RC1 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.

This paper uses a novel combination of back-trajectory analysis and machine learning models to investigate aerosol processes in the Antarctic region, with a focus on sources and sinks of aerosol measured at Trollhaugen as well as the seasonal cycle. I congratulate the authors for this work, which includes a large range of data sources and complex, machine learning-based methodology.

The results show that a machine learning model trained on air-mass history, meteorological data and various proxies for aerosol sources and removal processes can outperform a global climate model form the CMIP6 ensemble when aerosol number concentration is compared to measurements.

Further analysis of the observational dataset and machine learning model using advanced statistical techniques reveals key processes that are important for controlling the aerosol size distribution at Trollhaugen throughout the year. Results from other locations show that the method can also be applied in other regions. The article therefore provides an important contribution both to our understanding of Antarctic aerosol and to the field of aerosol modelling more generally. I recommend that the article is published after minor comments below have been addressed.

General comments

Documentation of ACTRIS data: the time resolution of the raw data is not clear. Also, according to Table S1, 2014 appears to contain more flagged data than other years. Do the authors know possible causes for this, and are there any implications for a possible sensitivity to which years are selected for the testing vs. training datasets?

The first year of data from the Trollhaugen site in 2014 spanned 11 months, starting at the end of January, resulting in a smaller initial dataset compared to the other years. The first filtering step indicates removal of data across the whole timeseries with consecutive NaNs, which will encompass both flagged invalid and missing datapoints, thus for clarity we have separated the number of already empty datapoints to a separate column in Table S1.

We note that for 2014 and 2015 the smallest size distribution bin midpoint diameter 33.4 nm. The majority of data for these years was flagged as ‘0.390’ and ‘0.392’ which indicates that the data completeness was less than 50% or 75% (https://ebas-submit.nilu.no/templates/comments/fl_flag), however this is a listed as a valid measurement, so these were included in the analysis. In addition to this some data entries were flagged as 0.559 listed as “unspecified contamination or local influence, but considered valid” (https://ebas-submit.nilu.no/templates/comments/fl_flag), so these were also included in the analysis.

To investigate processes across the whole seasonal cycle, it is important to select a period that includes data samples for all months. We acknowledge that in Antarctica there will be interannual variability in the strength of aerosol sources and processes (Preunkert et al., 2007; Weller et al., 2011a, b), however in general the dominating processes acting from year to year are likely to be mostly consistent as one of the most pristine sites in the world. The difference between cross-valuation scores and test scores suggests the role of interannual variability or distribution of the data across the seasons compared to the test dataset. To investigate the interannual variability on the SHAP relationships we would require a longer timeseries of data to facilitate multiple test years. This requires consistent overlap of the in-situ measurements, reanalysis data and all satellite products. To facilitate a much larger dataset in order to investigate interannual variability, future studies could investigate the use of multiple measurement sites, in a pan-Antarctic or Antarctic regional study.

We have added and adapted text in the manuscript to discuss these points:

Lines 597-603: We acknowledge that in Antarctica there will be interannual variability in the strength of aerosol sources and processes (Preunkert et al., 2007; Weller et al., 2011a; Weller et al., 2011b), however in general the processes likely to be mostly consistent as one of the most pristine sites in the world. Comparing the seasonal cycle of the test year to the training years (Fig. 3), we note that the test year is generally consistent with the median and interquartile range of the training years. However, there is evidence of interannual variability, for example for the Aitken mode particle number concentrations in April the median falls to approximately the 25th percentile of the training dataset for that month (Fig. 3a).

Lines: 647-648: The difference between cross-valuation scores and test scores (Table 3) suggests the role of interannual variability or distribution of the data across the seasons compared to the test dataset.

Lines 1164-1176: Moreover, expanding the dataset through the addition of measurement sites would allow for investigation into interannual variability and differences in dominant processes across an environment.

Figures and tables: I suggest that for clarity and brevity, Table 3 could be reported directly in the text or combined with table 1, and figures 3 and 4 could be combined into 1 figure. Tables 1 and 5 could also be combined, discussed more below.

Use of different sites: it was not clear to me until later in the article (section 4.6) that results from multiple sites are presented, which makes references like “sites” or “for each site” confusing earlier in the text (e.g. Lines 421, 425, 442, title of section 4.1). Varrio and Mace Head could be briefly mentioned in earlier sections (abstract, introduction or methods) and Tables 1 and 5 could be combined.

We have updated the text referencing “sites” and “for each site” as well as adding mention of Värriö and Mace Head in the introduction and methods sections to improve the clarity around the use of different sites. Thank you for your suggestion regarding table formatting to improve clarity, as suggested we have combined Tables 1 and 5.

Line 196-197: We demonstrate the capability of this framework at Trollhaugen in the Antarctic; however, this framework has been developed to be globally applicable, and we showcase the generalisability of the framework by testing at a boreal forest site (Värriö) and maritime site (Mace Head) (Sect. 4.5)

Line 244-247: The same methodology was applied to in-situ PNSD measurements from the sites used to test the generalisability of the framework: Värriö (VAR) SMEAR I measurement station (67.767°N, 29.583°E, 390m a.s.l.), 120km north of the Arctic circle (Hari et al., 1994) and Mace Head (53.3267º N, 9.9046º W, 8m a.s.l.) on the coast of Ireland (O’Dowd et al., 1998) .

Discussion of UKESM bias: You show that UKESM has a significant low bias in aerosol number concentration and also show that the ML model outperforms UKESM. This is a very interesting result and you rightly say in the conclusions that the approach presented in this study should therefore be used to inform model developments. However, I found the recommendations for future work on this topic a bit vague. Since identifying and reducing sources of model bias seems to me to be one of the main sources of value of this approach, I feel that the article would benefit from more detailed discussion of this in the conclusion.

The first step was developing the framework that works for observations to understand the physical world and ensuring an accurate performance of the real-world ML model if it is to be used to provide GCM process constraints. Understanding key processes and variable proxies for the Antarctic, for example the proxy of Chlorophyll, can be used to target UKESM evaluation, to identify sources of bias.

This framework can be seamlessly applied to GCMs to facilitate identification of differences in relationships and dominant processes to provide targeted identification of potential sources of bias. We have a follow up paper in preparation for submission that demonstrates the application of the framework to GCMs allowing like for like comparison and thus clear insight into the differences in process representation. We have updated the text in the introduction and conclusion to make the discussion on this clearer.

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 1185-1214: 'We benchmark the framework against UKESM at the sites in a Eulerian frame of reference, highlighting the ability of the ML models to better replicate the magnitude of the aerosol number concentrations for both Aitken and accumulation mode. This highlighted the underprediction of UKESM for the Austral summer. By developing a framework that works for observations to understand the physical world we inform understanding of key observational proxies for Antarctic processes. These observational proxies, for example Chlorophyll-a, can then be used to target process driven GCM evaluation and identify sources of the biases. In future work upon expansion of the framework to further sites, consistent sources of model bias can be identified across different environments. This can then be used to improve representation of natural aerosol processes in GCMs tuned for best global representation, for example by updating representation of a proxies such as Chlorophyll-a as investigated in Bhatti et al. (2023). Applying this framework to GCMs to perform a transparent, process-based evaluation by combining GCM derived trajectories with GCM output for explanatory and target variables in a manner consistent with the set-up of the observational models would allow a like-for-like comparison, and thus holistic constraint on aerosol processes.'

Data and code: I note that the authors plan to release the data and code associated with the article upon publication. Because the article's results rely on the synthesis of several datasets and the implementation of complex numerical methods, I strongly recommend that the authors deposit the code and any novel, processed datasets in openly accessible repositories (e.g. Zenodo, GitHub, etc) with persistent identifiers as soon as possible.

Thank you we plan to publish the framework Github via Zenodo once the paper is accepted.

Specific comments

Lines 158-161 and section S1.1: please clarify the time resolution of the raw PNSD measurements used before 6-hourly means are taken. Currently, it is not clear how the filtering steps affects the data availability within 6-hourly windows.

Line 231: Thank you we have now specified that the raw PNSD resolution is hourly.

Lines 180-181: the size ranges for the target variables are given as approximate (~30-80, ~80-660nm), whereas I would imagine that they are defined precisely based on which bins are summed over. Please clarify this.

Line 307: We have now specified the precise size range for each site in table 1, this is based on the bins defined in Sect. 2.1.

Lines 192-193: do the authors plan to release the code for this bug fix, or is it documented elsewhere?

Line 278: This was a bug fix provided by NOAA (personal correspondence), and is included in the most recent versions of HYSPLIT. We have updated the text to clarify this.

Lines 298-302: should "geometric standard deviation" on line 298 be "geometric mean diameter" as on line 300? Please clarify, and if so, consider reformulating the first two sentences to avoid repetition and aid clarity.

Lines 407-410: We have updated the text to geometric standard deviation to fix this typo and reformulated the paragraph.

Line 479: "source" instead of "sources"

Thank you for noting this error we have updated the text.

Line 482: remove comma after "similarly"

Thank you for noting this error we have updated the text.

Line 561: in the text you reference seasonal behaviour: "As found with numerous previous studies, with less sea ice, in the warmer months, Antarctic sites have increased aerosol concentrations associated with more contribution from the ocean." However, this makes it somewhat unclear whether the data presented in Figures 8 and 9 is seasonal or for the full annual cycle – please clarify.

We have updated the figure captions for figures 8 and 9 to clarify that these are for the full test year 2017. And removed the phrase 'in the warmer months' for clarity.

Line 563: "Accumulated chlorophyll is also ranked highly for SHAP of both aerosol size range models (Fig. 7) and demonstrates a very consistent logarithmic relationship (Figs 8d and 9c)." Firstly, it seems like this should read "Figs 8c and 9c." Secondly, the phrase "demonstrates a very consistent logarithmic relationship" is not totally clear to me. What is the relationship consistent over Please specify.

We have updated the Figure letters and removed the phrase 'very consistent'.

Line 702-706: I am not following what physical process could explain the link between increased time in cloud and increased particle number in both mode. "Cloud processing" is given, but I would expect this to increase accumulation mode number at the expense of Aitken mode number, as described in lines 700 and 701. Please clarify this section.

To explore the difference between the SHAP-feature relationships for the different modal models (Aitken and accumulation mode concentration) we performed a model simulation with additional relative humidity and time in cloud variables. We added a non-weighted RH mean variable as well as additional time in cloud variables: the first 12 hours (along the back trajectory initialised at the receptor site), 48 hours, and the first 5 days to try to untangle the timing of the impact of these variables.

From Figure 1, we can isolate that the negative SHAP-feature relationship for RH exhibited for the Aitken mode model is associated with the weighted mean – i.e. weighted for transport close to the site (Figure 1a). Whereas there is a contrasting relationship, albeit non-linear, when considering the non-weighted RH mean (Figure 1b). For the accumulation mode both the RH features demonstrate mostly positive SHAP-feature relationships (Figure 1c and 1d). Thus, this means that high relative humidity close to the site is leading to low concentrations of Aitken mode particles, but high concentrations predicted for the accumulation mode model.

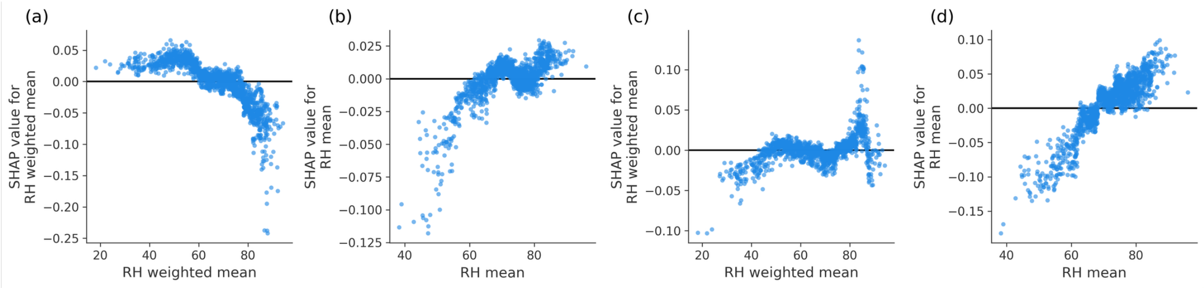

Figure 1: SHAP dependence plots for (a, c) RH weighted mean and (b, d) RH mean. Where (a and b) are for the 30-80nm model and (c and d) for the 80-660nm model.

The role of high RH close to the site is also demonstrated by the negative relationship found for the SHAP-feature relationship for time in cloud considering only the initial 12 hours, contrasting with the time in cloud over longer trajectory periods (Fig. S12). Thus, this substantiates the hypothesis that the growth into the accumulation mode, depleting Aitken mode occurs close to the measurement site, at least within the 12 hours prior to arrival.

In this study we use a 94% threshold to evaluate time in cloud as defined in prior studies (Isokääntä et al., 2022; Tunved et al., 2004) for which it is assumed that an airmass is inside cloud or fog, which is relatively close to the values used for critical RH for cloud formation in reanalysis data and large-scale models (Isokääntä et al., 2022). However, to test the potential for capturing evaporation post-fog detrainment, we have tested a 98% threshold. As shown in Figure 2 the resulting SHAP-feature relationships between for the variable ‘Time in cloud’ are not sensitive to the choice of RH threshold between 94% and 98%.

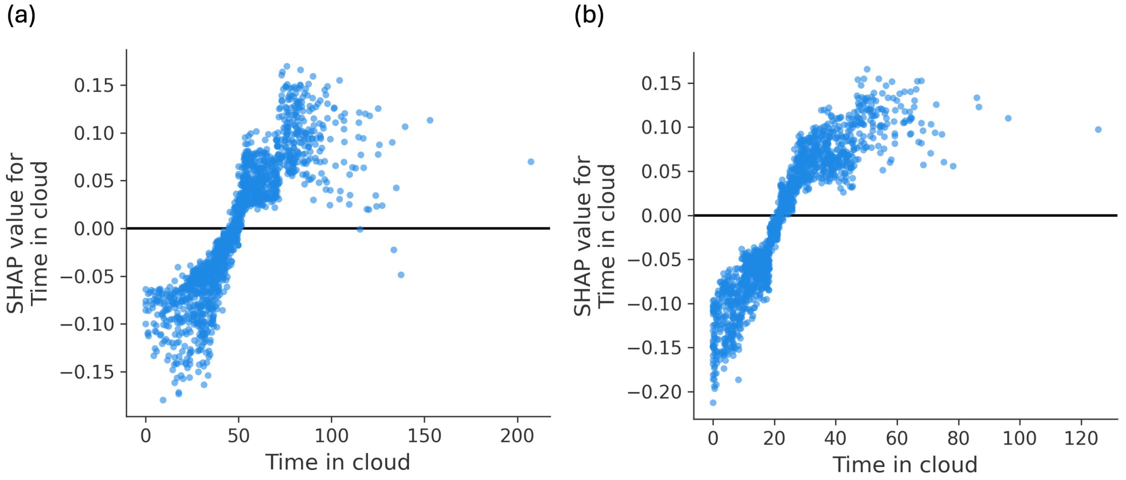

Figure 2: SHAP dependence plots for Time in cloud at (a) RH>94% and (b) PH>98% for the 30-80nm model.

We have updated section 4.4.5 as:

Lines 959-988: The contrasting relationships between RH and SHAP value for the two size range models (Figs 13a and 13c) highlights the importance of RH for growth into the accumulation mode, leading to a reduced concentration in the Aitken mode.

'Time in cloud' is not a weighted variable and therefore, represents the total experienced cloud and averaged relative humidity during the 10-day back-trajectory and shows consistent relationships for both model size ranges (Figs 13b and 13d). With increased time in cloud during transport, there is increased SHAP for both accumulation and Aitken mode models. Additional simulations were conducted including trajectory slices to calculate the additional features: 'Time in cloud – initial 12 hours', 'Time in cloud – 48 hours', 'Time in cloud – initial 5 days' and 'Time in cloud'. This revealed that sign of the SHAP-feature relationship is dependent on the section of trajectory used (Fig. S14). If only considering the transport 12 hours prior to arrival at the station, then there is a negative SHAP-feature relationship between 'Time in cloud' for predicted Aitken mode number concentration, which is consistent with the negative relationship observed with increased 'RH weighted mean'. The change of sign suggests a change in dominating mechanism for the Aitken mode represented by the RH and 'Time in cloud' variables.

As 'Time in cloud' is defined based on a relative humidity threshold of 94%, this will encompass cloud edges and high humidity regions, not solely cloud. In the Arctic aerosol concentrations have been found to be strongly affected by evaporation post-fog detrainment (Kecorius et al., 2023), which could be associated with the positive SHAP-feature correlation for 'Time in cloud'. In the Southern Ocean, McCoy et al. (2021) found that Aitken aerosol, that subsides or is entrained from the free troposphere, grow to accumulation mode sizes in-cloud and sub-cloud. The study presents evidence for a hypothesised Aitken-buffering mechanism: scavenging of CCN by precipitation increases peak super saturation, activating some Aitken aerosol, thus maintaining high summertime Southern Ocean droplet number concentration through activation and growth of boundary layer Aitken particles (McCoy et al., 2021). From the SHAP analysis for Trollhaugen we find that the SHAP-feature relationship for 'Time in cloud' has a large contribution from the experienced surface solar radiation for the Aitken mode (Fig. S8b). The increase in predicted Aitken mode concentrations with time in cloud is particularly strong for periods of low insolation (Fig. 13). Considering the SHAP interaction values (the impact of two features together), there are contrasting relationships for periods of high insolation compared to periods of low insolation (Fig. S15). This could suggest that the role of the Aitken-buffering mechanism in the summer is depleting Aitken mode concentrations. An important factor to consider is the fixed size ranges utilised in this study, as modal diameter limits are not static and vary diurnally and seasonally across the Antarctic (Brean et al., 2025; Järvinen et al., 2013; Kim et al., 2017), thus the threshold of 80nm could conflate Aitken mode and accumulation mode particle number concentrations.

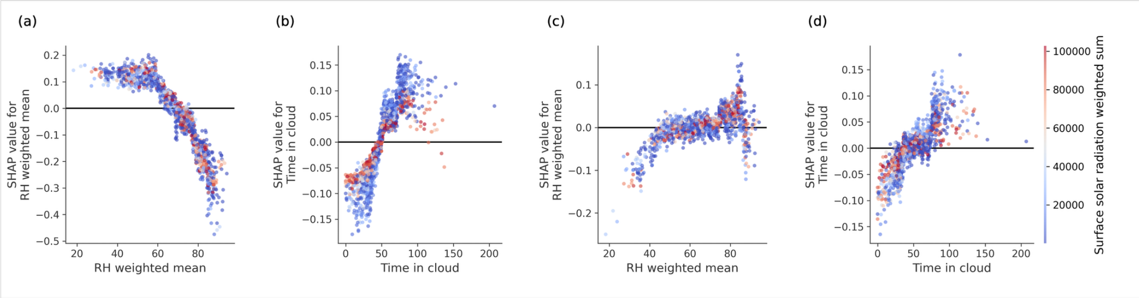

Figure 13: SHAP dependence plots for (a, c) RH weighted mean and (b, d) Time in cloud. Where (a and b) and for the 30-80nm model and (c and d) for the 80-660nm model, with the corresponding values for surface solar radiation weighted sum indicated by the colour of points.

We have added the following figures to the Supplement:

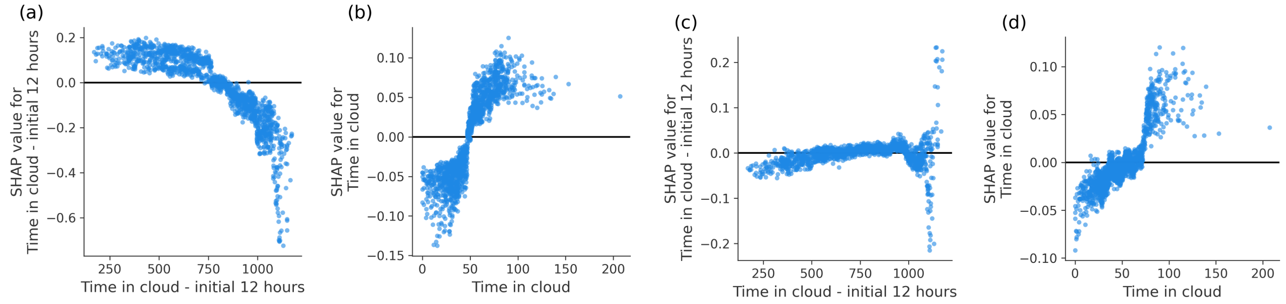

Figure S14: SHAP dependence plots for (a, c) Time in cloud – initial 12 hours and (b, d) Time in cloud (along the full 10-day trajectory). Where (a and b) and for the 30-80nm model and (c and d) for the 80-660nm model.

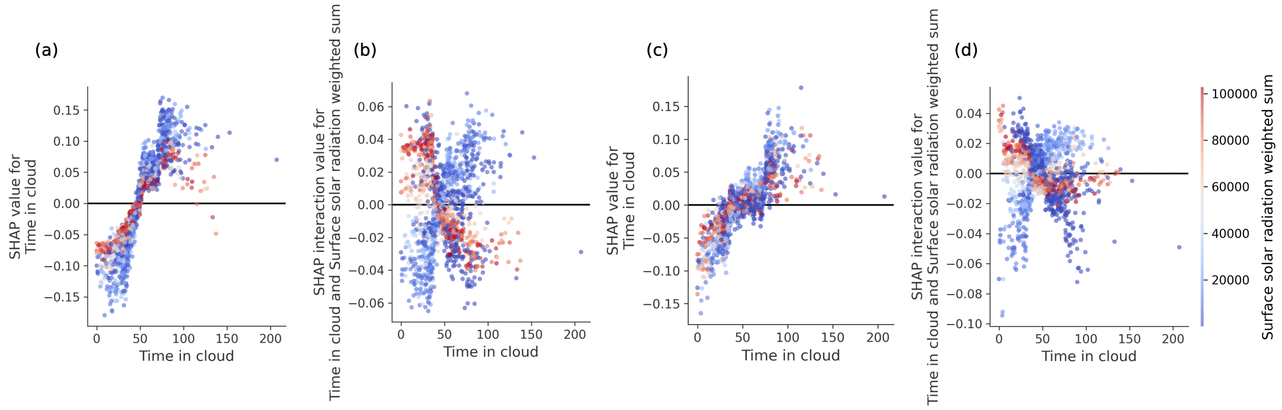

Figure S15: SHAP dependence plots for (a, c) Time in cloud. SHAP interaction value dependence plot for Time in cloud and surface solar radiation weighted sum. Where (a and b) and for the 30-80nm model and (c and d) for the 80-660nm model, with the corresponding values for surface solar radiation weighted sum indicated by the colour of points.

Lines 802-804: “consistent sources of model bias can be identified, to improve representation of natural aerosol processes in GCMs tuned for best global representation” although this study does indeed present UKESM underestimation of aerosol number and shows that the ML models outperform UKESM in this regard, the rest of the results focus only on insights into the observations and ML model. Please add more detail about how this approach can be used to identify sources of model bias, such as the bias presented in Figure 4.

The first step was developing the framework that works for observations to understand the physical world and ensuring an accurate performance of the real-world ML model if it is to be used to provide GCM process constraints. Understanding key processes for Antarctic, for example the proxy of Chlorophyll, can be used to target UKESM evaluation, to identify sources of bias.

This framework can be seamlessly applied to GCMs to facilitate identification of differences in relationships and dominant processes to provide targeted identification of potential sources of bias. We will be submitting a subsequent paper demonstrating the application of the ML framework to GCMs focused on revealing which processes drive the observed biases in aerosol properties. We have updated the text in the conclusion to make the discussion on this clearer.

Lines 1185-1214: 'We benchmark the framework against UKESM at the sites in a Eulerian frame of reference, highlighting the ability of the ML models to better replicate the magnitude of the aerosol number concentrations for both Aitken and accumulation mode. This highlighted the underprediction of UKESM for the Austral summer. By developing a framework that works for observations to understand the physical world we inform understanding of key observational proxies for Antarctic processes. These observational proxies, for example Chlorophyll-a, can then be used to target process driven GCM evaluation and identify sources of the biases. In future work upon expansion of the framework to further sites, consistent sources of model bias can be identified across different environments. This can then be used to improve representation of natural aerosol processes in GCMs tuned for best global representation, for example by updating representation of a proxies such as chlorophyll-a as investigated in Bhatti et al. (2023). Applying this framework to GCMs to perform a transparent, process-based evaluation by combining GCM derived trajectories with GCM output for explanatory and target variables in a manner consistent with the set-up of the observational models would allow a like-for-like comparison, and thus holistic constraint on aerosol processes.'

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 0s to reduce removal of trajectories spending all the time over the continent. This results in a larger test and train dataset. 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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