

Reviewer comments: Coral reef–influenced air masses promote new particle formation over the Great Barrier Reef by Okuljar et al. (2026)

Okuljar et al. describes measurements conducted on Heron Island, in the Great Barrier Reef (GBR) region over the course of 6 weeks in November and December of 2024. The authors present measurements of particle number concentrations in different size regimes from several instruments alongside gas-phase measurements from a PTRMS, particle composition measurements and meteorological data. In the first part of the paper, the sampling period is separated into two different regimes, where one shows higher particle number concentrations and trajectories passing over the GBR. In the second part, an algorithm for identifying new particle formation (NPF) events is applied to particle size distribution data from the NAIS and NPF event days are compared with non-event and undefined days.

While the underlying dataset seems promising and valuable and the topic is within the scope of ACP, there are some major problems in the way the analysis is conducted, and the results are interpreted and presented. The main underlying issue is that the authors perform statistical analysis on a very small dataset, which does not support this approach; hence, the conclusions drawn here are not supported by the data. Additionally, the overall structure of the manuscript is unclear, with a weak connection between the first and second parts of the analysis. Values or parameters are often presented without proper discussion or relevance to the main story. On the other hand, some interesting and important plots have been moved to the SI. The quality of the figures is often poor, and they are difficult to parse. In the following, we list the major problems and support them with line-by-line comments before pointing out some of the minor issues.

Based on these issues, we cannot recommend the manuscript for publication in ACP. Ample changes to the analysis would be necessary, which are outside the scope of major revisions and should lead to an entirely new submission.

Major comments:

An important problem is the lack of up-to-date literature references. Often old studies are cited, where newer ones are available and would allow a clearer understanding of the underlying processes. This indicates a lack of thorough literature research and understanding, which is crucial for the interpretation of measurement results. In the methods section, relevant citations are often missing completely.

Line 47: The references for the interaction between aerosols and cloud properties are from 1989 and 1974. Newer studies (Rosenfeld et al. 2019, Chen et al. 2022, Li et al. 2025, Gordon et al. 2023) have been conducted and should be added.

Line 62: Broadgate et al. 1997 only measured isoprene in the North Sea and the Southern Ocean. References for emissions from the reef and additional aerosol precursors are missing.

Line 65-68: The oxidation of DMS and its role in NPF are not discussed in sufficient detail. DMSO and sulfuric acid are only two of many different compounds formed from DMS. Methanesulfonic acid (MSA) can be present in comparable concentrations to SA and has been shown to contribute to particle growth and even nucleation (Beck et al. 2021, Baalbaki et al. 2026, Johnson and Jen 2023). HPMTF is another important product, especially under the low-NO_x conditions of the remote marine BL. Its uptake in cloud droplets can reduce the production of gas-phase SO₂ from DMS oxidation (Novak et al. 2021). Additionally, aqueous-phase processes play an important role in the BL, with most DMS-derived sulfate being

produced in droplets (Chen et al. 2018, Jongebloed et al. 2025). Potential pathways for NPF should be discussed more thoroughly. While sulfuric acid is a crucial compound for NPF, it cannot nucleate without the presence of bases under the warm conditions of the tropical marine BL (Dunne et al. 2016).

Line 70: The strong negative feedback proposed in the CLAW hypothesis has mostly been abandoned. While individual links were supported by later papers, the overall feedback loop does likely not exist (Quinn and Bates 2011). The latest IPCC report in fact estimates a small positive feedback loop; however the uncertainties remain large (Szopa et al. 2021). Mentioning the CLAW hypothesis without further discussion is inadequate.

Line 129-167: References should be added for instruments that were used in the analysis.

Line 148: According to which formula or reference was the hygroscopic growth calculated?

The separation of the data based on aerosol concentrations and origin is in principle valid; however, the conclusions drawn from the differences in measured parameters are often not compelling. In many cases, small differences are overinterpreted, and a statistical significance measure is missing. The authors identify the first period as 'emission affected' based on the higher aerosol number concentrations, but it remains unclear whether this is related to emissions from the GBR or local influences. Strongly anthropogenic influenced airmasses were excluded based on a threshold in BC, but it is not clear what the basis of this threshold value is and the BC values in the first period are still elevated compared to the second.

Line 234: The threshold for excluding anthropogenic influences seems arbitrary. On what basis was it selected, and would the results look significantly different if the data were included?

Fig. 2: The two periods are identified as clean-marine and emission affected, while the reason for this becomes clear only after the aerosol measurements are analysed in Fig. 3. If the two periods are identified based on the time series, this should be shown in the figure instead of the averaged values. Based on Fig. 2 alone, both air masses have a purely marine origin and the variables only vary slightly. No statistical analysis is applied to assess these differences, nor are they strongly different on visual inspection.

Line 285: The identification of the air masses as clean marine/emission affected requires a more solid foundation. Were BC or VOCs elevated? Could the difference be caused by meteorological conditions, e.g. precipitation during the past days? Could the aerosols in the first period be a result of long-range transport (e.g. does the height of the trajectory indicate origin in the free troposphere or the BL)?

Line 306-311: This is a vague conclusion of the analysis. It is not clear whether the aerosols originate from the reef or local island sources. Can these be separated further based on BC measurements or anthropogenic VOC emissions?

Line 324: For DMS, monoterpenes and acetone, statistical significance is reported but not for any of the other parameters in Fig. 2-4. Does that mean that the test was not conducted for the others or that the differences were not significant?

Line 325-326: Does the time spent over the island really play a role for the measured DMS concentration? This would mean that DMS is oxidised so rapidly that the transport time over the island is enough to reduce it significantly. If this was the case, then emissions from the GBR reef would be oxidised before they reach the station, since the distance between the

reef and island appears longer than between the coast and measurement station. The higher DMS concentrations in the second period are interpreted as “suggesting biogenic marine origin such as corals or phytoplankton”. However, previously the argument was that period 1 has a stronger reef impact. This seems contradictory.

The NPF classification was performed only on a subset of the data, where NAIS data was available. This subset lies entirely in the ‘emission affected’ period identified previously, which should be stated more clearly. It is questionable whether the applied scheme for the identification of NPF days is suitable for application to such a small data set. The number of days in each category (event day, non-event day and undefined) are only 8, 5 and 3, respectively. Based on these, several comparisons are performed, which would require a higher sampling size to yield compelling results. The finding that 3 NPF events occur on event days is rather surprising and should be discussed in more detail. The size distribution plot shown for the NPF events does not show the typical ‘banana’ shape and growth from the smallest sizes expected for NPF. The argument for the GBR as a source for NPF is not supported by the data shown in the manuscript.

Line 332-335: In Aliaga et al. 2023 the method for NPF ranking was applied to a 5-year data set. It relies on the enhancement of nucleation mode particle concentration compared to background values and assumes a log-normal distribution of this enhancement for both event and non-event days. Applying this method to such a small data set is questionable. The authors of the study clearly state: “While there is no strict minimum on the number of days required to implement this method, we advise a baseline of 1 month to increase the likelihood of obtaining a representative sample of NPF intensity and occurrence. However, more extensive datasets are preferable, with multi-year and multi-season time series being optimal.” (Aliaga et al. 2023) Fig S5 shows the Gaussian fits but not the underlying histogram. We expect this to be very scattered and not well represented by the fits. If this method is applied, it should be compared to other ranking methods (e.g. Del Maso et al. 2005, Dada et al. 2018) or at least a visual inspection of the days should be conducted. In such a small data set, a visual characterisation is feasible and might even be more accurate than the applied method.

Line 346-347: The conclusion that “(...) the evening mode (...) does not originate from a nucleation process but rather from the growth of existing particles, potentially linked to local combustion emissions.” is not well supported by either black carbon or PTRMS measurements. It could just as well be nucleation that occurred some distance away and was transported to the station.

Line 349-351: Do all 5 of the NPF days show 3 NPF events? This is very unusual and not even observed in field sites with frequent NPF (Del Maso et al. 2005, Dada et al. 2018). Is this an artefact of averaging days with single NPF events at different times? If the data really shows multiple NPF events per day, this should be discussed in more detail and compared to other measurements of NPF in marine environments.

Line 353: The sudden termination of the last event is explained here by sunset. However, sunset during this time of year should be around 6 pm, almost 2 hours before the end of the event.

Line 357: Here, the period is defined as “clean marine background”. However, it lies entirely in the “emission affected” period of the previous analysis, which is inconsistent.

Fig. 5: Which measurement of the NAIS is plotted here (ion or particle mode, positive or negative)? If three instruments were combined for the analysis in 3.1, why is this combined

size distribution not also shown here? Does the colour scale really show number concentration or $dN/d\log D_p$, which is usually used in surface plots of size distributions? We recommend checking the upper limit of the colour scale; it seems like large portions of the plot are saturated, and could get more detail by increasing the upper limit of the colour scale. The NPF events in this plot are not very clear and lack the characteristic 'banana-shape'. Why are there sharp cutoffs at the edges of the events? This appears poorly smoothed and nonphysical. It should be confirmed that these are really NPF events and not transport of small particles by looking at the individual days.

Line 381: The statistical difference in cloudiness in Fig. S6 appears minimal and is shared by both undefined and NPF event days, which seems to indicate a lack of uniqueness in the separated g3 mode.

Line 383: This does not appear to be explanatory. Much is being made of fairly small differences in solar radiation, especially given that the diurnal solar radiation cycle of non-event and NPF days is overlapping closely.

Fig. 7: Non-event and NPF days do not show significant differences in most of these plots. The wind direction component is unnecessary and better described by the previous trajectory plot.

Fig. 8: Why was the framework of non-event, undefined and NPF days abandoned here but used in the previous figures? Was an attempt made to calculate absolute concentrations of DMSO and MeSH?

Fig. 9: This figure is difficult to understand. According to Fig. 5 the NPF events are barely 6 hours apart, but the time series here goes back more than 15 hours and would therefore include events twice? a-d do not show clear trends leading up to NPF, at least not on the y-scale shown. (e) seems compelling but has the same information already shown in Fig. 6.

Line 446-447: The gradual decrease of DMSO is not very clear, and why does it increase again after the event?

Line 452-455: The increase in sulfate and NH_4 is not clearly related to NPF. It was not shown that the particles actually grow into the size range of the AMS. Additionally, the change in the 12-hour reef fraction after the NPF event would indicate a change in the origin of the air masses associated with higher sulfate/ammonium levels.

Line 467: The accumulation of DMS over night is interesting and the diurnal cycle should be shown in the main text. How does it compare to other measurement studies?

Chemical analysis of the aerosol composition was performed using the AMS data. However, it is unclear how this relates to the NPF event, as the aerosols do not seem to grow into the size range of the AMS. Additionally, the difference between the periods seems marginal and the interpretation is vague. The gas-phase measurements of VOCs by the PTR do not help to form a clear picture for the NPF mechanism either. The data is noisy and does not seem to support the claims made in the manuscript.

Line 486-494: The triangle plot in Fig. S9 is incorrect. It is not the appropriate F43/f44 triangle shown in Ng et al. 2010. Both Fig. S9 and S10 are unclear in their formulation and

description. Furthermore, it was not shown that particles from the NPF events grow to sufficient sizes and mass for strong AMS detection. If this is not the case, then the observation of the larger particle population measured by the AMS is not related to the observed NPF events. Finally, the measured aerosols mostly consist of sulfate; an analysis of the contribution of SA and MSA (e.g. Chen et al. 2019) could be more insightful than analysing the organic fraction.

Line 496-497: Given that PTR-MS compound-specific sensitivities can vary wildly, why are values in Fig S8 presented in ions/second? Direct calibrations were performed and can be used to generate extended calibration to non-targeted analytes, as in Holzinger et al. 2019.

Line 499-501: The diurnal cycle presented here in Fig S12 is incredibly noisy, and making claims based on a single sharp peak that could be the result of a singular erroneous outlier seems odd.

Line 504-505: According to the methods section, NO_x was measured directly. Can that support the claim that NO_x interferes with NPF? The references given here only show the impact of NO_x on NPF/SOA formation driven by alpha-pinene. Based on the measurements here, AP does not seem to play a major role in the region.

Line 508-509: This claim is not well supported. Differences in the concentrations of oxygenated compounds are not clear in Figure S8, and while the timing of peak concentrations does differ, it is not clear why nucleation would occur at one time and not the other (especially if the authors expect nucleation to occur at multiple times on the median NPF day).

Minor comments:

Line 86: Sulo et al. 2026a and b are the same citation.

Line 153: To what temperature was the PTR inlet line heated?

Line 324: How do the measured DMS mixing ratios compare to literature values or satellite data? CAMS provides DMS estimates in the region and could be used for a comparison.

Line 339-340: Do the spikes in the particle distribution during non-event days occur in all of those or is this a feature of averaging 5 days?

Fig. 6: Panel (a) is useful, but the quality, especially the coloured trajectories, could be better. Panels b-d do not add anything that can't be seen in (a) and can be put in the SI.

Line 387: The pressure argument is unclear. What mechanism leads to better mixing and how does this impact NPF? The solar radiation does not differ strongly between non-event and NPF.

Line 425: Is the higher IP concentration related to NPF at all? Under BL conditions, IP oxidation should not initiate NPF.

Line 429: The lifetime of DMS and DMSO should be discussed here. Is the transport time between the reef and the measurement station long enough for DMS to be significantly reduced?

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