Second review of Coral reef exposure increases aerosol and cloud condensation nuclei over the Great Barrier Reef, Sulo et al.,

Thank you for the revised manuscript as well as detailed responses to my comments. I consider that the paper can be published after some minor comments are addressed. In general, the added text requires editing, to improve readability. There are some errors and repetition of words.

We thank the reviewer for their detailed comments on the revised manuscript, and we appreciate the thorough comments on the improvements on readability. We have added supplementary analysis based on HYSPLIT relative humidity and precipitation to strengthen the argument of cloud processing frequently occurring in the marine air masses arriving to the reef. Further, we have edited the text to improve readability and flow. Detailed answers to each comment can be found below.

Page 2, 164 leading to "an' increase in CCN concentration"s"

We have amended the "an" as per suggestion: "*leading to an increase in CCN concentration"*

Page 3, 250 (and again page 12, section Cloud processing): For the third objective. Cloud processing. I consider that basing this analysis only on particle size distributions is difficult. This bi-modal distribution can also exist depending on the emission of primary and secondary sources.

As part of your HYPLIT analysis, or through ECMWF analysis, it is possible to determine the % of time that the airmass was exposed to cloud liquid water (or ice water). This would provide a more robust indicator of the time the airmass spent in cloud.

We agree that size-distribution bimodality alone can be unconvincing. We therefore added an independent HYSPLIT-based cloud/precipitation-history analysis. It supports frequent cloud exposure and shows physically consistent differences in aerosol populations. We have also softened our language because neither method uniquely establishes cloud processing. We did not add the substantially more involved ECMWF LWC trajectory analysis. We have explicitly added: *"However, bimodality alone cannot uniquely identify cloud processing, as distinct primary and secondary aerosol populations can also produce multimodal size distributions. To independently assess whether the observed size-distribution structure was consistent with recent cloud exposure, air-mass histories were classified using HYSPLIT relative humidity and precipitation along the preceding 24 h of each trajectory (Isokääntä et al., 2022). Overall, 66.9% of air masses experienced precipitation, while a further 1.5% encountered non-precipitating cloud conditions; 31.6% showed neither (Table S2). Air masses with precipitation history exhibited lower total, Aitken-mode, accumulation-mode and CCN concentrations, consistent with aerosol removal by wet scavenging (Fig.S8). These* *results provide independent meteorological support for frequent cloud processing of aerosol reaching the GBR, although the trajectory-based classification and Hoppel minimum represent complementary rather than equivalent indicators of cloud history."*

Page 3 254: Repetition of line 250.

We have removed the repetition "*Lastly, we investigate variability in cloud processing and local aerosol particle sources over the GBR*"

Page 4, 305: repetition of the word different in the same sentence. A reorganization of this sentence is necessary.

We have modified the sentence to be : "*These campaigns have large spatial variability along the reef, and varying parts of the reef have been monitored over different years (Fig. S1)"*

Page5, 390: Please define what "agree fairly well" is in terms of correlations.

We have added a correlation coefficient along with a reference to specify how well the two instruments correlate temporally: *"The two instruments have been shown to agree fairly well in temporal variation (R2 = 0.93, Marto et al. 2020)"*

Page 6, 458: are assumed to "be" active as...

We have amended the text as per the suggestion to: "*particles are assumed to be active as the CCN*"

Page 6, 467: Both critical diameter and CCN activation ratio can only be calculated whenever data is available. This information was already mentioned in line 461.

To streamline the text and avoid repetition, we have amended the text to: *"CCN activation ratio is defined as the ratio between CCN and total aerosol number concentrations."*

Page 6, 469: For consistency, "only" the CCN...

We agree with the reviewer and have edited the text to: "*For consistency, only the CCN concentrations at 0.5% supersaturation were considered."*

Page 6, 475: Reword to indicate that a specific algorithm was developed to detect and remove ship plume sampling aboard the RV investigator (Humphries et al..). As it is not the RV itself that uses this algorithm.

This has been clarified to: *"An algorithm developed for detecting when the ship plume is affecting the measurements aboard the RV Investigator was used for the relevant campaign datasets aboard the vessel (Humphries et al., 2019)."*

Page 6, 481: , "that have" values larger (this sentence needs to be rephrased).

We have clarified this sentence to: *"The specific limits were selected to account for outliers, that is, values exceeding these limits fall above the 95th percentile of the respective datasets."*

Page 6, 484: "remove" per hour or rephrase the sentence.

We have removed "per hour" from the sentence: *"In order to determine the effect of both continental and reef sources on the airmasses, 72-hour back trajectories were calculated using the HYSPLIT (Stein et al., 2015) Lagrangian dispersion model by the National Oceanic and Atmospheric Administration (NOAA),"*

Page 7, 586: Specify which high concentrations you are referring to. It might read better if this sentence was removed or placed at the end of the paragraph (line 592).

We have reorganized this paragraph to increase clarity and specified that the high concentrations refer to aerosol and CCN concentrations: "*There are a number of uncertainties related to our analysis. The uneven spatial and temporal distribution of the data makes it difficult to infer spatiotemporal variability in the analysis. Years 2021-2023 occurred during the four-year La Nina event from 2020 to 2023, while 2016 was an El Niño year and 2019 was a neutral El Niño Southern Oscillation (ENSO). The different phases of ENSO likely contribute to the increased variability in our measurements as variations in ENSO affect not only the meteorology and precipitation over the reef, but also sea surface temperatures. ENSO can influence CCN concentrations through changes in sea surface temperature, marine biological activity, DMS emissions, sea spray production, and boundary layer dynamics, as well as through altered transport, convection, and wet scavenging. These processes affect both aerosol precursor availability and the growth and survival of particles into the CCN size range. The unusually high aerosol and CCN concentrations observed in 2016 may be linked to the exceptionally strong El Niño conditions during that year, which was the only clear El Niño period in the dataset. However, the available data do not allow a direct attribution, and the cause of the elevated 2016 concentrations remains uncertain."*

Page 8, Line 645: rephase this sentence, specifically "exceeded a factor of several ....orders of magnitude (?)"

A factor of several means to be larger by more than to but by less than an order of magnitude, which is a power of ten. We recognize that the wording is easily misunderstood and have therefore amended the text as follows: *"Nevertheless, the observed particle concentrations within individual campaigns commonly varied several-fold, substantially exceeding expected instrumental uncertainty, with variability observed across both Aitken and accumulation modes. This suggests that the major observed patterns are unlikely to be dominated by measurement artefacts."*

Page 11, Line 810: How was the difference between the 2016 and the 2023 data determined? What statistic tool was used to determine the significance? MannWhitney?

Mann-Whitney was used to determine statistically significant differences between datasets, with corrections for multiple tests accounted for. This has been now specified in the manuscript as: *"The 2016 CCN concentrations are statistically different from the 2023 measurements (Mann-Whitney U test, $p < 0.01$), but otherwise concentrations are similar campaign-to-campaign (Fig. 1a)."*

Page 12: 810: As mentioned above, I feel that discussing cloud processing based only on size distribution is not sufficient and would require a more in-depth analysis on airmass history based on HYSPLIT reanalysis or ECMWF. Also in this section, low, medium, and high activation ratios are discussed without being defined.

Regarding the cloud processing, see above. We have clarified the activation ratios in the figure caption as follows: *"Aerosol size distributions are coloured by the activation ratio Low (0.03–0.53; purple), Medium (0.53–0.69; black), and High (0.69–0.99; green)."*

Page 21: Figure 6: The green and black areas in A to D need to be defined. The axis to these figures also requires labeling.

We have added labels to the maps (Fig. 6 a-d), and added a description of the green and black areas in the maps to the figure caption:

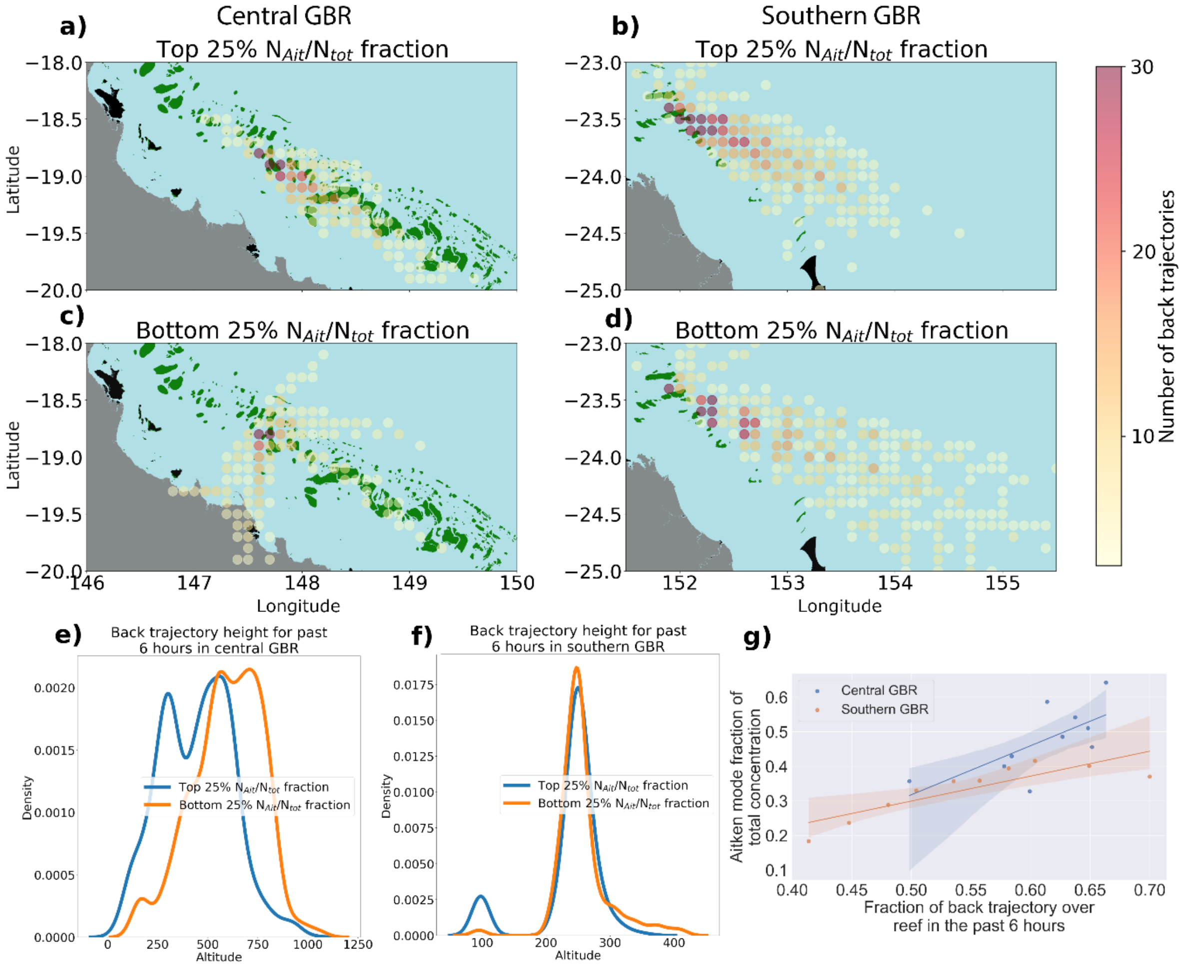

Figure 6: Spatial distribution of 6-hour back trajectories for data with top 25-percentile (a, b) and bottom 25-percentile (c,d) of Aitken mode fraction in proportion to total particle concentration in Central GBR (left) and Southern GBR (right). The green areas are reefs, black areas are islands and the grey area is the Australian mainland. Kernel density estimates for the back trajectory altitude for top 25% (blue) and bottom 25% (orange) Aitken mode fraction in central (e) and southern (f) GBR. Fraction of the total aerosol number concentration in the Aitken mode plotted against the proportion of the past six hours during which the air mass was over the reef at Central and Southern GBR (g). The lines are linear fits to calculated Aitken mode fraction quantiles and their respective reef fractions. Shaded areas represent the line fit uncertainty.

Page 22: For the conclusion, it is mentioned twice in the first part of the conclusion that long term measurements at a fixed station are required. It might be better to separate the conclusion and perspectives more distinctly and include these long term measurement requirements at the end of the conclusion.

We agree with the reviewer that there is unnecessary repetition regarding the long-term measurements. However, we do not see the need to separate the conclusion and perspectives, as the current text progresses from results, to interpretation to perspectives. By moving the need for long-term measurements to the final paragraph, we feel that this improves the flow towards what the reviewer is suggesting: "*Aerosol concentrations over the GBR are typical of a clean coastal environment (Dall'Osto et al., 2011), sitting somewhere between a clean marine (Humphries et al., 2021a, 2023) and Australian land environment (Chen et al., 2019; Milic et al., 2017; Tessendorf et al., 2013). While the North Atlantic and GBR are not directly equivalent marine environments, the comparison with Mace Head (Dall'Osto et al., 2011) provides useful context for clean coastal aerosol conditions. The comparison to Southern Ocean (Humphries et al., 2021b, 2023) represents a substantially more pristine Southern Hemisphere marine environment showing that aerosols in the GBR are higher than that baseline. The air masses observed at the GBR frequently went through cloud processing resulting in a particle number size distribution that often contains a Hoppel minimum (92%). While mode values of most particle parameters, N_{tot}, CCN concentration, accumulation mode particle concentration, and Hoppel minimum, are comparable between most measurements, the hygroscopicity parameter exhibits significant variability between data. This suggests that we are capturing a variety of different aerosol particles in terms of chemical composition. The heavily organic particles measured at Heron Island in 2023 are of particular note, as the stark difference suggests local organic emissions could play a key role in aerosol loading over coral cays and islands.*

As it stands, it appears that aerosol loading over the GBR is primarily influenced by long-range transport of aerosol particles. It is also apparent that there is a non-negligible effect from local sources. Air masses traveling over the reef are enriched in small particles, and their contribution to CCN concentration is up to 6% (Fig. S10). It is important to note that this enrichment is clearly not the dominant process, but rather occurs in the background. The process is likely similar to silent NPF described in Kulmala et al. (2022), in which it is mostly unobservable in surface plots. This is one of the key reasons why the effect can only be inferred through statistical modeling and back trajectory analysis.

Meteorology, sea surface temperature in particular, appears to play a role as well. However, although meteorological conditions do vary over the reef, their effect on CCN concentration is notably small. It could be that the meteorological effect is contained in the changing aerosol loading and the residual effect in our statistical modelling is small. We can hypothesize that long-term changes in climate and large-scale weather patterns such as ENSO and the Southern Oscillation affect CCN concentrations primarily through changes in aerosol sources, sinks, and transport pathways. Variations in sea surface temperature influence marine biological activity, DMS emissions, and sea spray production, all of which alter the availability of aerosol precursors and primary particles. At the same time, ENSO-driven changes in boundary layer height, convection, *precipitation, and large-scale circulation modify vertical mixing, wet scavenging, and the transport of aerosols and precursor gases. Together, these processes influence both the production of new particles and the survival and growth of existing particles into the CCN size range. However, confirming this hypothesis is not possible with the available data.*

Previous multi-site observational studies have demonstrated strong relationships between CCN concentrations and aerosol number concentrations across a range of environments (Andrews et al., 2025; Schmale et al., 2017), and our results agree well with these observations. However, direct quantitative comparison of fitted coefficients between studies is complicated by differences in regression formulation, supersaturation conditions, and variable transformations. Within the GBR dataset itself, the clean marine subset exhibited both a weaker power-law dependence and lower explained variance than reef- and/or land-influenced air masses. This suggests that under very clean marine conditions, accumulation-mode number concentration alone becomes a less complete parametrization of CCN variability, likely because aerosol composition, growth state, activation efficiency, and recently formed particles exert a proportionally larger influence. Overall, accumulation mode parametrizations work reasonably well overall, but clean marine environments may require additional treatment of growth processes and aerosol composition.

Coral reef contributions to aerosols and CCN are typically not explicitly parameterized in climate models. This analysis highlights the importance of resolving atmosphere-biosphere processes at fine spatial scales to capture subtle but climatically relevant sources of CCN. By directly isolating and quantifying the effect of coral reef emissions on CCN concentrations, our study provides, to our knowledge, the first direct observational evidence of a reef-origin aerosol contribution to cloud-relevant particle populations. While likely negligible at global scales, this reef signal represents a unique natural marine source that can influence regional CCN budgets and cloud microphysics. It is important to note that we cannot distinguish between various sources within the coral reef ecosystem, and therefore quantifying the exact sources within the ecosystem itself remains a subject of future work. These results offer a novel constraint for regional modelling and a foundation for incorporating biogenic aerosol processes from reef systems into Earth system models. Furthermore, this analysis method could be extended to cover other biologically active systems that are potentially important regional sources of CCN.

Our results have important implications for the RRAP Cooling and Shading subprogram. The observed sensitivity of CCN concentrations to air-mass conditions, and the complex aerosol dynamics having an impact specifically in clean marine air masses suggests the effectiveness of cooling and shading intervention over the GBR is likely notably variable in space and time. Consequently, consistent long-term atmospheric monitoring at fixed locations, together with robust characterization of baseline aerosol-cloud interactions, is essential both for resolving long-term variability in natural GBR aerosol processes and for *evaluating the potential effectiveness and environmental impacts of future solar radiation management interventions over the reef.”*