Response to Reviewers of egusphere-2023-2319 "Prominent role of organics in aerosol liquid water content over the south-eastern Atlantic during biomass burning season" by L. Zhang et al.

We sincerely thank the reviewers for the time and effort invested in assessing this manuscript. Your constructive comments and insightful suggestions have been very helpful in improving the clarity and overall quality of our work. We have carefully considered each of your comments and have made revisions accordingly. Below, we provide a point-by-point response, addressing all concerns raised. For clarity and ease of reference, the reviewers' comments are presented in blue, our responses in red, and relevant revisions to the manuscript are highlighted in black where applicable.

Reviewer 1:

The manuscript by Zhang et al. (Prominent role of organics in aerosol liquid water content over the south-eastern Atlantic during biomass burning season) reported hygroscopicity/liquid water content of aerosol particles at the eastern Atlantic ocean measured during the ORACLES aircraft campaign. The campaign was conducted during biomass the biomass burning season in Africa. Humidity-controlled nephelometers were employed for estimating water contents. The data were combined with the AMS measurement results and ISORROPIA thermodynamic models for retrieving contributions of organic aerosol on water contents. The topic is within the scope of the interests of readers of the journal. Such an aircraft observation in the region is relatively rare. The research community will be benefitted by the publication of this manuscript. However, the reviewer has some concerns, as detailed below.

Major comments
1. Section 2.1 Aircraft Instrumentation and Data Analysis
The section should provide further details about the measurement. The authors cited Titos et al. (2016) and Zieger et al. (2013) as references for the technique. However, neither of them is for aircraft observations/research groups of the authors. The reviewer believes that the authors developed the humidification setup by themselves, and tested it. All the technical details of the setup should be described in the manuscript, at least as a part of SI. Calibration results of the setup also need to be provided for convincing the readers about the data quality. Both the precision and accuracy of RH should be provided, as they are the key parameters for measuring hygroscopic growth. For comparing the nephelometer data with the AMS results, it is important to make sure that the two instruments measured particles in the same size range. Some information about the inlet design and size-cut of aerosol particles will need to be added for addressing the concern. The authors mentioned that PNSD was measured using the UHSAS and APS. Did the data from these instruments agree reasonably well with the AMS results? The comparison would help checking if chemical species that are unmeasurable by the AMS occupied a substantial fraction of aerosol particles.
We thank the reviewer for the thoughtful comment. We have rewritten Section 2.1 to provide a more detailed description of the measurement setup. Specifically, we have 1. added technical details of the setup of the humidification system (which measures the scattering enhancement factor f(RH)), including its design, calibration, and accuracy, 2. included a description of the inlet of the P-3 aircraft (including the cut-off size), 3. expanded the details

on the measurement of particle size distribution and chemical composition. This section now reads as follows:
"

2.1 Aircraft Instrumentation

We analyse in situ airborne data measured over the SEA region from the ORACLES 2016 and 2018 campaign (Redemann et al., 2021). The ORACLES 2016 and 2018 campaign took place from 31st August to 24th September and from 30th September to 21st October 2018, respectively. All instruments were mounted on the NASA P-3 aircraft. Aerosol particles up to 4.0 μm were introduced into the P-3 via a solid diffuser inlet. More details of the inlet can be found in McNaughton et al. (2007) and Dobracki et al. (2023). The scattering enhancement factor, which was defined as the ratio of scattering coefficients at humidified conditions to those under dry conditions, was measured by a humidification system. The system consists of two Radiance Research M903 integrating nephelometers, RRDry and RRWet. They operated in parallel at 5 lpm in the rear of the plane. The RH in RRDry was not controlled, though the cabin was warmer than ambient, so the RH was typically below 35%. The RRWet RH was maintained at around 80 % RH via a humidity controller upstream (Howell et al., 2006). It has been tested that the RH differences between the inlet and outlet of RRWet for this humidification system were generally within 2%, suggesting a small RH change in the nephelometer (Howell et al., 2006). RH probes in both nephelometers have been corrected based on lab calibrations. Temperature errors are about 0.5 ℃ and RH errors are roughly 3%. Both nephelometers were calibrated in the field with refrigerant R-134A (1,1,1,2-tetrafluoroethane) and with CO_2 before deployment. The nephelometers' stock filters were replaced with Corion CA-550 80 nm bandpass filters for an effective operating wavelength of 540+/-3 nm. Data with scattering coefficients below 10 Mm^{-1} are not included. The final f(RH) used in this study has an average RH of 79 ± 0.5 % for the RRWet and RH < 30 % for the RRDry. The uncertainty of f(RH) was estimated to be ~ 20 % for RH < 85 % (Titos et al., 2016; Zieger et al., 2013).

Non-refractory submicron aerosol compositions were measured by a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS, Aerodyne Research Inc.), and the refractory BC was determined by a single particle soot photometer (SP2, Droplet Measurement Technology). The particle number size distribution (PNSD) was from an ultra-high-sensitivity aerosol spectrometer (UHSAS), which measures particles between 60 and 1000 nm in optical diameter. It was converted to mobility diameter according to Howell et al. (2021). Since supermicron particles account for a minimal fraction (<1.5% for 85% of the time), they were neglected in this study. The aerosol/plume age was simulated with a two-week forecast utilizing the Weather Research and Aerosol Aware Microphysics (WRF-AAM) model (Thompson and Eidhammer, 2014). All measurements were averaged over 15 s and adjusted to STP values at 273.15 K and 1013 hPa. The ALWC has only been calculated when temperature > 0 °C. Measurements from 21 flights, totalling approximately 134 flight hours, were analysed in this study.
"

2. Figure 3
The reviewer does not understand what the color scales for LV-OOA, SV-OOA, BBOA, and HOA mean in the figure. Please clarify it.
L239-240

The reviewer's understanding on the concepts of LV-OOA and SV-OOA is that these categorizations were developed following factor analysis of the aerosol mass spectra. Did the authors conduct factor analysis?

L241

Is this statement for LV-OOA, SV-OOA, or oxidized BBOA?

Since these three comments are all related to the concepts of LV-OOA, SV-OOA, BBOA, and HOA, we have merged them. In this study, the classification of LV-OOA, SV-OOA, BBOA, and HOA was based on the oxidation state (OSc). It was proposed by Kroll et al. (2011, Figure 2) and summarized in Donahue et al. (2012). The specific classification is as follows: −2.0– −1.5 for hydrocarbon-like OA (HOA), −1.75– −0.75 for BBOA, −1.0–0.0 for semi-volatile OA (SV-OOA) and 0.0–1.0 for low volatility OA (LV-OOA). The authors acknowledge that the definition and classification of LV-OOA, SV-OOA, BBOA, and HOA are missing in the manuscript. Therefore, we have added that information in Line 232 (original manuscript):

"

The color scale represents different types of OA based on oxidation state (OSc): −2.0– −1.5 for hydrocarbon-like OA (HOA), −1.75– −0.75 for BBOA, −1.0–0.0 for semi-volatile OA (SV-OOA) and 0.0–1.0 for low volatility OA (LV-OOA). These classifications are proposed by Kroll et al. (2011) and summarized in Donahue1 et al. (2012).

"

And Line 241 (original manuscript):

"

The OSc, defined as $2O/C−H/C$ (Kroll et al., 2011), concentrates around 0.1 for 2016 and -0.7 for 2018, indicating that the majority of the OA belongs to LV-OOA (low volatility OA, OSc between 0 and 1.0) in 2016 and SV-OOA (semi-volatile OA, OSc between -1.0 and 0) in 2018 (Kroll et al., 2011; Donahuel et al., 2012).

"

3. L243

Why does volatility matter for the present study? Please clarify.

Many factors that can influence OA hygroscopicity. Due to the chemical complexity of atmospheric OA, up to now, there is no good representation of OA hygroscopicity. Models usually assume OA to be hydrophobic or give a constant and relatively small κ_{OA} value. Investigation of OA hygroscopicity is still very much in the developmental stage. Nakao et al. (2017) found that volatility variation plays a vital role in OA hygroscopicity. Kuang et al. (2020) rewrote the κ-Kohler theory with an empirical relationship between OA density and O/C and H/C ratios (Kuwata et al., 2012) and found that under the same O/C ratio, OA hygroscopicity is theoretically inversely correlated to OA volatility. However, this relationship does not always hold or can be discernible for particles in the real atmosphere, for example, in this study. Besides, the unique volatility nature of OA in this region compels us to present it in the study. Usually, more aged aerosols are linked with lower volatility, however, in the BB season in SEA, we found the opposite, that more aged aerosols can exhibit a semi-volatile nature, possibly through fragmentation of carbon chains. This unique nature of aged BB aerosols from African fires is worth mentioning, which might be helpful for future studies on OA hygroscopicity. Therefore, we have provided a comprehensive description of OA oxidation and volatility in the manuscript.

4. L247

Kappa_OA of 0.00 is extremely low, suggesting that OA was practically water-insoluble. Is it an accurate number, or is it due to the measurement uncertainties? If the low value is not due to the measurement uncertainty, is the idea supported by the AMS mass spectra observed at the same time (e.g., low O:C or f44)?

The authors believe the very low and small amount of negative values of κ_{OA} come from uncertainty. As can be seen from Fig. S3, those low values do not show a correlation with OA oxidation levels. However, the authors noticed that it would be more convincing if the uncertainty analysis of κ_{OA} could be provided. Therefore, we have added a section on the uncertainty of κ_{OA} in the supplement. Please find below the added section:
"

S1 Uncertainty analysis of κ_{OA}

In this part, we analysed the uncertainty of $\kappa_{f(RH)}$ and κ_{OA}. The Monte Carlo uncertainty analysis is applied here due the calculation complexity.

In the Monte Carlo uncertainty analysis, we used the uncertainty in each input variable to generate an array of scale factors to represent the variability of the variable may have when measured a large number of times. Specifically, we first generated a scale factor array that follows the Gaussian distribution with a mean of 1 and a standard deviation of the uncertainty of each input. The scale factor array was then multiplied by the corresponding input variable to generate an array of variables representing the likelihood of this input variable if it were measured a large number of times, 10000 was used in this study. Variables considered in this analysis include the scattering enhancement factor f(RH), the BC mass from SP2, the mass concentration of submicron non-refractory species from AMS, and PNSD from UHSAS. The uncertainty of f(RH) is 20 % (Titos et al., 2016). For the mass concentration of AMS chemical species, we used the 1-σ uncertainty of 19 % for SO_4, 17 % for NO_3, 18 % for NH_4, and 18 % for OA (Bahreini et al., 2009). An uncertainty of 4% was given to the real part of refractive index of the coating following Taylor et al. (2020). An uncertainty of 30 % was given to PNSD. Input variables and corresponding uncertainties are summarized in Table S1. We chose a straight and level run from the 12th October 2018 flight measurements as it was relatively short and thus relatively fast to calculate.

The $\kappa_{f(RH)}$ and κ_{OA} show uncertainties of 47 % and 68 %, respectively. This large uncertainty of κ_{OA} partly explains the physically unreasonable negative κ_{OA} values. The uncertainty of $\kappa_{f(RH)}$ and κ_{OA} from f(RH) is larger than those from the cloud condensation nulcei, which is measured under supersaturated conditions, or g(RH), which is only for one specific diameter. The hygroscopicity parameters obtained from f(RH) can provide a hygroscopicity for the whole aerosol population under subsaturated conditions. As aerosol hygroscopicity varies with size and RH (Petters and Kreidenweis, 2007), such information is important for direct radiative forcing estimation.

Table S1. Mean and standard deviation of physical parameters involved in the Monte Carlo analysis and relative standard deviations of $\kappa_{f(RH)}$ and κ_{OA}.

	Uncertainty	Instrument/calculation
f(RH)	20 %	Humidification system
NO3 mass	17 %	AMS

SO4 mass	19 %	AMS
NH4 mass	18 %	AMS
OA mass	20 %	AMS
BC mass	20 %	SP2
PNSD	30 %	UHSAS
RH	3 %	RH sensor
$\kappa_{f(RH)}$	47 %	Mie retrieval
κ_{OA}	68 %	ZSR method

"

5. L252

What kind of uncertainties are available in the calculation? Please clarify. Is there a possibility that the issue is induced by measurement uncertainties?

Yes, the authors believe that the 5% negative values can come from the uncertainty of both measurement and calculation. To analyze it, we have added a section on the uncertainty analysis of κ_{OA} in the supplement. Due to the calculation complexity, the Monte Carlo uncertainty analysis is employed. This sentence now reads:

"

The 5 % negative κ_{OA} might be caused by the uncertainty in the calculation (refer to section S1 in the supplement).

"

The added section of κ_{OA} uncertainty can be found in detail in the response to comment 4.

6. L254-L259

Has such a phenomenon been observed for sub-micrometer particles (sub-micrometer particles should be the major focus of the present study, considering that the AMS is used for measuring chemical composition).

According to our TEM analysis, which is reported by Dang et al. (2022), particles with organic shells exist, though are rare. For example, there is one organic particle with potassium and sulfur inclusions in RF06_1 in ORACLES as shown in Table 4 in Dang et al. (2022).

7. L265

It is better to show the corresponding figure, rather than only showing the correlation coefficient.

We have added a figure of κ_{OA} vs. plume age and κ_{OA} vs. $f44$ in Fig. S3 in the supplement. The sentence has been revised as:

"

with a Pearson correlation coefficient of 0.14 with the plume age and 0.04 with f_{44} (Fig. S3).

"

The following figure has been added in the supplement:

"
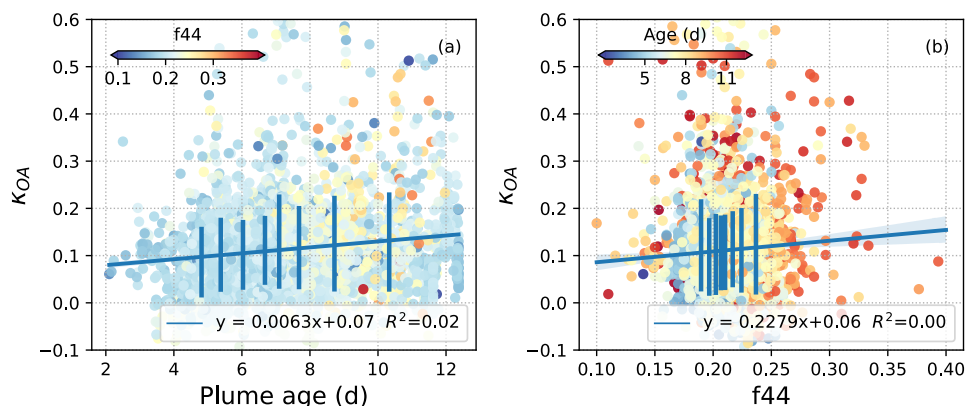
Figure S3. Correlation between κ_{OA} and OA oxidation level – (a) plume age and (b) $f44$ in ORACLES 2016 and 2018 campaigns.

"

8. L267

Why is there a limitation in relating kappa_OA and OA oxidation level? What would be the thermodynamic reason behind it?

There are many possible thermodynamic reasons. As organic aerosols undergo oxidation, there will be more chances for polar functional groups (e.g., hydroxyl, carboxyl) to be introduced into the molecules, enhancing their ability to attract and absorb water, thereby increasing OA hygroscopicity. However, The oxidation process can produce a wide range of functional groups, not all of which contribute equally to water uptake. Some oxidation products may form oligomers or highly functionalized low-volatility species, may have limited solubility in water, and contribute minimally to hygroscopic growth. This complexity means that a higher OA oxidation level does not straightforwardly translate to increased κ_{OA}. In addition, solubility determines whether a compound will partition into the aqueous phase of an aerosol particle. This phase state of OA (liquid, semi-solid, or solid) also affects water diffusion and therefore, hygroscopic growth. Besides functional group and solubility, surface activity and environmental parameters such as temperature and relative humidity all has an impact on OA hygroscopicity, highlighting the complexity of linking κ_{OA} to the OA oxidation level.

9. L275

The paragraph which starts from L275 was unclear to the reviewer, as various types of kappa appeared without clear definitions. As a result, what the authors are trying to mean by Figure 4 was also unclear to the reviewer. Please update the corresponding descriptions.

The description of various kappa have been provided in Line 126-140. However, we have noted that they can be more clearly defined. Therefore, we have revised section 2.2, and more clear information on the definition and calculation is included. The revised part now reads:

"

2.2 Calculation of hygroscopic related parameters - $f(RH)$, $\kappa_{f(RH)}$ and κ_{OA}

The aerosol scattering enhancement factor, $f(RH)$, is calculated as:

$$f(RH) = \frac{\sigma_{sp}(RH)}{\sigma_{sp}(RH_{dry})},\tag{1}$$

where $\sigma_{sp}(RH)$ and $\sigma_{sp}(RH_{dry})$ represent the scattering coefficients at humidified and dry RHs (RH and RH_{dry}), respectively. Previous studies usually take RH_{dry} as 30-40 % assuming aerosols are dry at/under such RHs (Burgos et al., 2019; WMO/GAW, 2016; Titos et al., 2016). In this study, to facilitate comparison with previous studies and minimize the influence of water, we only included the $f(RH)$ with RH_{ref} equal to or smaller than 35 %.

Aerosol hygroscopicity parameter κ can be retrieved from $f(RH)$ (Zieger et al., 2010), which is usually denoted as $\kappa_{f(RH)}$. It represents the hygroscopicity of the overall aerosol population and can be considered as the scattering coefficient weighted average κ (Kuang et al., 2021). The detailed procedure of $\kappa_{f(RH)}$ retrieval can be found in Chen et al. (2014). The Python package PyMieScatt (Sumlin et al., 2018), an implementation of Mie theory, was applied in this study to retrieve $\kappa_{f(RH)}$. For PM_1 (particulate matter with an aerodynamic diameter of less than 1 μm), $\kappa_{f(RH)}$ is proven to be a good representation of κ_{chem}, which is the κ that is calculated from various chemical compositions following the ZSR (Zdanovskii–Stokes–Robinson) mixing rule (Petters and Kreidenweis, 2007).

Therefore, the hygroscopicity of OA, κ_{OA}, can be calculated as

$$\kappa_{OA} = \frac{\kappa_{f(RH)} - \sum_{inorg} \kappa_i \varepsilon_i}{\varepsilon_{OA}},\tag{2}$$

where κ_i and ε_i represent the hygroscopic parameter and volume fraction of the inorganic component i. The κ and density of inorganics can be found in Table S2 in the supplement. The detailed calculation can be referred to Zhang et al. (2024).
"

We have also added a table of the hygroscopicity parameter and density of inorganics, OA, and BC in the supplement:
"

Table S2. Density (ρ) and hygroscopicity parameter (κ) of inorganics, OA, and BC used in this study.

	$(NH_4)_2SO_4$	NH_4HSO_4	NH_4NO_3	KCl	OA	BC
ρ (g cm^{-3})	1.77 (Lide, 2008)	1.78 (Lide, 2008)	1.72 (Lide, 2008)	1.98 (Kuang et al., 2021)	1.4 (Alfarra et al., 2006)	1.8 (Bond and Bergstrom, 2006; Liu et al., 2017)
κ	0.47 (Topping et al., 2005; Gysel et al., 2007; Kim et al., 2020)	0.56 (Kuang et al., 2021)	0.58 (Topping et al., 2005; Gysel et al., 2007; Kim et al., 2020)	0.89 (Kuang et al., 2021)	n/a*	0 (Kuang et al., 2021; Topping et al., 2005; Gysel et al., 2007)

* κ_{OA} is determined in the study.

"

10. L281
What kind of mixing state was assumed? Could it really happen under the ZSR approximation?
In the retrieval of $\kappa_{f(RH)}$ and calculation of κ_{OA}, the ZSR (Zdanovskii–Stokes–Robinson) mixing rule was applied. The ZSR rule is a widely used approximation in climate modeling due to its simplicity and computational efficiency. However, its accuracy depends on the mixing state of the aerosols, which can vary based on many factors, such as the aging process.

For aerosols in the south-eastern Atlantic (SEA) region during the BB season, Zhang et al. (2022) suggest that the homogeneous mixing assumption is generally appropriate, except for aerosols with extremely thick coatings. Given this finding, we consider homogeneous mixing to be the most suitable approach for this study.

11. L288-289
The description about the phase state of aerosol particles is highly speculative. Do the authors have any data to support the statement?
The authors acknowledge that the description of the phase state is speculative, as direct measurements of aerosol phase state were not available in this study. However, our assumption of a metastable state is based on several atmospheric factors that influence aerosol water uptake and retention.

These aerosols originate from African biomass burning, a process that emits not only large amounts of particulate matter but also substantial water vapor. During transport, these particles are further influenced by the African Easterly Jet, which carries additional moisture. Under such conditions, aerosols can initially take up water and transition into a metastable liquid state.

Once water uptake occurs, it is generally difficult for aerosols to return to a completely dry state due to several mechanisms. For example, the Efflorescence Relative Humidity (ERH) constraint: the RH must drop well below the deliquescence RH for crystallization to occur, and even then, particles must overcome a nucleation energy barrier to transition into a solid state. Besides, some organic coatings can stabilize the liquid state by suppressing water evaporation. Considering these factors, we speculate that the presence of solid particles should be minimal.

12. Figure 5
The sums of values in the PDF are larger than unity. How was the data normalized?
We thank the reviewer for the careful examination of Figure 5. We have recalculated the sum of the probability density function (PDF) values and confirmed that it sums to unity.

The observed discrepancy may arise from the logarithmic scale used on the x-axis. Since we used a log-scale histogram, the bin widths are not the same, which can give the impression that the total exceeds unity when viewed visually. We appreciate the reviewer's diligence in checking this, and we have double-checked the calculation to ensure accuracy.

13. L311

Would it be possible to use the AMS or size distribution data here for representing mass loading?

It is possible to use the aerosol mass from AMS or UHSAS to represent the aerosol loading. However, this study is based on the measurement of scattering coefficients. Therefore, we found it is more relevant to use aerosol scattering to represent the aerosol loading in this study.

14. L316-322

It is better to show the corresponding figures, rather than only showing the correlation coefficients.

We thank the review for the comment. We have added the figure in the supplement. Now the sentence reads:

"

We observed a positive correlation between relative humidity (RH) and ALWC with a Pearson correlation coefficient of 0.7 (Fig. S4a). The Aerosol dry scattering coefficient shows a much less positive correlation with a Pearson correlation coefficient of 0.3 (Fig. S4b).

"

The figures shown below have been added in the supplement:

"
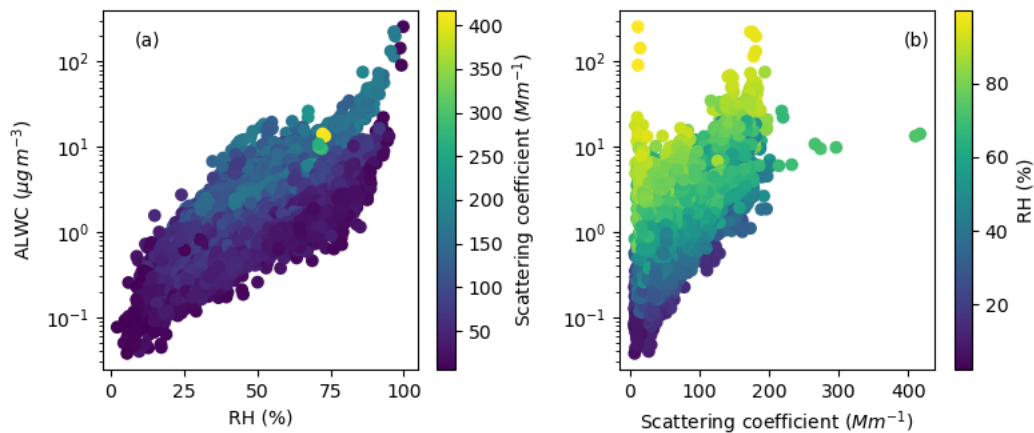
Figure S4. Correlation between ALWC and (a) RH and (b) scattering coefficient.

"

15. Figure 6

The reviewer was confused to see this figure, as the reviewer thought that RH of the nephelometer was fixed during the observation. Is it an estimation that employed the thermodynamic model and kappa_OA? If so, do the authors have any evidence to support the idea that kappa_OA does not change with RH?

The ratios, aerosol liquid water content (ALWC), and particle mass shown in Figure 6 are all under ambient conditions. Specifically, the ALWC and ALWC$_{OA}$ were calculated following equations 3 and 4, using ambients RHs. The ALWC$_{Inorg}$ is from ISORROPIA II,

where the ambient RH is one of the inputs. This figure shows the important contribution of OA to ALWC for aerosols during biomass burning season in the SEA region.

16. L350

What does 'hydrated' mean here? Does it imply formation of hydrates, or formation of aqueous phase following hygroscopic growth?

The authors realize that the word "hydrate" is not accurate, therefore, we revised the sentence and now it reads as follows:

"

However, Jin et al. (2020) assumed a stable state, where the ISORROPIA-II algorithm starts by assuming a completely dry particle. As RH increases, the algorithm sequentially dissolves each salt based on its DRH (Fountoukis and Nenes, 2007). In reality, particles modeled as solid below their DRH might not be completely dry in the atmosphere. For example, for particles that have already absorbed water before being exposed to low RHs, the RH needs to be below the ERH, rather than the DRH, for the particles to transition into a solid state. This discrepancy can lead to an underestimation of $ALWC_{Inorg}$ and, consequently, an overestimation of the ratio $ALWC_{OA}/ALWC$, as discussed in Jin et al. (2020).

"

17. L361-364

Jin et al. (2020) and Gao et al. (2015) observed totally different types of air masses than the present study. Is there any reason why the authors selected these two references from the numerous literature about hygroscopicity?

The authors compared the contribution of organic aerosol (OA) to aerosol liquid water content (ALWC) in this section. To our knowledge, limited studies are reporting on this specific topic, and we have included a substantial portion of the relevant literature in our comparison. The studies by Jin et al. (2020) and Gao et al. (2015) were specifically highlighted because they reported a significant contribution of OA to ALWC, which is among the highest observed globally. Our findings, which show values slightly higher or comparable to theirs, further emphasize the importance of OA in this region. However, the authors acknowledge that the significance of this high contribution was not sufficiently emphasized in the original manuscript. Therefore, we have revised the text to better highlight this point. Now the sentence reads:

"

The mean contribution of $ALWC_{OA}$ to the total ALWC in both campaigns was around 38±16 %, a value notably higher than what is commonly reported in many studies. For instance, some studies suggest that the water uptake of aged organics accounts for only a few percent of the total aerosol water uptake (e.g., Gysel et al., 2007; Engelhart et al., 2011), which contrasts with our findings. Carlton and Turpin (2013) and Hennigan et al. (2008) even reported negligible OA-related ALWC in the US and Africa, respectively. In contrast, Li et al. (2019) found a significant fraction of $ALWC_{org}$ to ALW in Beijing, ranging from 18 to 32 %. Similarly, Jin et al. (2020) reported a contribution of 30±22 % in Beijing, and Guo et al. (2015) obtained a comparable value of 35 % in the Alabama forest and the south-eastern US. These values represent some of the highest contributions of OA-related ALWC reported globally. Our results align closely with these high-end observations, highlighting

the substantial role of OA in driving ALWC in this study, which has important implications for radiative forcing and aerosol-cloud interactions in the SEA region during the BB season. ”

Minor comments

1. L61 thermal equilibrium

Thermal equilibrium or thermodynamic equilibrium?

We have changed it to thermodynamic equilibrium.

2. L139

The values of the densities that were used for the study should be provided in the manuscript, rather than referring a previous manuscript by the authors.

We have added a table of the density and hygroscopicity parameters of inorganics, OA, and BC in the supplement:

“

Table S2. Density (ρ) and hygroscopicity parameter (κ) of inorganics, OA, and BC used in this study.

	$(NH_4)_2SO_4$	NH_4HSO_4	NH_4NO_3	KCl	OA	BC
ρ (g cm^{-3})	1.77 (Lide, 2008)	1.78 (Lide, 2008)	1.72 (Lide, 2008)	1.98 (Kuang et al., 2021)	1.4 (Alfarra et al., 2006)	1.8 (Bond and Bergstrom, 2006; Liu et al., 2017)
κ	0.47 (Topping et al., 2005; Gysel et al., 2007; Kim et al., 2020)	0.56 (Kuang et al., 2021)	0.58 (Topping et al., 2005; Gysel et al., 2007; Kim et al., 2020)	0.89 (Kuang et al., 2021)	n/a*	0 (Kuang et al., 2021; Topping et al., 2005; Gysel et al., 2007)

* k_{OA} is determined in the study.

”

3. L240 LV-OOA/SV-OOA

These abbreviations are not defined in the manuscript.

Added.

4. L279

Figure 4a does not exist.

We have changed it to Figure 4.

5. L309

What does '38% of the RH' mean? RH = 38%?

Sorry for the confusion. We have changed it to

“

38% of the RH values were below 30%.

”

6. L344
What does 'the compound state' mean?
We have changed it to aerosol state, i.e. physical state of aerosols.

Reviewer 2:

This study investigated the contribution of organics to ALWC during the 2016 and 2018 ORACLES campaigns which was conducted over the south-eastern Atlantic Ocean during the biomass burning season. Although the observation data is valuable, I don't think this study is worth publishing as an independent manuscript, as many of the main findings is already implied in the authors' another study (Zhang et al., 2023, https://doi.org/10.5194/egusphere-2023-2199). Trying to split one study into too many pieces heavily diluted the creativity of this study, as detailed below.

(1)   In the abstract, the authors highlighted three major conclusions. The first one, that there's high contribution of ALWC from organics during the biomass burning, is worth reporting but not surprising. However, the other two points, that the hygroscopicity of OA are positively correlated with ALWC_OA/ALWC and that hygroscopicity of OA can vary much, are not surprising at all. Actually, they're well recognized fact and should be applied to explain some other observed phenomenon.

Thank you for your thoughtful comment. We acknowledge that the positive correlation between OA hygroscopicity and $ALWC_{OA}/ALWC$, as well as the variability in κ_{OA}, are well-recognized concepts. However, studies have been suggesting a small sensitivity of κ_{OA} to global climate forcing and the adequacy of a constant κ_{OA} value (Pöhlker et al., 2023). We emphasize that our study contributes by demonstrating the impact of these factors in a climatically significant region where such analyses have been limited. Our findings highlight two key conclusions:

1. Prominent OA Contribution to ALWC: Aerosols in the south-eastern Atlantic (SEA) region during the biomass burning (BB) season exhibit a notably high OA-related ALWC contribution, among the highest reported globally. Given that ALWC plays a crucial role in aerosol direct radiative forcing, this finding underscores the importance of accurately estimating OA-related ALWC in climate models.
2. Need for Real-Time κ_{OA} in ALWC Estimation: Current models often assume OA as hydrophobic or assign it a constant κ_{OA}, potentially leading to substantial biases. Our comparison between $ALWC_{OA}$ derived from real-time κ_{OA} and that obtained using a campaign-mean κ_{OA} reveals a significant discrepancy. While we have not yet identified a robust parameterization for κ_{OA} in this region, our findings emphasize that using a constant κ_{OA} may introduce a major source of uncertainty in climate models, particularly in the SEA region during the BB season.

We recognize that our original abstract may not have clearly emphasized these key points. To enhance clarity, we have revised the abstract (original manuscript Line 28-39) as follows:
"

Notably, the $ALWC_{OA}$ accounts for 38±16 % of the total ALWC, a remarkably high contribution of OA-related ALWC that aligns closely with some of the highest values reported globally. This underscores the significance of OA in ALWC and, therefore, its role in aerosol direct radiative forcing in this climatically significant region. The strong correlation between κ_{OA} and $ALWC_{OA}/ALWC$ ($R^2 = 0.72$) underscores the necessity of accurately estimating κ_{OA} for reliable ALWC calculations. The substantial difference in $ALWC_{OA}$ when using real-time versus campaign-mean κ_{OA} further demonstrates the limitation of assuming a constant κ_{OA}, a common practice in climate models. These findings reinforce the need to account for the variability in OA

hygroscopicity, as it has important implications for aerosol-water interactions and direct radiative forcing, particularly in the SEA region during the BB season.
"

(2)   I was expecting that more pattern analysis of OA hygroscopicity during the campaign would be provided, but then realized that those are the main part of Zhang et al. (2023), and Fig. 2 in this study actually expressed the same idea as the Fig. 5 in Zhang et al. (2023). Similarly, major findings in section 3.2 is already somehow implied and discussed in Zhang et al. (2023).

The authors thank the reviewer for the time and effort in reading these two manuscripts. We acknowledge that there are similarities between this manuscript and Zhang et al. (2024) regarding the discussion of OA hygroscopicity. To minimize overlap and enhance clarity, we have carefully revised both manuscripts to ensure each study maintains a distinct focus.

Zhang et al. (2024) primarily examine the hygroscopicity of total aerosols, including vertical variation, influencing factors, and parameterization during the BB season in Africa. While the vertical trend of OA hygroscopicity is discussed in that study, this is done in the context of total aerosol hygroscopicity and its subsequent parameterization. In contrast, this study places greater emphasis on the factors influencing κ_{OA}, specifically focusing on its variability and its relationship with OA oxidation levels. Given that vertical variations have already been addressed by Zhang et al. (2024), we have shifted our emphasis to a more detailed exploration of the patterns and drivers of κ_{OA} variability. We have rewritten this part and provided additional pattern analysis of OA hygroscopicity during the campaign in the revised manuscript. Now this section reads:
"

The vertical distributions of κ_{OA} in both years' campaigns have been discussed by Zhang et al. (2024). This study focuses on the factors influencing κ_{OA}. The relationship between OA oxidation levels and κ_{OA} has been widely reported, with many studies finding significant positive correlations between κ_{OA} and OA oxidation markers such as f_{44} and O/C (Kuang et al., 2020b; Lambe et al., 2011; Mei et al., 2013). Li et al. (2019) used O/C to estimate κ_{OA} and further calculated ALWC for total aerosols. However, such a relationship was not consistently observed across all flights in our study, except for RF03Y18 (which took place on 7th October 2018).

For RF03Y18, κ_{OA} exhibited the highest Pearson's correlation with altitude (r=-0.82), followed by temperature (r=0.79), plume age (r=0.72), and $f44$ (r=0.64). The strong negative correlation between κ_{OA} and altitude suggests that altitude-related factors significantly influence OA hygroscopicity. While plume age remains an important factor, its lower correlation with κ_{OA} indicates that additional altitude-related factors, independent of aging, may also play a role. The even weaker correlation with $f44$ suggests that the addition of CO_2^+ alone does not fully capture the complexity of OA hygroscopicity changes during aging, other aging processes also contribute.

In most other flights from both campaigns, no single dominant factor was identified as the primary driver of OA hygroscopicity, suggesting a more complex interplay of multiple processes. While oxidation level and plume age have been widely linked to OA hygroscopicity, their relatively weak correlations across most flights imply that additional processes must be considered. One possible explanation is fragmentation during oxidation. Functionalization typically leads to higher hygroscopicity, whereas fragmentation may reduce it. In this study, evidence of fragmentation was found (this study, Dang et al. (2022), Dobracki et al. (2023)). In highly oxidative environments, fragmentation can lead to the formation of volatile or semi-volatile compounds that may evaporate from the particle, potentially decreasing hygroscopicity if the

remaining material is less water-soluble. Such effects could explain the lack of a clear trend between OA hygroscopicity and aging. These findings underscore the need for a more comprehensive understanding and improved representation of OA hygroscopicity, particularly during the BB season in the SEA region.
"

Since the vertical distribution of κ_{OA} is not the focus of this manuscript, we have removed it from Figure 2:
"
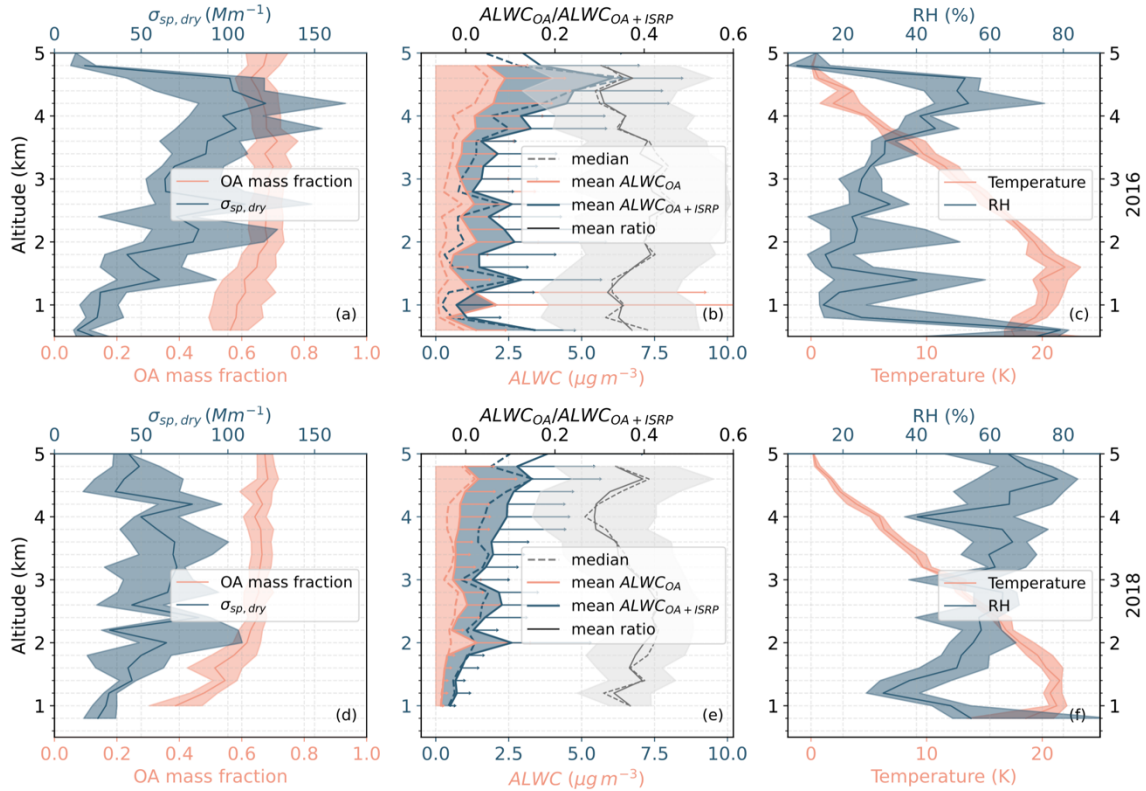
Figure 2. The vertical distributions of various aerosol properties. (a, d) Vertical distributions of OA mass fraction (pink) and scattering coefficient under dry conditions (blue) in the 2016 and 2018 ORACLES campaigns, respectively. The lines and shades represent the mean and standard deviation in every 200 m bin, respectively. (b, e) Vertical distributions of OA-related liquid water content $ALWC_{OA}$ (pink), the aerosol liquid water content $ALWC_{OA+ISRP}$ (blue), calculated as the sum of $ALWC_{OA}$ and $ALWC_{inorg}$ from ISORROPIA-II model, and the fraction of $ALWC_{OA}$ in $ALWC_{OA+ISRP}$ (grey) in 2016 (b) and 2018 (e) ORACLES campaigns, respectively. The solid and dashed lines represent the mean and median in every 200 m bin, respectively. Error bars and shades represent 1 standard deviation. (c, f) Vertical variations of ambient temperature (pink) and RH (blue) in 2016 (c) and 2018 (f) ORACLES campaigns, respectively. The lines and shades represent the average and standard deviation in every 200 m bin, respectively.
"

(4)   The Fig. 1 in this study is the same as in Zhang et al. (2023), which needs to be clarified.

The authors acknowledge that the original Figure 1 closely resembles the one in Zhang et al. (2024) and was not optimally suited for this manuscript. To better align with the focus of this study, we have revised the figure to enhance its relevance and clarity. Specifically, we have: 1. added the countour of u-wind at 600 hPa, providing a clearer representation of the easterly transport of aerosols. 2. Incorporated the ambient RH along the flight tracks, due to its importance in this study. Please find the revised Figure 1 in below:
"
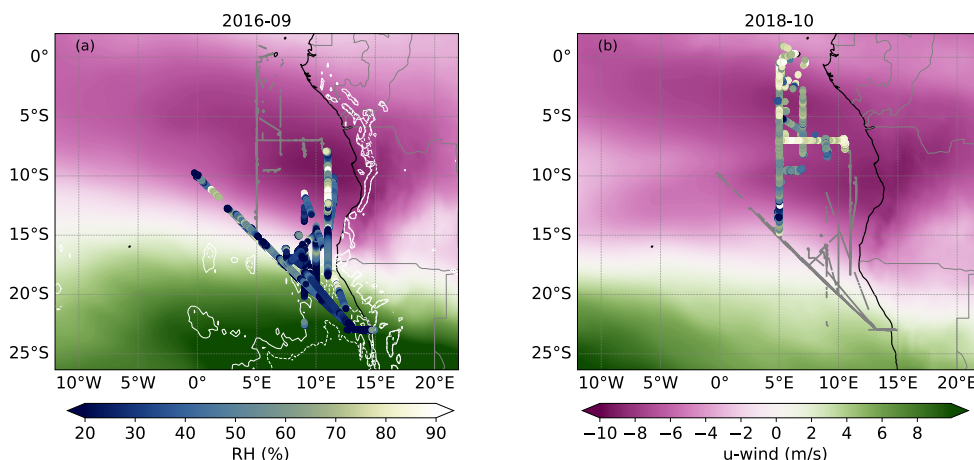
Figure 1. Maps of the (a) September mean and (b) October mean zonal winds at 600 hPa and flight tracks colored by the ambient RH in 2016 (a) and 2018 (b) ORACLES campaigns, respectively. The easterly transport north of around 15° is evident. Flight tracks in grey are drawn for reference. White contours in (a) are the 2016 September mean vertical velocity, omega, at 800 hPa. Solid and dashed lines represent the subsidence of 55 and 65 hectopascals per day (hPa d^{-1}), respectively.
"

(5)   The comparison as shown in Fig. 4 is kind of circular reasoning. As the kappa_OA is derived from the kappa_f(RH) and kappa_INORGANICS, it was not surprising at all that the ALWC_f(RH) agrees well with the ALWC_ISOR+OA. In fact, the good agreement would only prove that the kappa of inorganic species is well represented in ISORROPIA -- which has already been proved during the validation of ISORROPIA.

Thank you for your insightful comment. Since κ_{OA} is derived from $\kappa_{f(RH)}$ following the ZSR rule, according to the definition of κ proposed by Petters and Kreidenweis (2007), the comparison between $ALWC_{\kappa_{f(RH)}}$ and $ALWC_{OA+ISRP}$ primarily evaluates the water content predicted by ISORROPIA II for inorganic aerosols against the water content estimated for inorganic aerosols using the method outlined in Petters and Kreidenweis (2007, Eq. 3). For the calculation of inorganic aerosol water content, the κ values for individual inorganic species are taken from the literature, while their volume fractions are determined using a modified ion-pairing scheme (Zhang et al., 2022; Gysel et al., 2007). The strong correlation observed in our results provides further validation of both the κ values applied and the ion-pairing scheme used in our analysis.

However, it is important to note that good agreement between these two methods is not always expected, as discrepancies commonly arise, particularly under low RH conditions. ISORROPIA II

has been primarily validated for RH > 60% (Fountoukis and Nenes, 2007), whereas notable deviations have been reported at lower RHs. Jin et al. (2020) similarly found a strong correlation between the two methods for RH > 60%, yet observed discrepancies at RH < 60%, with an overall coefficient of determination R^2 of 0.92. In contrast, our study shows an exceptionally high R^2 of 0.99. This exceptionally high correlation strengthens our confidence in the accuracy of the inorganic aerosol water content estimation, ultimately leading to lower uncertainties when calculating the organic aerosol liquid water content. The reduced uncertainty enables a more robust estimation of the organic aerosol liquid water content. However, we acknowledge that this explanation was not clearly articulated in the original text. To clarify this point, we have rewritten the relevant section in the revised manuscript.
"

In this section, we examine the aerosols and OA-related liquid water content. First, we compare the ALWC obtained from the aforementioned two methods, $ALWC_{\kappa_{f(RH)}}$, calculated from $\kappa_{f(RH)}$, and $ALWC_{OA+ISRP}$, which is the sum of $ALWC_{inorg}$ from ISOPPROPIA-II and $ALWC_{OA}$ calculated from κ_{OA}. Both methods regard BC as hydrophobic and assume aerosols to stay in a metastable state. Since κ_{OA} is derived from $\kappa_{f(RH)}$ following the ZSR rule, the comparison between $ALWC_{\kappa_{f(RH)}}$ and $ALWC_{OA+ISRP}$ essentially evaluates the consistency between the water content predicted by ISORROPIA II for inorganic aerosols and the water content for inorganic aerosols calculated using the method in Petters and Kreidenweis (2007, Eq. 3). Figure 4 illustrates the comparison of $ALWC_{\kappa_{f(RH)}}$ and $ALWC_{OA+ISRP}$, which shows an excellent agreement with an R^2 of 0.98. This high consistency between the two methods provides a robust baseline for estimating OA-related ALWC.
"

(6)   Overall, I suggest merging this study into other companying studies, otherwise the major points and unique opinions of this study needs to be reframed and highlighted more clearly.

We appreciate the reviewer's feedback and understand the concern regarding the distinct contribution of this study. In response, we have carefully revised and refined the manuscript to clearly highlight its unique findings and ensure that it presents a distinct perspective from the accompanying studies. Specifically, we have 1. Clarified the study's primary focus on the high contribution of OA-related ALWC and the necessity of accurately estimating κ_{OA}. 2. Refined the text to explicitly distinguish this study's scope from the related work.

We hope these revisions have addressed the reviewer's concern. However, we remain open to further refinement and are happy to make additional improvements if needed.

Refence in the response:

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