

Referee feedback in black, author responses in blue

The authors have made extensive revisions to the manuscript, which is now much better organised and easier to follow. This is to be commended. However, I do think that the conclusions section needs to be strengthened. I have made General comments below regarding the Conclusions in Section 6, as well as some minor Technical comments, and once these have been addressed I recommend that the manuscript is published.

We thank the referee for thoughtful feedback and positive assessment of the revisions. We appreciate your constructive suggestions regarding the Conclusions section and the technical comments, and have now addressed these points carefully in this revision.

In addition to the requested changes, two technical corrections were applied,

1. the author affiliations were corrected
2. explanation for CCN (cloud condensation nuclei) was added when first mentioned (in page 20)

General comments:

Conclusions regarding Objective 1.

I think the sentences in e.g. L843-844, L853-854 do not summarise the study's findings clearly.

From my interpretation of Figure 4 the models do predict the observed trajectory-based relationships between particle mass and precipitation for total aerosol and the three aerosol species. However, there are model biases in trajectory-based relationships between particle number and precipitation that are different for the model and season.

So I think that the Conclusion should state that: UKESM exhibited significant loss in particle number via precipitation compared to the observations (and ECHAM) in summer. The lack of boundary layer nucleation (BLN) likely contributes to more, large particles that rain out more easily. Figures 7 and 8 also show that the strong relationship between activated fraction and updraft velocity in UKESM in summer may also contribute to the model's bias in number concentration.

We do agree with the referee regarding this conclusion, and apologize that our writing was not yet appropriate enough such that the message would be clear. Please see our reply at the end of these general comments on how the referred text was revised.

However, in winter both models overpredict particle number concentration with accumulated precipitation compared to the observations. The lack of BLN may contribute to UKESM's bias, but I'm curious if both model's overprediction of number concentration could be due to underprediction of accumulated precipitation <2mm - where the largest model biases in number concentration occur (Fig 4c)?

We thank the referee for this very careful observation regarding the differences in number of trajectories with certain number of trajectories. Indeed, the overprediction during winter could be partly due to the difference in the smaller number of trajectories with accumulated precipitation below 2mm (as can be seen from Fig. 4c where purple bars i.e., observations, have larger count compared to the models). This could cause the fact that the particle number removal is not as efficient in the models as in the observations during wintertime. On the other hand, with larger accumulated precipitations the models show larger trajectory counts, which in turn can contribute to the fact that the normalized number concentrations approach each other's (Fig. 4b) with increasing accumulated precipitation. Therefore, the differences in the precipitation are unlikely the only explanation,

however, this, indeed, could contribute to the differences. We have now added a sentence to results section 4.4.3 acknowledging this (starting from line 632): *“However, some of the winter differences may also be attributed to variations in the number of trajectories with specific amounts of accumulated precipitation (Fig. 4c). Observations show a higher frequency of trajectories with low accumulated precipitation (<2 mm), whereas the models produce slightly more trajectories with larger precipitation totals.”*

This is now also brought up in the revised conclusions which were also adjusted as suggested by the referee. For this, please see our reply below.

The role of activated fraction, updraft velocity, and the relationship between them (Figures 7 and 8), on model biases in trajectory-based relationships between particle number and accumulated precipitation in winter are less clear to me. For example, I don’t follow that: *“The seasonal differences we observed in these variables, along with changes in particle chemistry during the transport, were consistent with the aerosol-precipitation relationships.”* (L853-854). Are the model biases due to too much large aerosol, or a too-weak relationship between activated fraction and updraft velocity – perhaps because the GCM’s do not resolve the local meteorology well?

The model biases arise from both effects (aerosol number & relationships between activated fraction and updraught velocity) the referee mentions. With the current set up in our work, however, it is not possible to perfectly distinguish the actual roles of these factors such that we would be able to determine which has the largest effect—these are also impacted by each others.

Regarding the referred lines in the previous version of the manuscript (L843-844, L853-854) we have now revised the second and third paragraph of the conclusions completely and arranged them into three paragraphs for clarity. We also added a reference to a very recent study by Virtanen et al., 2025, which highlights the inter-model differences (and differences to observations) in the relationships between droplet number and updraughts.

“Our first objective was to investigate whether trajectory-based relationships between aerosol mass, number, and precipitation differ between observations and GCMs. For aerosol mass, the observed removals generally fell between those simulated by ECHAM-SALSA and UKESM1 across seasons, indicating that both models captured the observed mass–precipitation relationship for total aerosol and individual species (OA, SO₄, BC). In contrast, aerosol number revealed clear model biases that varied by season. In summer, UKESM1 exhibited a pronounced loss of particle number via precipitation compared to both observations and ECHAM-SALSA. This bias likely stems from the absence of boundary layer nucleation, which produces fewer small particles and leaves a larger fraction of particles susceptible to wet removal.

Key variables influencing the wet removal processes, such as number of potential cloud condensation nuclei (N_{80}) and updraught velocities, were also examined to evaluate the observed removals. In UKESM1, a strong summer correlation between activated fraction and updraught velocity (Figs. 8) may further increase particle number removal. However, analogous study examining droplet number/CCN versus updraught (Virtanen et al., 2025) show substantial variability across models, highlighting that the relationship between updraught and particle activation remains uncertain and warrants further investigation. In winter, both models overpredicted particle number removal relative to observations. This overprediction may in part reflect differences in precipitation statistics, with models simulating fewer low-precipitation trajectories (<2 mm) than observed (Fig. 4c). However, other factors such as particle size distributions, activation efficiencies, and limitations in the representation of subgrid-scale

meteorology are also likely to contribute. Overall, our results emphasize the need for better representation of particle number size distributions (PNSDs) in GCMs.

Earlier work has indicated that aerosol activation into cloud droplets followed by rainout is the dominant wet removal process. Our results support this, with UKESM1 showing nucleation followed by rainout as the largest contributor. Supplementary analysis comparing a wider ensemble of GCMs indicated that these two models were broadly representative, with their aerosol–precipitation relationships generally falling near the middle of the inter-model spread. Overall, our method using normalized submicron mass and number as a function of accumulated precipitation proved to be effective in comparing removal across models, though it lacks details on particle size evolution—an important topic for future work.”

Technical comments:

Section 1: Introduction

1. L141-142:

“Do the GCMs exhibit similar increase in sulfate mass due to in-cloud production as the observations and are the observed effects reasonable when reflected to model parametrizations?”

Suggest compared instead of “reflected”

The word “reflected” is now changed to “compared” as suggested.

2. L143:

out -> our

We thank the referee for noting this typo. It has now been corrected.

3. L145:

Please define the SMEAR II acronym here or refer to e.g. ‘the measurement station’.

Indeed, this is the first time when the acronym is used. We have now adjusted the sentence to “The aerosol properties at the measurement station (Hyytiälä, Finland) are given...”.

Section 2: Data and Methods

4. L190:

“...new particle formation in the boundary layer is not yet implemented in UKESM1 (Mulcahy et al., 2020).”

While not a correction as such, boundary layer nucleation is now available in UKESM1.1 and will be released with the next UKESM version.

This is a good point indeed. We adjusted the sentence to reflect this “..new particle formation in the boundary layer is not implemented in this version of UKESM1 (Mulcahy et al., 2020)”

5. L192-193:

“...were ran longer to cover years from 2005 to 2018...”

I suggest: “...were extended for the period 2005 to 2018...”

The given suggestion is now implemented.

6. L242:

GCMoutput -> GCM output

We thank the referee for noting this typo. The missing space is now added.

Section 4

7. L438:

“...thus unlikely driving differences...” -> “...thus unlikely to be driving differences...”

This has been adjusted as suggested.

8. L509:

Figure 4 and 4.-> Figure 4 and 5.

We thank the referee for noting this typo, it has now been corrected.

9. L850:

“Aerosol activation into cloud droplets followed by rainout appears to be the dominant removal process”

Dominant wet removal process

The word “wet” has now been added to the sentence as suggested.

References:

Virtanen, A., Joutsensaari, J., Kokkola, H., Partridge, D. G., Blichner, S., Seland, Ø., Holopainen, E., Tovazzi, E., Lipponen, A., Mikkonen, S., Leskinen, A., Hyvärinen, A.-P., Zieger, P., Krejci, R., Ekman, A. M. L., Riipinen, I., Quaas, J., and Romakkaniemi, S.: High sensitivity of cloud formation to aerosol changes, Nat. Geosci., 1–7, <https://doi.org/10.1038/s41561-025-01662-y>, 2025.