Towards an improved understanding of the impact of clouds and precipitation on the representation of aerosols over the Boreal Forest in GCMs

Manuscript overview

This manuscript presents research that investigates aerosol-cloud interactions at a Boreal forest location. The study focusses on how cloud and precipitation processes affect aerosol properties including aerosol concentration, size distribution, and chemical composition. The study compares model output from two global climate models to observations made at the Boreal forest location. The study then uses a Lagrangian framework to investigate the aerosol cloud interactions along trajectories prior to their arrival at the measurement site in summer and winter conditions.

The study presents detailed results describing aerosol-cloud interactions in the Boreal forest location and provides useful insights into the global climate model's behaviour, for which the authors are to be commended. However, the manuscript requires improvements so that the results, and their significance, are clear to the reader. I have made some general and technical comments below.

General Comments

The manuscript presents quite a lot of detail across the results sections, supplementary material and the appendices, and I found it difficult to follow and understand. I think that the results of the research need to be better focussed on the study's main objectives (1-2).

For me, the novel aspect of this study was the model-model-observation comparison, why there are differences between the models, where there are model biases compared with the observations and how the models might be improved. I think this 'story' needs to be made clearer throughout the manuscript.

1. I think the additional research questions (a, b) detract from the focus of the manuscript. The relevant results could be reported in the supplementary material. I also do not think additional research question (b) is necessary. This is because the analyses only looks at if precipitation in UKESM1 and ECHAM-SALSA is representative of AeroCom GCMTraj. However, in this study aerosol behaviour in the models is also very important for explaining differences between the models and observations.
2. I think that the manuscript could be better organised to help the reader understand the main results and their significance.
   a. I suggest that the authors decide on what order to present their results to best tell their story and keep this consistent throughout the manuscript. This should then be summarised at the end of the Introduction to guide the reader through the manuscript. E.g. "In Section 2 we present…." etc , etc.
   b. Sections and sub-sections could also be organised more clearly to guide the reader. For example, in the Introduction, the Lagrangian analysis is discussed across Paragraphs 1 and 3.
   c. The manuscript's supplementary material is very useful to understand some of the study details. However, I think that the main manuscript, including figures, needs to 'standalone'. For example, where results have been presented for one aspect of the analysis (e.g. for one season) a summary of the rest of the analysis and reference to the Supplementary materials could be given at the end of the section.
   d. Some longer sub-sections could be split up further. For example by aerosol type (organic aerosol and sulphate) in Section 4.2.

2. I think the models should be referred to as global <u>climate</u> models or Earth System models (ESMs). UKESM is definitely an Earth system model.
3. Isokääntä et al., 2022 is regularly referenced. This manuscript appears to use observational data from Isokääntä et al. and extend their analysis techniques to GCMs. In the Introduction the authors could consider including a paragraph clearly summarising of Isokääntä et al. and how this study builds on it.

Technical comments

<u>General technical comments</u>

- Colloacation => co-location
- Airmasses => air masses
- Use of 'hexagon'. Is this analogous to a model grid box?
- Please consider using "particles cm^{-3}" instead of "# cm^{-3}".
- For clarity please consider using "updraft velocity" rather than "updraft".

<u>Abstract</u>

- This needs to include the main study objectives and the main conclusions, i.e. what does the study tell us about how precipitation impacts aerosol-cloud interactions in GCMs?

<u>Introduction</u>

- L53: **and therefore** partly masking….
- L67: "Such frameworks facilitate ~~the way for~~ the development…" OR "Such frameworks **pave** the way for the development…."
- L85-117: Paragraph 3 is very long and would benefit from being broken up.
- L126-129: *"In this work, the effects of wet processing (wet removal and aqueous phase processing) along air mass trajectories on modelled aerosol size distributions are compared with long-term observations of aerosol size distributions in Hyytiälä, Finland. The observations are combined with ERA-Interim reanalysis trajectories, and the trajectories to be utilized with the GCM variables are calculated with the GCM simulation (nudged to ERA-Interim reanalysis) output meteorology"*

  These sentences are really important because they describe the research, but the second sentence is difficult to understand.

- L98-99: *"…more efficient removal on the number concentration of the accumulation mode particles with increasing accumulated precipitation…"*

  Suggest: "…more efficient removal of the accumulation mode particles with increasing accumulated precipitation…"

- L100: environment => environments

<u>Methods:</u>

- L168-169: "*The dataset used in this study is reduced compared to Isokääntä et al. (2022) and extends to the end of 2018 to facilitate comparisons to the simulation period of the GCMs.*"

  Do you mean a shorter time series? Or have other constraints been applied to the data for quality control purposes? What is the time period used for this study – is it 2005-2018?

- L174-178: This sentence needs rewriting for clarity. I suggest describing one model component per one sentence.
- L181-185: Please edit this sentence for clarity. I suggest using shorter sentences.
- L207-208: "…chemical components including mass mixing ratios of sulfate noted here as SO4 (extracted as sulfuric acid H2SO4 and then converted, see Sect. S1.1),…" See also Section 2.4.3.

  Could the authors please explain why they have taken this approach? UKESM does output mass mixing ratios of sulphate aerosol (SO_4^{2-}) for the different aerosol modes (nucleation, Aitken etc) and I'm not sure why these were not used. UKESM's H_2SO_4 diagnostic is sulphuric acid in the gas phase.

- Section 2.2: Could the authors please include a mention of how were the GCM simulations extended beyond 2014 and up to 2018? Was a future simulation used from one of the SSP scenarios used? Although there is little expected difference in SSPs for the 2014-2018 period, it would be useful to see what was used.
- L241 By "4 d" do you mean four day?
- L277: "…allows to retain the…" => "…permits retention of the…"
- L277-279: While I understand the reasons for the author's approach here, I'm not sure 'ensuring consistency' is the right term because the precip is ERA, which the aerosol variables are GCM.
- Section 2.4.1: Were the hourly observations averaged to co-locate with the 3 hourly model data?
- L300: Please define DMPS here
- L300: Please define ACSM here

Results sections

- In general, at the start of a sub-section or paragraph, please try to give an overview of what the relevant figure is showing. The sub-figures can then be referenced as each result is described. Please see the Section 3.1 comments for a suggestion.

Section3:

- In paragraph 1 consider indicating which variables are be compared between the model and observations - Aerosol particle number size distributions and chemical composition
- L340-342: "For the GCMs, fewer studies looking into aerosol properties at single sites exist, but Leinonen et al. (2022), for example, conducted an extensive study comparing long-term aerosol particle seasonality and trends in observations and GCMs in multiple locations, also including SMEAR II."

  This could be in the Introduction so that the Results sections can focus on the study's analyses

- L347-354: I suggest that this could be a short subsection. Something like "Comparison of transport for ERA and GCMs". Figure S4 could be included in the main manuscript with the GCM output shown as difference plots compared with the ERA data. This would also help illustrate the study location.

<u>Section 3.1</u>

- L356-363: I suggest re-writing the first paragraph as follows to help guide the reader. Sub-figures can be referenced as the results are described. Information about the time periods plotted and which percentiles are used for the statistics can be clearly stated in the Figure caption.

  In Figure 1 particle number size distributions from the GCMs is compared with observational data at the SMEARII. Median aerosol number concentrations for nucleation, Aitken and accumulation mode are shown in Table S5. ECHAM360 SALSA data in Figure 1c is presented in its native resolution for size bins falling between $dp = 3.0 - 1700$ nm and those size bins are positioned within the y-axis to the geometric mean of the ECHAM-SALSA size bins (see Table S3). To calculate the difference in Figure 1e, the measured size distribution is regridded to the ECHAM-SALSA bins by integrating between the upper and lower limit of each ECHAM-SALSA size bin.

- L375: *"…this could indicate from other processes which dominate the accumulation mode."*

  I suggest re-writing as: "…this could indicate that there are other processes that dominate the accumulation mode."

- Figure 1: Consider reducing the number of sub-figures. For example, omit 1b and 1c and just show the difference plots (1d and 1e).

<u>Section 4</u>

- I suggest that Section 4 is re-written to help guide the reader through it and to help the reader understand the main results. I suggest that the analysis of the impact of extracting the vertically resolved liquid precipitation from UKESM1 described in paragraphs 1 and 2 is described in its own section, e.g. 4.1. I then think that including the analysis of the representativeness of precipitation in UKESM and ECHAM-SALSA compared to the AeroCom GCMTraj ensemble here is distracting. I think this analysis, together with Appendix B, could be moved to the supplementary material.
- L440: "smallest" => smaller
- L445-451: I suggest removing the sentence "*The analysis is simplified by removing the size dependent component noted in previous literature (see e.g., Figure 3 in Isokääntä et al., 2022 and Figure 4 in Khadir et al., 2023)…*"

  I don't understand what the sentence is referring to here, but it could be included when the relevant results are shown later.

- L445-451: Based on the above comments I then suggest starting Section 4 with the following paragraph:

  "In this section we use a Lagrangian trajectory framework to…. In Section 4.1 we examine the impact of using vertically resolved liquid precipitation, which has not previously been done for Lagrangian trajectory analyses. Then the investigation inspects the relationship between accumulated precipitation and aerosols for the two GCMs used in this study: UKESM1 and ECHAM-SALSA. In Section 4.2 we focus on total aerosol mass and number. In Section 4.3 we focus on the OA, BC, and SO4 portions of the total mass for submicron-size aerosols. Then, in Section 4.4, the processes controlling the precipitation-aerosol relationships are investigated, and the differences are discussed in detail between the GCMs and within each GCM."

- Section4.1: To help the reader understand the results in this section I think it would be helpful to describe them after Figure 4. Why you have looked at seasonal periods (as in L466-474) can then be discussed as you discuss the results shown in Figure4. Additional results such as those in Figures S7-9 can be mentioned at the end of the Section. I also think that statements like *"Non-normalized mass and number concentrations are shown in Figures S8 and S9, and those are similar to Isokääntä et al., (2022) which employed whole year data and total precipitation"* (L475-476) are not useful. How are the mass and number concentrations similar to Isokääntä et al., (2022) and why is this important? The novelty of this study is the model-observation comparison. If it is important the observational data used here is similar to Isokääntä et al., (2022), there should be an additional analysis.
- L471-474:
  (i) "…which further cause differences…" => "…which causes further differences…"
  (ii) I suggest splitting this sentence up for readability.
- L477: In describing Figure 4 here and elsewhere in the manuscript the authors refer to "removal of the normalized masses…" However, the plots are not actually showing removal processes. I think it would be more correct to refer to e.g. reductions in normalized masses.
- L480-483: Why would there be a seasonal effect of UKESM's missing boundary layer nucleation process?
- L539 and Figure 5 caption: "…number of data rows for the bin was 10 or larger." What does this mean?
- L575: Activated fraction? Is this the fraction of N_{80} particles that have been transported upward?
- L609-610 "These regions coincide with the high trajectory frequencies; thus, the high updraughts are being reflected on the averages irrespective of transport direction in Figure 7j."

Could the authors please clarify this statement.

- L614: "…where the north of Norway…" The region of very high activated fraction appears to extend over the Arctic Ocean.
- L621-623: *"Assuming the solubility of OA compensates for the missing portion of SO4 in summer, these differences could increase the particle activation potential even further during summer in UKESM1 (compared to winter)."*

I think this is a bit difficult to follow.

- L624-626: Please consider re-wording as "Figure 8 shows the relationship between mean activated fraction and mean updraft velocity that the air mass experienced before arriving at SMEAR II for the summer and winter periods."
- Figure 8: Please consider resizing the text annotations on the plots.
- L634: Here, I think it should be "lower concentrations" rather than "smaller concentrations"
- L649: "…which is steeply…" => "…which it steeply…"
- L650-651: *"As these differences are taking place relatively close to SMEAR II, it is likely that they are driven by the seasonal differences in the transport very close to SMEAR II."*

Could the authors please explain why that might be?

- L722: I suggest re-wording *"…UKESM1 updraughts are below and above 0.2 m s-1…"* to "…UKESM updraught velocities range from 0.2-0.4 m s^{-1}…"
- L749-768: Please consider putting this text in a new sub-section with a heading something like "Other reasons for model differences"

- L826-829: Possibly, but this would be balanced against likely decreases in SO2 emissions due emission control policies in many regions.
- L837-846: Please consider putting this paragraph before the preceding discussion on SOA
- L835-834: In this study region or in general?
- L844-846: Are the same (e.g. CMIP6) emissions data sets being used in both models? Would rule out differences in emissions.
- Figure 11: I think that it would be useful to include the observations for the for corresponding period and sector.
- L896-904: Please consider re-wording this paragraph because I cannot understand what it means.
- L902-904: Both models have very limited aqueous phases schemes – do the results imply that this does not matter too much, at least at the Boreal forest location?

Section 5

- In common with Section 4, there are many references to the supplementary materials. I think the text could generally be improved by clearly summarizing the main findings as shown in the main manuscript's figures.

Conclusions and outlook

- I suggest that this section is edited to ensure that the main results and their impact is clear to the reader. I also think that the authors should include some discussion on the impact of their results for GCMs. For example:
  - What were the main model biases against observations?
  - Are there any processes that should targeted for model development?
  - Are the results likely to be applicable to different locations? This is difficult, but important because GCMs are used for global climate model prediction.

Supplementary

- Tables S1 and S3: I'm missing a description of "GSD" and "GMD" in column 4.
- Figures S4 and S5: Difference plots would help to illustrate the differences, or lack thereof, between ERA and the model simulations.