Dear Prof. Petters,

Please find attached our revised manuscript "Optimizing CCN predictions through inferred modal aerosol composition – a boreal forest case study" by Ranjan et al.

The work has substantially improved as a result of the thoughtful feedback from the reviewers. Motivated by this feedback, we have applied another inverse closure method (DREAM-MCMC) to ensure the robustness of the earlier conclusions and demonstrate the sensitivity of the results to uncertainties in the input parameters. The addition of this new method has resulted in substantial revisions in the Abstract, Methods and Results sections in the revised manuscript. Furthermore, besides improving the manuscript based on the specific suggestions by the reviewers, we made one more round of edits for clarity and readability. All changes as compared with the original submission are marked in the tracked manuscript file.

The point-by-point responses (in blue) to all the reviewer comments (in *black*), and the associated changes in the manuscript (in red) are given below.

**Reviewer #1:**

***Review on the Manuscript entitled: "Optimizing CCN predictions through inferred modal aerosol composition – a boreal forest case study"***

*Aerosol hygroscopicity and CCN activity, both depending on particle size and chemical composition, play a key role in the aerosol indirect climate effects. Aerosol hygroscopicity and CCN activity can be probed by specialized instrumentation, which can also offer size resolved measurements, like for instance the Hygroscopic Tandem Differential Mobility Analyzer (HTDMA) or the Differential Mobility CCN counter (DMA-CCNc). However, such instrumentation is not widely used due to various issues (e.g., bulkiness, purchasing and operating costs). By exploiting the dependence of aerosol hygroscopicity/CCN activity on particle size and chemical composition (both measured at higher spatial resolution), one can in principle overcome this limitation. Aerosol chemical composition and size distribution are also used in atmospheric/climate models for estimating aerosol hygroscopicity/CCN activity and for deriving potential cloud droplet number concentration and cloud dynamics using different parameterization schemes. While particle size distributions are measured and/or modelled nowadays accurately and with adequate resolution, aerosol chemical composition is most commonly measured and/or modelled for the bulk submicron aerosol population. This can reduce the accuracy of the estimated, based on the bulk chemical composition, aerosols hygroscopicity/CCN activity, especially in complex environments where the aerosols exhibit different compositions at different sizes and/or are externally mixed. The latter refers to particles of the same size that exhibit different chemical composition. The identified by many studies discrepancies between the measured hygroscopicity/CCN activity and that estimated based on the aerosol bulk chemical composition was the main motivation of the authors of this manuscript. In more detail, the authors exploit long-term observations of submicron particles size distributions, bulk chemical compositions and CCN activity conducted at the boreal forest site of SMEAR II (Hyytiälä, Finland) for their study. They investigate the discrepancies between the measured aerosols CCN activity and that estimated from measured particle size distributions and the bulk chemical composition derived aerosol hygroscopicity, expressed by the aerosols hygroscopic parameter κ. In addition, they study the discrepancies between the measured aerosol CCN activity and that estimated by the measured particle size distributions but assuming a time-constant aerosol hygroscopicity, expressed as a constant hygroscopic parameter κ of 0.18. Furthermore, they suggest a method for improving the estimated CCN activity by assigning different chemical compositions (and hygroscopic κ parameters) at different size ranges (i.e., modes). In order to achieve this, they made some assumptions/simplifications, like treating the whole aerosol population as internally mixed (i.e., particles of the same size, share the same chemical composition), assigning similar hygroscopicities to inorganic species and assume that Black Carbon (BC) concentration fraction is the same at all particle sizes.*

***General comments***

*While size-resolved aerosol hygroscopicity/CCN activity can be probed with adequate instrumentation (HTDMA, DMA-CCNc, Scanning Mobility CCN Analysis; i.e., CCNc coupled to an SMPS; Moore, Nenes and Medina, 2010), this manuscript presents the very important aspect of suggesting a method for deriving modal chemical composition from (bulk) CCN and ACSM measurements. For this reason, I suggest its publication in Atmospheric Chemistry and Physics, after a minor revision.*

**Response:** We thank the reviewer for this positive assessment.

*In more detail, by using adequate instrumentation, like for instance one CCNc downstream a DMA, one can measure the CCN activity spectrum for monodisperse particles residing in Aitken and accumulation modes. Two monodisperse sizes and 5-7 super-saturations would perhaps be adequate for performing these observations. This would result in a more accurate estimation of the aerosol hygroscopic parameter κ at these two modes (i.e., Aitken and accumulation). Adding a neutralizer and a DMA in front of an existing CCNc does not require a major effort and/or cost. In addition, the time resolution of such measurements will be still adequate for studying aerosol CCN activity/hygroscopicity at rural sites and comparable to the one used in this study. However, the authors present a method that associates the modal hygroscopic parameter κ to the modal chemical composition, using bulk chemical composition measurements (i.e., ACSM); something innovative according to my best knowledge. This aspect of their work significantly increases the importance of this manuscript.*

**Response:** We thank the reviewer for their kind attention to the innovative nature of this work. We also agree regarding the suggestions for potential improvements for direct sampling of the size-dependent CCN activity. Our approach – using the inverse closure methods – simply presents a relatively cost-efficient alternative to this, and is of course also applicable on data sets where the measurements of size-dependent hygroscopicity are not available. In the revised manuscript we have further improved the inverse closure methodology by introducing an approach based on the DREAM-MCMC algorithm, which allows also accounting for the variability of the particle number size distribution during the 2-h long CCN measurement cycle. The results from this more sophisticated approach improve the closure further and corroborate the conclusion on size-dependent hygroscopicity as a key explaining factor for obtaining successful closure. In this approach, we not only optimize the modal chemical composition but also account for the variability of the size-distribution lognormal parameters during the 2-hour CCN cycle.

**Changes to the revised manuscript:** We have added the following statement to the Conclusion (L844-847 of the revised manuscript): "If modal or size-resolved κ (in addition to just having bulk chemical composition) were available, our approach could be extended to derive more detailed size-dependent chemical composition–for example, size-dependent organic hygroscopicity–while also helping to constrain κ values by identifying those that best reproduce observed CCN concentrations." For the description of the added MCMC results, see the Abstract, Methods and Results sections of the revised manuscript.

*A) I suggest that the authors emphasize more on this aspect of their work (i.e., deriving the modal chemical composition from CCN activation spectra).*

**Response:** We agree that estimation of modal aerosol chemical composition from CCN spectra is the most important part of the manuscript. As discussed above, we feel that the addition of another inverse closure method (which allows for optimization with respect to more variables) has really strengthened our conclusions on this.

**Changes to the revised manuscript:** To emphasize these points even more, we have rewritten the Abstract, added text to the Introduction (see L66-72, L159-169 and L177-179 in the revised manuscript), added further descriptions to Methods (particularly on the MCMC approach, but also including a work flow chart of the inverse methods, see Fig. 3 in the revised manuscript and below), Results and Conclusions.

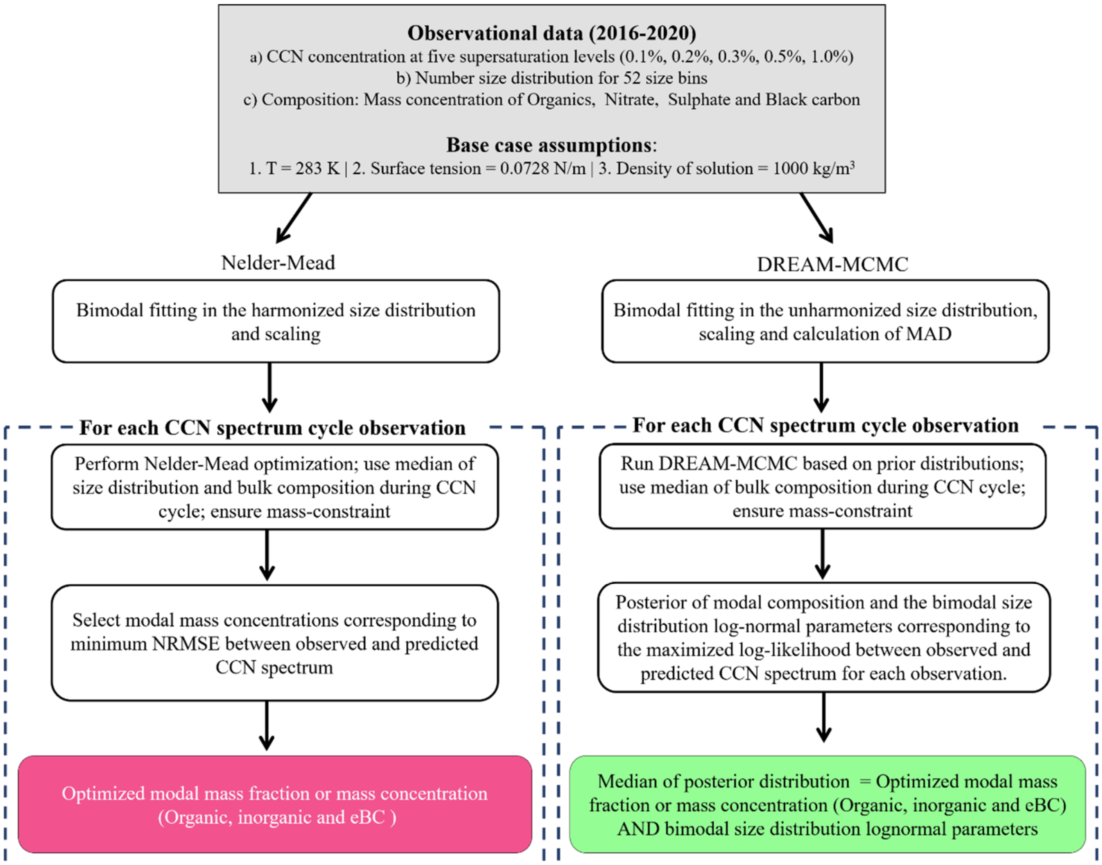


**Figure R1.** Workflow of the two inverse closure methods: the Nelder–Mead algorithm (left) and the DREAM-MCMC (right) approach. Bimodal fitting - representation of the aerosol size distribution as two lognormal modes. Harmonized size distribution - size distribution data harmonized to CCN data; data thus obtained has 2-hour resolution. Unharmonized size distribution - raw size distribution data with 10 min resolution. Scaling - adjustment of number concentrations of reconstructed lognormal size distribution from bimodal parameters to match observations. Mass-constraint - conservation of total aerosol mass (sum of mass in two modes) of each species during optimization. NRMSE - normalized root mean square error, a metric of model–observation agreement. MAD - median absolute deviation, used to quantify variability in size distributions during CCN spectrum cycle period. Prior distribution - initial parameter ranges provided to the MCMC sampler. Log-likelihood - statistical measure of consistency between observed and modeled CCN spectra.

*B) The authors should comment (and perhaps describe/mention in the discussion/conclusion sections) if their method for deriving the modal aerosol chemical composition can be used in the case(s) where modal (or even size resolved) hygroscopic parameters κ are available.*

**Response:** Thank you for this comment – which is naturally also linked to the way that the sampling is done as mentioned above. If size-dependent hygroscopicity values would indeed be available, they could be used together with similar composition data as here to infer even more detailed insights on the size-dependent chemical composition – perhaps through a similar optimization procedure as here, but perhaps allowing for e.g. variability in the properties of the organic mixture (which were assumed to be constant here) or internal vs. external variability (the former assumed to be the case here throughout the data set). Even if modal (or size-resolved) hygroscopicity parameters ($\kappa$) are available, there's of course always the possibility that these values carry uncertainties or do not fully represent the actual hygroscopic behavior of the aerosols in each mode. Our method can help constrain these $\kappa$ values by identifying the set that leads to the best agreement between predicted and observed CCN concentrations. In other words, such measurements would allow for a more detailed studies on the topic.

**Changes to the revised manuscript:** We have added the following statement to the Conclusion (L844-847 of the revised manuscript): "If modal or size-resolved κ (in addition to just having bulk chemical composition) were available, our approach could be extended to derive more detailed size-dependent chemical composition–for example, size-dependent organic hygroscopicity–while also helping to constrain κ values by identifying those that best reproduce observed CCN concentrations."

*C) Their methodology, assumptions/simplifications/limitations should be more clearly described in order to be more understandable by other aerosol scientists and to be easier to replicate in other sites/studies.*

**Response:** We agree that the methodology, assumptions, simplifications, and limitations should be described more clearly to enhance transparency and reproducibility.

**Changes to the revised manuscript:** In the revised manuscript, we have made a concerted effort to improve the clarity of the information flow and to explain the steps of the method in more detail (see the Methods section of the revised manuscript).

***Specific comments:***
*1) Abstract (lines 33-35): — Our study demonstrates the potential for utilizing CCN measurements for inferring information on the parts of the aerosol size distribution that are beyond the reach of traditional online composition measurements.*

*This sentence needs to be better written in a way to more clearly convey the important message that bulk CCN and (perhaps; see my comment #22) size resolved hygroscopicity/CCN activity together with bulk chemical composition measurements can be used for estimating the modal chemical composition. In addition, the term — traditional online composition measurements can be replaced by the more accurate — online bulk chemical composition measurements.*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have rewritten the Abstract.

*2) Introduction (line 57): — $N_{CCN}$ and CDNC are primarily determined by aerosol properties and the drivers of $SS_{max}$ fluctuations…*

*Please define the abbreviation $SS_{max}$ prior of its first use in the manuscript. While this abbreviation is well known to aerosol scientists studying aerosol – cloud interactions, the authors should not assume that other aerosol scientists are familiar with this abbreviation.*

**Response:** Thank you for pointing it out.

**Changes to the revised manuscript:** $SS_{max}$ is now defined appropriately when mentioned for the first time (see L42) in the revised manuscript.

*3) Introduction (lines 96-97): — Importantly, some organic aerosol properties beyond hygroscopicity may enhance the likelihood of an Aitken mode aerosol particle to serve as CCN (Lowe et al., 2019).*

*The authors could elaborate a bit more on which properties of Aitken-mode organic aerosols, besides their hygroscopicity, can enhance their CCN activation.*

**Response:** Thanks for this suggestion. We agree.

**Changes to the revised manuscript:** We have revised the sentence to read (L97-98 of the revised manuscript): "Importantly, some organic aerosol properties beyond hygroscopicity such as solubility or surface activity, may enhance the likelihood of an Aitken mode aerosol particle to serve as CCN (Lowe et al., 2019)."

*4) Introduction (lines 101-103): ― Studies incorporating organic aerosol effects demonstrated significant improvements in closure as compared with attempts considering inorganics alone (e.g., Broekhuizen et al., 2006; Rose et al., 2008; Ervens et al., 2009; Guenther et al., 2009; Bougiatioti et al., 2009; Jurányi et al., 2010).*

*To which ― organic aerosol effects are the authors pointing at? Surface tension changes to organic compounds, solubility effects or just to the fact that by omitting the organic component particle hygroscopicity and CCN activity are overestimated? Please be more specific here.*

**Response:** In this context, we refer specifically to the inclusion of organic compounds in the chemical composition when predicting CCN. The cited studies demonstrated improved closure primarily by accounting for organics — rather than omitting them or assuming them to be insoluble — which led to more accurate representations of particle CCN activity. In previous studies the way that organics have influenced the results and / or improved the closure varies, but we feel elaborating too much on these reasons is beyond the scope of the present work – as the purpose of this part of the Introduction was simply to highlight the important role that organics play in determining the CCN properties of an aerosol population.

**Changes to the revised manuscript:** We hope the response clarifies what was meant. No changes were made to the revised manuscript.

*5) Introduction (line 161): ―...using a constant hygroscopicity value of 0.18 throughout the study period, as recommended by Sihto et al. (2011).*

*Please use the more appropriate term ― hygroscopic parameter κ of 0.18.*

**Response:** Thank you for pointing this out.

**Changes to the revised manuscript:** Modified according to the reviewer suggestion (L168 of the revised manuscript).

*6) Section 2.1.1 (lines 213 – 214): ―However during the winter time more black carbon is also observed (Luoma et al., 2019), which tends to decrease the overall hygroscopicity.*

*While black carbon it's a known hydrophobic species it would be better to explicitly mention it in the sentence. For example: However, during the winter time the increased contribution of black carbon, which is hydrophobic, in the particles decreases their overall hygroscopicity, or something along these lines.*

**Response:** Thank you for this helpful suggestion. We agree that explicitly mentioning the hydrophobic nature of black carbon adds clarity.

**Changes to the revised manuscript:** Modified according to the reviewer suggestion (L226 of the revised manuscript).

*7) Section 2.1.2 (line 238-239): ―For the inverse closure, we used a Python version (Khadir, 2023) of the algorithm by Hussein et al. (2005) to fit two modes into the measured aerosol size distributions.*

*The way that this sentence reads seems quite misleading. The algorithm suggested and described in Hussein et al. (2005) is aimed at performing modal analysis on the particle size distributions measured with scanning mobility particle sizers (SMPSs) and can be applied on other instruments that probe particle size distributions at equivalent size ranges and with adequate resolution. This algorithm is not* *related to any closure studies between aerosol chemical composition and CCN activity. I understand that the authors used a similar (or perhaps the same) algorithm for performing the modal analysis, which however is only the first step for performing the inverse closure. This sentence needs to be written in a clearer way.*

**Response:** Thanks for mentioning a lack of clarity here.

**Changes to the revised manuscript:** We have now modified beginning of the appropriate paragraph (L251-259 in the revised manuscript) in Sect. 2.1.2 to read: "As a first step toward the inverse closure (see also Sect. 2.2.3), we applied a Python implementation (Khadir, 2023) of the modal-fitting algorithm described by Hussein et al. (2005) to decompose the measured aerosol size distributions into two modes." Please see also response to comment #8 below.

*8) Section 2.1.2 (lines 239 – 242): — The algorithm takes size distribution as input and returns the lognormal parameters (number concentration, geometric standard deviation, geometric mean diameter) of different modes as output. While the algorithm would allow fitting up to four modes, bimodal fits (Aitken and accumulation mode, respectively; Fig. S1a) were selected to avoid overfitting.*

*According to my opinion, this part of the procedure should be described in more detail (perhaps in the supplement, before figure S1). When reading it, some questions arise. For example, is the number of fitted modes (e.g., unimodal, bimodal, trimodal) decided by the user (as an input parameter) in the algorithm employed by the authors or is it an automated process? In Hussein et al. (2005) a number of criteria for reducing the number of fitted modes (e.g., from a trimodal to a bimodal fitting) are described. Did the authors use those criteria or they choose the bimodal fittings due to improved Pearson's r correlation in respect to a unimodal fitting? Was the bimodal fitting optimum for all the measured size distributions or there were cases when a unimodal or even a trimodal fitting would be preferable? For instance, during a new particle formation (NPF) event, particles residing in the size range <25 nm would exhibit increased number concentrations, thus making necessary a trimodal fitting (i.e., nucleation, Aitken and accumulation modes) to better describe the measured particle size distribution.*

**Response:** This is a great point. In fact, some of the co-authors have supervised a BSc thesis (Liwendahl, 2023) that focused on the performance of the Hussein et al. (2005) algorithm and the optimal number of fitted modes for the particle number size distributions measured in 2012-2017. The study found (expectedly) four fitted modes to represent the measured size distribution about 30% better than just two modes. There was, however, no clear seasonality or annual trends in this which would indicate that our choice of fitting two modes (which simplifies the inverse closure procedure considerably) would significantly distort the trend analysis or even the closure itself. This is also because we scale the fitted size distributions to match the observed values, which we explain in detail in Supplementary note 2 of the revised manuscript (see also Fig. 3 and L448-454 in the revised manuscript). In simple words, in scaling we equal the number concentration of particles in each bin in the fitted size distribution to the observed size distribution.

**Changes to the revised manuscript:** We have added a reference to the BSc thesis mentioned above (Liwendahl, 2021), and added edits to the paragraph (L447-459) where the scaling to measured concentrations is explained.

*9) Section 2.1.3 (lines 253 – 256): —The CCNc consists of a saturator unit and an Optical Particle Counter (OPC). The saturator includes a vertical flow tube where aerosol samples are introduced alongside filtered sheath air under laminar flow conditions, creating a central flow path. The tube's inner surface is kept moist to generate a supersaturation gradient.*

*The sentences describing the operating principles of the CCNc can be better and more clearly written. For instance, the sheath air flow is saturated at the inlet temperature. A positive temperature gradient* *is maintained at the saturator column, inducing a quasi-constant supersaturation profile for a specific temperature difference.*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have rewritten the description of the CCNc in the revised manuscript (L274-280). We have also included a description of the measurement cycle to obtain the complete CCN spectrum (L286-289).

*10) Section 2.1.4 (lines 278 -280): —An Aerosol Chemical Speciation Monitor (ACSM; Ng et al., 2011) was used to retrieve long-term observations of the non-refractory sub-micron particulate matter (NR-PM1; i.e., organics, sulfate, nitrate, ammonium and chloride) at SMEAR II. —*

*This sentence can be written in a clearer way that better describes what the ACSM is measuring. For example, the ACSM measures the mass concentrations of ions originating from non-refractory organic and inorganic atmospheric species. The results are provided as mass concentrations of ammonium, sulfate, nitrate and chloride ions, as well as a total organic mass.*

**Response:** Thank you for this suggestion.

**Changes to the revised manuscript:** We have made suggested change in the revised manuscript (L305-307).

*11) Section 2.1.4 (Data Coverage and seasonal classification): This should be section 2.1.5.*

**Response:** Thank you for pointing this out.

**Changes to the revised manuscript:** We have corrected the section numbering in the revised manuscript.

*12) In the same section (lines 321 – 322): —As mentioned earlier, SOA formation and NPF events lead to higher particle number concentrations during spring and summer.*

*During these observed NPF events did the authors still use a bimodal fitting? Would a trimodal fitting (i.e., including nucleation, Aitken and accumulation modes) be more appropriate during the cases that NPF events were observed (see also my comment #8)? Would a trimodal fitting during NPF events affect the inverted closure (CCN-ACSM) procedure described in the manuscript? The authors should clarify these aspects. In addition, in the case that they have used bimodal fittings for all the measured particle size distributions they should justify that by omitting the nucleation mode during NPF events the inverted closure procedure is not significantly affected. They can add briefly this justification to the manuscript.*

**Response:** Thank you for the comment – please see our response to comment #8 and the associated changes to the revised manuscript.

*13) Section 2.2.2 (lines 390 – 392): — We acknowledge that the assumption that sulfate is present solely as AS can cause underestimations of aerosol hygroscopicity at SMEAR II, because aerosols can be more acidic at the site (e.g., Riva et al., 2019).*

*What do the authors mean by more acidic aerosols? Do they mean that perhaps there are cases that particles may contain ammonium bisulfate or sulfuric acid as well? Please be more specific here. In addition, why did the authors not employed a simplified ion-pairing algorithm, similar to the one described in Gysel et al. (2007)? They could employ this simplified ion-pairing scheme, after calculating the organic nitrate content (as they have already done).*

**Response:** We thank the reviewer for this valuable comment. By "more acidic aerosols," we refer to the presence of species such as un-neutralized ammonium bisulfate ($NH_4HSO_4$) and sulfuric acid ($H_2SO_4$), which are more likely under ammonium-limited conditions. To evaluate the impact of this assumption, we additionally calculated $\kappa$ using the simplified ion-pairing approach described by Gysel et al. (2007). The comparison of $\kappa$ values obtained using both methods — (i) assuming full neutralization to ammonium sulphate and nitrate, and (ii) using the Gysel et al. (2007) ion-pairing scheme — is shown in Figure R2 and will be added to the supplement to the revised manuscript. The resulting $\kappa$ values were very similar, with median values of 0.21 and 0.23, respectively. This difference is insignificant for our inverse-closure output.

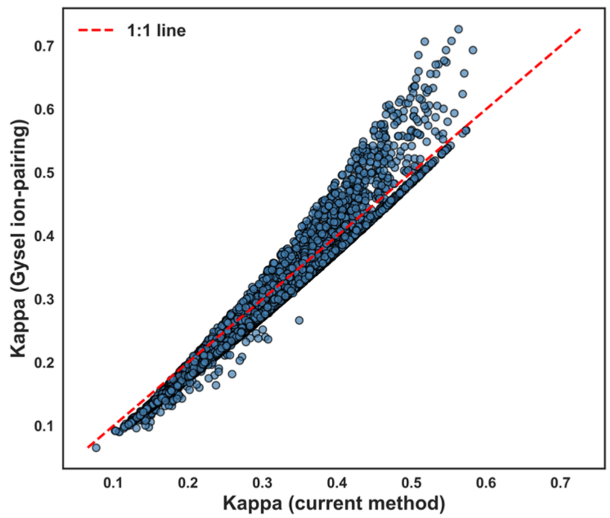


**Figure R2.** Comparison of $\kappa$ values calculated using two different methods: (i) a simplified full neutralization approach assuming complete conversion of $SO_4^{2-}$ and $NO_3^-$ to ammonium sulfate and ammonium nitrate, and (ii) a more chemically detailed ion-pairing scheme based on Gysel et al. (2007), which allows for the formation of acidic species such as ammonium bisulfate ($NH_4HSO_4$) and sulfuric acid ($H_2SO_4$). The red dashed line denotes the 1:1 reference.

*14) Section 2.2.3 (Inverse closure): This section can be complemented with additional information (and perhaps equations) in order for the inverse closure procedure to be clearer and easier to reproduce or even being improved. The authors may use the supplement for including the additional information (and perhaps explanatory figures) for this scope, if they want to avoid —overloading the manuscript.*

**Response:** Thank you for this valuable suggestion. We hope the approaches are clearer in the revised manuscript.

**Changes to the revised manuscript:** As discussed above, we have added a great deal of more details on the inverse modelling approach to both, the main manuscript and the supplement. In particular, Sect. 2.2.3 has been rewritten to describe the two inverse modelling approaches in detail, new Fig. 3 has been added, along with new Supplementary notes 4-6.

*iii) In section 2.2.1, equation 3, the authors (correctly) provide the equation for the volume fraction of each species, accounting for the mass and the density of each species. However, in section 2.2.3, they use the bulk density (derived by the bulk chemical composition measurements). While this is perhaps a necessary simplification, I wonder if they could further optimize this aspect. If the system of equations solved for the inverted closure procedure was provided, it would be clearer if this assumption (and potential limitation) is indeed necessary or if two different apparent densities (i.e., one for the particles residing in the Aitken mode and one for those residing in the accumulation mode) could be estimated, further improving the results.*

**Response:** We thank the reviewer for this comment. Indeed, for the inverse closure procedure this assumption is key for making conversions between the dry diameters and compositions. It would indeed be possible to the role of the density assumption for improving the inverse closure. However, with the DREAM-MCMC approach, we were able to mitigate the overprediction in CCN concentrations by accounting for size-dependent chemical composition and incorporating variability in the size distribution's lognormal parameters within each CCN spectrum cycle. Introducing further optimization of mode-specific densities could add unnecessary complexity and increase the risk of overfitting, particularly given the limited observational constraints. Nonetheless, we recognize this as a valuable point for future study – for instance in cases where size-dependent hygroscopicity values would provide further constraints for the inverse closure.

*iv) It would be easier for the reader to deeply understand the inverse closure procedure if some explanatory images were added in the supplement. For instance, figure S6 helps a lot in understanding the scaling process of the fitted lognormal distributions. Similar figures could be added below figure S6, showcasing the process step by step (e.g., converting the scaled fitted size distributions to mass distributions and then to fractional volume distributions, which in turn will be used for estimating modal hygroscopic parameters, etc).*

**Response:** Thank you for this valuable comment.

**Changes to the revised manuscript:** We hope that the combination of new Fig. 3, the reworked versions of Sect. 2.2.3 and L251-259 help address this comment.

*15) Section 3.1 (lines 467 – 469): —The activation diameters decrease with increasing supersaturation and when all seasons are taken into account median $D_{act}$ (see Table S1) being generally higher than reported in earlier studies using similar methodology (e.g., Sihto et al., 2011; Paramonov et al., 2015).*

*It is not very clear what the authors refer to as the median $D_{act}$ when all seasons are taken into account. Do they mean the yearly median $D_{act}$, which is not depicted in Table S1 or that $D_{act}$ for every season is generally higher than that reported in earlier studies? In addition, it would be better to report the median $D_{act}$ from those earlier studies for comparison reasons.*

**Response:** Here we meant to say that in every season, the $D_{act}$ is greater than previously reported.

**Changes to the revised manuscript:** We have modified the revised manuscript to clarify this issue and added the quantitative comparison requested (L575-578 of the revised manuscript, and Table S1).

*16) Section 3.1 (lines 475 – 476): — While the median activation diameters show almost no seasonality, looking in more detail (see Fig. S4), an increase in the $D_{act}$ is observed during the transition from winter to spring.*

*Figure S4 does not depict $D_{act}$ values. Please correct accordingly (figure S3 seems to be the correct one). In addition, the increase in $D_{act}$ is more pronounced for the lower supersaturations (0.1 and 0.2%).*

**Response:** Thank for pointing this out.

**Changes to the revised manuscript:** We have revised the manuscript accordingly (see L592-601 in the revised manuscript).

*17) Section 3.1 (lines 478 – 481: —After autumn, there is an increase in $D_{act}$ toward winter, despite a decrease in BVOC emissions and the resulting lower organic mass fraction alongside a higher inorganic fraction (see Fig. S9). This suggests the influence of another factor, possibly the higher eBC fraction observed during winter (see Sect. 3.3).*

*From figure S3 it seems the opposite (i.e., $D_{act}$) decreasing for the lowest supersaturation (i.e., 0.1%) from November and until April (i.e., last month of autumn and the whole winter). For all the other supersaturations a clear trend for autumn and winter months cannot be seen, with the exception perhaps* *of 0.5% supersaturation. For the lowest supersaturation (0.1%) the decrease of $D_{act}$ during the winter period is consistent with the lower contribution of the organics, observed during the same period from the bulk chemical composition (figure S9). That said, $D_{act}$ for 0.1% supersaturation is well within the accumulation mode and in the size range where the chemical composition measured by the ACSM should match that of these particles. On the other hand, for the higher supersaturations (0.5 and 1.0%), where $D_{act}$ resides well within the Aitken mode, the differences in the median $D_{act}$ values between autumn and winter do not seem significant to justify a higher contribution of BC in this mode.*

**Response:** Many thanks for this suggestion. We acknowledge that there has was a mistake in the description of the figure.

**Changes to the revised manuscript:** We have revised the manuscript accordingly (see L592-601 in the revised manuscript).

*18) Section 3.2 (paragraph starting from line 536 ending in line 553): In this paragraph the authors provide some plausible explanations for the discrepancies between the estimated (based on the different closure methods) and measured CCN number concentrations. According to my opinion, they should include in addition some sentences discussing the implication(s) of particles mixing state. In section 2.2.3, the authors correctly point out that for performing the closure studies they had to assume internally mixed particles. However, what would be the effects of sampling externally mixed particles? In addition, the authors could perhaps use the HTDMA measurements (Hämeri et al., 2001; cited in the manuscript; or other more recent HTDMA measurements if available) for qualitatively investigating if the particles residing in the Aitken mode are externally mixed and if yes, if this happens in most of the cases or just in some.*

**Response:** In our analysis, we assumed internally mixed particles within each fitted mode (Aitken and accumulation), as outlined in Section 2.2.3. Unfortunately, we did not have concurrent HTDMA measurements to directly verify mixing state during our study period. However, existing literature provides useful insights into typical mixing behavior at Hyytiälä. According to Paramonov et al. (2015), the aerosol in Hyytiälä shows clear seasonal and size-dependent mixing state characteristics. Specifically, they report that particles in the ~75–300 nm range are internally mixed during late spring and early summer (May–July), with a very small CCN-inactive fraction (~0.2%). For the rest of the year, the aerosol becomes partially externally mixed, with the CCN-inactive fraction increasing to ~6.6%. Moreover, the study also presents a distribution of $\kappa$ which shifts significantly between 0.2% and 0.4% supersaturation — potentially reflecting an external mixing between particles above and below 100 nm. However, within each size range — either below or above 100 nm — the $\kappa$ distributions are relatively consistent, suggesting that particles are mostly internally mixed within those size classes. Due to this, we do not expect the assumption of internal mixing to significantly bias our inverse closure results as our analysis optimizes chemical composition and thus hygroscopicity parameter separately for the Aitken and accumulation modes, rather than assuming a single bulk composition. This partially compensates for possible differences in mixing state between modes. We acknowledge that if externally mixed particle populations (e.g., internally non-hygroscopic subfractions) were consistently present and active in the CCN size range, they could influence the closure. However, incorporating such effects would require a more advanced inverse-modeling framework that includes the mixing state as an explicit parameter, as well as supporting observational constraints (e.g., HTDMA or SP-AMS measurements). Developing such an approach would be a valuable next step but is beyond the scope of the current study.

**Changes to the revised manuscript:** We have added a few sentences commenting on the internal mixing assumption to the end of Sect. 2.2.1 in the revised manuscript (L394-400).

*19) Section 4 (lines 641 - 642): — However, all of the applied methods tend to overpredict CCN concentrations to varying degrees.*

*A more clear —take home message can be conveyed to the reader if the authors could be more specific. For instance, they may add some percentages, in order for the reader to better understand the magnitude of the overprediction.*

**Response:** Thank you for this comment.

**Changes to the revised manuscript:** We have modified the Abstract and Conclusions of the revised manuscript to include more clear and quantitative take-home messages (see also response to specific comment #1).

*20) Section 4 (lines 657 – 659): The Aitken mode has the lowest κ values in winter while summer features higher Aitken mode hygroscopicity (lowest accumulation mode κ) possibly due to decreasing BC content which was not accounted for in the calculations.*

*This sentence can be written in a clearer way. I suggest that the authors should conclude separately for the κ values of the Aitken and of accumulation mode particles, since the reasons for the observed seasonal variability in their hygroscopicities are most probably different, based on the discussion in the previous sections. In addition, if I understood correctly, BC content was accounted during the estimation of the particle hygroscopicities and in the different closure methods. What was not accounted for, was a size-dependent BC content. The authors need to describe this in clearer way.*

**Response:** Great point.

**Changes to the revised manuscript:** We have now rewritten the paragraph in question (L817-829), also to account for the new insights from the additional inverse closure method.

*21) Section 4 (lines 678 – 680): — Our study uses this approach, leveraging routine monitoring instruments to estimate size-dependent composition; with the inverse closure method it takes only a few seconds to determine the composition of Aitken and accumulation mode particles for a given time.*

*Do the authors refer here to the computation time of the inverse closure method or to the necessary measuring time by the ACSM, CCNc and DMPS? To my understanding, the time resolution of these instruments is in the order of an hour or longer, especially when accounting for the time that the CCNc needs in order to step 5 supersaturations. Considering this, the estimation of size-dependent composition by combining these instruments would take far more than few seconds. The authors should distinguish and more clearly report the necessary time resolution of the measurements from the computational time of their software routine(s).*

**Response:** We thank the reviewer for this comment. Our results indicate that the 2-hour resolution of the CCN cycle is often enough to get a reasonable idea of the modal composition of the aerosol population.

**Changes to the revised manuscript:** We have removed this confusing statement from the revised manuscript – particularly also because of the more nuanced and quantitative picture given by the MCMC calculations. We hope that the additional detail given in the manuscript helps to further address this comment.

*22) Section 4 (lines 682 – 684): — Moreover, the aerosol particle size distribution should remain relatively stable during a CCN measurement cycle, as the accuracy of predicting CCN spectra is more sensitive to variations in size distribution than to changes in chemical composition (see e.g. Lowe et al., 2016).*

*The combination of the instruments described in this work for estimating one data point of size-segregated chemical composition results to time resolution in the order of one hour or more (see my comment above). However, perhaps the same (or similar) instruments with a different mode of* *operation can be employed for reducing the necessary measuring time. For example, could a Scanning Flow CCN Analysis (SFCA, Moore and Nenes, 2009) or a scanning mobility CCNc Analysis (SMCA, Moore, Nenes and Medina, 2010) be used for significantly reducing the necessary measuring period? Can the above two CCN methods be used with the inverse closure method and software routine(s) developed by the authors?*

**Response:** Indeed, as also shown by the added analysis with the second inverse closure method, the time-resolution of the CCN measurement makes a significant difference for how accurately the modal composition can be constrained.

**Changes to the revised manuscript:** The manuscript has been extensively revised to clearly illustrate this final important point.

**Reviewer #2**

*This manuscript presents a thorough CCN closure study at the SMEAR II station in Hyytiälä, Finland, leveraging long-term observational data (2016–2020) of aerosol size distributions, chemical composition, and CCN concentrations. The authors compare three CCN prediction methods using κ-Köhler theory with different inversion schemes: i) constant kappa parameter, ii) based on bulk chemical-composition, and iii) inferring size-resolved (modal) composition consistent with total mass and CCN observations.*

*They demonstrate that allowing the Aitken and accumulation modes to have distinct chemical compositions ($\kappa_{opt}$) significantly improves the closure, especially at higher supersaturations (>0.5%), where Aitken mode particles contribute more to CCN. Seasonal trends show organic enrichment in the Aitken mode and a higher inorganic fraction in the accumulation mode, with implications for understanding biogenic contributions to CCN. This study addresses key limitations in CCN prediction models related to the lack of size-resolved chemical composition data and provides a novel methodology to infer this information from inverse modeling. The work is especially timely as climate models increasingly rely on more accurate aerosol-cloud interaction representations, particularly in biogenically influenced environments like boreal forests.*

*The manuscript is well-structured, the methodology is sound, and the conclusions are meaningful, therefore, the manuscript should be accepted in ACP. However, before publication some minor revisions are recommended to enhance clarity and strengthen the study.*

**Response:** We thank the reviewer for their positive view on this manuscript.

*Minor comments:*

- ***Lines 26–27:*** *"This optimization improved the CCN closure primarily at supersaturations above 0.5%..."*

  *Please quantify the improvement in closure — e.g., percent error reduction or NRMSE change — so the reader can assess the magnitude of the model enhancement.*

  **Response:** Table 2 presents the NRMSE values for all the closure approaches (see Table S2 for the corresponding GMB values). We believe it is straightforward to calculate the requested differences from these tables.

  ***Lines 29–31:*** *"The mass fractions of inorganics in the two modes vary with season..."*

  *Consider reporting the absolute inorganic mass fractions in addition to the percent* *difference. This would clarify the physical relevance of the enrichment, particularly for radiative implications.*

**Response:** Thank you for this comment.

**Changes to the revised manuscript:** We have included Tables S3-S6 providing the requested information to the supplementary information accompanying the revised manuscript:

- ***Line 182, Section 2.2***

  *The method assumes the total submicron composition remains constant while redistributing it between modes. Please justify whether this assumption holds during periods of intense NPF or cloud processing, which may differentially affect modal composition.*

  **Response:** Thank you for this insightful comment. Indeed, the composition might vary during the CCN measurement cycle due to e.g. the processes mentioned in the comment. It is however beyond the scope of the study the estimate the magnitude of such variability due to the limited observational data in this regard.

- ***Line 213, Section 2.1.1:****"...more black carbon is also observed, which tends to decrease the overall hygroscopicity."*

  *Since eBC is treated as size-invariant in the inverse approach, this assumption might bias winter κ estimates. It can be interesting evaluating the sensitivity of $\kappa_{opt}$ to this fixed eBC partitioning.*

  **Response:** Thank you for this suggestion. We have now conducted an inverse closure study testing the sensitivity of the results to this assumption, and found the effects to be minor for our conclusions.

  **Changes to the revised manuscript:** The Supplementary note 10 and the description in L685-686 of the revised manuscript now describe the sensitivity test with respect to this assumption and its results.

- ***Table 1 and Eq. 4, κ values and densities:***

  *The use of fixed κ for organics (0.12) and organic nitrate might oversimplify temporal variability. Consider discussing the expected range of $\kappa_{org}$ from literature (e.g., 0.05–0.2) and its potential influence on $\kappa_{bulk}$ accuracy.*

  **Response:** Thank you for this suggestion. We have now conducted an inverse closure study testing the sensitivity of the results to these assumptions, and found the effects to be unlikely to provide an alternative explanation to our observations.

  **Changes to the revised manuscript:** The Supplementary note 11 and the description in L776-783 of the revised manuscript now describe the sensitivity test with respect to these assumptions and its results.

- ***Figure 4, Forward and Inverse Closure:*** *The figure would benefit from adding shading or markers to show uncertainty (e.g., interquartile range) of observations. Right now, it's difficult to assess fit quality beyond the median.*

**Response:** Thank you for the comment – we do not understand what is meant here however as the whiskers corresponding to the quartiles are already present in the figure. Perhaps there has been a misunderstanding or an issue with the local reproduction of the figure?

- ***Lines 532–535, κ Discussion:*** *The increase in Aitken mode organic fraction during summer aligns with biogenic SOA production. This would be strengthened by directly comparing seasonal $\kappa_{opt}$ to expected κ values from known BVOC oxidation products (e.g., monoterpenes, sesquiterpenes).*

  **Response:** Thank you for this useful comment. The assumed organic hygroscopicity of about 0.1 corresponds generally well for most organics, including known oxidation products from BVOCs (see e.g. Siegel et al., 2022 and references therein).

  **Changes to the revised manuscript:** We have added a reference to Siegel et al. (2022) and a sentence about the typical hygroscopicity values of organics in L776-783 of the revised manuscript.

- ***Lines 536, CCN overestimation:*** *The consistent overestimation of CCN concentrations across all supersaturations may point to a limitation of the internal mixing assumption, especially during periods of aerosol complexity. Previous studies suggest that such overestimations are exacerbated during times of mixed aerosol sources (e.g., biogenic + anthropogenic + aged background), where internal mixing assumptions break down. I recommend the authors to explore whether episodes of high bias correlate with increased variability in PNSD, eBC, or chemical markers—and to discuss the implications of external or mixed-state aerosols on the robustness of κ-based predictions.*

  **Response:** Thank you for raising this excellent point. See response to the specific comment #18 by Reviewer #1.

- ***Supplementary Table S2:***

  *Although GMB is a useful summary metric, standard deviation or interquartile ranges of GMB values could help assess variability and robustness across time.*

  **Response:** Thank you for this useful suggestion.

  **Changes to the revised manuscript:** We have added Supplementary note 9 and Fig. S16 to provide the requested statistics, and a brief description of the results to L674-675 to the revised manuscript.

**Reviewer #3:**

***Reviewer Report on Manuscript: "Optimizing CCN predictions through inferred modal aerosol composition – a boreal forest case study"***

*The manuscript provides a thorough and insightful exploration of aerosol-cloud interactions, focusing specifically on cloud condensation nuclei (CCN) closure studies in boreal forest environments. Using a robust, multi-year dataset from SMEAR II (2016–2020), the authors employ forward and inverse modelling approaches to evaluate the impact of size-dependent chemical composition and hygroscopicity on CCN predictions. The study addresses key uncertainties in climate modelling and attempts to constrain modal aerosol composition using CCN observations — an approach with significant scientific merit. The manuscript presents an extensive dataset of ~6,200 concurrent ACSM,*

*CCN and size distribution observations from Hyytiälä, Finland. The manuscript evaluates three methods for predicting CCN concentrations and explores seasonal variability in aerosol hygroscopicity and composition, with a particular focus on the differences between the Aitken and accumulation modes. This work addresses a significant challenge in the field of aerosol-cloud-climate research, particularly in the context of future scenarios involving declining anthropogenic emissions and increased contributions from natural aerosol sources. The authors implement a novel inverse closure technique using optimization to estimate mode-specific κ values. This comprehensive study is highly relevant to the atmospheric sciences community.*
*The study is well structured, with a clear methodology and appropriate referencing to prior work. However, there are some areas where the clarity could be improved, potential ambiguities could be resolved and minor corrections could be made to enhance the quality of the manuscript. The dataset is extensive and the topic is timely. Nevertheless, clarification or expansion of several methodological and interpretational aspects is required before the manuscript can be considered for publication.*

**Response:** We thank the reviewer for their kind attention to the innovative nature of this work and considering it timely.

**Changes to the revised manuscript:** We have revised the manuscript extensively to improve clarity. We have also added another inverse modelling technique to strengthen the robustness of the conlusions and allow for optimization with multiple variables. We hope these changes help address the issues raised by this reviewer.

***Major Comments***
*1. The optimization of size-resolved composition is central to this study, yet the method remains a bit opaque. It is not very clear: What parameters are varied during optimization? Are any physical constraints or priors imposed (e.g., known hygroscopicity bounds for organics/inorganics)? Is the optimization performed independently per time point, season, or SS level?*

**Response:** We agree that the methods section could be improved. This was also pointed out by the two other reviewers (please see our responses to them above).

**Changes to the revised manuscript:** We have now extensively revised the Methods section, particularly Sect. 2.2.3 on the inverse modelling, and added new Supplementary notes 4-6 to add more detail.

*2. While the optimized CCN predictions agree better with observations, the inferred composition remains unvalidated. Have the authors compared the mode-specific organic/inorganic fractions with any available independent chemical data (e.g., AMS, offline filters, or PTR-MS)? Without this, there could be a risk of overfitting the CCN closure. Furthermore, the large seasonal variability in composition (e.g., +156% inorganic in winter) should be discussed in the context of known aerosol processes—such as wintertime transport, boundary layer dynamics, or nucleation suppression.*

**Response:** Thanks for this insightful comment that contains several important points. We agree that there is indeed always a risk of overfitting. Largely inspired by this comment we have now added another inverse modelling application (the MCMC) that allows for better accounting for the variability and uncertainties in the input values used in the optimization. We did not have a data set with size-dependent AMS measurements available to us, but presented a comparison to past studies (particularly Allan et al., 2006).

**Changes to the revised manuscript:** The manuscript has been extensively revised to better account for the uncertainties mentioned by the reviewer (among others). These revisions can be found in the Abstract, Methods, Results and Conclusions. In particular, the seasonal variability of composition and its links to atmospheric processes is discussed extensively in Sect. 3.3 of the revised manuscript (see also L592-601 of the revised manuscript).

*3. The improvement is mainly observed above 0.5% supersaturation. The authors should also discuss the implications at low SS and whether the optimization technique could be adapted or constrained to better handle this regime.*

**Response:** Thank you for this insightful comment. Indeed, the addition of the MCMC calculations provide further insights into the supersaturation-dependence of the bias – suggesting the bias at 0.1% to be mainly of instrumental origin, while the bias at 0.2%-1.0% supersaturations could be mitigated when the variability of the PNSD during the CCN measurement cycle was accounted for.

**Changes to the revised manuscript:** The dependence of the bias of the supersaturation is now extensively discussed in Sect. 3.2 of the revised manuscript (see particularly L646-666).

*4. The optimization suggests that accumulation mode particles are more enriched in inorganics while the Aitken mode is more organic-richer. This is plausible, but a more mechanistic explanation is needed. For example: Is this pattern consistent with condensation of oxidized VOCs on smaller particles and aqueous-phase processing on larger particles? How does this compare with seasonal biogenic activity or anthropogenic influence? Including a more detailed interpretation supported by prior literature would strengthen the conclusions.*

**Response:** Thank you for raising this important point.

**Changes to the revised manuscript:** We have extensively revised the manuscript to address for this comment: please see particularly the new and more detailed results and discussion in Sect. 3.3 of the revised manuscript, and their discussion in the Conclusions section. We have also added references to more recent prior studies (e.g. Lance et al., 2013; Spiteri et al., 2023; Massling et al., 2023).

*5. While the research questions are outlined at the end of the introduction, the manuscript would benefit from explicitly stating the working hypothesis earlier (perhaps around line 55 or 70). Suggest rephrasing and condensing the goals for better readability and alignment with subsequent methodology.*

**Response:** Thank you for the good suggestion.

**Changes to the revised manuscript:** We have now added the following sentence to the relevant part of the Introduction (L71-72 of the revised manuscript): "In this study we intend to use a CCN closure study as a means to infer information on size-dependent chemical composition of CCN-sized aerosol particles, to enhance bulk chemical composition measurements."

*6. The inverse modeling framework is a major novelty in this work but is not adequately introduced in terms of assumptions, mathematical implementation, or validation strategies. Clarify what "inverse aerosol-CCN closure" means in practical terms—e.g., optimization method, objective function, constraints used.*

**Response:** Thanks for this suggestion.

**Changes to the revised manuscript:** As discussed in the response to Comment #1 above, we have added substantially more details on the inverse closure methods used to the revised manuscript.

*7. The manuscript discusses organic aerosol extensively but does not explain how the complex properties of organics (e.g., surface tension depression, limited solubility) are accounted for in κ parameterization or closure attempts.*

**Response:** Thank you for this observation. In our analysis, the $\kappa$ parameter is used as an effective hygroscopicity parameter that implicitly accounts for various influences, including limited solubility and possible surface tension effects. However, we do not treat surface tension depression or solubility limitations explicitly. It is worth noting that incorporating surface tension depression into the $\kappa$-Köhler framework would typically reduce the activation diameter, leading to higher predicted CCN concentrations. Given that our closure results based on bulk chemical composition already tend to slightly overpredict CCN compared to observations, explicitly accounting for surface tension depression would likely worsen the agreement.

**Changes to the revised manuscript:** To account for this comment (and similar comments from the other reviewers), we have now conducted an inverse closure study testing the sensitivity of the results to the assumptions of organic molecular properties. We have described these results (which we feel strengthens our conclusions) in the revised manuscript in Supplementary note 11 and L776-783 of the revised manuscript.

*8. There's an implicit assumption that Hyytiälä data can be generalized to other forest regions or clean continental environments. This assumption should be stated explicitly and discussed in the limitations.*

**Response:** Thank you for this comment.

**Changes to the revised manuscript:** We have now added a sentence stating this assumption ("Through assuming that the SMEAR II station represents a remote continental site with a reasonable accuracy, we aim to provide useful insights on the role and dependencies of natural aerosol on CCN loadings.") to the Introduction (L181-182 of the revised manuscript) and its limitation to the Conclusions ("In the future, the method applied here should be tested at other locations with varying aerosol chemical compositions – also to mitigate the inherent representativity issues related to using data from a single station.") (L850-853).

*9. The study notes persistent overprediction errors not resolved by optimized κ values. It would strengthen the work to more directly explore model structural assumptions such as: constant surface tension, neglecting semi-volatile partitioning, mixing state (internal mixing assumption for size modes).*

**Response:** Thank you for this excellent comment. Motivated by this suggestion and the suggestions from other reviewers we have now indeed included a more detailed inverse analysis with an additional method (namely the DREAM-MCMC calculation) which allows for letting also the particle number size distribution vary within its uncertainty limits during the CCN measurement cycle. Including the size distribution variability during the CCN measurement cycle improves the closure considerably.

**Changes to the revised manuscript:** As discussed above, we have now extensively revised the manuscript to describe the results of the additional MCMC with allows for a multivariate optimization and hence gives a more robust but nuanced picture of the results.

*10. The assumption of stable size distributions during the CCN cycle is critical. Was this stability verified using size-resolved time series? Otherwise, this assumption should be treated more cautiously.*

**Response:** Thank you for this excellent comment, see also our responses to the comment #9 above.

**Changes to the revised manuscript:** We have extensively revised the manuscript to describe the new inverse closure methodolody and results, which mitigate this critical issue.

*11. The text mentions calibration frequency for CCNc and invalidation criteria for aethalometer data (e.g., RH > 40%). Please clarify how data gaps or invalid data points were handled in the analysis. Were interpolation or gap-filling methods used? What fraction of data was excluded due to quality control?*

**Response:** Thank you for your comment. For the CCNc data, we did not apply any gap-filling or interpolation methods. Only valid, quality-assured data were used in the analysis. Regarding the equivalent black carbon (eBC) data, approximately 92% of the study period is covered. Data gaps mainly resulted from periods when the instrument was undergoing maintenance or experienced technical issues. As part of our quality control, we excluded data points when the relative humidity inside the aethalometer exceeded 40%, as well as occasional clear outliers. Similar to the CCNc data, no interpolation or gap-filling was performed. All analyses were based solely on available, quality-checked data.

***Minor Comments***

*Line 43: "Aerosol particles are important in the formation…" consider rephrasing as "Aerosol particles play a critical role in the formation…"*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have modified the manuscript accordingly (L40).

*Line 44: Check the phrasing. Suggest: "...by lowering the energy barrier for the heterogeneous nucleation of water, thus promoting cloud droplet activation…"*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have modified the manuscript accordingly (L41).

*Line 46-47: Rephrase: "thereby changes in the CCN concentration" to "thus, changes in CCN concentration"*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have modified the manuscript accordingly (L43-44).

*Line 57: "drivers of SSmax fluctuations". Define "$SS_{max}$" explicitly on the first use for clarity.*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have modified the manuscript accordingly (L55).

*Line 68: Suggest moving the sentence "These inverse approaches…" earlier to clarify the inverse model's novelty and importance.*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have rewritten the respective paragraph for clarity (L66-72).

*Line 93: "Still, organic aerosol plays a significant role…". Consider beginning with "Nevertheless," to better connect to prior sentence.*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have modified the manuscript accordingly (L94).

*Line 123: Confusing sentence. Suggest: "Specifically, median $\kappa$ was 0.41 at 0.1% SS (corresponding to larger activation diameters), and 0.14 at 1.0% SS (smaller activation diameters)..."*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have modified the manuscript accordingly (L125).

*Line 129: Add a clarifying phrase on what "systematic overprediction" means quantitatively.*

**Changes to the revised manuscript:** We have added substantial detail on the quantiative statistics of the goodness of the closure for the various methods. We hope that this helps address this comment.

*Lines 196-214: The site description is thorough, but additional discussion on how representative the SMEAR II site is for boreal forest aerosols under varying seasonal anthropogenic influence would be valuable.*

**Response:** Thanks for this comment. We believe that the current site description already explains how the sources influencing SMEAR II vary seasonally and how this affects aerosol composition.

**Changes to the revised manuscript:** To improve the site description further, we have now added a few clarifying sentence to Sect. 2.1.1.

*Line 310: "with by a Nafion dryer" to "with a Nafion dryer" (remove "by").*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have modified the manuscript accordingly (L321).

*Lines 314-315: The ACSM and eBC data are averaged over 1-hour intervals but converted to a 2-hour median to match CCN measurements. Please discuss potential impacts of this temporal averaging on capturing short-term variability in aerosol composition and CCN. Were any tests performed to ensure this does not bias the results?*

**Response:** We did not perform a separate test for this, as the variability of the size distribution was deemed to be the most important source of uncertainty in this regard. The original ACSM data were already averaged to 1 h, since the overall aerosol loading at Hyytiälä is quite low and shorter averaging would not provide meaningful statistics. For comparison with CCN, we further converted these to 2 h medians to match the CCN cycle. The sensitivity tests conducted with the MCMC approach and described in the new Supplementary note 11 at least partly address this comment however.

*Line 659–661: Consider rephrasing for clarity: "The relative difference in the median Aitken and accumulation κ…" to Perhaps "The seasonal variability in median κ between Aitken and accumulation modes is most pronounced in winter (~162%)…"*

**Response:** Thank you for this comment. We would, however, prefer to keep the sentence as is to keep it clear that the 162% refers to the difference in the Aitken and accumulation mode $\kappa$ instead of the amplitude of the seasonal variability. We hope this is acceptable.

*Line 676–678: Repetition – consider merging: "observed CCN concentrations are a valuable tool… Our study uses this approach…" to avoid redundancy.*

**Response:** Thank you for highlighting this repetition.

**Changes to the revised manuscript:** We have removed the repetitive paragraph.

*Line 687–688: Suggest citing more recent or diverse κ-related parameterization studies for broader context.*

**Response:** Thank you for the suggestion.

**Changes to the revised manuscript:** We have included a few more studies, such as Lance et al. (2013), Ray et al. (2023), and Siegel et al. (2022) to the revised manuscript.

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