Editor Revisions: Author Response to Peer Reviewer Report 1 from RC3

The authors would like to express their gratitude to the editor and reviewers for their thoughtful and constructive feedback throughout the review process. Their comments and suggestions have significantly contributed to improving the clarity and quality of this manuscript.

Reviewer comments are reproduced in **bold**, and our responses follow each comment in **plain text**. Text from the previous Author response is produced in ***red***. Line numbers refer to the revised manuscript unless otherwise stated.

- Start of Comments -

- The basic 10 subitems of comment 4

Comment: **4. Are the scientific methods and assumptions valid and clearly outlined?**
To some extent. Some parts of the methods section are not clear to me:

- line 112: what are ”specific dimensions”?

The authors addressed this comment in the last round of reviews (Author Response RC3), and adjustments were made accordingly. The response read as: The authors see that this was incorrectly described, where we were intending to refer to the size and log-normal structure of M7 here for the modal bins. This has been corrected and now reads on line 115: “However, this modal system in M7 limits the size distribution appearance to fall within specific log-normal modes at fixed sizes, …“

- line 127: what does ”explicitly modelled” mean?

The authors recognize the unclear language here of ”explicitly modelled” and have made changes to this description of the model process in the text. This line 115 now reads as:

“The KK function in M7 determines the particle formation rate using available gas phase ELVOCs and H2SO4 concentrations for estimated particle survival through condensational growth. Only after this growth to 5 nm are the aerosols introduced into the modal size distribution (Bergman et al., 2022).”

Previously this line read as:

“The KK function in M7 determines the particle formation rate using available gas phase ELVOCs and H2SO4 concentrations for estimated particle survival through condensational growth, and the resulting particles then enter the explicitly modelled size distribution in the nucleation mode (Bergman et al., 2022).”

- line 138: how are the five dimensions of the lookup table discretized?

The authors have addressed this further by adding the following description for the lookup table clarifying what was brought up in this comment. Following Line 139 now reads as:

"The lookup table contains nucleation rates for all combinations of the five variables that define the rate, and rates at specific conditions are determined by multivariate interpolation. This approach ensures accurate rates, avoiding the typical problem of nucleation rate parameterizations, namely that the rate may not be reproduced under all different conditions of "the parameter space"."

- line 139: how is the "cluster scavenging sink" computed?

The authors addressed this comment in the last round of reviews (Author Response RC3), and adjustments were made accordingly. The response read as: The authors have added this as a clarifying Line 143: "The (5) molecular cluster scavenging sink in CLUST is calculated from sulfuric acid condensation sink which is scaled for different cluster sizes (Yazgi and Olenius,2023b; Lehtinen et al., 2007). In the present EC-Earth3 setup, the input total condensation sink of sulfuric acid to CLUST is calculated from all 7 aerosol modes at every model time step."

- line 142: what are the dimensions of the IPR lookup table?

Authors have included a further description of the IPR table in the Methods section, on line 154:

"An IPR lookup table with global coverage of galactic cosmic rays and soil radon is used (Yu et al., 2019). It reads model pressure (203 altitude levels), magnetic latitude (91 bands), and the model grid land cover fraction (for ^{222}Rn) to generate the ion production rates, analogous to the implementation in Svenhag et al. (2024)."

- line 169: there seem to be different emissions for EC-Earth3 and ADCHEM. how does that influence the results?

The authors have included a further discussion addressing this point made by the reviewer. Adding onto the discussion between the models, line 343 now reads:

"There are also some differences in the emission inventories used by ADCHEM and EC-Earth3. However, these discrepancies are unlikely to be the primary source of emission-related differences observed at the two stations. A more significant factor is the spatial resolution at which each model reads the emissions. EC-Earth3 uses areal mean emissions from the CMIP6 inventory, interpolated over a coarse $2° \times 3°$ (longitude × latitude) grid. In contrast, ADCHEM utilizes the CAMS inventory at a much finer resolution of $0.1° \times 0.1°$ extracting emission values directly from individual grid cells."

- line 167: how is the trajectory discretized in time? how is the column discretized in space?

The authors recognized that clearer model description of ADCHEM was necessary here, and these following lines have been adjusted to clarify in the Method section (2.4):

Line 179: ‘‘Back-trajectories were simulated seven days backwards in time using the HYSPLIT default output interval of 1 hour for the Hyytiälä and Hyltemossa field stations. The trajectory coordinates were then linearly interpolated to a temporal resolution of 10 minutes when calculating the emissions of gases and particles from the urban areas, ocean, and forested regions surrounding the stations.’’

Line 181: ‘‘In this version of ADCHEM, the one-dimensional column model consists of 20 vertical layers spaced logarithmically, starting from the first layer that representing the lowest 10 meters up to the 20th layer which represents the atmospheric layer between 1900 m and 2100 m above ground level.’’

Line 186: ‘‘ADCHEM used a main model time step of 60 seconds when solving the atmospheric chemistry, aerosol dynamics and vertical mixing.’’

- line 181: ”Particles and gasses were mixed by use of the GDAS” how does that work in ADCHEM?

The authors have again included a further description of the ADCHEM model to answer the ‘‘**-line 183 & -line 181**:’’ questions:

Now, a longer paragraph on line 198 reads as:

‘‘For the gases, the model simulates the gas-phase and aqueous-phase chemistry of 5005 species via 13062 reactions. Strong inorganic acids (H2SO4, HNO3, HCl, HIO3), ammonia and organic oxidation products with a pure liquid saturation vapour pressure less than 10^-2 Pa at 293 K (in total 873 species) were treated as potentially condensable vapours and represents the particle size dependent condensation and evaporation dynamics. Other water-soluble gases such as SO2 and the DMS oxidation product MSIA are further oxidized in the aerosol and cloud droplet aqueous phase, forming lower volatility products like sulfate and MSA that likewise help to growth the particles.

The model solves the atmospheric diffusion equation in the vertical direction. The vertical diffusion coefficients (Kz) were calculated based on a slightly modified Grisogono scheme (Jericevic et al., 2010; Öström et al., 2017), where the Kz depends on the height above ground, the friction velocity and the height of the atmospheric boundary layer, which is sourced from the GDAS meteorology. “

And on line 205:

“A more detailed description of the model along with specific cases where the model has been used can be found in Roldin et al. (2019) and Wollesen de Jonge et al. (2024).”

These added lines also include two added references in the main text:

Öström, E., Putian, Z., Schurgers, G., Mishurov, M., Kivekäs, N., Lihavainen, H., Ehn, M., Rissanen, M. P., Kurtén, T., Boy, M., Swietlicki, E., and Roldin, P.: Modeling the role of highly oxidized multifunctional organic molecules for the growth of new particles over the boreal forest region, Atmos. Chem. Phys., 17, 8887–8901, https://doi.org/10.5194/acp-17-8887-2017, 2017.

Jericevic, A., Kraljevic, L., Grisogono, B., Fagerli, H., and Vecenaj, Ž.: Parameterization of vertical diffusion and the atmospheric boundary layer height determination in the EMEP model, Atmos. Chem. Phys., 10, 341–364, https://doi.org/10.5194/acp-10-341-2010, 2010.

- line 183: ”The ADCHEM model thereby attempts to reproduce the concentration of gasses and particles” how does that work in ADCHEM?

See text above and the provided additional references to previous ADCHEM publications.

- line 231: what grids are used in EC-Earth3?

Authors here repeat the previous comment from our Author Response to AC3, pointing to what is already described in the paper, in method section, line 101:

” The IFS model time step is 45 minutes and set to generate output every 3 hours on 100 a 0.7°spectral truncation grid. TM5 uses hourly time steps and is set to produce hourly model output with 2° × 3° (latitude× longitude) resolution. The vertical resolution in TM5 is represented by 34 hybrid sigma pressure levels, and IFS have the same hybrid pressure levels, but extrapolated to 91 layers.”

11. Is the language fluent and precise?
Mostly yes. Some sentences are rather long and complex and should be rephrased for a better readability.

Following both the initial and the current round of the review process, we examined the manuscript for instances of long or complex sentence structures highlighted by the reviewers. Where appropriate, we have rephrased and simplified these sentences to clarify and improve overall readability.

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?
Mostly yes. Mathematical symbols sometimes appear in text font instead of math font. For example, particle diameter d on lines 111 and 112 or nucleation rate J on line 124. The long terms on lines 176-178 should be simplified.

The authors have adjusted these text fonts on line 124, 111 and 112 to mathematical font accordingly.
The authors recognize these terms are long. However, the terms: “*DLPNO-CCSD(T)/aug-cc-pVTZ//ωB97X-D/6-31++G(d,p)*”, “*DLPNO-CCSD(T)//M06-2X*”, and “*RI-CC2//ωB97X-D*” are description terms decided by model development to signify specific versions and enable simpler reproducibility and consistency.

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?
Some figures should be revised. There are no subplot labels in figures 3, 5, A1, and A6. The font sizes in figures 3, 4, 5, 6, A4, A5, A6, and A7 are too small. The captions of some figures are not self-explanatory. In figures 3 and A6, it is unclear why the ADCHEM base case has a non-zero value of 10 cm-3. In figures 4, 6, and A7, its is unclear what ”modelled layer 1 and 2” or ”level 1” and ”level 2” mean.

Following both the initial round of review revisions, figures have been adjusted, combined and clarified. The following are the main changes for the figures referenced above:

- The non-zero value for ADCHEM in Figure 3 has been addressed more clearly both in the method section and in the figure caption. The caption for Figure 3 and 4 (combined) now reads as: “**Figure 3.** The top two figures show the surface aerosol number size distribution over the springtime for the 5 modelled EC-Earth3 cases, with the DMPS measured aerosols at Hyytiälä and Hyltemossa. ADCHEM simulations have no available hourly data outside the Summer months for Hyytiälä. The bottom section shows EC-Earth3 modelled layer 1 and layer 2 Hyytiälä and Hyltemossa springtime cases showing (a, b, g, h) daily maximum particle formation rate, (c, d, i, j) H2SO4, (e, k) ELVOC, (f, l) and NH3 gas concentration. The concentrations for ELVOC and NH3 level 2 are shown as dotted lines in e, f, k, l. The missing particle formation rates below 10-4 in panel a, b, g, and h are considered practically zero.”
- On line 272 the terms “model level 1” and “model level 2” are described.
- The font sizes in Figures 3, 4, 5, 6, A4, A5, A6, and A7 have been slightly adjusted to avoid obscuring any information. We remain fully open to making further refinements prior to final submission, if needed.

Editor Revisions: Author Response to Peer Reviewer Report 2 from RC1

The authors would like to express their gratitude to the editor and reviewers for their thoughtful and constructive feedback throughout the review process. Their comments and suggestions have significantly contributed to improving the clarity and quality of this 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 Comments -

Minor comments
I previously wrote: '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.'
I still think the abstract should be revised so that the sentence doesn't imply that the ADCHEM simulation provides insights into the whether or not the organic-H2SO4 nucleation parameterization can explain aerosol number concentrations (even though I accept that the observations could provide these insights).

The authors agree with the concern and have revised the sentence in the abstract to avoid implying that the ADCHEM simulation provides insights into the organic–H_2SO_4 nucleation parameterization. The updated wording now more accurately reflects that the insights are based on comparison with observations, as suggested. The referenced line 10 in the abstract now reads as:

"When comparing diurnal EC-Earth3 model results with in-situ observations at an hourly temporal resolution, we establish that using solely organic−H2SO4 nucleation parameterization will underestimate the aerosol number concentrations. The new added NH3−H2SO4 nucleation parameterization in this study improves the resulting aerosol number concentrations and reproduction of particle formation events with EC-Earth3. However, from March to October, the EC-Earth3 still underestimates particle formation and growth."

L255: "unaffected by altitude" – I think this would be clearer if it were qualified, for example "approximately unaffected by altitude due to compensating effects"

The authors agree and have altered this sentence accordingly, line 263 (previously L255) now reads as:
"In contrast, the CLUST nucleation rate remains approximately unaffected by altitude because of compensating effects, as its formation is governed by temperature, ionization, and cluster scavenging sinks."

L344: “Assuming the…” sentence can be improved with more scientific language

The authors regognized the unclear language here and have revised the sentence, Line 339 now reads as:
“The inclusion of some detailed chemical processes, as represented in the ADCHEM model, may yield results of sufficient relevance to climate impacts to justify the associated increase in computational expense when coupled with EC-Earth3.”

L372: sentence starting “It” can be improved – perhaps “It” -> “This artefact” and “aerosols…influence” or “aerosol..influences”

The authors have adjusted this sentence accordingly. Line now reads:
“This artifact may also introduce uncertainty regarding the influence of near-surface aerosols on cloud–aerosol interactions in EC-Earth3 within the 6-hour coupling window.”

L385: “suggests”->”suggest”
This word has been adjusted accordingly.

L387: “more NPF”- specify what "more" is relative to
The authors see this language error, and this word have been adjusted to “higher NPF rates” which should be the correct term here.

L390: “the inclusion”->”should be included”
This sentence was adjusted in the **L344** comment above.

L394 “potential false representation from” phrasing could be improved

This sentence has been adjusted to clarify the conclusion, line now reads:
“This study further underscores the potential for misrepresentation when relying solely on annual median values to evaluate the performance of EC-Earth3 (and other ESMs), due to pronounced seasonal variability.”

L397: would be good to add a reference to support the assertion that the climate forcing from aerosols has large seasonal variation
For this statement, the authors have now included the reference to support this, the two (also previously cited) sources to corroborate this claim on this line:
”Forster et al. (2021)”(CMIP6 report, Chapter 7) and ”Carslaw et al. (2013)”.