

Reply to Referee #1

This paper reports on the use of what the authors claim is a simplified experimental system based on two nephelometers to determine the hygroscopicity parameter (κ) of ambient PM1 organic aerosol (κ_{OA}) for RH in the range of 85-95%. The system is essentially two relatively low price nephelometers that are used to measure aerosol mass concentration, both measuring ambient air one at close to ambient conditions the other is dried. The results are contrasted for different aerosol sources and include comparison to the AMS-measured O/C ratio. The nephelometers also report mass concentration and so difference in the wet and dry neph reported particle mass concentrations are interpreted as equal to the liquid water concentration due to the differences in the RH of the two nephs. There are some limitations noted by the authors, such as differences in particle size ranges when comparing masses from the dry neph to the AMS, that the AMS is not a comprehensive measurement of even PM1 mass, and uncertainty in the calibration of the nephs for converting scattering to mass. Furthermore, the sampling is done within an (I assume) airconditioned trailer which will result in biases when trying to determine actual ambient particle water concentrations, although that is not the goal of this study. For someone who has not read the first Zang et al paper on the pDRs, what these instruments actually are is not clear. Maybe a photo in the Supp, or a small description of what they are typically used for and stating the cost (\$10k) early in the manuscript, not just in the Conclusions, would help to explain why this is claimed to be a simple method early in reading the paper.

One major issue lacking in this paper is a discussion comparing the specific method used here to the $f(RH)$ method to infer particle water. Both use a wet and dry neph. The $f(RH)$ method has a substantial history, yet is never noted in this work (see description in Guo et al and a list of references therein; www.atmos-chem-phys.net/15/5211/2015/)

Overall, the paper is of interest and suitable for publication in ACP but there are unclear sections in this paper that need to be addressed.

R: We thank the reviewer for the detailed, helpful, and overall supportive comments. We have revised the manuscript to account for each comment. Responses to the individual comments are provided below. Below is our point-by-point response to each comment. Author responses are in Bold black. Modifications to the manuscript are in our normal font. Line numbers in the response correspond to those in the revised manuscript text file (tracked version).

For this major concern, we add more discussion to compare this specific method to the previous studies, as “A combination of dry and wet nephelometers has been used to estimate (1) aerosol liquid water content (ALW) (Guo et al., 2015; Kuang et al., 2018) and hygroscopicity (Kuang et al., 2017), replying on the measured aerosol light scattering enhancement factor (f_{RH}) (Fierz-Schmidhauser, et al., 2010; Titos, et al., 2016). When combined with aerosol chemical composition data, this approach also allows for the determination of κ_{OA} (Kuang et al., 2020;

Kuang et al., 2021). These advancements have significantly promoted the application of nephelometers in aerosol hygroscopicity studies, and they also open up possibilities for using currently very popular, inexpensive optical scatter particle monitors for same purpose (e.g., Thermo pDR-1500, priced around \$5,000; even more affordable options like Purple Air, costing a few hundred dollars, and Plantower PMS series, available for tens of dollars). These inexpensive devices, based on single-wavelength nephelometric technology, could potentially be used to infer aerosol hygroscopicity and associated ALW. However, unlike the commonly dry/wet nephelometers that measure particle scattering coefficients to calculate f_{RH} , these inexpensive particle monitors directly report particle mass concentration as a bulk measurement, essentially functioning as “black boxes”. Unfortunately, there are very few studies that explore the potential of these optical particle monitors for such applications.” (Line 50-61)

Thank you again for this comment, which has helped strengthen the manuscript.

Specific comments

#1. In section 2.2 System setup, lines 93 to 100 where particle losses in sample lines are discussed it would be useful to add the flow Reynolds numbers. For line 99, what particle sizes does this less than 1% loss apply to?

R: Thank you for this comment. We added these related information, as “The air flow was expected to be turbulent based on the calculated Reynolds Number (RN=30234, as determined from <https://www.omnicalculator.com/physics/reynolds-number>), and the estimated particle loss of the ambient aerosol, with a size between 100 nm to 1000 nm, from the van inlet to each instrument was less than 1% (<https://www.mpic.de/4230607/particle-loss-calculator-plc>).” (Line 116-119)

#2. Line 91, is RH of 45% sufficient to assume that particles do not contain water, which is, I believe, the assumption here in this calculation?

R: Thank you for this comment. We added more information, as “During the deployment, the RH in pDR_{dry} ranged between 30% and 45%. We used 45% as the upper RH threshold for self-calibration, based on the following considerations: (1) ISORROPIA II model calculations indicate that aerosol liquid water associated with inorganics (ALW_{IOA}) is zero for all data below 45% RH, and (2) submicron internally mixed inorganic-organic particles do not exhibit hygroscopic growth until they reach their deliquescence point, which occurs at approximately 77% RH (Pope et al., 2010; Jing et al., 2016; Bouzidi et al., 2020).” (Line 105-109)

#3. Line 118, what about the fact that the AMS only measures non-refractory species, so it is not

a comprehensive measurement of particle mass concentration, not even considering the size of particles sampled. I.e., this should also be noted in this part of the paper, since it is also discussed later on, along with the PM_1 vs $PM_{2.5}$ issue.

R: Thank you for this comment. We added more information, as “Aside from the uncertainty due to size differences, the AMS only measures non-refractory aerosols and has limited sensitivity to refractory aerosols (e.g., sea salt), which introduces additional uncertainty and will be discussed further in Section 2.3.” (Line 160-162)

#4. Line 140, why is the chemical composition data not used to estimate density of OA instead of assuming a constant value of 1.4 g/cm^3 .

R: Thank you for this comment. As not accurate species information of OAs, we used the 1.4 g cm^{-3} for ρ_{OA} following the commonly used value. We added more discussion about this, as: “In this study, we used 1.4 g cm^{-3} for ρ_{OA} following the commonly used value (Hallquist et al., 2009; Shakya and Griffin, 2010; Nguyen et al., 2016; Riva et al., 2017; Jiang et al., 2019). However, the ρ_{OA} can vary significantly depending on the sources and formation pathways of organic aerosols, with a range between 1.2 and 1.6 g cm^{-3} based on a recent chamber study (El Mais et al., 2023), introducing some uncertainty into our results.” (Line 191-195). Thanks for your understanding.

#5. Line 145, note that if these data are used to estimate ambient air LWC in this study there are issues with the ambient measurements (wet) being made indoors. This is why many past studies on using HTDMA or $f(RH)$ run the ambient (wet) instrument outdoors.

R: Thank you for this comment. Agree with this, and we add more information into the text, as “Meanwhile, it also emphasized the possibility of using this system for using direct ambient measurements, very similar to the innovative outdoor dry/wet nephelometer system described by Qiao et al. (2024), without drying aerosols first before analysis as the HDMA (Tang et al., 2019) and without worrying about altering their actual phase state in ambient air (Qiao et al., 2024).” (Line 200-204)

#6. Line 150, what is the basis for assuming a constant fine/coarse mode mass ratio? Doesn't the fine and coarse mode chemical composition vary? Not sure how one assesses the impact of this assumption. The reasoning in lines 149 to 152 (“By simply assuming a constant ...”) is not clear. My interpretation is that the authors assume that the chemical composition of the coarse and fine modes is the same and invariant throughout the study and so the ratio of particle water in the fine and coarse modes will equal the ratio of fine and coarse mode dry mass concentration. This assumes no nonlinearities, such as the Kelvin effect.

R: Thank you for this comment. We reword them as: “The basic assumption here is the chemical composition of the coarse and fine modes is similar to each other throughout the study (Sun et al., 2020), and the ratio of particle water in the fine and coarse modes will equal the ratio of fine and coarse mode dry mass concentration. So that, the estimated M_{ALW} here based on the calibrated aerosol mass concentration from the pDRs using AMS as reference can represent the liquid water in non-refractory PM_{10} . However, significant uncertainty will be introduced in the estimation of κ_{OA} , particularly due to the presence of sea salt and other high- κ refractory components in coarse aerosols (AzadiAghdam et al. (2019)), which can greatly increase their hygroscopicity. Due to the limited information on the chemical composition (including refractory components) of fine and coarse aerosols, we can only provide a rough estimate of this uncertainty as a bulk, as shown below.” (Line 208-216)

#7. Line 163, the standard deviation is given as 0.08, but this is somewhat meaningless without knowing the typical (mean) κ_{OA} . Maybe the range in the standard deviation divided by the mean could be given for all the bins to get an idea of the relative error estimated by this method. (do some calculation, to add more)

R: Thank you for this comment. Considering the uncertainty on a quantity is generally quantified in terms of the standard deviation, we used the maximum standard deviation of all bins to represent the uncertainty for this method. For clarity, we add information of mean value, as “The maximum standard deviation of κ_{OA} across all bins of the identified three groups was determined to be 0.08 with the mean value of κ_{OA} for this bin as 0.18, which was expected as the upper limit of the uncertainty for κ_{OA} .” (Line 230-232). **Thank you so much for your understanding.**

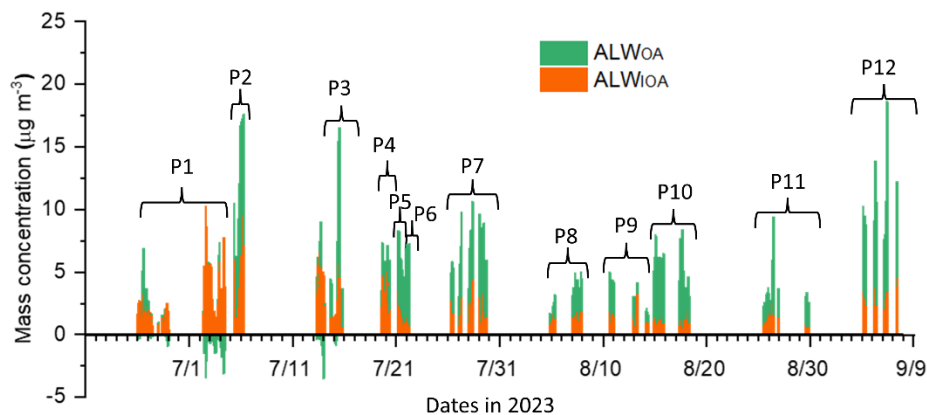
#8. In Fig 2b define what the given ratios are (slope?). The associated text is not clear (lines 172-174, ie what is the 2.5 referring to, and [24]).

R: Thank you for this comment. We add the related information to Figure 2 caption, as “The dashed lines represent the ratio lines of PDR_{wetc} to PDR_{dryc} at 1:1, 1.3:1, 2:1, and 4:1”. **Meanwhile, we have revised the previous statement, as** “As shown in Fig. 3b, the mass growth factor ($=M_{pDRwetc}/M_{pDRdryc}$) was mainly between 2 to 4 under RH range of 90% to 100%, with an averaged value of 2.5, which was generally higher than the value under the RH range of 80% to 90% with an averaged value of 1.3.” (Line 262-265)

#9. Fig 3, the x-axis has no label. This is somewhat stated in the fig caption but seems poor form. What is the year? Are the data shown in Fig 3 added (stacked) or each (ALW_{OA} and ALW_{IOA}) go to zero on the y axis? (correct it)

R: Thank you for this comment. We add the x-axis label. It is a stacked column plot, and

we added it in the caption.



P1: 89 points on June 26, 27, 28, 29 and July 02, 03, 04
P2: 14 points on July 05, 06
P3: 33 points on July 13, 14, 15
P4: 17 points on July 19, 20
P5: 13 points on July 21
P6: 13 points on July 21, 22

P7: 41 points on July 26, 27, 28, 29
P8: 30 points on Aug. 04, 05, 07
P9: 28 points on Aug. 10, 11, 12, 13, 14
P10: 38 points on Aug. 14, 15, 17, 18
P11: 32 points on Aug. 25, 26, 29
P12: 21 points on Sep. 04, 05, 06, 07

Figure 4. The time series of stacked column of the ALW_{OA} and ALW_{IOA} with a time resolution of 1-hr. (P1-P12 denote the different sub-periods mentioned in the following text with the data points and time periods of each subperiod indicated.)

#10. Line 195, is derived $Kappa_{OA}$ from equation 3, if so state it.

R: Thank you for this comment. We added it, as “The box and whiskers distribution of the derived κ_{OA} based on Eq. (4) for each sub-period is shown in Fig. 5a” (Line 290)

#11. Typo in line 200 ,,

R: Thank you for this comment. We corrected it.

#12. Line 196 and Fig 4b, define mass concentration, ie is it dry PM_1 ? (Not sure what total mass concentration means).

R: Thank you for this comment. We added the information, as “alongside the HR-ToF-AMS measured PM_1 mass concentration in Fig. 5b”. (Line 291)

#13. Line 214 to 216. Doesn't burning conditions, smoldering/flaming affect $Kappa_{OA}$, or is

this washed out the in highly averaged nature of smoke transported over long distances?

R: Thank you for this comment. We added the information, as “it is reasonable to infer the wildfire κ_{OA} could be strongly affected by the burning time of the original forests, the related burning conditions (i.e., smoldering vs. flaming, etc.), the transport time from west to east, etc. (Garofalo et al., 2019), resulting in significant variation between different cases, warranting further investigation.” (Line 310-312)

#14. Would it be useful to plot Kappa_OA to Mass_ALWOA? They are related by equation 3.

R: Thank you for this comment. As they are directly related to each other based on new Eq. (4), we did not to plot it again. Thank you so much for your understanding .

#15. Line 237 starting with “ It also shows.... What is being referred to, Fig 5b? (change to: It shows to Fig 5b shows...?”

R: Thank you for this comment. We corrected it, as “Meanwhile, the near-constant trends of κ_{OA} are showed for each period affected by the wildfire plumes (Fig. 6b)” (Line 334-335)

#16. First line of Conclusions, why not call them inexpensive single wavelength nephelometers instead of optical scattering systems, the latter could include a single particle optical particle counter, which these are not (I assume). (not hard)

R: Thank you for this comment. We corrected it from the comment.

#17. Line 263, not only is the slope different but the magnitude is significantly different between urban and rural (the curves are nowhere near overlapping). Doesn't this have implications for using O/C to estimate Kappa_OA.

R: Thank you for this comment. We added this information as “Meanwhile, the magnitude of κ_{OA} of rural aerosol is much higher than the value of urban aerosol.” (Line 361)

and,

“These large different κ_{OA} vs. O:C relationships, including both slopes and magnitudes, for each group imply the necessity of estimation of κ_{OA} through direct measurements, rather than through a simple dependent relationship based on one kind of aerosol other properties (i.e., O:C ratio).” (Line 364-366)

#18. Line 279, typo, varication?

R: Thank you for this comment. We corrected it, and moved it to new section 2.3, as “It is important to note that the derived κ_{OA} values in this study were not continuous, as we could only obtain them under high relative humidity (RH) conditions (85% to 90%). Additionally, our current inability to maintain aerosol under such high RH conditions limited the laboratory calibration and verification of this method using substances with known hygroscopic parameters (Fierz-Schmidhauser, et al., 2010; Zieger et al, 2013; Han et al., 2022), even though this method is theoretically feasible.” (Line 236-240)

#19. A final comment: It is curious to me why one does not compare water soluble organic carbon to κ_{OA} . (no hard)

R: Thank you for this comment. During the field measurements, we did not have an instrument (i.e., PILS) to report the water soluble organic carbon. Thanks for your understanding.

#20. Lines 277 and on where it is noted that there the measurements were not continuous.... This is not clear. The schematic shows that the wet measurement was straight ambient. It then seems that the gaps in the data are due to only periods of high ambient RH were analyzed in this study. So the authors are suggesting that adding a humidification system to the ambient leg to maintain an RH in a specific range, such as 85-95% would allow continuous measurements – is this the point? (make it more clear)

R: Thank you for this comment, and sorry for the confusion. You are totally right, and we rewrite it as, “It is important to note that the derived κ_{OA} values in this study were not continuous, as we could only obtain them under high relative humidity (RH) conditions (85% to 90%). Additionally, our current inability to maintain aerosol under such high RH conditions limited the laboratory calibration and verification of this method using substances with known hygroscopic parameters (Fierz-Schmidhauser, et al., 2010; Zieger et al, 2013; Han et al., 2022), even though this method is theoretically feasible. To resolve this issue, one possible update of this system could be adding a humidifier system to the pDR to get wet aerosol with RH between 85% to 95%, and the possible set-up for humidifier system could include a Perma Pure MH-series humidifier, water pumps and tanks (red dash box in Figure 2). This will make this system more be similar to the widely used humidified nephelometer system (Guo, et al., 2015; Burgos et al., 2019, Fierz-Schmidhauser et al., 2010; Kuang et al., 2017,2018,2020, 2021).” (Line 236-244).