

Author Response RC1

We thank the reviewers for their thorough and constructive comments, which have helped to improve the quality and clarity of our manuscript. We have carefully considered all suggestions and made corresponding revisions in the manuscript. Below, we provide detailed responses to each comment, with changes clearly indicated in the revised version of the manuscript.

Reviewer comments are reproduced in **bold**, and our responses follow each comment in **plain text**. Line numbers refer to the revised manuscript unless otherwise stated.

- Start of RC1 Comments: -

In a previous paper, Svenhag et al (GMD 2024) implemented a mechanism for new particle formation (NPF) from H₂SO₄ and NH₃ in EC-Earth and found minor differences in CCN, but despite this they found a change to the radiative effect of clouds of 0.28-1Wm⁻². They performed a model evaluation, checking the aerosol size distribution at 12 surface stations worldwide.

In this paper, the authors focus on simulating NPF events at two surface stations using (I think) the same EC-Earth simulations, and compare them to an ADCHEM model run, presumably one of those published in de Jonge et al (E, S & T 2024). A new EC-Earth simulation without NPF is included, but only features in part of the analysis.

The simulations overestimate primary emissions in winter. ADCHEM has some different NPF mechanisms including iodine and dimethylamine, and no organic NPF, and performs better.

There's an interesting set of plots comparing simulated and observed size distributions and "bananas" at two sites. However, given that most of the simulations (as far as I can tell) are already published and a very similar evaluation of ADCHEM at these sites was already published by de Jonge et al (2024) e.g. their Figure 3, it left me wondering whether the paper currently satisfies the "substantial new concepts, ideas, methods, or data" review criterion for ACP. I think it could, if the analysis were broadened and deepened to probe the model more comprehensively and replace some of the speculations in Section 3.2 with additional detailed analysis or sensitivity studies. This is likely to need new simulations.

Major comments

In my assessment, the paper relies for its novelty on two aspects

- 1. The evaluation of simulated size distributions as a function of time ("banana plots") in EC-Earth, which is interesting and not often done with aerosol-climate models.**
- 2. The comparison between ADCHEM and EC-Earth.**

One could argue that the value of (1) is limited for people outside the EC-Earth community because the size distributions are dominated by the effects of the coupling to the IFS meteorology, and it's hard to disentangle the odd behaviour resulting from this this from the effects of the NPF. However, it's still interesting to see.

The value of (2) is currently limited since the authors only show one ADCHEM simulation with very different nucleation mechanisms to the EC-Earth simulations. Perhaps there is an opportunity here to analyse the other ADCHEM simulations published by de Jonge et al,

which include simulations that only include H₂SO₄-NH₃ NPF that would (at least to an outsider) seem to be a fairer comparison to the EC-Earth CLUST simulations.

If the sentence in the abstract “When comparing diurnal EC-Earth model results with ADCHEM and observations, we establish that using solely organic-H₂SO₄ nucleation parameterization will underestimate the aerosol number concentrations” is not to be misleading, the authors should test what happens when ELVOC nucleation from Riccobono et al (2016) is included in ADCHEM. Currently ADCHEM does not tell us about the organic-H₂SO₄ nucleation parameterization.

We thank the reviewer for their insightful comments and appreciate the opportunity to clarify our work. Our primary aim with this study is to present a detailed characterization of the EC-Earth3 model’s aerosol size distribution, particularly highlighting the diurnal, seasonal, and vertical variability introduced by the newly implemented nucleation mechanism. Previous evaluations have largely relied on long-term averages, potentially masking important sub-daily and seasonal dynamics that are crucial for understanding the performance and behavior of aerosol-climate models. Our results indicate that performing detailed comparisons between Earth system models and observations at a high temporal resolution reveal previously unexplored insights into model structures. We believe that this process-level focus, e.g. nucleation events, adds unique information to existing model evaluation efforts, even though the results may be especially relevant to those working within the EC-Earth community.

Concerning the comparison to ADCHEM, we fully agree that such comparisons can be powerful tools for identifying model behavior and constraints. In this work, our intention was to use ADCHEM as a reference for diurnal variability, as well as to validate the general trends in particle number concentrations and growth. However, we acknowledge the reviewer’s point that the comparison is limited by the fact that the ADCHEM simulation shown uses different nucleation mechanisms from the EC-Earth3 CLUST configuration. We agree that additional ADCHEM simulations (including those with simpler nucleation mechanisms e.g., H₂SO₄-NH₃ only) could provide more model insight.

Nevertheless, performing such comprehensive work of additional ADCHEM simulations, including detailed sensitivity experiments, would extend beyond the scope of this paper. The primary reason for this is that ADCHEM and EC-Earth3 are fundamentally different in terms of model architecture, spatial resolution, and process representation. So, changes in nucleation parameterizations in ADCHEM are unlikely to yield results that are directly analogous to modifications in EC-Earth3, limiting the interpretability of such a comparison within the context of this study.

With that in mind, our study aims to assess differences between a coarser Earth system model and a detailed process model rather than to reproduce identical nucleation conditions in both models. The ADCHEM simulation included in our study was chosen to highlight the contrast in nucleation parameterizations and model frameworks. In response, we have revised parts of the text and added new figures, which include further analysis, seen in the new manuscript version.

Minor comments

–In Figure 2e, NPF is so non-linear that the annual mean NPF rate must be dominated by values close to zero. Would something like the 95th percentile make more sense instead?

That is an interesting point. After testing this, the authors have adjusted and analyzed this plot using the 95th percentile, this elevates the disparity between the CLUST and Control case in the free troposphere further. The plot now shows the 95th percentile, including the added Fig. A1 of 1000–800 hPa.

–Using the Jokinen et al (2015) yields for ELVOC with the Riccobono et al 2014 nucleation mechanism likely leads to high uncertainty. More discussion of this might be useful in the light of developments to organic aerosol schemes in ADCHEM by the paper’s coauthors (e.g Roldin et al, Nat Comms 2019).

The authors have now addressed this in the Further discussion section on Line 327 as: “Additional uncertainties in the EC-Earth3 aerosol chemistry can also arise from the simplifications in modeling organic oxidation yields (from Jokinen et al. 2015) and growth processes. However, incorporating more complex chemical mechanisms is often constrained by the computational costs associated with most Earth system models.”

–A table of the NPF mechanisms in ADCHEM, and in general a more detailed description of how ADCHEM simulates particle formation and growth and how fair the comparison with a climate model is, would be useful.

While this is a valid point, adding further ADCHEM description does not contribute much to our paper’s focus here, the authors however, would refer the reader to our references Wollesen de Jonge et al. (2024) and Roldin et al. (2019) where the model mechanisms are described in detail.

–The differences between the CLUST schemes are described, but the value of including two EC-Earth simulations with different CLUST NPF mechanisms would be greater if differences (or similarities) between the behaviour of the simulations (ie the number of particles formed) with these schemes were discussed in the text.

Good point, the authors do acknowledge that a more detailed discussion on the specific behavior of the simulations, such as the number of particles formed, could provide valuable insight. Svenhag et al. (2024) focuses on a deeper analysis of the CLUST-High and CLUST-Low differences, where we study the behavior of these two schemes in more detail.

–I liked Figure 3 in principle, but it could be tidied up a bit with larger labels, especially the colorbar. Also just reading the caption I was a bit confused why ADCHEM had no aerosols at Hyytiala but did have the data at Hyltemossa (maybe just add to the caption “, while for Hyltemossa the simulation was run from to ”).

We appreciate these beneficial suggestions; the authors have adjusted Fig. 4 to account for the ADCHEM with a box saying: “No Data”. Additionally, larger labels have been added to these figures showing weekly trends.

–Figure 4 also needs larger labels.

The Authors have adjusted Fig. 4 and made the labels larger.

–The sudden discontinuities in the aerosol size distribution around midday each day are attributed to the coupling of the chemical transport model to IFS. This is interesting. Because NPF is nonlinear, you would expect a systematic bias to result. Can this be studied further? How hard would it be to increase the frequency of the coupling?

We agree that this coupling introduces potential for systematic biases, particularly given the nonlinear nature of NPF processes. On one hand, It would indeed be interesting to conduct a further study focused on the impact of this coupling error on the simulation results, especially considering the variability in particle formation that could result from the temporal resolution of the coupling. However, we would like to note that future model versions of EC-Earth (currently under development) in OpenIFS48r1, the coupling between the chemistry and meteorology will occur within in every time step (30-60 min), which is expected to significantly reduce the error introduced by this temporal discrepancy.

–Line 315 and elsewhere: sentences could be improved, a number of typos to fix.

The authors have made changes to multiple sentences and sections, including Line 315.