Response to Anonymous Referee #1

Review of the paper "Insights into the real part of natural sea spray aerosol refractive index in the Pacific Ocean" by Fan et al.

The paper presents an interesting study of the real part of the refractive index of sea salt measured using an optical trap and proposes a linear fit of the data. The authors also extrapolate the fit outside the data range and provide some evidence of why that might be justifiable. The motivation of the work is good and the paper is mostly well written but some additional details need to be provided and the same clear caveats need to be explicitly mentioned before the paper can be published.

Response: We would like to express our deepest gratitude for taking the time to review our manuscript. In response to your constructive feedback, we have carefully revised our manuscript. Below, we will provide a detailed and point-by-point response to your comments. All the changes have been included in the newest version of our manuscript.

General Comments

1. The work provides the RRI of salt at a narrow range of specific wavelengths (around 650 nm). This important limitation should be evident from the beginning, so the authors should mention the wavelength in the abstract and at the end of the introduction. This issue is explicitly mentioned only at the very end of the paper while should be evident at the beginning already.

Response: Thank you for your suggestion. We have now clarified the limitation regarding the narrow wavelength range of the RRI measurements in both the abstract and the introduction of the manuscript, as suggested. The specific wavelength (650 nm) is now explicitly mentioned to ensure that this limitation is evident from the beginning.

2. The size range generated by the nebulizer and the size range of trapping of the AOT should be discussed in more detail and critically compared to "real" sea salt particles.

Response: Thank you for your valuable comment. In fact, the ultrasonic nebulizer generates fine bubbles within the seawater solution through high-frequency ultrasonic vibrations. These bubbles grow periodically with the ultrasonic vibrations and eventually burst once they reach a certain size. The bubble burst creates small pits on the liquid surface, and the surrounding liquid flows toward the burst point to fill the gap, splashing out jet drops. This process simulates the jet drop mechanism involved in forming real sea salt aerosols, generating micron-sized particles (Richter and Veron, 2016). What's more, the particle size range captured by our optical tweezers is approximately 3-7 microns in radius, which aligns with the size of particles generated by the nebulizer. This can further support the consistency of particle sizes produced by the nebulizer.

For real sea salt aerosols, particles are typically classified as small SSA, medium SSA, and large SSA, with the medium SSA size range being 1-25 microns (Lewis and Schwartz, 2004). These are mainly produced by the jet drop mechanism and contribute significantly to the total sea salt aerosol mass and optical scattering. Therefore, using the particles generated by the nebulizer as a surrogate for real SSA to study their radiative properties is a reasonable approach. Relevant explanations and discussions have been added to both the introduction and methodology sections.

3. The linear parameterizations provided by equations (1) and (2) are calculated from data at RH>60%. Is there a specific reason why data at lower RH were not directly measured? Is that because the assumption of sphericity must be satisfied to use Mie and only above an RH of 60% one can make that assumption confidently (the salt fully deliquesced and therefore spherical?)? This point should be clarified.

Response: Thank you for your valuable suggestion. Data at lower RH were not directly measured because SSA particles shrink as they lose water, eventually becoming too small to be stably captured by the AOT system and falling off. In practice, for particles like sucrose, which have low hygroscopic growth factors and water diffusion coefficients, the radius changes less significantly and more slowly with RH, and they can still be stably captured and measured even at RH = 10%. Additionally, WGMs indicating a uniform spherical particle shape remained present throughout the capture of SSA particles. This passage has been added to section 3.2:

"Data at lower RH were not directly measured because SSA particles shrink as they lose water, eventually becoming too small to be stably captured by the AOT system. Additionally, WGMs indicating a uniform spherical particle shape remained present throughout the capture of SSA particles."

4. Perhaps I missed it, but I could not find what the authors mean by "artificial sea salt". What is it? Later in the paper they mentioned that artificial sea salt is a good surrogate, but sodium chloride is not (lines 227-228), so artificial sea salt is not sodium chloride, what is it then?

Response: We apologize for the confusion. "Artificial sea salt" refers to a synthetic substitute for real sea salt, which is denoted as "standard sea salt" in the original manuscript. We have revised it to "artificial sea salt" at the end of section 2.1. The main components of artificial sea salt are 58.49% mass of sodium chloride, 26.46% mass of magnesium chloride, and 9.75% mass of sodium sulfate, along with calcium chloride, potassium chloride, and other salts.

Specific Comments

Line 34, page 2: Are these values a radiative effect? If so, where, at the surface, at the top of the atmosphere, for the atmosphere itself? Also, why is the range so large?

Response: Thank you for your comment. Yes, the values here refer to radiative effects at the top of the atmosphere (TOA).What's more, the large range is mainly due to uncertainties in global aerosol distributions, sea salt load, which varies with wind speed and regional factors, and the lack of comprehensive optical property data for SSA, such as RRI (Haywood et al., 1999; Aldhaif et al., 2018). We revised the sentence in the introduction part as:

"These particles directly scatter solar radiation at the top of the atmosphere (TOA), with global average scattering radiation ranging from -0.08 to -6 W/m^2. The large uncertainties in this range arise from variability in sea salt load and the limited availability of optical property data for SSA, such as the RRI (Haywood et al., 1999; de Leeuw et al., 2011; Aldhaif et al., 2018)."

Lines 41 and 42, page 2. I understand how that can be a limitation of thin disk bulk measurements, potentially, but why is that a limitation in electrodynamic balances, that should provide similar measurement options to AOT?

Response: Thank you for your question. The limitation of the electrodynamic balance (EDB) method lies in the fact that the first physical quantity measured is the particle's mass. To calculate the particle's volume, assumptions about the particle density or empirical relationships are required. Only then can the refractive index be derived by combining scattering light signals (Tang et al., 1997). In contrast, the AOT system directly measures the particle's radius and refractive index using whispering gallery modes (WGMs) and Mie fitting, allowing for more accurate and direct measurements (Preston and Reid, 2013). This makes the AOT method more advantageous in terms of precision and reliability.

Line 43, page 2: "fixed value" fixed with respect to what?

Response: Thank you for your comment. The term "fixed values" refers to the refractive indices of pure sea salt particles, which were measured in previous studies. Additionally, the word "or" should be replaced with "and" in this context. These "fixed values" are typically used in volume-weighted mixing rules to estimate the overall refractive index of sea spray aerosols (SSA) (Shettle and Fenn, 1979; Wang et al., 2019). We revised the sentence in the introduction part as:

"Moreover, model calculations typically estimate the refractive index of SSA using the RRI of pure sea salt and apply volume-weighted mixing rules based on known component properties (Shettle and Fenn, 1979; Wang et al., 2019)."

Lines 57-58, page 2: How similar does the process of generation by an atomizer resemble that of sea salt formation in the sea (from bubble bursts?). Might that affect the size and perhaps also the chemical composition of the aerosols produced?

Response: Thank you for your comment. The nebulizer simulates the jet drop mechanism of real SSA formation by generating a jet through bubble bursts and liquid flow. The particles generated in this way have a similar size distribution (micron-sized) to supermicron SSA that dominates scattering. Additionally, while organic components can account for more than 50% of the dry mass of submicron SSA, their proportion is typically less than 5% in supermicron SSA particles (de Leeuw et al., 2011). For the micron-sized particles generated by the nebulizer in our experiment, lowering the environmental RH to 70% still showed no C-H peaks in the Raman spectra (Fig. S1), indicating that the organic content in these particles is very low, similar to that of real supermicron SSA. Therefore, considering that the solution used for generation was real seawater and that the organic content is minimal, the chemical composition of the aerosol particles generated by the nebulizer should not differ significantly from that of real SSA. We have revised the sentence in the introduction part as:

"This system overcomes the limitations of bulk measurement methods by allowing us to isolate individual SSA particles produced by an ultrasonic nebulizer through the pattern of jet drops (Richter and Veron, 2016). It provides accurate, in situ measurements without the need for density assumptions and the particles generated in this manner are micron-sized, exhibiting a similar size and composition to supermicron SSA."

and in the manuscript, the following paragraph was added in section 2.2:

"It is important to note that the nebulizer generates fine bubbles in the seawater solution through ultrasonic vibration. When these bubbles grow and burst, the resulting surface cavities induce surrounding liquid flow and splashing, creating jet drops. This process simulates the jet drop mechanism in forming real sea spray aerosols, producing micron-sized particles. In our experiments and subsequent fitting calculation, the AOT system usually stabilized particles within the size range of 3-7 microns, aligning with the expected particle size generated by the nebulizer. Real sea salt aerosols are typically categorized into small, medium, and large SSA, with the medium SSA size range being 1-25 microns (Lewis and Schwartz, 2004). These particles, mainly produced through the jet drop mechanism, contribute significantly to sea salt aerosol mass and optical scattering. Therefore, using the particles generated by the nebulizer as surrogates for real SSA to study radiative properties is a reasonable approach. Regarding chemical composition, while organic components can make up over 50% of the dry mass of submicron SSA, they account for less than 5% of supermicron SSA particles (de Leeuw et al., 2011). In our study, reducing the environmental relative humidity (RH) to 70% still showed no detectable C-H peaks (around 2800-3100 cm^{-1}) in the Raman spectra (Fig. S1), indicating that the organic content in the micron-sized particles is minimal, similar to that of real supermicron SSA. Given that the solution used for aerosol generation was real seawater and the organic content is negligible, the chemical composition of the particles generated by the nebulizer is also unlikely to differ significantly from real supermicron SSA."

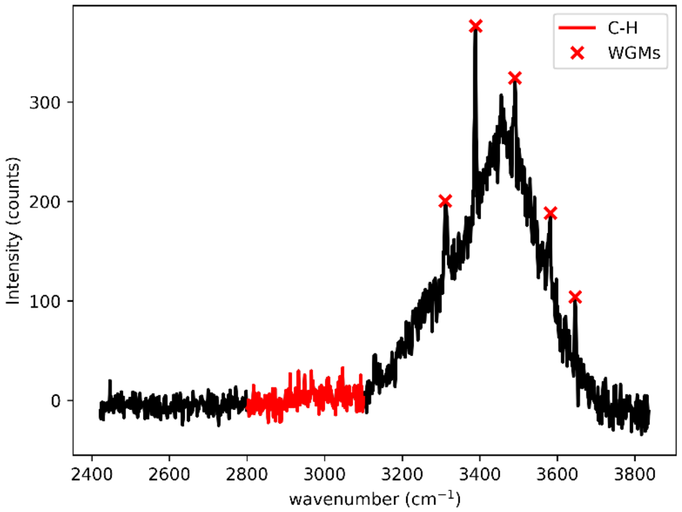

Figure S1. The Raman spectrum of the particles generated by the nebulizer at RH = 70%. The red part represents the typical peak position of the C-H peak, and the red cross marks indicate the particle’s significant Whispering Gallery Modes (WGMs).

Lines 86-87, page 3: Are there any concerns related to salt exclusion (brine) while freezing the sample? This detail should be at least briefly discussed.

Response: Thank you for your valuable comments. The salt exclusion phenomenon has indeed been considered and we added relevant discussion here:
“It should be noted that salt exclusion may occur during freezing, concentrating dissolved salts in the remaining liquid as the ice forms. To minimize this effect, we allowed the samples in sealed bottles to thaw completely at room temperature and mixed the samples thoroughly before measurement. We believe this slow thawing process ensures minimal impact on sample composition and subsequent results.”

Line 109, page 4: Again, how well does the nebulizer reproduce sea particle sizes similar to those found in marine air?

Response: Thank you for your comment. The ultrasonic nebulizer generates micron-sized particles by simulating the pattern of jet drops in the formation of real SSA. The particle size of these generated particles closely matches the size range of the medium SSA, which plays a dominant role in total light scattering of the sea salt and is well-reflected by our experimental particles. In the manuscript, relevant explanations and discussions have been added here.

Line 122, page 5: How are the samples unfrozen and brought to 20 C before nebulization? Can the details of the tawing process and rate affect the end results?

Response: Thank you very much for your comment. Perhaps you were referring to the thawing process and rate. Since salt exclusion may occur when the samples are frozen, too fast thawing might lead to less uniform mixing of the salt and water, which could alter the concentration of the solutes when the sample is nebulized and measured, resulting in measurement results that do not represent the original sample.

In our experiments, the seawater samples were allowed to thaw at room temperature for several hours to ensure uniform thawing. After thawing, the samples were thoroughly mixed before nebulization. We believe this approach minimized the potential impact of the thawing rate on the refractive index measurements. The relevant description has been added along with the discussion of salt exclusion in the manuscript.

Figure 3. I am not sure the term "stimulated" for the peaks is correct because it is still a spontaneous although narrow Raman peak, at least that one reaches optical densities that might result in nonlinear effects within the droplet, which might not be the case here.

Response: Thank you for your valuable comment. In my case, WGMs are indeed associated with spontaneous Raman scattering, where narrow peaks are observed in the Raman spectrum resulting from multiple total internal reflections inside the droplet. These peaks are not induced or stimulated by an external source but are naturally enhanced due to the optical cavity effect inside the droplet. We revised Fig 3. as:

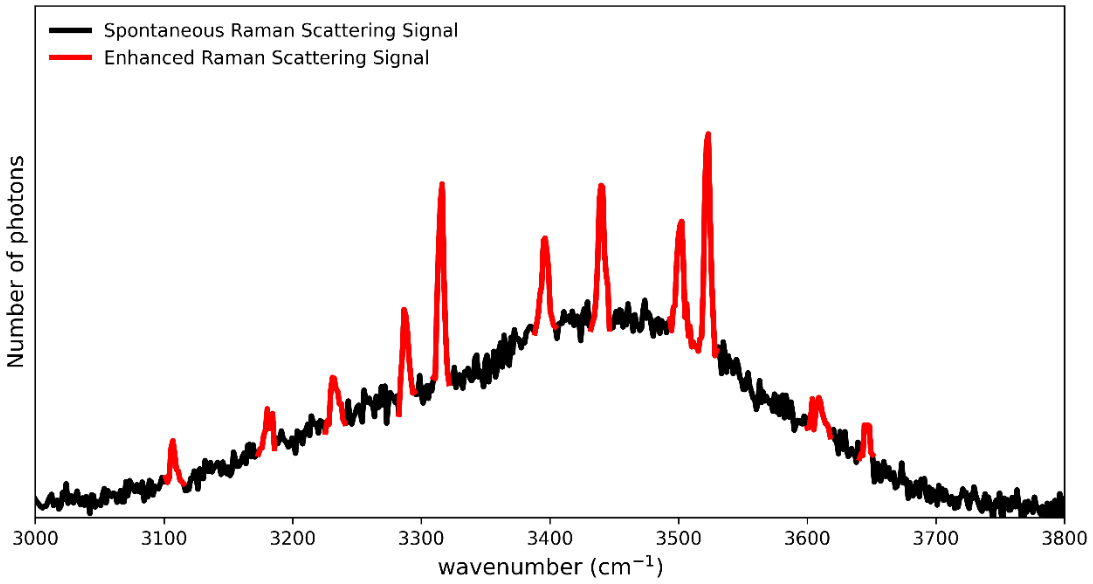

Figure 3. Example of Enhanced Raman scattering signal on the broad spontaneous Raman scattering background.

Line 140, page 6: that's the case for spherically-symmetric particles.

Response: Thanks for your comment, this sentence has been revised as:

"Each WGM is characterized by three parameters for spherically symmetric particles: mode number, mode order, and polarization state (either Transverse Electric (TE) or Transverse Magnetic (TM) mode), which corresponds to Mie scattering theory."

Line 141: Although the fitting method apparently is described in Preston and Reid (2013), it would be useful to provide a brief overview here.

Response: Thank you for your comment. We have added the following description in section 2.3 as you suggested:

“Briefly, a table of resonance size parameters is constructed for different refractive indices and WGMs. Experimental spectral peak values are then converted into size parameters for the given radius. Next, a parameter space of radius and refractive index is explored to minimize the total error between the calculated and observed peak positions under dispersion-free conditions. Following this, a narrow range of dispersion is considered, and a new table is generated for a smaller parameter space, with fitting performed to minimize the overall error and obtain the best parameters. By selecting appropriate parameter boundaries and intervals, the Mie scattering series can be optimized using a recursive approach, enabling the calculation of particle size and the RRI.”

Line 144, page 6: “this method can reach…” can in ideal conditions or did actually achieve this precision/accuracy in the specific work presented here? This also brings up the question, are these precisions or accuracies?

Response: Thank you for your comment. The accuracy described mentioned in the reference can only be achieved under ideal conditions. In practice, due to fluctuations in relative humidity and occasional lower numbers of WGMs, the ideal accuracy is not always reached. The reported values in our experiments are averages of the fitting results from each frame at equilibrium, emphasizing the consistency of the measurements, so "precisions" is the appropriate term here. In the experiment, the typical precision for the radius and refractive index was ±20 nm and ±0.005, respectively. We revised the precision in our manuscript and an example of fitting is shown in Fig. S2.

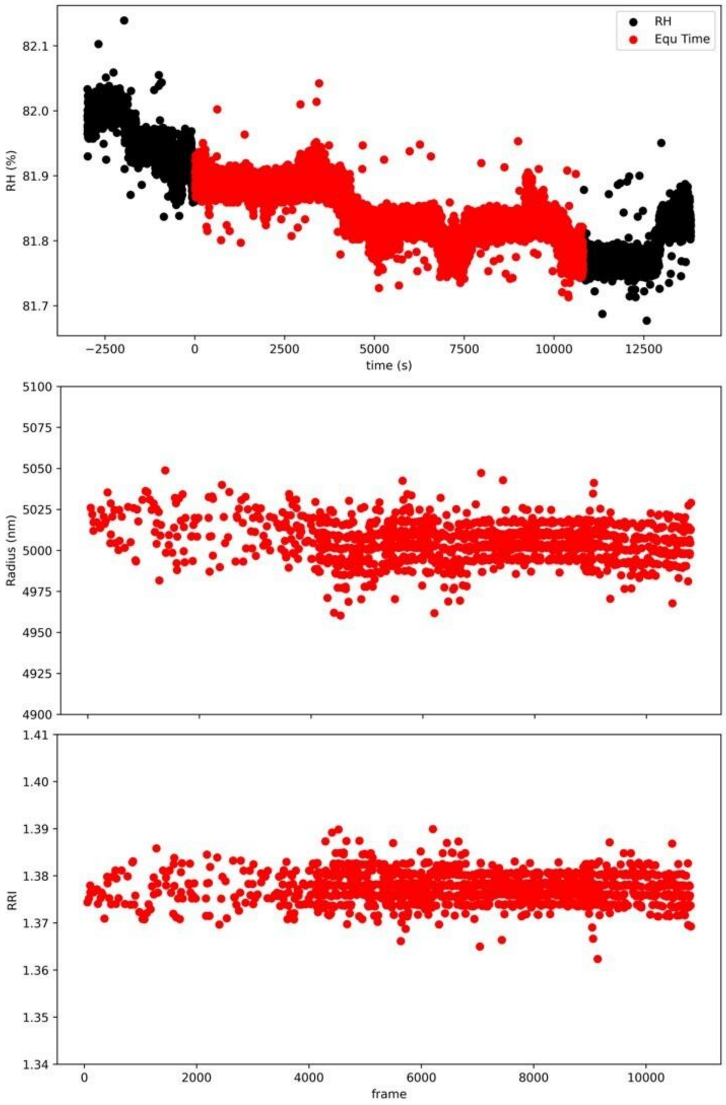

Figure S2. An example of the relative humidity (top) when an artificial sea salt particle is in equilibrium at RH = 81.8%, with the fitted radius (middle) and RRI (bottom). The mean values are 5005.4 nm and 1.3773, with a standard deviation of 10.7 nm and 0.0029, respectively.

Line 155, page 7: It is not clear why 0.7% was chosen if the error is 1.3%. The criteria for this choice should be discussed.

Response: Thanks for your comment. Though the maximum error for the molar refraction method is ±1.3%, the average absolute error for values determined by the molar refraction method, refractometer, and ellipsometer is within 0.75% (Wang and Rood, 2008). Therefore, 0.75% is chosen in the new version of the manuscript:
"The average error for the molar refraction method is within 0.75% and is adopted as uncertainty here (Wang and Rood, 2008)."

Figure 4: Even though each of the AOT and E-AIM points seems to fall within the error bars of each other, there still seems to be a consistent positive bias in the AOT data. Are there any hypotheses of why that might be?

Response: Thank you for your valuable comment. We recalculated the refractive index of the particle composition provided by the E-AIM model, and the calculation result is presented in Table S2 and the revised Figure 4.

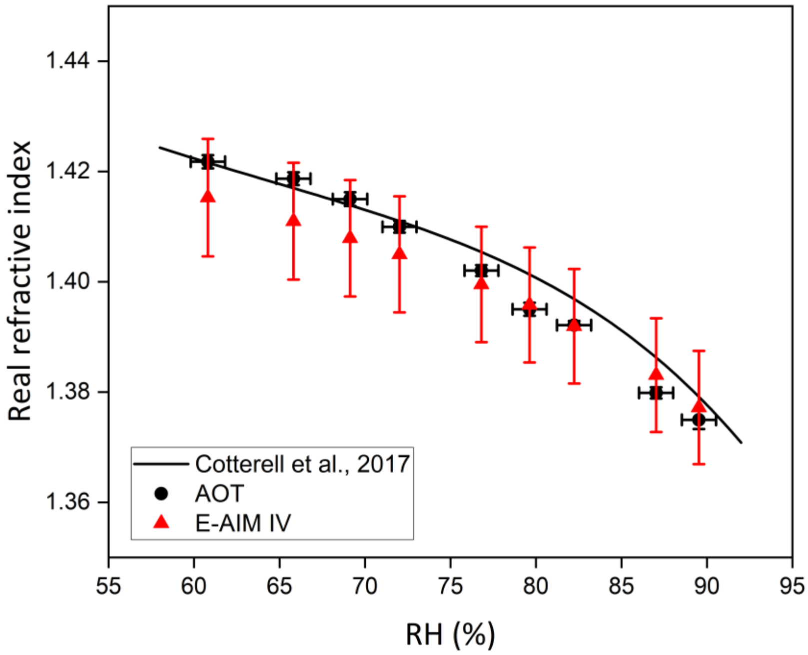

Figure 4. Variation of the real part of the refractive index of ammonium sulfate particles with RH. Data from the E-AIM IV model at 293.15 K and the previous parameterization scheme by Cotterell et al. (2017) are shown for comparison.

The RRIs obtained by the AOT and the E-AIM IV model still remain consistent within the uncertainty range, and they match very well at RH values above 75%. Although the E-AIM IV model underestimates the RRI to some extent at RH below 75%, this may be due to the data in the E-AIM IV model being extrapolated from unsaturated solutions, which can introduce a certain bias when predicting the composition of supersaturated particles. This, in turn, leads to discrepancies between the calculated and measured RRI values. Additionally, errors in the temperature and humidity probes may contribute to this deviation. The Relevant discussion has been added to the manuscript:

"Although the E-AIM IV model underestimates the RRI to some extent at RH below 75%, this may be because the data of the E-AIM IV model are extrapolated from unsaturated solutions, which can introduce a certain bias when predicting the composition of supersaturated particles. This, in turn, leads to discrepancies between the calculated and measured RRI values."

Lines 163-166: Similarly, is there a possible explanation for the positive bias of the parameterization by Cotterell?

Response: Thanks for your comment. The positive bias of the parameterization was also observed in Cotterell et al. (2017), and attributed to the lack of measurement data at wavelengths longer than 600 nm, which leads to inherent overestimations in the parameterization at these wavelengths. Additionally, errors in the humidity-temperature probes and inconsistencies in environmental conditions may also contribute to this deviation. We have added the above analysis to the latest version of our manuscript.

Line 173: Is there any evaporation of the droplet happening over such a long period of time? If so, how is the corresponding change in the index of refraction and size accounted for? I would guess that one could calculate the index of refraction for different subsections of these 3 hours and compare for example, the results at the beginning, at a few intermediate times, and at the end to see if there is a consistent trend.

Response: Thank you for your comment. An example of a three-hour measurement can be found in Fig. S2. When the RH reaches equilibrium, there is no significant change in RH, except for a slight downward fluctuation. The corresponding droplets experience minor evaporation or condensation, which is reflected in the fitted radius. Overall, the radius and refractive index show good consistency during the equilibrium period, with no obvious change trends.

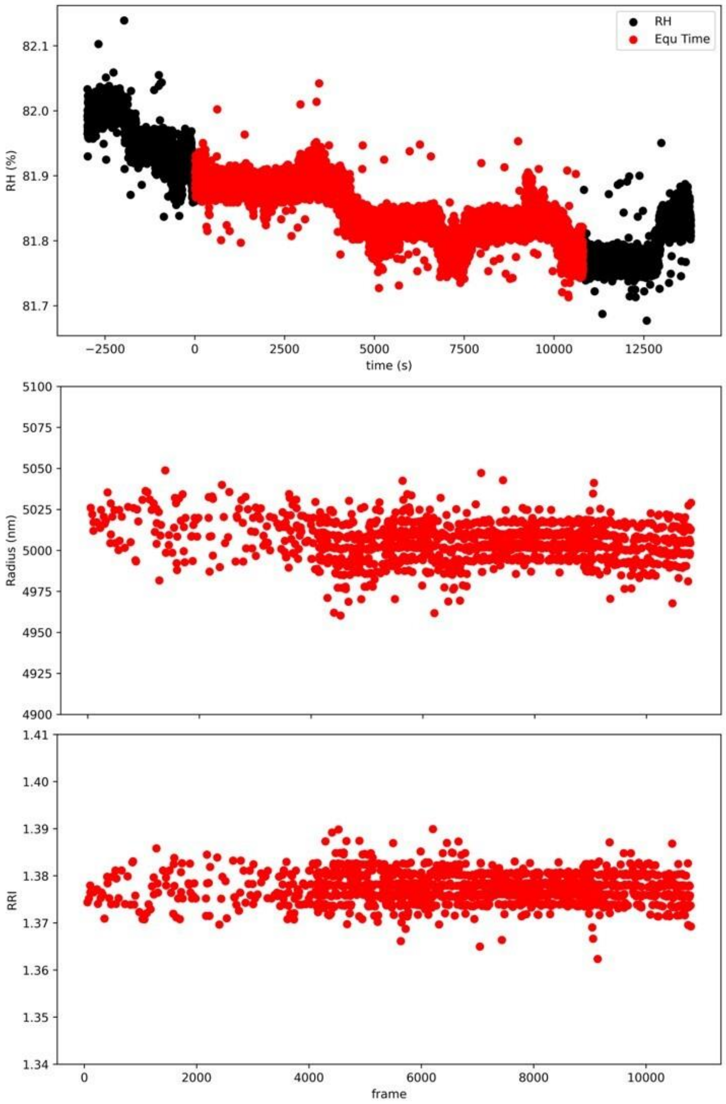

Figure S2. An example of the relative humidity (top) when an artificial sea salt particle is in equilibrium at RH = 81.8%, with the fitted radius (middle) and RRI (bottom). The mean values are 5005.4 nm and 1.3773, with a standard deviation of 10.7 nm and 0.0029, respectively.

Figure 5 and corresponding discussion: the range of salinity seems very narrow from about 33.9 to 35.2 ‰, a change of just a little bit more than 1‰. Is it reasonable to extrapolate any conclusion (as in line 190) from such a narrow range? Also, what is the error in salinity?

Response: Thank you for your comment. The salinity measured by the onboard instrument has a precision of 0.0001‰, as shown in Table S1. Although the salinity range is narrow, the key factor affecting particles' hygroscopic and refractive index is the relative composition of the dry sea salt, rather than the salinity of the seawater. Reich et al. (2023) reached a similar conclusion, noting that variations in seawater salinity do not affect droplets' hygroscopic properties when ion relative abundances are the same and small changes in the relative abundance of ions in seawater have no significant effect on the hygroscopicity of the particles, which maybe be the same case of the RRI. Therefore, it is reasonable to perform a sound extrapolation and even extend the measurement results to broader sea areas. This passage has been added to the latest manuscript:

"Reich et al. (2023) reached a similar conclusion, noting that variations in seawater salinity do not affect droplets' hygroscopic properties when ion relative abundances are the same and small changes in the relative abundance of ions in seawater have no significant effect on the hygroscopicity of the particles. This may be similar to the RRI, given the nearly identical hygroscopic growth factors and solute refractive indices for both types of SSA."

Equation (1): The authors should provide the uncertainties on the slope (they do provide that for the intercept).

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

"The 95% confidence interval for the slope and intercept ranges from -1.375e^{-3} to -1.220e^{-3} and 1.478 to 1.490, respectively."

Line 202 (and then again on lines 211 and 225), please provide more digits (the first non-zero) for the p-values

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

"The coefficient of determination, R^2, for this scheme, is 0.974, and the p-value for slope and intercept is 3.59e^{-26} and 1.19e^{-61} respectively, indicating a statistically significant relationship."

Lines 203-206: The agreement is interesting and encouraging, but one should clearly caution that the intercept has been calculated from a fit of data only at the high end of the full RH range (>60%), so it is a significant extrapolation and cautions should be required. Isn't deliquescence and efflorescence playing any role here (which I would expect to be non-linear)?

Response: Thanks for your valuable comment. Indeed, the intercept was obtained by fitting the data with RH > 60% and extrapolation. In the latest manuscript, we pointed out that although the intercept is obtained through extrapolation, it is an objectively existing value that can provide a basis for subsequent research. Moreover, we emphasized that the parameterization scheme and the linear relationship are valid in marine environments.

On the other hand, the effects of deliquescence and efflorescence may indeed lead to non-linearity. We assume a molten state at room temperature in the latest manuscript, as mentioned in Clegg and Wexler (2011) and Cai et al. (2016). That is, we assume that sea salt particles do not undergo efflorescence during water loss and remain in a dissolved molten state to avoid the possible impacts of deliquescence and efflorescence. The manuscript has been revised as:

"The intercept, corresponding to RH = 0, is interpreted as potential RRI of molten SSA particles at 20 °C if we assume that the sea salt particles do not undergo efflorescence and remain in a dissolved molten state. Although the assumed molten state may not occur in the real atmosphere, the extrapolated values still provide a basis for further analysis. Furthermore, the linear relationship and parameterization scheme, which can be incorporated into climate models, are valid and align well with measurements in marine environments with RH > 60%. On the other hand, while structural changes may occur, the RRI of molten sea salt appears to closely match the dry sea salt refractive index of 1.49 reported in previous studies (Shettle and Fenn, 1979)."

Figure 6: what is the symbol for artificial sea salt? Is the artificial sea salt represented by the light brownish dots indicated in the legend as "Sea salt"?

Response: Thanks for your comment. Yew, the artificial sea salt represented by the light brownish dots indicated in the legend as "Sea salt". We revised the legend to "Artificial sea salt" in the latest manuscript to avoid confusion.

Lines 215-216: These are good lines of evidence, but I would not be so categoric about the validity of the fit.

Response: Thank you for your comment. In field measurements, 40% RH is likely chosen as the dry condition because most inorganic salts experience efflorescence at this relative humidity, resulting in little difference in properties compared to 0% RH. However, the linear parameterization scheme can't capture sudden efflorescence and possible changes in the RRI. Thus, we regarded the extrapolated value at 0% RH as the RRI of dry particles, instead of the value at 40% RH as in field observations. However, in the latest manuscript, as the molten state at room temperature is a hypothesized condition lacking verification and comparison with measured data, we have removed the part about using ammonium sulfate as a reference substance.

Line 228: is the RRI of dry sodium chloride reported by Wang and Rood at 0% RH or 40% RH, if at 40% then, as discussed by the authors with regard to ammonium sulfate in lines 214-216 then would not be a fair comparison, and the comparison for published values of sodium chloride should be made at the same RH.

Response: Thank you for your comment. The RRI of dry sodium chloride reported by Wang and Rood was indeed measured at nearly 0% RH. However, the RRI may not differ significantly between 0% RH and 40% RH for effloresced or non-deliquesced sodium chloride. In the latest manuscript, we compare the RRI of molten sodium chloride extrapolated from high temperatures to 20 °C with natural sea salt, which is a fairer comparison:

"However, the RRI of molten sodium chloride, extrapolated from high-temperature data to 20 °C, is 1.513 (Janz, 1967), and is 1.42% higher than that of SSA. This is likely due to the presence of lower refractive index compounds like sodium sulfate in natural sea salt and indicates that sodium chloride is not a perfect substitute for SSA in studies of radiative properties when greater accuracy is required."

Line 234: is no the dry RRI, meaning at 0% or 40% RH?

Response: Thank you for your comment. The dry RRI means the RRI measured at 0% RH as well as the dry radius. The RRI may also not differ significantly between 0% RH and 40% RH for effloresced or non-deliquesced sea salt.

Table 1: how is rd calculated?

Response: Thanks for your comment. The dry radius of SSA was estimated from the empirical relationship that the particle radius at RH = 80% is approximately twice the dry radius (Lewis and Schwartz, 2004; Qiu et al., 2024). The value of SSA's hygroscopic growth factor κ calculated from this empirical relationship is reasonable and in agreement with previous studies (Qiu et al., 2024).

Response to Anonymous Referee #3

Fan et al. show a method to quantify the real part of natural sea spray aerosol refractive index and develop a linear regression relation to estimate the real part of the refractive index under different relative humidity. Overall, this study is well designed and has potential important impacts on reducing uncertainties of aerosol climate effects. However, there are some points I cannot fully support. Moreover, some critical information is missing. I have my comments below. Thus, I recommend major revision before considering it for publication.

Response: Thanks for your valuable comments, which really helped improve the manuscript. Below, we will provide a detailed and point-by-point response to your comments. All the changes have been included in the latest manuscript.

General comments:

The authors claim that this method can measure particle sizes. However, the discussion of particle sizes is missing. This is critical for calculating the refractive index. Please add a discussion about how you measure particle sizes and what sizes of particles you used in your refractive index calculation. Also, what is the upper and lower limit of particle sizes your optical tweezer can measure?

Response: Thank you for your suggestions. We calculated both the particle radius and refractive index based on the WGMs signals of the particles captured by the AOT system and the Mie fitting algorithm in Preston and Reid (2013). From multiple capture processes and the corresponding radius fitting results, the radius range stably captured by our optical tweezers is approximately 3-7 microns. Relevant discussions have been added to Sections 2.2 and 2.3.

It should be noted that the particles used in this manuscript are crystals, which have delinquency points. Since the significant fraction of these particles should be NaCl,

I expect the delinquency point to be around 75% RH at 293 K. Therefore, it raises two questions. First, the particle should not be spherical below the delinquency point, which will lead to uncertainties by using the Mie simulation. Second, the linear correlation between n and RH in this manuscript is impractical from the RH range below the delinquency point. In addition, the delinquency point also depends on the temperature. Therefore, the proposed correlation might not be useful in the real world.

Response: Thank you for your valuable suggestions. Although the deliquescence point of NaCl is around 75% RH, since the particles are in a non-contact optical levitation state, a supersaturated state will form, which does not affect the spherical shape of the particles and the Mie fitting. Moreover, as shown in Fig. S1, the WGMs signals indicating that the particles are uniformly spherical remained present throughout the capture of SSA particles. On the other hand, although the efflorescence process occurs randomly, NaCl measured by electrodynamic balance (EDB) usually effloresces at 45% RH. Therefore, in experiments and typical marine environments with RH > 60%, SSA is generally considered to remain liquid (Lewis and Schwartz, 2004).

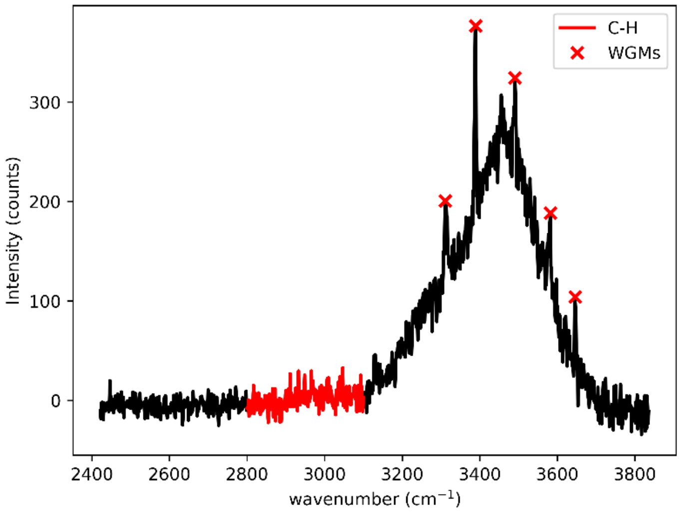

Figure S1. The Raman spectrum of the particles generated by the nebulizer at RH = 70%. The red part represents the typical peak position of the C-H peak, and the red cross marks indicate the particle’s significant Whispering Gallery Modes (WGMs).

On the other hand, the effects of deliquescence and efflorescence may indeed lead to non-linearity of the linear correlation. We assume a molten state at room temperature in the latest manuscript, as mentioned in Clegg and Wexler (2011) and Cai et al. (2016). That is, we assume that sea salt particles do not undergo efflorescence during water loss and remain in a dissolved molten state to avoid the possible impacts of deliquescence and efflorescence. Therefore, the linear parameterization scheme can keep linear with no need for capturing sudden efflorescence and possible changes in the RRI. Moreover, we emphasize the linear relationship and parameterization scheme, which can be incorporated into climate models, are valid in the real world and align well with measurements in marine environments with RH > 60%. The manuscript has been revised as:

"The intercept, corresponding to RH = 0, is interpreted as potential RRI of molten SSA particles at 20 °C if we assume that the sea salt particles do not undergo efflorescence and remain in a dissolved molten state. Although the assumed molten state may not occur in the real atmosphere, the extrapolated values still provide a basis for further analysis. Furthermore, the linear relationship and parameterization scheme, which can be incorporated into climate models, are valid and align well with measurements in marine environments with RH > 60%. On the other hand, while structural changes may occur, the RRI of molten sea salt appears to closely match the dry sea salt refractive index of 1.49 reported in previous studies (Shettle and Fenn, 1979)."

It will be useful to provide a chemical composition of particles analyzed in this study, which is important for understanding these particles' hygroscopicity and refractive index. Considering the relation between RH and scattering, the hygroscopic growth of SSA, which is chemical composition dependent, is important. Please discuss how the different chemical compositions can affect the derived relation.

Response: Thank you for your comments. We agree that chemical composition can help understand particle hygroscopicity and refractive index. We have added an analysis of the fitted slopes, suggesting that the stronger hygroscopic growth capacity of the components leads to a larger negative slope. On the other hand, while the AOT system cannot provide particles' precise chemical composition, a comparison of the refractive index of open-sea and offshore SSA under varying relative humidity reveals that differences in salinity and minor compositional variations between these regions do not significantly affect the RRI properties. Finally, given the similar RRI-RH relationships, we approximate the composition of real sea salt particles with that of artificial sea salt for subsequent analysis. The following passages have been added to the latest manuscript:

"The slope reflects the hygroscopic growth ability of particles to some extent. A stronger hygroscopic ability of the components leads to greater water uptake with increasing RH, resulting in a more pronounced decrease in refractive index, and consequently, a larger absolute value of the slope."

"The RRI values for offshore and open-sea SSA are consistent, showing no significant dependency on the salinity of the source seawater. This finding aligns with previous research showing that the hygroscopic properties of offshore and open-sea SSA are nearly identical (Qiu et al., 2024). Reich et al. (2023) reached a similar conclusion, noting that variations in seawater salinity do not affect droplets' hygroscopic properties when ion relative abundances are the same and small changes in the relative abundance of ions in seawater have no significant effect on the hygroscopicity of the particles. This may be similar to the RRI, given the nearly identical hygroscopic growth factors and solute refractive indices for both types of SSA."

"Additionally, assuming that SSA components can be represented by artificial sea salt due to the similar RRI across various RH, we considered the main components, sodium chloride, magnesium chloride, and sodium sulfate only, with original mass fractions of 58.49%, 26.46%, and 9.75%, respectively.”

How representative is the size of particles measured in this study compared to the real-world SSAs? Moreover, it is also unclear to me how you generate SSA particles. The authors mentioned they used an atomizer. Which type did you use? The atomizer is not suitable for generating coarse-mode particles. Typically, scientists use nebulizers to generate coarse-mode particles.

Response: Thank you for your valuable comments. We used a medical ultrasonic nebulizer (Yuyue 402AI model) to generate SSA particles, and the misused word “atomizer” has been revised to “nebulizer”. The ultrasonic nebulizer generates fine bubbles within the seawater solution through high-frequency ultrasonic vibrations. These bubbles grow periodically with the ultrasonic vibrations and eventually burst once they reach a certain size. The bubble burst creates small pits on the liquid surface, and the surrounding liquid flows toward the burst point to fill the gap, splashing out jet drops. This process simulates the jet drop mechanism involved in forming real sea salt aerosols, generating micron-sized particles (Richter and Veron, 2016). What’s more, the particle size range captured by our optical tweezers is approximately 3-7 microns in radius, which aligns with the size of particles generated by the nebulizer. This can further support the consistency of particle sizes produced by the nebulizer.
For real sea salt aerosols, particles are typically classified as small SSA, medium SSA, and large SSA, with the medium SSA size range being 1-25 microns (Lewis and Schwartz, 2004). These are mainly produced by the jet drop mechanism and contribute significantly to the total sea salt aerosol mass and optical scattering. Therefore, using the particles generated by the nebulizer as a surrogate for real SSA to study their radiative properties is a reasonable approach. Relevant explanations and discussions have been added to both the introduction and methodology sections.

Specific comments:

The introduction section misses the discussion of the nephelometer, a well-established instrument to measure scattering coefficients.

Response: Thank you for your comment. The following passage has been added to the introduction section:
“In contrast to another well-established nephelometer method for measuring aerosol scattering coefficients, optical tweezers circumvent the potential inconsistencies arising from ensemble averaging and multi-particle interactions by capturing and measuring individual particles. This ensures that each measurement reflects the intrinsic optical properties of individual particles and reveals the physicochemical characteristics of particles from a distinct perspective, particularly the influence of humidity on the refractive index.”

Section 2.2: As mentioned in L56, this is a real-time measurement. How many particles/droplets can you measure every set, and how long does it take to measure

1 set sample?

Response: Thank you for your comment. Our AOT is a single-beam Gaussian optical tweezer, which can only capture and measure one particle at a time. For 1 set sample, each relative humidity level requires a three-hour equilibrium time for data collection, with more time for the changed relative humidity to reach equilibrium. Therefore, it generally takes two to three days to measure 1 set sample.

L112: What is the smallest size the CCD camera can measure?

Response: Thank you for your comment. The CCD camera is mainly used to confirm the capture of particles and the approximate outline of particles, and it cannot be used to measure the size of particles. The accurate measurement of particle size is provided by the WGMs signals of the particles and Mie fitting. The particle range from 3-7 microns can be stably captured by the AOT system and the particles that can be recognized and observed by the CCD camera are about 1 micron.

L150: What is the E-AIM IV model? Please provide some basic information and the input parameters you used in the SI.

Response: Thank you for your comment. The E-AIM IV model (Extended Aerosol Inorganics Model, version IV) is a thermodynamic model used to predict the properties of aerosol solutions, including their chemical composition, water content, and density, under varying environmental conditions, particularly relative humidity (Wexler and Clegg, 2002). In this study, we used the E-AIM IV model to estimate the composition and density of ammonium sulfate aerosols at specified RH. The input parameters for the model including the temperature, relative humidity, and subsequent calculation are provided in detail in Table S2.

Table S2. Input parameters for the E-AIM IV model, results, and the RRI calculated using the molar refraction method

Temperature (K)	RH (%)	$x_{(NH_4)_2SO_4}$	ρ	M_e [a]	R_e [b]	n
293.15	60.81	0.173	1.340	37.72	7.05	1.415
293.15	65.82	0.151	1.319	35.26	6.64	1.411
293.15	69.12	0.138	1.305	33.72	6.37	1.408
293.15	72.02	0.126	1.291	32.39	6.15	1.405
293.15	76.81	0.107	1.266	30.23	5.78	1.400
293.15	79.62	0.096	1.249	28.97	5.57	1.396
293.15	82.23	0.086	1.232	27.77	5.37	1.392
293.15	87.03	0.065	1.194	25.49	4.98	1.383

a $M_e = x_1M_1 + x_2M_2$, where M_e is the relative molecular mass of the mixture. The relative molecular mass of ammonium sulfate and water are 132.14 g/mol and 18.02 g/mol, respectively.

b $R_e = x_1R_1 + x_2R_2$,where R_e is the molar refraction of the mixture. The molar refraction of a pure substance is calculated by the following formula:$R = \frac{n^2-1}{n^2+2}\frac{M}{\rho}$ and the molar refractions of ammonium sulfate and water are 23.06 and 3.71, respectively.

L165: What is the "inherent overestimation in the parametrization" here?

Response: Thanks for your comment. The positive bias of the parameterization was also observed in Cotterell et al. (2017), and attributed to the lack of measurement data at wavelengths longer than 600 nm, which leads to inherent overestimations in the parameterization at these wavelengths. Additionally, errors in the humidity-temperature probes and inconsistencies in environmental conditions may also contribute to this deviation. We have added the above analysis to the latest version of our manuscript:

"At RH above 75%, the values provided by the parameterization are slightly higher than those measured by the AOT (about 0.5%). This discrepancy was also observed by Cotterell et al. (2017) and attributed to the lack of measurement data at wavelengths longer than 600 nm, which leads to inherent overestimations in the parameterization at these wavelengths. Additionally, errors in the humidity-temperature probes and inconsistencies in environmental conditions may also contribute to this deviation."

L188: Are you ignoring the organics? If yes, please state the potential uncertainty in your results.

Response: Thank you for your comment. Since the organic content in supermicron particles is typically less than 5% (de Leeuw et al., 2011), and the C-H peak characterizing organic material is not visible in Fig. S1, the impact of organic material on the analysis is negligible. Additionally, since the refractive index of organic material is generally higher than that of water, the analysis suggesting an increase in refractive index as the particles lose water is still valid. Furthermore, whether or not organic material is present, Mie fitting accurately determines the particle's refractive index. Therefore, it does not introduce additional uncertainty into the results theoretically.

L235 eq 4: Please explain in the text what Vi, rd , and rw are.

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

L243: How did you calculate AOD?

Response: Thank you for your comments. We did not directly calculate AOD in this study. Instead, we utilized the findings of Aldhaif et al. (2018), which indicate that a 0.18 change in RRI for marine aerosols results in approximately a 41.9% change in AOD. Therefore, based on our finding of a 0.032 underestimation in RRI, we estimated a corresponding 7.4% underestimation in AOD through proportional scaling.

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