Authors' response to reviewers' comments for "Cloud droplet number enhancement from co-condensing NH_3, HNO_3, and organic vapours: sensitivity study" by Yu Wang, Beiping Luo, Judith Kleinheins, Gang I. Chen, Liine Heikkinen, and Claudia Marcolli

We thank the reviewer for the suggestions that have significantly improved our manuscript. Below, we provide our responses and summarise the changes that we have made with line numbers referring to the uploaded document with tracked changes. Other minor revisions were also made to improve the manuscript.

Summary

This paper uses a non-ideal cloud parcel model (with AIOMFAC/Pitzer activity treatment) to quantify CDNC changes from co-condensation of semi-volatile organics and inorganics for a Hyytiälä boreal case. The core findings are: (i) combined co-condensation raises CDNC by up to ~52% in the non-ideal case (vs 131% if ideal mixing is assumed), and (ii) the combined effect exceeds the sum of organics-only and inorganics-only contributions, with the largest boosts at intermediate updrafts. The setup uses a single observed size distribution and fixed ambient state (≈8 °C, 80% RH).

We thank the reviewer for their interest in our study.

Assessment: interesting, well-motivated case study; methods are appropriate; conclusions are supportable with some clarifications. I recommend accept after minor revision.

Strengths

- Clear demonstration that non-ideality matters and that assuming ideality overestimates the organic contribution to co-condensation and thus CDNC.
- Sensible separation of organic vs inorganic roles and a transparent VBS framing, including the importance of higher-volatility bins (log C* ≈ 4) near activation.
- Mechanistic analysis across updrafts showing a 21–52% CDNC increase for combined organics+inorganics and a non-linear dependence on w.

Essential clarifications

1. **Title scope**
   Current title reads more general than the experiments (single size distribution; fixed T).
   Action: tone down to "boreal case study" in title or abstract.
   Example: "Synergistic organic–inorganic co-condensation enhances CDNC in a boreal case study with non-ideal mixing."

   The title was also a concern of Reviewer 2. We therefore revise it to:
   "Cloud droplet number enhancement from co-condensing NH_3, HNO_3, and organic vapours: boreal case study"

2. **Parcel-model upper bound / entrainment context**
   Field CDNC is often lower than parcel-model CDNC because entrainment, w-variability,

and turbulent quenching reduce Smax and can deactivate marginal droplets; semi-volatiles taken up near activation can re-evaporate upon mixing.
Action: add 2–3 sentences in Discussion stating that reported CDNC enhancements are an upper bound for in-cloud conditions and that entrainment could buffer these effects.

We thank the reviewer for pointing out this aspect. We add the following sentence to the conclusions on lines 351–353:
"Note that our simulations describe cloud droplet activation at cloud base and do not include turbulence and entrainment-mixing, which can also lead to deactivation of cloud droplets thus affecting CDNC and droplet size distributions (Morales et al., 2011; Yang et al., 2018; Oh et al., 2023)."

Initialisation & computational shortcut
The manuscript uses 1.2 m s^{-1} below 98% RH then switches to the target w, claiming negligible impact because most uptake occurs above 98% RH. This is fine, but please make this explicit in explaining the control case too (if that's what was done)
Action: state clearly that control (no co-condensation) runs use the same 80%→equilibration step (I know it is simpler to equilibrate the aerosol when there are no other co-condensing vapours) and the same pre-98% RH shortcut, and add one sentence reporting that a no-shortcut check (e.g. for at least one of the co-condensing cases) produced CDNC within X%.

We improved the description of the initialisation procedure by revising starting from line 245:

"The concentrations of inorganic salts and gaseous ammonia and nitric acid are taken from Table 1. To match the total mass concentration according to the size distribution, the aerosol composition was complemented with Na+, mineral dust and black carbon as described in Sect. 2.4.2. This composition initialisation was applied to all simulations, including the control runs."

Non-ideality framing
You nicely document that ideality inflates CDNC changes (131% vs 52%). Consider reporting the range too, rather than just the maximum for ideal vs non-ideal CDNC (and Smax) with error bars for the range (e.g. the data in figure 9 suggests 25 to 131% (ideal) vs 25 to 52% (non-ideal)) for quick reader digestion.

Thanks for this suggestion. We now refer to the range by revising (line 451):

"...(CDNC changes are 14–53% and 20–44% for ideal and non-ideal cases, respectively)..."

Note that the revised numbers are considerably smaller than the ones reported in the manuscript. This is because we realized an error in our calculation for the ideal case during the revisions. Namely, we had used a too short equilibration time (only 2 min instead of 30 min). Because of this, there was too little partitioning to the condensed phase at 80% RH.

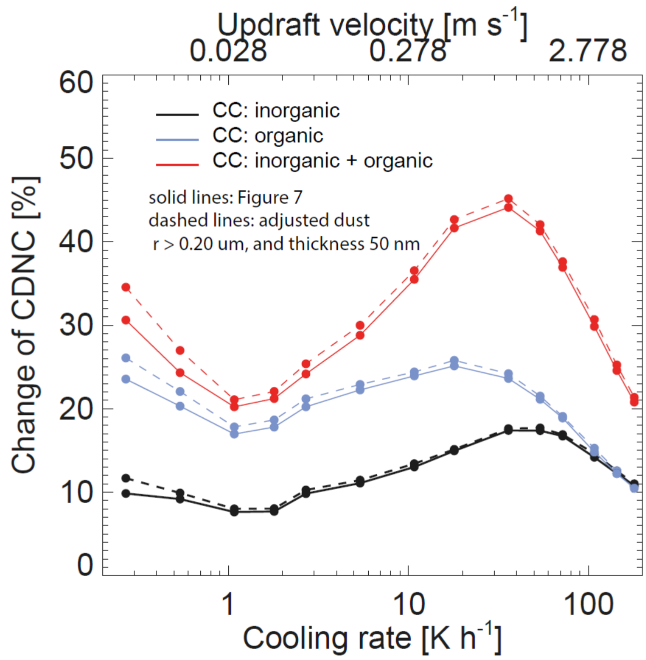
Due to the high gas-phase concentrations of organics still available for condensation, the CDNC increase resulting from co-condensation was overpredicted in Fig. 9. Correcting this, the difference between ideal and non-ideal simulations became significantly smaller.

Large-particle/composition completion

You add mineral dust (10% v/v; r > 250 nm with a 10 nm coating) and BC (3.6% v/v) to close mass/volume. Briefly say whether toggling this coarse tail alters partitioning/CDNC (expected: small).

Action: add a clause like "Removing/halving the coarse tail changed CDNC by <X%, indicating little influence on the results."

We introduced the mineral dust to reach consistency between the aerosol mass measured by the Q-ACSM and the aerosol volume derived from the size distribution measured by the DMPS. As mineral dust typically belongs to the coarse aerosol fraction, the assumption that it forms the tail of the particle size distribution is justified. We assumed an organic coating on the mineral dust to allow absorptive partitioning. Following reviewer 1, we performed an additional run where we distributed the mineral dust more evenly among the particles. The figure below compares the change of CDNC as a function of cooling rate for the assumption that all particles with radius r > 0.2 µm possess a coating of 50 nm thickness with the assumption in the paper, namely that particles with r > 0.25 µm possess a coating of 10 nm thickness. It can be seen that the difference in the mineral dust distribution hardly affects the CDNC.



Detectability statement

The abstract suggests the magnitude should be detectable in closure studies. Please outline a practical closure strategy for observations.

We add the following statement to the conclusion.

"Note that our simulations describe cloud droplet activation at cloud base and do not include turbulence and entrainment-mixing, which can lead to deactivation of cloud droplets thus affecting CDNC and droplet size distributions (Morales et al., 2011; Yang et al., 2018; Oh et al., 2023). This will also render the observation of this effect in field measurements more difficult. To detect it in closure studies, the measurements should be performed close to cloud base, while cloud edges where entrainment is most likely to affect CDNC (Freud et al., 2011) should be avoided."

Specific:

- Line 35: avoid citing Köhler (1936) twice in the same sentence. We removed one reference.
- Line 136: fix "Hyyitälä" → Hyytiälä. fixed
- Line 137: extrapolation. What justification is there for this extrapolation? And how was the extrapolation done? What method? straight line? Last two bins, or some kind of mass closure? I know you say bins 0-3 but more detail needed.

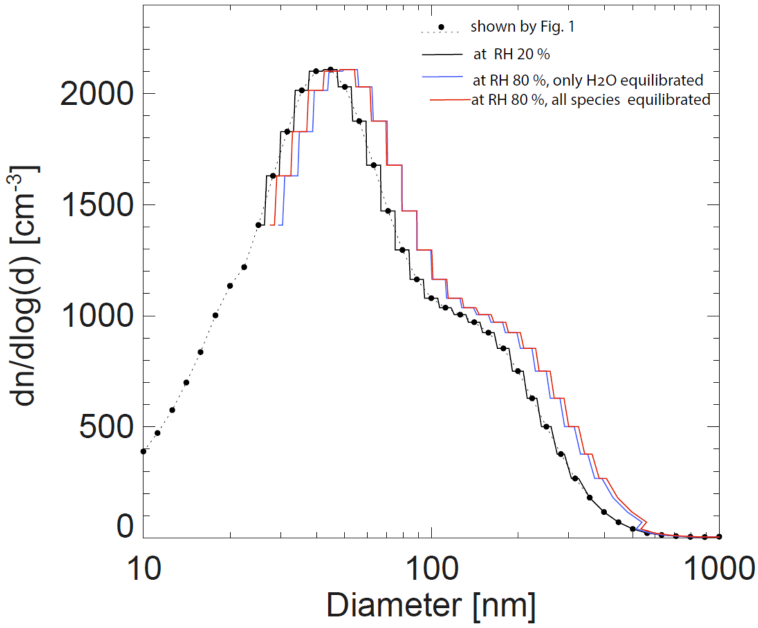
  We agree with the reviewer that this assumption has a notable impact. We think that a high mass in volatility bin as a result of secondary organic aerosol formation is justified as this process starts from volatile products, which are oxidized stepwise in a cascade of reactions to first intermediate, then semi-volatile to finally low volatility compounds. A suitable reference for this notion is Stolzenburg et al. (2022), which we cite now in the revised manuscript. We add after "VBS that we derived":
  "Note that this extrapolation leads to the highest mass fraction in the volatility bin with log(C*) = 4. This high mass fraction is justified considering the cascade process of secondary organic aerosol formation starting from volatile compounds (Stolzenburg et al., 2022)."

- Line 272: explicitly state whether assumptions about large particles/coatings influence partitioning and CDNC

  See the response to comment "Large-particle/composition completion".

- Lines 275–280: show (or state) that the post-equilibration size distribution still matches DMPS within uncertainty; justify the high-w shortcut with the quick check for one or two cases.
- We did this simulation and show it in the figure below. As the aerosol is dried before the measurement of the size distribution, we tailored our initialisation to match the measurements at RH = 20% (black dashed line for measurements and solid lines for simulation). The size distribution at 80% was obtained by first equilibrating the aerosol with water vapor so that the vapor pressure including Kelvin effect above the

particles corresponds to 80% RH (blue line). In a second step, both, co-condensation of water vapor and semi-volatile species was performed again at 80% (red line). As expected, the size distribution shifts to larger diameters mainly due to the water uptake.



- Line 428 (“Key factors”): list exactly what you varied (VBS, w, non-ideality) and note temperature/size-distribution variability as likely important but not explored here.

  We did not come to this summary of key factors through direct variation of parameters, but through the analysis of Figs. 6-8. We make this clear by revising the sentence to (lines 444–446):

  “Overall, the analysis of Figs. 6–8 has shown that updraft velocity, condensed and co-condensable mass, and aerosol size distribution are key factors controlling the CDNC enhancement due to co-condensation.”
  Moreover, we extend the conclusion starting from line ...:
  “Overall, systematic variation of updraft velocity during cloud droplet activation allowed elucidating how the aerosol size distribution influences the enhancement of CDNC due to co-condensation. Simulations with two different VBS showed that the mass in the bin with log(C*) = 4 has a large influence on the magnitude of the co-condensation effect. Inclusion of non-ideality proved to be relevant for a realistic estimate of the co-condensation effect especially for the less oxidized organics. Studies in other regions with different aerosol size distribution and composition are required to establish more comprehensively the role co-condensation plays in cloud droplet activation.
  Note that our simulations describe cloud droplet activation at cloud base and do not include turbulence and entrainment-mixing, which can lead to deactivation of cloud droplets thus affecting CDNC and droplet size distributions (Morales et al., 2011;

Yang et al., 2018; Oh et al., 2023). This will also render the observation of the co-condensation effect in field measurements more difficult. To detect it in closure studies, measurements should be performed close to cloud base, while cloud edges where entrainment is most likely to affect CDNC (Freud et al., 2011) should be avoided."

References

Freud, E., Rosenfeld, D., and Kulkarni, J. R.: Resolving both entrainment-mixing and number of activated CCN in deep convective clouds, Atmos. Chem. Phys., 11, 12887–12900, https://doi.org/10.5194/acp-11-12887-2011, 2011.

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