonymous referee #2

General comment: If the focus of the manuscript, as stated in the title, is the presentation of a novel method they need to pay more attention to the explanation of the method itself and the validation of the method using additional data (ideally from different measurement sites) and quantify the uncertainties of using the look-up-table to retrieve aerosol hygroscopicity. Otherwise, the authors are just presenting relationships between variables but not actually a new (usable) method. There are many redundancies in the paper that should be omitted as well as typos and grammar spelling errors. Split sentences into two or more individual sentences to improve readability.

Response: Thanks for your comment. We agree with the reviewer. In the revised manuscript, the look up table is produced based on PNSD and BC measurements from four different field campaigns. Meanwhile, datasets about PNSD and BC during Wangdu campaign when measurements from the humidified nephelometer system are available are not used for simulating the look up table. In addition, datasets from a different field campaign which is conducted at another site of the NCP in autumn is also used to validate the proposed method. That is, the produced look up table is verified with measurements from two different sites in different seasons. Please refer to Fig.7 and Fig.8 of the revised manuscript for more details. The results demonstrate that the look up table is applicable in different sites and seasons. As to uncertainties of R_k under different Angstrom exponent and κ_{sca} conditions, the standard deviations of R_k within each grid of the look up table are shown in Fig.6b.

Specific comments

Comment: Line21: avoid redundancies like “newly proposed novel approach”

Response: Thanks for your suggestion. We revised the manuscript accordingly.

Comment: Line22: Replace by “ . . . is that $\kappa_{f(RH)}$ can be estimated without any additional information...”

Response: Thanks for your suggestion. We revised the manuscript accordingly.

Comment: Line34: “. . .most important factors affecting these . . .” Introduction: there are too much methodological information in the introduction, that should be moved to the methodology section.

Response: Thanks for your suggestion. This sentence reflects the significance of aerosol

hygroscopicity, and this is the motivation of this research.

Comment: Line104: “similar to”

Response: Thanks for your suggestion. We revised the manuscript accordingly.

Comment: Line107: “based on”

Response: Thanks for your comment. We revised the manuscript accordingly.

Comment: Section2: This section is very bad organized. Include a table with information about the campaigns (dates, sites, data used here from each campaign, etc). What is the time resolution of the PM2.5 filter samples? 24 hours? How often is the sampling performed?

Response: Thanks for your suggestion. We revised this section according to your suggestions.

Comment: More information on HH-TDMA measurements and inversion routine should be presented. Same applies for the nephelometers tandem. Include information on nephelometers correction and calibration, humidogram schedule, RH range in the dry neph, where were the RH sensors located in the system?, how often were the sensors calibrated?

Response: Thanks for your suggestion. We added information about the nephelometer system in Section 2 of the revised manuscript. The instrument set-up of HH-TDMA and inversion routine of κ from measurements of HH-TDMA are introduced in detail in Liu et al. (2011).

Comment: Line120: replace dot by comma

Response: Thanks for your suggestion. We have revised the manuscript.

Comment: Section 3.1: Further details on the methods used to derive the κ parameter should be given even though the methods were published before. At least the basic information to allow the reader to understand the manuscript. Concerning the $\kappa_{f(RH)}$ method, which chemical species have been considered apart of BC? A table including the chemical species, refractive indices, densities and contribution during the measurement period must be included.

Response: Thanks for your comment. The flow chart of retrieving $\kappa_{f(RH)}$ is provided in the supporting information. A simplified aerosol model was applied to aerosol optical calculations. In the model, aerosol components are divided into two classes in terms of their optical properties: the light absorbing component (BC) and less absorbing components (comprising inorganic salts and acids such as sulfates, nitrates, ammoniums, as well as most of the organic compounds). We have added this statement in Section 3.1 of the revised manuscript.

Comment: In the Mie routine, is the chemistry considered as constant during the campaign? See my previous comment on PM2.5 sampling schedule.

Response: Thanks for your comment. In this paper, a simplified aerosol model was applied to aerosol optical calculations. In the model, aerosol components are divided into two classes in terms of their optical properties: the light absorbing component (BC) and less absorbing components (comprising inorganic salts and acids such as sulfates, nitrates, ammoniums, as well as most of the organic compounds). We have added this statement in Section 3.1 of the revised manuscript.

Comment: Section 3.2: The reference of Quinn et al. (2005) is not appropriate here. The gamma parameterization was first introduced in Kasten (1969) and Hanel (1980). Kasten, F., 1969. Visibility forecast in the phase of pre-condensation. *Tellus* 21 (5), 631-635 Hanel, G., 1980. Technical Note: an attempt to interpret the humidity dependencies of the aerosol extinction and scattering coefficients. *Atmos. Environ.* 15, 403-406.

Response: Thanks for your comment. We have revised the reference accordingly.

Comment: Line 194: avoid redundancy, this sentence “more details . . .” could be omitted.

Response: Thanks for your comment. We have revised the manuscript accordingly.

Comment: Results: Line 207-221: This paragraph could be omitted since basically is a repetition of the results presented in Kuang et al., (2016) and does not provide any additional/useful information.

Response: Thanks for your comment. We have deleted these sentences.

Comment: Line 207: information about nephelometer correction should be moved to the instrument section

Response: Thanks for your suggestion. We revised the manuscript accordingly.

Comment: Lines 212, 216 and somewhere else: “a lot” is not very scientific, be more quantitative and avoid colloquial expressions.

Response: Thanks for your comment. We have revised the manuscript accordingly.

Comment: Line 257: This paragraph should be rewritten. What is the aim of including these two additional campaigns?

Response: Thanks for your comment. The aim of including PNSD and BC information from different campaigns is to simulate variations of R_k under different conditions. We have added this sentence in the revised manuscript.

Comment: Line 297: “The fitting performance . . . values” could be omitted. Again, avoid redundancy.

Response:

Comment: Line 300: The γ -Method and κ -Method are just different ways of fitting the experimental $f(\text{RH})$ -RH relationship. Which method is better or worst depends on your specific data, and many other equations have been previously proposed in the literature (Titos et al.,2016). The discussion in lines 300-306 and figure 4 about which fitting is best do not add much and could be omitted.

Response: Thanks for your comment. We have deleted these sentences.

Comment: Line 316: “pretty good linear relationship” does not sound very quantitative neither scientific Try to be more specific . . .

Response: Thanks for your comment. We have revised the manuscript accordingly.

Comment: Line 333: This is the first time that κ_{chem} and κ_{ext} are introduced.

Response: Thanks for your comment. We revised the manuscript.

Comment: Line 347 and somewhere else: Avoid repetitions like “which is introduced in Section

...”

Response: Thanks for your comment. We have revised the manuscript accordingly.

Comment: Line 359-360: “and then it turns out”, “much more complex”... this is not very appropriate for a scientific paper...

Response: Thanks for your comment. This sentences is revised as the following: “A robust linear relationship is found between $\kappa_{f(RH)}$ and κ_{sca} in Sect.4.2 , however, results of further analysis suggest that R_k varies a lot”

Comment: References of Titos et al., 2016 and Zieger et al., 2014 are not used appropriately here. The Angstrom exponent was first introduced by Angstrom!

Response: Thanks for your comment. We have deleted the reference.

Comment: Line 377: Keep in mind that the Angstrom exponent is not a measure of the PNSD, it provides information on mean predominant aerosol size so values close to 2 denote a predominance of fine particles while values below 1 denote a predominance of coarse particles.

Response: Thanks for your comment. We have revised the sentence as the following: “Based on results shown on Fig.6a, the different impacts of aerosol hygroscopicity and dry scattering Ångström exponent on R_k can be distinguished to some extent”.

Comment: Line 393 and Figure 7: This comparison exercise is interesting but it is not appropriately done. The predicted R_k values using the look-up-table are compared with the measured R_k values. However, these measured R_k were used before to generate the look-up-table. Thus, it is clear that a high correlation is expected. A different dataset, with additional R_k values not used to generate the look-up-table should be used for validation of the proposed model. Otherwise, the same data that is used to generate the model is used to validate it, which is meaningless.

Response: Thanks for your comment. In the revised manuscript. The dataset about PNSDs and mass concentrations of BC when measurements from the humidified nephelometer system are available are not used in the processes of producing the look up table shown in Fig.6a. Thus, the look up table is

independent of measurements during periods when $f(\text{RH})$ measurements are available. In addition, $f(\text{RH})$ measurements from another campaign is also used to verify the manuscript.

Comment: If the authors really expect researchers to use their method, they should provide them with an uncertainty range for R_k as a function of the Angstrom exponent and κ_{sca} . Probably, higher errors are expected at higher κ_{sca} values? This is certainly needed if they expect people to use the look-up-table. In general, the manuscript lacks of an appropriate treatment of errors despite the large expected errors for the hygroscopicity parameters.

Response: Thanks for your comment. We agree with the referee. The uncertainty range of R_k based on the simulative results is shown in Fig.6b. The results is consistent with the referee's point that higher errors are expected at higher κ_{sca} values. The maximum κ_{sca} of the look up table is 0.4, if R_k is 0.8 (close to the simulated highest R_k shown in Fig.5b), the corresponding $f(80\%)$ is 2.6. According to the review of Titos et al. (2016), most of $f(80\%)$ for continental aerosols are lower than 2.6. This look up table already covers most situations for continental aerosol types.

Liu, P. F., Zhao, C. S., Göbel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W. Y., Deng, Z. Z., Ma, N., Mildenberger, K., Henning, S., Stratmann, F., and Wiedensohler, A.: Hygroscopic properties of aerosol particles at high relative humidity and their diurnal variations in the North China Plain, *Atmos. Chem. Phys.*, 11, 3479-3494, 10.5194/acp-11-3479-2011, 2011.

Titos, G., Cazorla, A., Zieger, P., Andrews, E., Lyamani, H., Granados-Muñoz, M. J., Olmo, F. J., and Alados-Arboledas, L.: Effect of hygroscopic growth on the aerosol light-scattering coefficient: A review of measurements, techniques and error sources, *Atmospheric Environment*, 141, 494-507, <http://dx.doi.org/10.1016/j.atmosenv.2016.07.021>, 2016.