

Item-by-Item Responses to Reviewers

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We sincerely thank both reviewers for their time, effort, and insightful comments, which have greatly helped improve our manuscript. We have carefully considered all feedback and made the recommended revisions.

Below, we provide detailed, itemized responses to each comment. In this response document, reviewer comments are highlighted in light **blue**, our responses are in **black**, and the corresponding revisions made to the manuscript are shown in **green**.

Reviewer #1

The paper has been improved significantly after addressing the reviewers' comments point to point. Regarding this, there are still few things I'm not sure I have understood well. Once these issues are concerned, this manuscript can be accepted for publication.

Many thanks for your feedback. We have carefully considered all of your suggestions and have addressed them in the revised manuscript.

1. Line 135-136, why the particle size of 20-35 nm is defined and addressed here? In my opinion, the aged biomass burning aerosols and sea salt aerosols should be larger.

In this section, we aim to estimate the critical particle diameter at which aerosols activate as CCN at 1% supersaturation. This critical diameter represents the smallest particle size expected to activate, allowing for a more accurate comparison between CCN and integrated aerosol concentrations measured by SMPS.

We determined this critical diameter using κ -Köhler theory. At 1% supersaturation, particles as small as 20 nm would require an unrealistically high κ value (~ 2.24) to activate. Conversely, a critical diameter larger than 40 nm would suggest an too low κ value (0.2). Such low κ values do not align with the aerosol composition influenced by marine sources (e.g., DMS).

Therefore, we constrained the critical activation diameter within a realistic range of 20–40 nm. Within this range, we selected $\kappa = 0.6$ as representative of sulfate-dominated aerosols, leading to a calculated critical activation diameter of 31 nm. Sensitivity tests (detailed in Supplementary Notes 1) demonstrated that choosing a lower κ value of 0.3—reflecting increased organic aerosol content in this size range—had negligible impact on our results.

In summary, we adopted $\kappa = 0.6$ as a representative aerosol hygroscopicity value, allowing us to define a justified critical activation diameter (31 nm) for integrating SMPS data and

comparing it with CCN measurements at 1% supersaturation. The manuscript has been revised accordingly to avoid confusion.

we assigned a representative κ of 0.6 for particles in the lower part of the Aitken mode range ($\sim 20\text{--}40\text{ nm}$)—similar to that of sulfate aerosols.

2. Line 175: This numbers of closure procedure steps should be revised to avoid confusion, as numbers 1–4 are already designated for primary headings.

Thanks for the comment. We've replaced the numeric labels for the closure-procedure steps with lettered labels (a–e) to avoid conflicting with the primary heading numbers.

3. Line 185, please clarify why the criterion of 0.02 is chosen?

We consider κ_{CCN} and κ_{chem} to be in agreement whenever $|\kappa_{CCN} - \kappa_{chem}| < 0.002$, where 0.002 represents the uncertainty from calculations. Over the full campaign, κ_{CCN} ranged from 0.2 to 0.8, so this threshold corresponds to roughly 1% of the lowest measured κ value (0.2).

To clarify, we revise the text as the follow:

If $\kappa_{residual}$ exceeded 0.02 (equivalent to 1% of the lowest campaign-wide κ_{CCN}), small increments of NaCl volume (1% of the total non-NaCl aerosol volume) were iteratively added. After each addition, κ_{chem} was recalculated, and the $\kappa_{residual}$ was re-evaluated.

Reviewer #2

They authors provide a thorough revision of their original manuscript. My two main points of criticism, the omission of the contribution of sea salt and the counting discrepancy between CCN counter and SMPS, have been addressed.

The counting discrepancy and its correction was another critical comment in my previous review. The clarification in the manuscript text and the additional information in the Supplement Notes provide the necessary background to understand how this instrument artefact is handled and what impacts can be expected on the reported kappa values. Further, the detailed monthly data, the comparison with other instruments measuring particle concentrations, and the analysis of the temporal evolution of the artefact show that while the instrument performance was suboptimal, the true differences between the seasons and the two years will still be visible above this bias. i.e., the authors showed that the observed differences cannot be explained by the instrument artefact.

The inclusion of the thorough analysis of the role of sea salt in addition to the biomass burning related effects has now shifted the conclusions of this study. While there is a lack of direct measurements due to the set of instruments, the authors go now to some length to estimate the sea salt concentration as well as one can. They also present ranges/uncertainties to show how their approximations/assumptions would affect the interpretation. This is highly commendable and leads to a much more robust manuscript where the interpretations and claims are now well

supported. The overall conclusion, that the CCN activity of aerosol particles in the SEA region are affected by the interplay of sea salt from the ocean and sulphate and organic aerosol mainly from biomass burning on the African continent are very useful for the atmospheric science community.

One remaining weak point lies with the unfortunate absence of composition measurements for 2016. Using the MERRA-2 reanalysis data can serve as a proxy to detect general differences. The findings highlight the need for direct NaCl/sea salt measurements even under BB dominate conditions in such marine environments.

In the new section 3.6, one new question arose for me:

The MERRA-2 data in Fig 7 shows that there are clear differences in sea salt fraction between 2016 and 2017. But in Section 3.6, the authors assume that the sea salt fraction is constant between the years. This seems to be a contradiction.

Once this very minor clarification has been provided, I recommend prompt publication.

We sincerely thank the reviewer for the thoughtful feedback and constructive comments, which have significantly improved this manuscript. We fully agree that, despite the dominance of biomass-burning aerosols, direct measurements of aerosol chemical composition—including sea salt—are essential in marine environments.

Regarding to Section 3.6: because no composition measurements are available for 2016, Section 3.6 relies on the 2017 regression between sulfate mass fractions with and without NaCl to infer the total (NaCl-containing) sulfate fraction for 2016. This implicitly treats the sea-salt fraction as unchanged from one year to the next. As the reviewer notes, the observations instead show higher sea-salt loadings in 2017. Extra sea salt in 2017 would dilute the sulfate fraction, so our method likely underestimates the 2016 sulfate fraction.

Most of the sea-salt, however, appears in the $PM_{2.5}$ size range, whereas our analysis is confined to PM_1 aerosol. The sea-salt change should therefore be less pronounced—and the associated bias smaller—in PM_1 . Because the uncertainty bounds in Section 3.6 were derived from conservative upper and lower shifts of the 2017 sulfate-BC/ ΔCO line, we are confident that bias introduced by inter-annual sea-salt variability remains within the stated uncertainty range.

To clarify this, we have revise the text as below:

By comparing these estimated sulfate mass fractions between 2016 and 2017, we quantified the year-to-year differences. Multiplying these differences by the hygroscopicity of sulfate ($\kappa = 0.6$) enabled us to estimate the potential contribution of sulfate variations to the observed κ differences between the two years. This procedure probably underestimates the 2016 sulfate fraction, because Figure 7 suggests sea-salt were higher in 2017, which would dilute sulfate in that year relative to 2016. Nevertheless, the sea-salt increase is less pronounced in PM_1 than in $PM_{2.5}$, so the bias in our PM_1 -based estimate is likely within the uncertainty range evaluated below.