

Response to anonymous referee #3

Main comments: The authors present 2 methods for calculating the κ aerosol hygroscopicity parameter: from 1) Mie calculations that use the aerosol dry size distribution, BC mass concentration and the aerosol scattering hygroscopic growth (fRH) and 2) aerosol size-dependent hygroscopic growth, gRH. The authors use an empirical relationship between the fRH fit parameter, the scattering Angstrom exponent and the ratio fit values from fRH : method 1 kappa fit to create a look up table to predict kappa values.

The paper needs revision to better organize the paper, clarify the different hygroscopic fit calculations as well as discuss differences between the three kappa values.

In general: Break down run-on sentences into two or more sentences. Remove irrelevant information and words. Try not to repeat information. Reduce use of expressions such as “although, therefore, however, widely, especially and traditionally” as these terms usually don’t carry meaning and make the paper more difficult to read. Try to use precise words rather than generalities.

Response: Thanks for your comment. We have revised the manuscript according to your suggestions.

Specific comments

Comment: Lines 38-42: rewrite as “...,water usually constitutes half of the aerosol mass at a relative humidity of 80% with substantially higher water mass fractions existing at RH values above 90% for most ambient aerosol (Bian et al.,2014). The water content of aerosol and cloud droplets depends on both the ambient RH and hygroscopicity of the aerosol chemical constituents.”

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

Comment: rewrite as “ In order to account for the mixed organic and inorganic composition of ambient aerosol Petters and Kriedweiss (2007) proposed a modified version of Kohler theory called κ -Kohler theory to describe a single aerosol hygroscopic growth parameter, κ . The κ -Kohler equation, expressed in terms of the diameter growth factor, g(RH), is given in equation 1 below.”

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

Comment: Equation 1: Please change “S” to RH/100. “S” is associated with droplet activation and

may confuse readers. Remove “g” from the equation as it doesn’t belong

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

Comment: Line 58: remove sentence “In recent ten years, this....” as it states the obvious and doesn’t add to the paper.

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

Comment: Line 71: rewrite as “ The Humidified Tandem Differential Mobility Analyzer (HTDMA) measures the aerosol diameter hygroscopic growth as a function of RH.

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

Comment: Page 3: Remove reference to CCN measurements as it adds confusion and detracts from the discussion of diameter hygroscopic growth.

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

Comment: Line 84: Remove the Brock et al. reference as he uses a cavity ring down spectrometer and not a nephelometer.

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

Comment: Line 87-89: Rewrite as “The scattering enhancement factor $f(RH)$, defined as $f(RH) = \sigma_{sp}(RH, \lambda) / \sigma_{sp}(dry, \lambda)$, characterizes changes in the aerosol scattering coefficient with RH.

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

Comment: Line 92: Break into 2 sentences and rewrite as “Thus, κ calculated from $f(RH)$ measurements represents an optically weighted aerosol hygroscopic growth.

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

Comment: Line 95-99: Don’t start a new paragraph. Rewrite as “Traditionally, derivation of κ from $f(RH)$ measurements requires aerosol PNSD as well as black carbon (BC) measurements to determine the imaginary part of the refractive index. As PNSD and BC measurements are expensive their

availability in field campaigns are limited.

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

Comment: Clearly identify the 3 methods you use to determine κ by identifying them as Method 1, Method 2 and Method 3. Line 99: Start a new paragraph. “In this paper we use measurements from to derive κ values using 3 methods. The first 2 methods derive κ from aerosol diameter hygroscopic growth and the third method derives an aerosol optical parameterization of κ . Method 1, labeled as $\kappa_{f(RH)}$, derives κ from aerosol PNSD, BC and nephelometer $f(RH)$ measurements. Method 2, defined as κ_{250} , derives κ from aerosol diameter hygroscopic growth measurements, $g(RH)$, using a High-Humidity Differential Mobility Analyzer (HH-TDMA). Method 3, defined as κ_{sca} , is an empirical determination of κ using only nephelometer measurements of the aerosol scattering coefficient as a function of RH”.

Start a new paragraph to describe how you combine κ values from Methods 1 and 3 to devise a method to predict size-related κ values using only $f(RH)$ scattering measurements. You need to clearly identify and separate these 3 methods in the paper. Using the terms “Method 1, Method 2 and Method 3” or something similar will clarify and simplify much of the paper discussion.

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

Comment: You need to describe the nephelometer $f(RH)$ measurements. What was the RH range and did the instruments operated in parallel or in series? Describe the position of the RH sensors, the type of RH sensor and its uncertainty. How was the RH inside the nephelometer determined? Did the humidifier scan the hydration or dehydration branch of the aerosol RH growth? What range of RH values were used in calculating the fits?

Response: Thanks for your comment. We have added these information in the Section 2 of the revised manuscript.

Comment: Why are the HTDMA measurements done at such a high RH? At RH values $>90\%$, most RH sensors have an uncertainty $\pm 3\%$ or more. At high RH values the aerosol growth curve is particularly steep such that even a small error in RH would lead to a very high error in $g(RH)$. What

not measure $g(RH)$ at a lower RH such as 80-85%? What is the uncertainty in $g(RH)$ at 98%?

Response: Thanks for your comment. The basic principle of HH-TDMA is similar to that of HTDMA, however, its special feature is capable of operating stably under extremely high RH conditions (Hennig et al., 2005). The reason that this system operates at RH of 98% is the scientific focus of this instrument during this field campaign is hygroscopic properties of aerosol particles under extremely high RH conditions. Details about the uncertainties of RH and $g(RH)$ please refer to Hennig et al. (2005).

Comment: Line 172: Remove the first two sentences of section 3.2. Accurate measurement of $f(RH)$ depends on the uncertainty in aerosol scattering and RH. The empirical relationship of scattering to RH isn't difficult to measure or describe. What's difficult is modeling the size-dependent chemical composition of the aerosol, not the measurement itself. Page 7, line 187: Remove the sentence "Here, we give ...". As you haven't described curvature effects and these effects aren't apparent for equation 3, you should remove the sentence.

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

Comment: Line 189-198: Simplify the wording.

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

Comment: One assumption of the $f(RH)$ kappa parameterization is that $f_{RH=0}=1$ at $RH=0$. However, $f(RH)$ values are near constant for $RH < 40\%$, meaning $f(RH)=1$ at $RH \sim 40$. What this means is that the fits can't be forced through 1 at $RH=0$. The actual equation should be $f(RH) = b + \kappa(RH/100-RH)$. Equation 3 doesn't account for aerosol losses in the humidifier and nephelometer. These losses won't affect the gamma fit parameter in equation 2 (provided losses are a percent of the scattering), but will affect determination of κ in equation 3. For example a 10% aerosol loss will change κ by 10%, but multiplying equation 2 by this same 10% loss correction or 1.1 won't change gamma.

Response: Thanks for your comment. We agree with the referee that the humidified nephelometer system have the problem of aerosol losses in the humidifier. During Gucheng campaign (introduced in the revised manuscript), the control software of this system will let the humidifier do not humidify the sample air every two days and the period last about two hours. The purpose of doing so is to check the consistency of two nephelometers (Dry nephelometer and Wet Nephelometer). The results are shown

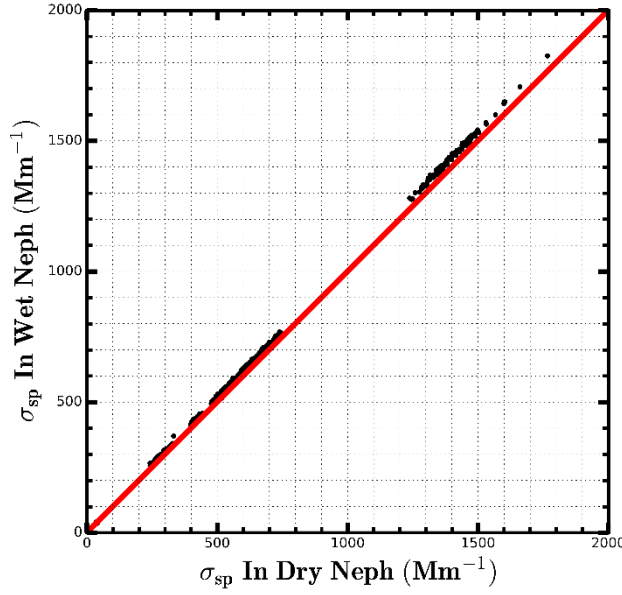


Figure 1. x-axis represents σ_{sp} at 525 nm measured by the dry nephelometer, y-axis represents σ_{sp} at 525 nm measured by the wet nephelometer, the red line is 1:1 line.

in Fig.1. The results demonstrate that σ_{sp} measured by the wet nephelometer is slightly higher than that measured by the dry nephelometer, the average relative difference is 3%. The reason that the higher σ_{sp} measured by the wet nephelometer might be attributed to the difference of RH in the dry nephelometer (about 8%) and wet nephelometer (about 15%). In addition, the relative difference between them is within the measurement uncertainty of nephelometer (Müller et al., 2011). This result indicates that aerosol losses in the humidifier have negligible influence on the σ_{sp} .

During processes of measuring $f(RH)$, the sample RH in the dry nephelometer (RH_0) is not zero. We have modified the fitting formula of measured $f(RH)$. According to equation (3) of the manuscript, the measured $f(RH)_{measure} = \frac{f(RH)}{f(RH_0)}$ should be fitted using the following formula:

$$f(RH)_{measure} = (1 + \kappa_{sca} \frac{RH}{100 - RH}) / (1 + \kappa_{sca} \frac{RH_0}{100 - RH_0}) \quad (4)$$

And in the revised manuscript, this equation is used for calculating κ_{sca} .

Comment: The gamma and kappa fit parameters are sensitive to the RH range of the fit. What range of RH was used in the fits? Note that RH values <40% and >90% will increase error in the fit as the growth curves don't conform to equations 2 or 3 in these RH regions.

Response: Thanks for your comment. We have added the information about RH range used in the retrieval algorithm in the revised manuscript. About 50% to 90% for cycles without deliquescence,

about 70% to 90% for cycles with deliquescence.

Comment: Line 225-226: rewrite: During deliquescence $f(RH)$ exhibits an abrupt increase between RH values of 60-65%. As such, only $f(RH)$ data points with $RH > 70\%$ were used in determination of κ when deliquescence was apparent.

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

Comment: Lines 237-240. Be more specific and describe the hygroscopic growth behavior during polluted times more quantitatively. What range of σ_{sp} values were categorized as polluted? Figure 1 shows that σ_{sp} was above 100 Mm^{-1} most of the measurement period. Can you show a plot of $f(RH)$ vs σ_{sp} ? Can you account for changes in $f(RH)$ with aerosol loading? How does aerosol size and absorption change with loading?

Response: Thanks for your comment. Periods with $\sigma_{sp} > 100 Mm^{-1}$ are categorized as polluted.

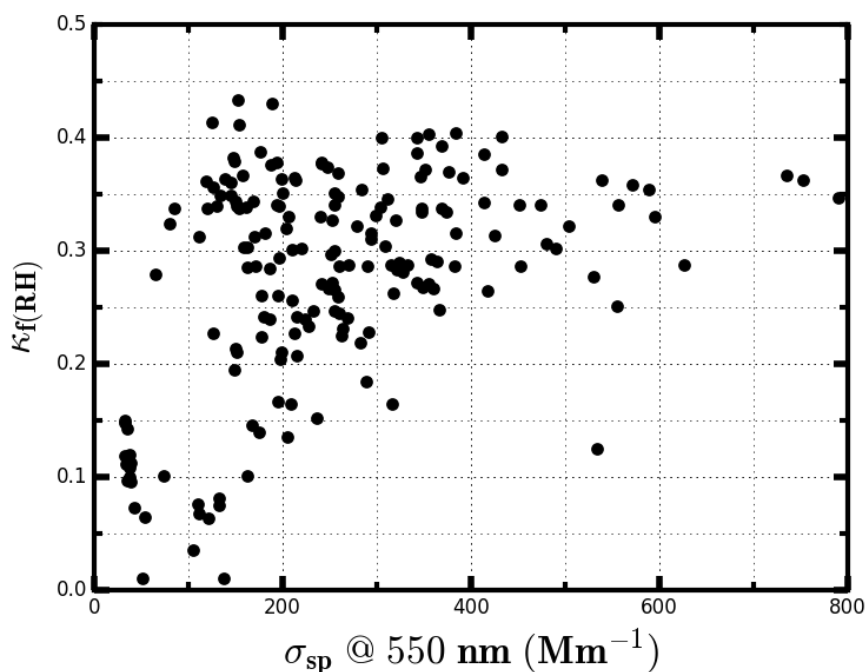


Figure 2. x-axis represents σ_{sp} @ 550 nm, y-axis represents retrieved $\kappa_f(RH)$.

During Wangdu campaign, the plot of $\kappa_f(RH)$ vs σ_{sp} is shown in Fig.2. The aerosol size and absorption changes with aerosol loading is a good scientific topic. Studies of aerosol size changes with aerosol loading can be found in previous studies, such as Shen et al. (2015). The absorption change with aerosol loading is highly variable in polluted region due to complicated emissions and aging

processes. Issues about aerosol size and absorption changes with loading are beyond the scope of this paper.

Comment: Line 271: Is $\kappa_{f(RH)}$ optically weighted or is it a size-dependent κ that is integrated over the entire size distribution? Method 1 varies the size-dependent κ until the Mie calculations equal the scattering $f(RH)$. Change “optically weighted” to “size-integrated”.

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

Comment: Page 10, Lines 266-285: Can you simplify the wording to make this paragraph easier to understand. It would help to distinguish the model κ values from Model 1 and Model 2 if you labeled it κ_{chem}

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

Comment: Page 11, Line 299: A comparison of the kappa and gamma fits to the measured value at a single RH of 85% isn't a good indication of the goodness of fit. Note in Figure 4a that both the gamma and kappa fits are higher than the measured value at 85%. A better indication of the goodness of fit would be a chi-square fit value or the sum of the square of standard deviation of the measured values from the fit line or variance. I suggest replacing Figure 4b with a plot of the probability distribution of the fit variances.

Response: Thanks for your comment. We agree with the referee. According to the suggestions of another referee, we have deleted this part.

Comment: Lines 313-321: This paragraph is unclear. The co-variance of $f(RH)$ fit parameters with OMF, SMF and NMF varies with aerosol type; e.g. source and oxidation state or aging. The fit quality depends on the measurement duration as well as the variability in the aerosol type. The chemical information can give an indication of the aerosol hygroscopic growth in the absence of scattering $f(RH)$ measurements. Gamma and kappa fit values in your comparison are both derived from nephelometer scattering measurements, so they should compare well. Remove the discussion of past comparisons of kappa with aerosol chemical composition

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

Comment: Line 338: The ratio R_k depends on the aerosol size-integrated scattering efficiency. This ratio will vary substantially with the aerosol type and size distribution. The variability of R_k of this study may not coincide with that of the Brock et al. paper as different aerosol types were sampled under very different conditions. Rewrite sentence as “... the ratio $\kappa_{sca}/\kappa_{f(RH)}$ may share a similar range of variability.”

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

Comment: Page 13, line 379: replace “PNSD at dry state” with “dry scattering Angstrom exponent”
Line 380: remove “nevertheless, aerosol hygroscopicity has non-negligible impacts”. The results indicate a strong size-dependence to the hygroscopic growth. Size and hygroscopicity are not separable, nor does one parameter dominate variation in R_k . Aerosol size would determine or dominate R_k only if the aerosol chemical composition had no size variation. As sulfate tends to be more prevalent in smaller sizes then R_k will be larger at higher Angstrom values.

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

Comment: Lines 378-384: Rewrite this section in terms of variation in the size-dependent chemical composition.

Response: Thanks for your comment. We agree with the referee that the size-dependent chemical composition also exerts influence on R_k . If PNSD is fixed, each size-resolved κ distribution corresponds to a certain $\kappa_{f(RH)}$, and $\kappa_{f(RH)}$ varies within certain range no matter how size-resolved κ distribution changes. Therefore, influences of size-dependent chemical compositions are already included in simulated results of producing the look up table by varying the $\kappa_{f(RH)}$ from 0 to 0.7 for a fixed aerosol PNSD. This discussion is added in the revised manuscript.

Comment: Lines 407-413: Note that the look up table only applies to aerosol on the NCP during the summer and can't be applied to other sites. Aerosol size distributions, secondary processing, and size-dependent composition vary widely with season and region. This method can be used as a tool for

other sites, however it requires measurements of nephelometer scattering, aerosol BC and particle number size distributions.

Response: Thanks for your comment. We agree with the referee that aerosol size distributions, secondary processing, and size-dependent composition vary widely with season and region. However, in the simulating processes of producing the look up table, no information about size dependent chemical composition is involved. The look up shown in Fig.6a of the manuscript is produced from measurements of four field campaigns (datasets from a new campaign are added) which were conducted at different seasons and sites. The small variation of R_k under different Angstrom exponent and κ_{sca} conditions shown in Fig.6b demonstrate good consistency exists between R_k produced from PNSD and BC measurements of these campaigns. In addition, in the revised manuscript, we have verified this method with datasets obtained from two sites of the NCP 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.

Hennig, T., Massling, A., Brechtel, F. J., and Wiedensohler, A.: A tandem DMA for highly temperature-stabilized hygroscopic particle growth measurements between 90% and 98% relative humidity, *Journal of Aerosol Science*, 36, 1210-1223, 10.1016/j.jaerosci.2005.01.005, 2005.

Müller, T., Laborde, M., Kassell, G., and Wiedensohler, A.: Design and performance of a three-wavelength LED-based total scatter and backscatter integrating nephelometer, *Atmos. Meas. Tech.*, 4, 1291-1303, 10.5194/amt-4-1291-2011, 2011.

Shen, X. J., Sun, J. Y., Zhang, X. Y., Zhang, Y. M., Zhang, L., Che, H. C., Ma, Q. L., Yu, X. M., Yue, Y., and Zhang, Y. W.: Characterization of submicron aerosols and effect on visibility during a severe haze-fog episode in Yangtze River Delta, China, *Atmospheric Environment*, 120, 307-316, <http://doi.org/10.1016/j.atmosenv.2015.09.011>, 2015.