Review: " A novel method to derive the aerosol hygroscopicity parameter based only on measurements from a humidified nephelometer system"
Authors: Y. Kuang et al.

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.

Comments by section

Introduction

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."

Line 45: rewrite as " In order to account for the mixed organic and inorganic composition of ambient aerosol Petters and Kriedweiss (207) 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."

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.

Line 58: remove sentence "In recent ten years, this…." as it states the obvious and doesn't add to the paper.

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

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

Line 84: Remove the Brock et al. reference as he uses a cavity ring down spectrometer and not a nephelometer.
Line 87-89: Rewrite as "The scattering enhancement factor *f(RH)*, defined as *f(RH) = σsp(RH, λ)/ = σsp (dry, λ),* characterizes changes in the aerosol scattering coefficient with RH.

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

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.

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 κ*fRH,* derives κ from aerosol PNSD, BC and nephelometer *f(RH)* measurements. Method 2, defined as κ250, derives κ from aerosol diameter hygroscopic growth measurements, *g(RH),* using a High-Humidity Differential Mobility Analyzer (HH-TDMA). Method 3, defined as κscat, is an empirical determination of κ using only nepehelometer 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.

Page 5:
You need to describe the nephelometer *fRH* 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?

Page 6: Methodology

Why are the HTDMA measurements done at such a high RH? At RH values >90%, most RH sensors have an uncertainty +/- 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 gRH. What not measure gRH at a lower RH such as 80-85%? What is the uncertainty in gRH at 98% ?

Line 172: Remove the first two sentences of section 3.2. Accurate measurement of *fRH* 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 sentance.
Line 189-198: Simplify the wording.

One assumption of the *fRH* kappa parameterization is that *fRH*=1 at RH=0. However, *fRH* values are near constant for RH <40%, meaning *fRH*=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 *fRH* = b + κ(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.

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.

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.

Lines 237-240. Be more specific and describe the hygroscopic growth behavior during polluted times more quantitatively. What range of σsp values were categorized as polluted? Figure 1 shows that σsp was above 100 Mm-1 most of the measurement period. Can you show a plot of frh vs σsp? Can you account for changes in *fRH* with aerosol loading? How does aerosol size and absorption change with loading?

Line 271: Is κ_{fRH} 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 *fRH.* Change"optically weighted" to "size-integrated".

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 1and Model 2 if you labeled it κ*chem.*

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.

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.

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 κ-*scat/* κ*f(RH)* may share a similar range of variability."

Page 13, line 379: replace "PNSD at dry state" with "dry scattering Angstrom exponent"
Line 380: remove "nevertheless, aerosol hygroscopicty has non-negligible impacts". The results indicate a strong size-dependence to the hygroscopic growth. Size and hygroscopicty 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.

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

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.