RC2 response

We are grateful to the reviewers for their insightful comments, which have strengthened the manuscript and improved the discussion of results. We present the reviewers' comments in black and our responses in red. All line numbers cited in this response refer to the revised manuscript and tracked changes have been applied throughout to help reviewers identify amendments.

Overall Assessment

This study proposes an innovative and insightful explainable AI framework that integrates Lagrangian airmass history with XGBoost and TreeSHAP for precise simulation and attribution of aerosol processes, providing in-depth analysis for the Antarctic case. However, the lack of rigorous physical verification for its key conclusions limits their persuasiveness. Additionally, the manuscript requires a thorough language revision to address recurrent grammatical errors.

We thank you for your comments we have addressed them below and updated the manuscript as appropriate, including a thorough language revision.

Major Comments

1. The study relies heavily on inferring dominant aerosol processes from feature importance rankings and SHAP-value relationships. However, the scientific robustness of these conclusions is limited by a lack of validation linking these statistical associations to physical causality.

The aim of this study was to provide a holistic statistical analysis, based on widely available particle number size distribution measurements at in-situ measurement sites. In the absence of process rate observations at the scales of interest, statistical relationships with available observations are fundamental to inform our understanding (Morrison et al., 2022). Previous studies have looked at individual aerosol sources and sinks for example of biogenic precursors from 'Time over forest' (Liao et al., 2014; Tunved et al., 2006) and accumulated precipitation (Isokääntä et al., 2022; Talvinen et al., 2025; Tunved et al., 2013), but a holistic understanding requires accounting for interactions between sources, sink and meteorology as all of these processes act on aerosol during its transport history to affect the aerosol lifecycle. In our study by including all variables we account for interactions between variables which could be masking true relationships. We acknowledge there are limitations to these correlation-based methods and have added additional text in the methods and conclusions to address this.

In future work we would look to test the potential of some recently developed causal SHAP methods and expand the framework to integrate these novel methods. The integration of these new causal SHAP approaches is out of scope for the current study which focuses on a developing a robust framework which can be integrated with novel techniques in the future.

We have updated and added to the manuscript test to address these points as follows:

Lines 57-96: The importance of airmass history for understanding aerosol processes has been demonstrated in many studies, including Sogacheva et al. (2005), Tunved et al. (2006) and (2013). Traditionally, these techniques have been applied by linking airmass trajectories derived from reanalysis data with in-situ aerosol observations from surface receptor stations to develop receptor models of potential aerosol source regions, for example using a concentration-weighted trajectory framework (CWT) (Hsu et al., 2003). As aerosol populations undergo significant transformations during transport, Lagrangian based airmass frameworks have also been exploited to provide significantly more detailed insights than can be achieved using Eulerian approaches into the processes controlling observed aerosol properties. This is achieved by considering the interplay between aerosols and the experienced conditions during the airmass history. Previous studies have provided insights into the role of specific processes controlling the aerosol lifecycle (e.g. potential emissions, precipitation and clouds) using one at a time (OAT) analysis, focusing on the relationship between a variable, particularly during transport, and the measured aerosol properties at a receptor site. For example, the role of removal via wet scavenging during transport (Khadir et al., 2023; Tunved et al., 2013) and the importance of emissions from the boreal forest as a source of secondary organic aerosols during transport (Liao et al., 2014; Tunved et al., 2006).

The strengths of these Lagrangian frameworks for understanding processes controlling observed aerosol has also been leveraged to perform process driven evaluation of aerosols in models. Recently a new modelling framework was developed to calculate airmass trajectories using the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT, Stein et al., 2015) from global climate models (GCMs) using data from the Aerosol Comparisons between Observations and Models (AeroCom) Phase III GCM Trajectory Experiment (GCMTraj, https://aerocom.met.no/experiments/GCMTraj, Duncan et al., 2026; Kim et al., 2020). This framework facilitates the calculation of airmass trajectories using meteorological fields from GCM simulations. The resulting GCM airmass trajectories can be combined with GCM simulated aerosol properties in a consistent manner to previous reanalysis–observational based studies. By combining GCM derived trajectories with GCM simulated diagnostics this methodology allows us to transparently evaluate and compare atmospheric processes acting on the aerosol during transport between the observations and GCMs in unprecedented detail. Studies have successfully applied this approach to perform process-based evaluation of the impact of clouds and precipitation on aerosols in GCMs (Talvinen et al., 2025) and sources of bias in PNSD response to an effusive volcanic eruption (Duncan et al., 2026).

Whilst these techniques are useful to improve understanding of model bias, these studies do not account for interactions or the potential for compensating errors in GCMs arising from other aerosol processes controlling aerosol properties not considered. Furthermore, the high dimensionality of aerosol modelling means that applying OAT approaches to constrain all processes would be prohibitively time consuming. Accordingly, new approaches are required that can capitalise on the strengths of Lagrangian evaluation of GCMs by automatically isolating the dominant atmospheric processes controlling the aerosol lifecycle in different environments. This is crucial to improve understanding based on observations and for future application to constrain current structural uncertainty in GCM representation of aerosol.

Lines 542-558: *SHAP is used for model interrogation, to investigate the relationships leading to predictions of the ML model in Sect. 4.4. SHAP values are calculated for each model prediction, which provides local explanations of predictions of the ML models. In this study we then aggregate the local SHAP analysis to investigate the overall impact of features on model predictions. The SHAP values provide a measure of the total contribution of a feature to model predictions in the ML model, including interactions with other features. We also utilise SHAP interaction values in which the total SHAP values are decomposed to the pair-wise interaction values which reflect the combined contribution of features to model predictions, and the individual contribution of a feature in the model described by the 'main effect'. The TreeSHAP interaction values reflect how the ML model partitions predictions in the learned structure for feature combinations, whilst interventional SHAP values would reflect feature contributions under the assumption of feature independence (Lundberg et al., 2020). Decomposing SHAP values into main and interaction effects can make dependencies between features in model predictions more explicit, whereas in many traditional statistical analyses these dependencies may remain implicit. However, it is important to note that neither interventional nor TreeSHAP methods, nor traditional statistical analysis identifies casual feature dependence or interactions (Silva et al., 2024). Instead, this methodology allows us to explore the ML model's representation of the airmass history and processes driving observed aerosol concentrations, and to infer the atmospheric processes that could underpin the learned relationships. To investigate the potential processes, as well as considering the SHAP-feature relationships from the SHAP analysis we consider the spatial distribution. To visualise the spatial distribution of SHAP values, we employ the CWT framework (Sect. 3.2), where each SHAP value for a measurement at the site corresponds to a trajectory.*

Lines 1165-1177: "Now the validity of the framework has been demonstrated, future studies can capitalise to explore attribution of physical causation of statistical relationships identified for ML model predictions. In future work alternative sites with chemical composition measurements could be used to facilitate more certainty and separation of aerosol sources alongside the current framework. It is important to note the limitation of XAI approaches for correlated variables, as discussed in Silva et al. (2024) for example. We note that neither model interrogation methods, including SHAP values and Sobol sensitivity indices, nor classical statistical approaches such as regression analysis establish causality. The framework developed in our study can provide an overview of potential dominating processes in a region to highlight areas for future study for a mechanistic physical process understanding. Approaches that incorporate causal networks with SHAP have been recently proposed as a method to differentiate correlation from causality (Heskes et al., 2020; Yee Ng et al., 2025). Causal SHAP methods require a causal graph which can be difficult to specify for complex systems. While recent approaches integrate causal discovery to circumvent this (Yee Ng et al., 2025), reliably inferring causal structure from data in high-dimensional systems with correlated variables remains challenging. Thus, these methods could be the subject of future endeavours, with a vision to integrate this with the current Lagrangian XGBoost framework."

For instance, the attribution of the log-correlation between chlorophyll-a and aerosol concentration to marine biogenic sources (Lines 564-566) is made without isolating confounding effects from co-varying drivers like temperature.

Thank you for your comment we have added to methods discussing the difficulty in separating the effects of confounding variables in these analyses, as with many common statistical approaches (e.g. Sobols, permutation feature importance, correlation-based methods). SHAP offers the benefit of decomposing SHAP values into main and interaction effects can make dependencies between features in model predictions more explicit, whereas in many traditional statistical analyses these dependencies may remain implicit. However, it we note that neither SHAP methods, nor traditional statistical analysis identifies casual feature dependence or interactions (Silva et al., 2024).
We have extended the analysis to including the decomposition of the SHAP values to 'main effect' and interaction effects with temperature and features of highest interaction.

We agree that there will be cofounding between variables in this complex atmospheric system, and whilst these methods cannot directly derive causality, SHAP offers the opportunity to make dependencies between features more explicit than other statistical methods. We have added the following text to the manuscript to discuss the cofounding factors for chlorophyll:

Lines 779-795: "*Decomposing the SHAP values to 'main effect' and interaction effects with the features of highest interaction, we demonstrate the contribution of solely the feature 'chlorophyll weighted sum' to ML model predictions. The SHAP-feature dependence plot exhibits a logarithmic relationship for the main effect of chlorophyll (Fig. S12a). The highest ranking SHAP interaction values are for each ML model are trajectory speed mean, and 'temperature weighted mean' (Fig. S9a). From the interaction values it is clear that high levels of chlorophyll weighted sum contributes positively to model predictions when paired with high temperatures and low trajectory windspeeds (Figs. S12 b and c). However, when normalising by the main effect (Fig. S9b), the main effect for chlorophyll sum dominates the total SHAP value.*

Intrinsically there will be a relationship between surface solar radiation, air temperature, sea ice fraction, and marine biogenic aerosol sources. As air warms and the ice melt begins due to solar radiation, winter-accumulated nutrient stocks and newly available light allows surface communities to bloom and release primary and secondary organics to the water column. Enhanced surface ocean biological activity due to the increasingly ice free ocean surface, coupled with lower gas solubility due to rising air and sea surface temperatures, is expected to drive an increase in marine biogenic contribution to aerosol in Austral spring and summer (Ferreira et al., 2024), along with higher nucleation rates (Park et al., 2023; Quéléver et al., 2022). Whilst these variables do exhibit SHAP feature interaction (Figs. S8a and S9a), these do not outweigh the main effect (Figs. S8b and S9b), and correlations between chlorophyll and surface solar radiation, air temperature, sea ice fraction are weak (0.098, 0.136 , and -0.136 respectively).

The following figures were added to the supplement:

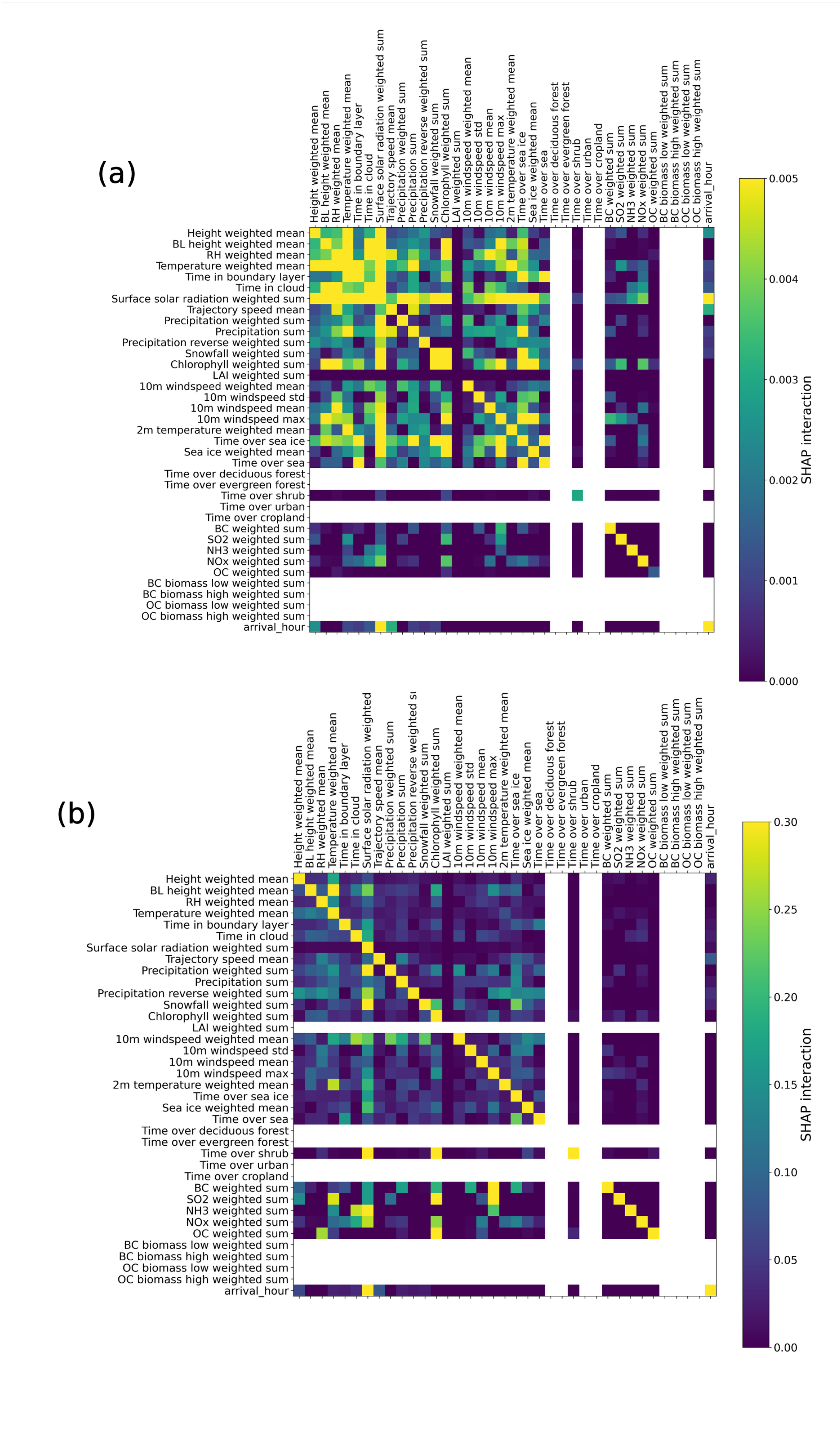

Figure S8: (a) Absolute mean SHAP interaction results for the 30-80nm model. (b) Absolute mean SHAP interaction results for the 30-80nm model, weighted by the main effect SHAP value for each feature. White indicates that the feature was not included in the final model.

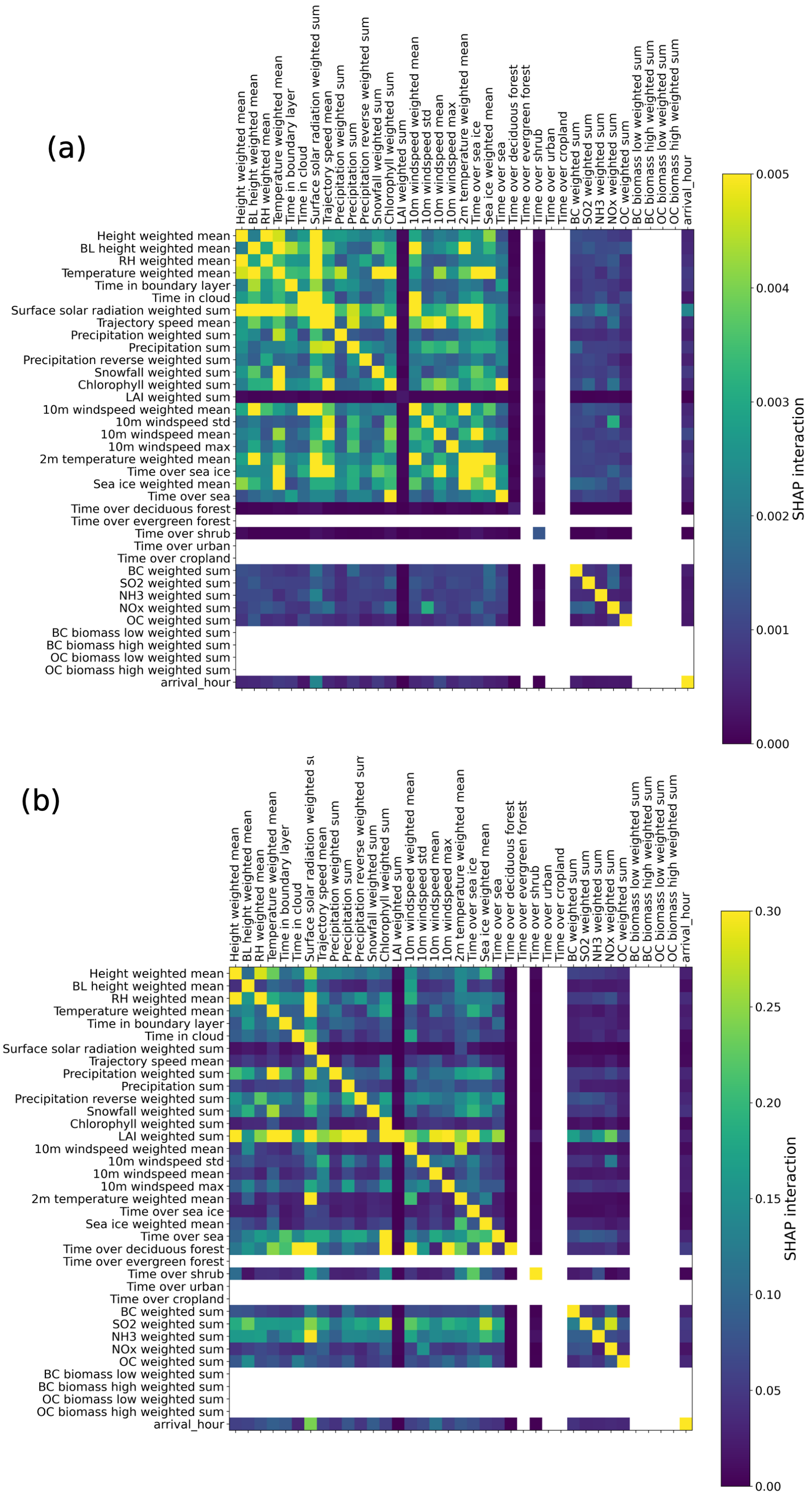

Figure S9: (a) Absolute mean SHAP interaction results for the 80-660nm model. (b) Absolute mean SHAP interaction results for the 80-660nm model, weighted by the main effect SHAP value for each feature. White indicates that the feature was not included in the final model.

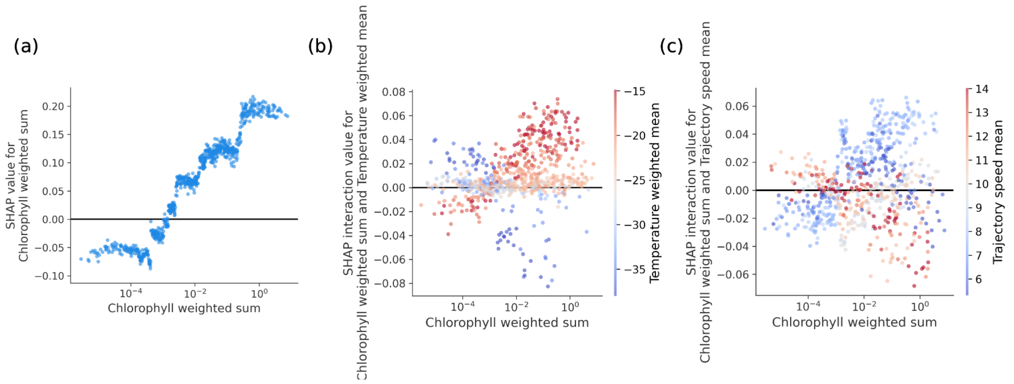

Figure S12: (a) SHAP-feature dependence plot for SHAP main effect of chlorophyll weighted sum, (b) SHAP interaction value dependence plot for Chlorophyll weighted sum and Temperature weighted mean. (c) SHAP interaction value dependence plot for Chlorophyll weighted sum and Trajectory speed mean. All figures for the 80-660nm model. Note the use of a logarithmic x-axis.

- Similarly, the dual role of precipitation (sink near site, source further away) is explained solely by opposing correlations of weighted vs. unweighted sums (Lines 711-721), lacking mechanistic validation against wet scavenging physics.

  Due to measurement availability, we cannot validate this mechanism with scavenging rates, however previous studies focused on Lagrangian transport have identified the role of precipitation removing aerosol populations during transport using these bulk diagnostics from reanalysis data (Isokääntä et al., 2022; Khadir et al., 2023; Tunved et al., 2013).

  We have added the following text to the manuscript comparing our results to previous studies to provide greater context:

  Line: 1016-1035: 'Indirect enhancement of NPF and subsequent growth, due to reduction of the pre-existing condensation sink has been identified previous studies (Browse et al., 2012; Dal Maso et al., 2002; Dall'Osto et al., 2017; Khadir et al., 2023; Zhang et al., 2012). This has precipitation enhancement also recently been confirmed by theoretical analysis (Zhao et al., 2024).
  Khadir et al. (2023) used a correlation-based approach between precipitation during transport and number concentration across the aerosol size distribution and showed, for several different environments, that the impact on aerosol number concentration is dependent on the timing during transport. For Arctic and boreal forest, above 30nm particle diameter, precipitation mostly acts to decrease aerosol number concentrations for the first 12 hours, which 'Precipitation weighted sum' relates to. However, beyond this point Khadir et al. (2023) found that precipitation before 12 hours along the back trajectory acts to increase particle number concentrations. The precipitation was found to constantly act as a sink for particles above a 100nm diameter for the Arctic. Our study uses a minimum diameter of 80nm for the accumulation mode and is based in the cleaner Antarctic environment which could explain why this is still seen for the N80-660nm ML model SHAP relationships.
  From the SHAP interaction values (Fig. S8a and S8b), precipitation sum interacts strongly with time over sea ice for the Aitken mode. The interaction values plot (the combined impact of the two variables) in figure S16 highlights that the precipitation removal and enhancement of particles is associated with lower time over sea ice and increased solar insolation – which substantiates the hypothesis that this is representing increase in NPF after precipitation reduces the condensation sink. Future studies using GCM output would be able to collocate scavenging diagnostics along trajectories to investigate these mechanisms further.'

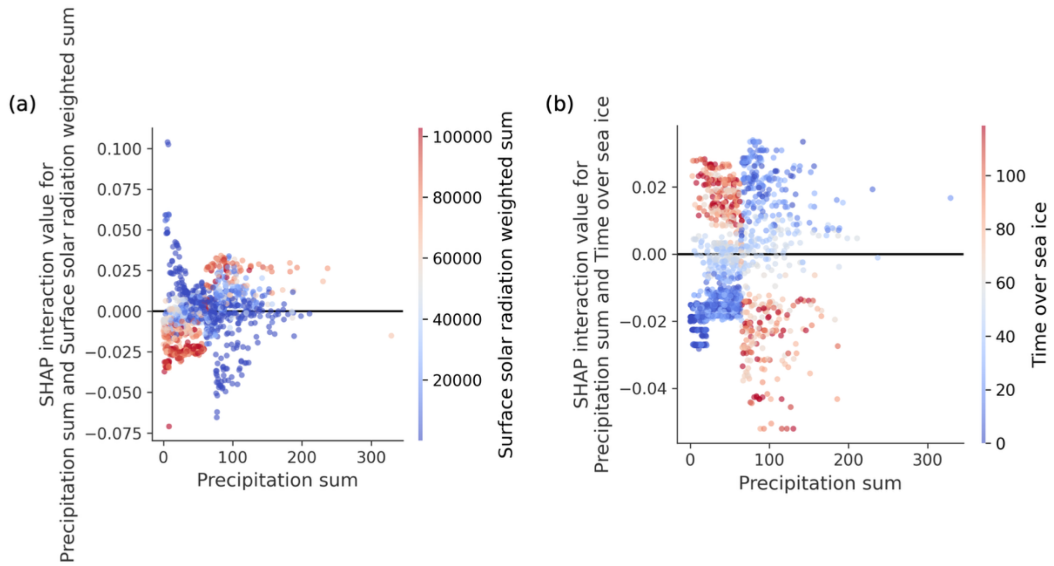

Figure S16: (a) The SHAP interaction values - feature relationship for precipitation sum and time over sea ice SHAP interaction values compared to precipitation sum. The colour of the points indicates the value of the time over sea ice. (b) as (a) but the SHAP interaction values for precipitation sum and surface solar radiation weighted sum and the colour indicates the surface solar radiation weighted sum.

- The proposed contribution of free-tropospheric transport to the Aitken mode rests primarily on SHAP analysis (Lines 621-625) and would benefit from direct support, such as aerosol composition measurements.

  Unfortunately, due to measurement availability (no aerosol composition measurements at the Trollhaugen surface station) we cannot investigate this further with direct support of aerosol composition measurements but instead can hypothesise based on the statistical analysis, based on this mechanism which has been found from previous studies (Brean et al., 2025; Fiebig et al., 2014; Lachlan-Cope et al., 2020). This highlights importance of continuous high-temporal resolution size-resolved chemistry for to inform physical process understanding. We would embrace future studies/ campaigns that provide measurements to substantiate this hypothesis in future studies. Future work applying the framework to other sites which include composition measurements would offer the opportunity to substantiate the statistical analysis.

  To link more clearly the SHAP analysis to transport in the free troposphere we used the variable 'Time in boundary layer' to derive the 'Time out of boundary layer' for each datapoint. We have added the following to the manuscript to add to the analysis:

  Lines 950-956:"Time out of boundary layer can be derived from time in boundary layer during model integration, which highlights the positive contribution of time above the boundary layer to model predictions and corresponds to high values of 'Height weighted mean' for positive SHAP associated with the feature 'Time in boundary layer' (Fig. S13). The weighting function is an exponential decay giving most weight to the first 12 hours (Fig. S3), which on average is associated with transport over the continent (Fig. S13b), where trajectory transport is high (Fig. 5b), associated with the region of positive SHAP (Fig. 11e). This indicates that airmass descending rapidly on the continent, as well as prolonged transport out of the boundary layer, lead to increased predicted aerosol concentrations, further suggesting contribution from the free troposphere.

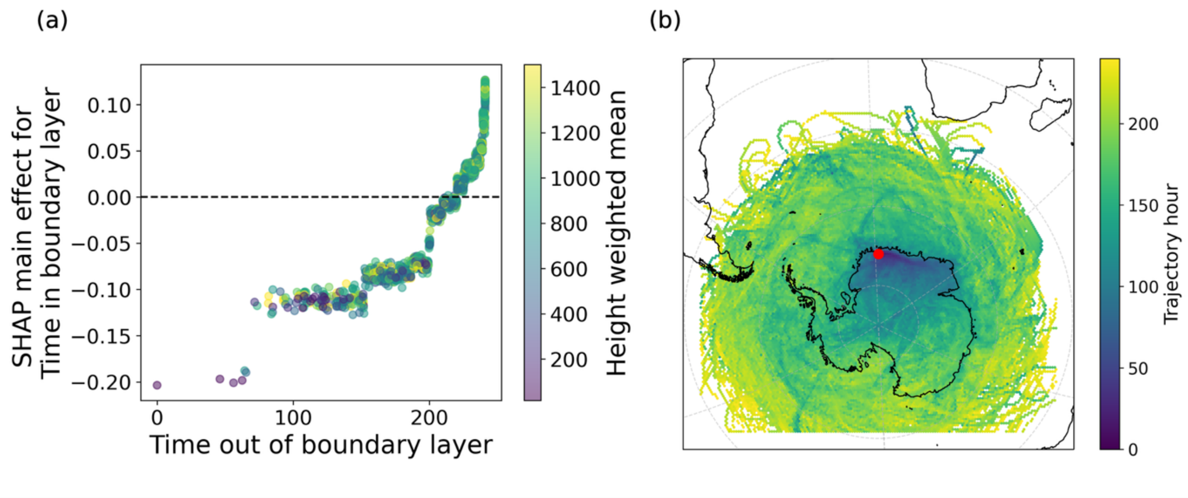

Figure S13: (a) SHAP dependence plots for Time out of boundary layer and the SHAP main effect for Time in boundary layer, with the corresponding values for Height weighted mean indicated by the colour. (b) A CVT receptor map for the average trajectory hour.

- Finally, the speculative interpretation of positive SHAP values at very high windspeeds (>20 m/s) as possibly from sea spray or blowing snow (Lines 611-618) remains unsubstantiated by concurrent observational evidence or targeted analysis.

  We agree with the reviewer that it is speculative because there are very few data points during the test year which exhibit the behaviour that we hypothesise to be associated with sea spray or blowing snow, i.e. positive contribution to model predictions of N80. This positive contribution occurs for 10m windspeed max values above 20ms^{-1}. There are very few points which exhibit this behaviour in the test year, which would result in a dataset too small to use to pursue further statistically robust analysis of these phenomena. Future studies focusing on a seasonal model would be able to investigate this mechanism further, perhaps with additional sites to add statistics or chemical measurements to perform a more targeted analysis. We have added to the text to highlight the speculative nature of this statement and the need for future work to corroborate the hypothesis:

  Lines 851-860:"Although the dominant relationships with the windspeed features are negative, there are a few outliers of positive SHAP values for the highest maximum windspeeds for the accumulation mode model (Fig. 10c) above 20 ms^{-1}, which we hypothesise could be indicative of sea spray production in gusty conditions. Blowing snow has also been found to be an important source of aerosol in polar regions (Frey et al., 2019; Lachlan-Cope et al., 2020; Yang et al., 2019). First direct observations of blowing snow events were recorded during a storm with wind speed maxima between 15 and 25 ms^{-1} (Frey et al., 2019), the upper end of which corresponds to the values for which we see positive SHAP. However, there are only a few outliers associated with positive SHAP at high windspeed (Fig. 10c); thus, it is not a statistically robust sample to investigate further or conclude that the model is able to represent this source. This could be due to the extreme concentrations associated with blowing snow events, comparative to the low concentrations in the winter (Frey et al., 2019) and the sparsity of events in the dataset (Fig. S5)."

2. The argument for the framework's out-of-distribution generalizability remains limited. While Section 4.5 presents proof-of-concept applications at the Värriö and Mace Head sites, the analysis does not sufficiently demonstrate its applicability to environments mechanistically distinct from the Antarctic case. For instance, the model is not tested to see whether the primary drivers identified at Trollhaugen (e.g., marine sources, free-tropospheric transport) remain valid or are superseded by different key factors (e.g., biogenic VOCs at Värriö, air mass history at Mace Head) in these new settings. A comparative analysis of how feature importance rankings shift across these diverse sites is needed to substantiate the claim of broad applicability.

We have now extended section 4.5 to include a SHAP summary figure for Värriö and Mace Head models as well as discussion surrounding the differences in feature importance and SHAP-feature relationships at the sites. As expected, there are clear differences in the primary drivers identified at Trollhaugen compared to the new environments which are in line with previous studies of those sites. Future studies will perform full analyses of these additional environments.

We have added the following text and a SHAP beeswarm summary figure for each station:

Lines 1067-1086: The SHAP summary for each site (Figs. 7b, 16a and 16b) demonstrates differences in the ranking of the features between sites, as well as the SHAP-feature relationships. The highest ranking SHAP features vary between the three sites, with meteorological features ranking most highly for Värriö, whereas for Mace Head, contrasting to the other two sites, the highest ranked features for the model are those associated with anthropogenic emission sources. Mace Head is frequently affected by transboundary pollution from the UK and continental Europe, which is driven by synoptic scale circulation (Jennings et al., 2003), which is likely driving the high concentrations in March for the test year (as seen in Fig. 15b). Mace Head also demonstrates a strong contribution from chlorophyll, highlighting the importance of marine organics at the marine site, as highlighted in previous studies, for example O'Dowd et al. (2014).

As well as changes in the ranking of variables between sites, there are also clearer differences in the SHAP-feature relationships. Some relationships are consistent, for example, the variable 'precipitation weighted sum', ranks highly across all the sites for the accumulation mode models and demonstrates negative relationships. Contrastingly, relative humidity which ranks highly for Värriö also demonstrates an inverse relationship compared to Trollhaugen. In the boreal forest NPF has been shown in previous studies to contribute to the accumulation mode size ranges and is less likely to occur during days with high RH (Bousiotis et al., 2021; Dada et al., 2017; Jokinen et al., 2022; Li et al., 2019; Zhu et al., 2024). Predicted Värriö accumulation mode concentrations also demonstrate a strong temperature dependence (Fig. 16a), with a non-linear SHAP-feature relationship. At Värriö, time over evergreen forest and chlorophyll rank within the top 10, implying the contribution of airmasses which experience transport over the forest and marine organics contributing to the accumulation mode aerosol burden. Previous studies have noted the contribution of aerosol from clean marine airmasses, travelling over the boreal forest, triggering NPF events (Jokinen et al., 2022; Tunved et al., 2006).

We have demonstrated that the ML framework is able to accurately predict concentrations and represent different dominating processes when applied to different environments, thus demonstrating its generalisability, full analysis of drivers in other environments will be the subject of future studies.

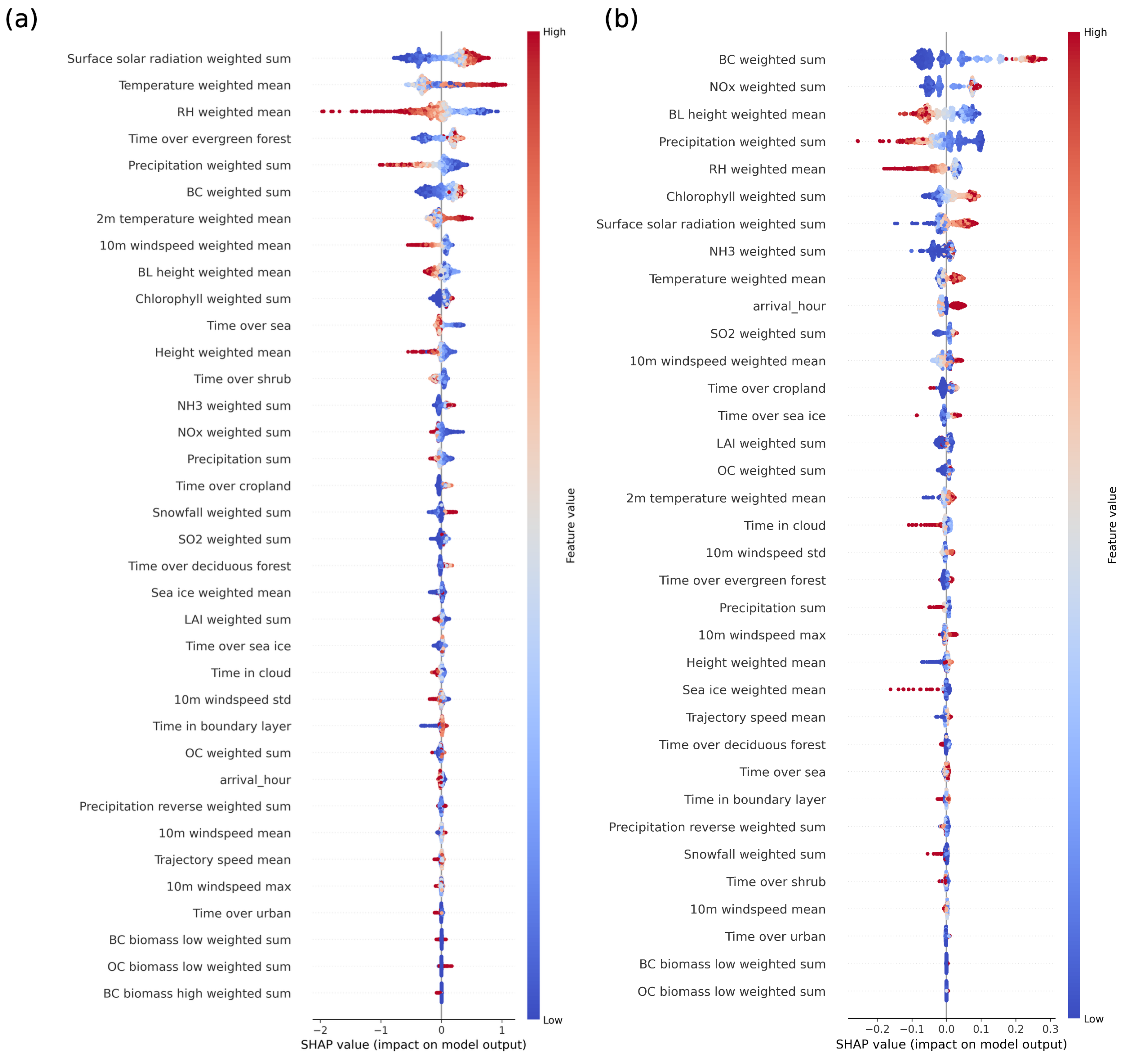

Figure 16: SHAP beeswarm for accumulation mode concentrations for (a) Värriö and (b) Mace Head. Plots are ordered by the rank of each feature as determined by the TreeSHAP analysis. The points are coloured by the feature value corresponding to the data point for the SHAP value, and the 'violin' shape of the distribution represents the density of points.

3. The inclusion of 35 explanatory variables is not accompanied by a clear description of the variable selection criteria. While the study correctly notes a high correlation (r=0.8) between trajectory speed and 10-m wind speed and acknowledges the limitation in separating their importance with TreeSHAP (Lines 595-598), it does not detail any preprocessing steps (e.g., filtering, regularization) taken to mitigate the impact of such collinearity on the SHAP-based interpretation. This leaves the feature importance rankings for these—and potentially other—correlated variables difficult to interpret confidently.

In this study as we use tree-based regression models we did not apply any regularization or normalization to the datasets. Tree-based models are invariant to monotonic transformation of input variables as they are based on recursive partitioning of the feature space. However, we have added to the hyperparameter tuning discussion to highlight the tuning of the L1 and L2 regularization in section 3.4. We do also perform feature selection during model training as detailed in section 3.4. We did not look at combining correlated variables as the critical point of this exercise was to ensure interpretability. This is a caveat of any studies considering meteorological or atmospheric analyses with machine learning or statistical approaches. There will be variables that are correlated for example Pernov et al. (2022) and Song et al. (2022), as there will always be correlation in these physical processes. We acknowledge this limitation and therefore take care to highlight the correlation between variables throughout the analysis.

We have also added additional text to the methods and conclusion to highlight the limitations of this approach:

Lines 542-558: *SHAP is used for model interrogation, to investigate the relationships leading to predictions of the ML model in Sect. 4.4. SHAP values are calculated for each model prediction, which provides local explanations of predictions of the ML models. In this study we then aggregate the local SHAP analysis to investigate the overall impact of features on model predictions. The SHAP values provide a measure of the total contribution of a feature to model predictions in the ML model, including interactions with other features. We also utilise SHAP interaction values in which the total SHAP values are decomposed to the pair-wise interaction values which reflect the combined contribution of features to model predictions, and the individual contribution of a feature in the model described by the 'main effect'. The TreeSHAP interaction values reflect how the ML model partitions predictions in the learned structure for feature combinations, whilst interventional SHAP values would reflect feature contributions under the assumption of feature independence (Lundberg et al., 2020). Decomposing SHAP values into main and interaction effects can make dependencies between features in model predictions more explicit, whereas in many traditional statistical analyses these dependencies may remain implicit. However, it is important to note that neither interventional nor TreeSHAP methods, nor traditional statistical analysis identifies casual feature dependence or interactions (Silva et al., 2024). Instead, this methodology allows us to explore the ML model's representation of the airmass history and processes driving observed aerosol concentrations, and to infer the atmospheric processes that could underpin the learned relationships. To investigate the potential processes, as well as considering the SHAP-feature relationships from the SHAP analysis we consider the spatial distribution. To visualise the spatial distribution of SHAP values, we employ the CWT framework (Sect. 3.2), where each SHAP value for a measurement at the site corresponds to a trajectory.*

Lines 1165-1177: "Now the validity of the framework has been demonstrated, future studies can capitalise to explore attribution of physical causation of statistical relationships identified for ML model predictions. In future work alternative sites with chemical composition measurements could be used to facilitate more certainty and separation of aerosol sources alongside the current framework. It is important to note the limitation of XAI approaches for correlated variables, as discussed in Silva et al. (2024) for example. We note that neither model interrogation methods, including SHAP values and Sobol sensitivity indices, nor classical statistical approaches such as regression analysis establish causality. The framework developed in our study can provide an overview of potential dominating processes in a region to highlight areas for future study for a mechanistic physical process understanding. Approaches that incorporate causal networks with SHAP have been recently proposed as a method to differentiate correlation from causality (Heskes et al., 2020; Yee Ng et al., 2025). Causal SHAP methods require a causal graph which can be difficult to specify for complex systems. While recent approaches integrate causal discovery to circumvent this (Yee Ng et al., 2025), reliably inferring causal structure from data in high-dimensional systems with correlated variables remains challenging. Thus, these methods could be the subject of future endeavours, with a vision to integrate this with the current Lagrangian XGBoost framework."

4. The criteria for removing extreme low-concentration data ($N80 < 4$ cm^{-3}) are not scientifically substantiated, being described only as "appeared anomalous" (Lines 177-179). This subjective filtering risks altering the training data distribution, potentially biasing the model's learning of wintertime aerosol processes and obscuring the unique source-sink balance under very dry polar conditions. The lack of a clear, objective threshold undermines the reproducibility of the analysis.

Thank you for your comment. Whilst we are aware best efforts are made to quality control these ACTRIS datasets, instrument errors and errors in processing of raw observations into temporally averaged datasets are possible. Due to the high temporal frequency of the framework, it is sensitive to large changes in observations, as with all machine learning studies is it essential to undertake due diligence checking any observational data used before any modelling training. We include figure 1 here to demonstrate the size distribution for dates that were removed – as these were deemed non-physical. For example, these measurements were consistent values for the last 20 days of the year of measurements in 2015 (from 8th of December 10:00) – no variation in concentration for 20 days is highly anomalous and suggests an instrumentation or recording failure at the site. Thus, the data from the days 8th December 2015 to 1st of January 2016 was removed to ensure only including robust data for the ML framework.

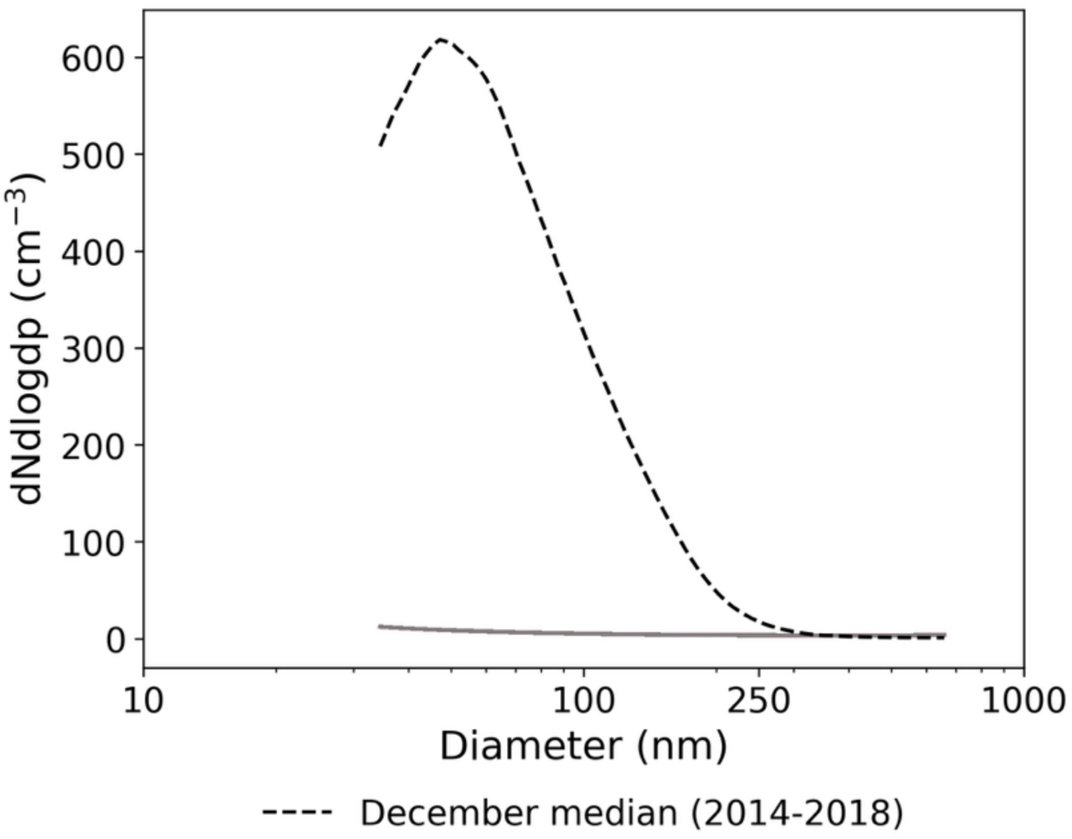

Figure 1: Size distribution for every hourly measurement between 8th December 2015 (10:00) to 31st December 2015 (23:00) (multicolour lines) compared to the December median size distribution (2014-2018) (black dashed line).

We additional removed distributions with concentrations <1cm-3 for (80-660nm) from other periods as again these demostrated very anomylous distributions with extrememly low concentrations (Figure 2).

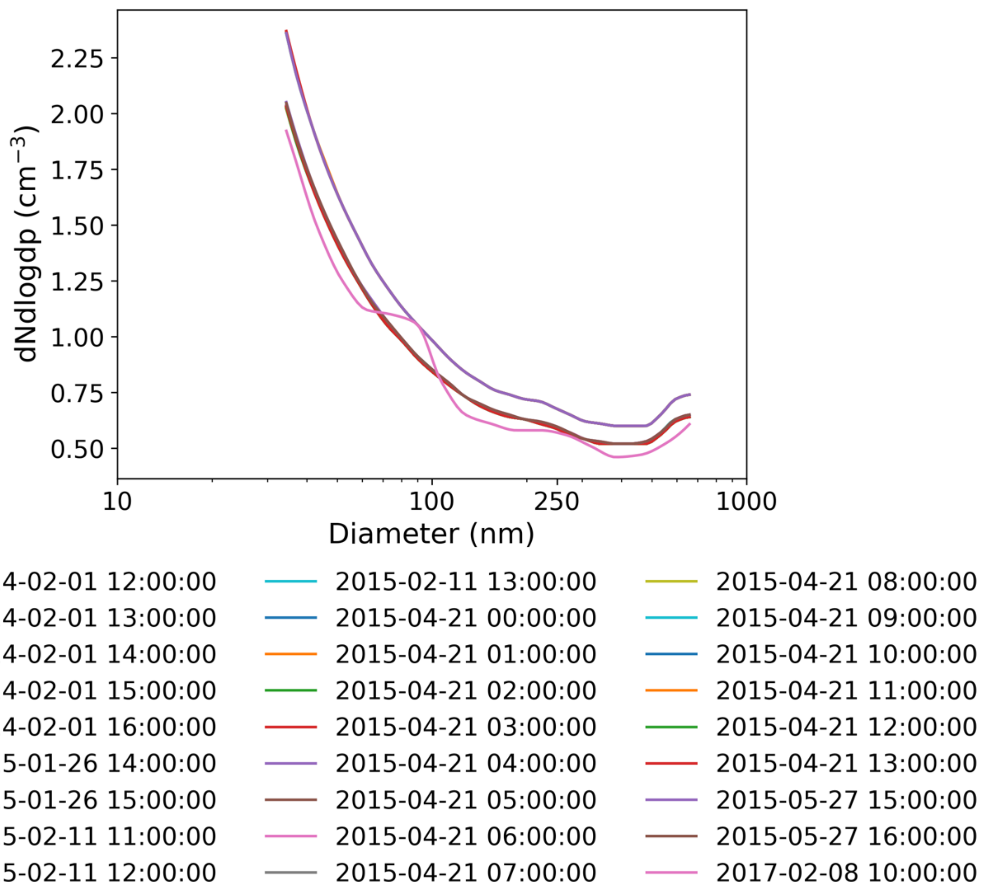

Figure 2: Size distribution for every hourly measurement where the N80 concentrations fall bellow 1cm-3.

We have added some text to the manuscript to demonstrate the necessity of removal of this data:

Lines:258-263: 'Additionally, some extremely low concentrations (N80 < 1cm-3), were constant over several days and upon inspection of the raw data these were deemed unphysical and were removed from analysis. For the last 20 days of the year of measurements in 2015 there was no variation in concentration for 20 days, which is highly anomalous and suggests an instrumentation or recording failure at the site, thus the data from the day containing the start of the anomalous data until the end of 2015 were removed to ensure a robust dataset for the ML framework. The results for this contamination filtering step are shown in Sect. S1.3."

Minor Comments

Thank you for your comments we have addressed these in the text.

1. Lines 11-12: Add a comma to separate the introductory phrase from the main clause.
Corrected and updated.

2. Lines 12-14: The sentence is grammatically confused due to its convoluted structure and a misplaced "which" clause.
Corrected and updated.

3. Line 41: Ensure a comma separates the author name(s) and year (e.g., Fiddes et al., 2024), and check for this throughout.
Corrected and updated.

4. Line 39: "e.g." should not be used before references. Please check elsewhere for similar issues.
This has been updated throughout the manuscript.

5. Line 44: Add a subject before "but" to create two complete clauses. For example: "...will have significant impacts by acting to enhance or dampen RF, but these feedbacks are currently poorly constrained...".
Corrected and updated.

6. Line 69: Add the missing verb. Correct to: "Whilst these techniques are useful to improve understanding of model bias, …".
Corrected and updated.

7. Line 75: Use a comma instead of a semicolon before "However". Check elsewhere for similar issues.
Corrected and updated.

8. Line 407: Insert a comma before "so".
Corrected and updated.

9. Line 426: Change the semicolon to a comma.
Corrected and updated.

10. Line 534: Change "ranking are" to "ranking is" or "rankings are".
Corrected and updated.

11. Line 539: Change "slightly changes" to "slight changes".
Corrected and updated.

12. Line 579: Change "not expect" to "not expected".
Corrected and updated.

13.Line 596: Change "demonstrate" to "demonstrates".
Corrected and updated.

14. Line 671: Change the comma before "therefore" to a period. For example: "...Southern Ocean (McCoy et al., 2021). Therefore, the negative relationship...".
Corrected and updated.

15. Line 683: Change "dilution aerosols" to "dilution of aerosols" or "aerosol dilution".
Corrected and updated.

Additional updates:
During revisions we noticed a minor bug in which the value for one of the parameters returned from the hyperparameter tuning (number of estimators) was not passed to the final model, this has been updated and the models rerun and figures updated throughout. An additional bug was fixed at the same time regarding the deaccumulation time of the ERA-interim precipitation dataset as well as reprocessing the GFEDs data to ensure stability of the processing during parallelised computation.
We additionally updated the calculation of the LAI summary statistics to allow NaN values over Antarctica to be represented as 0 to reduce removal of trajectories spending all the time over the continent. This results in a larger test and train dataset (a total of 6757 datapoints compared to 6497). We have added the following text to explain this:
Lines 451-451:"For LAI it was also necessary to account for the absence of this source proxy over the Antarctic continent."

Due to the stochastic nature of these models, this does result in some shift in the order of features by feature importance, however the impacts of these small changes were minor and none of the relationships or conclusions have changed.

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