## 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.*

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. More specifically:*

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. Based on comments and suggestion from other reviewers too, we would like to add few more analysis to the revised manuscript and we have attempted to mention the changes appropriately in this document. Just for clarity, please note that $\kappa_{\mathrm{opt}}$ refers to the method where we use Nelder–Mead optimization, focusing only on the optimization of the modal chemical composition. In the revised manuscript, we have included additional analyses, the most significant of which involves DREAM-MCMC simulations. In this approach, referred to as $\kappa_{\mathrm{MCMC}}$, 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.

*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).*

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. To improve on making these points even clearer, we have rewritten the abstract, part of conclusions, as well as elaborated the methods more in detail adding also a work flow chart (see Fig. R1 below) that shows how the size-dependent composition is inferred from the inverse closure methods.

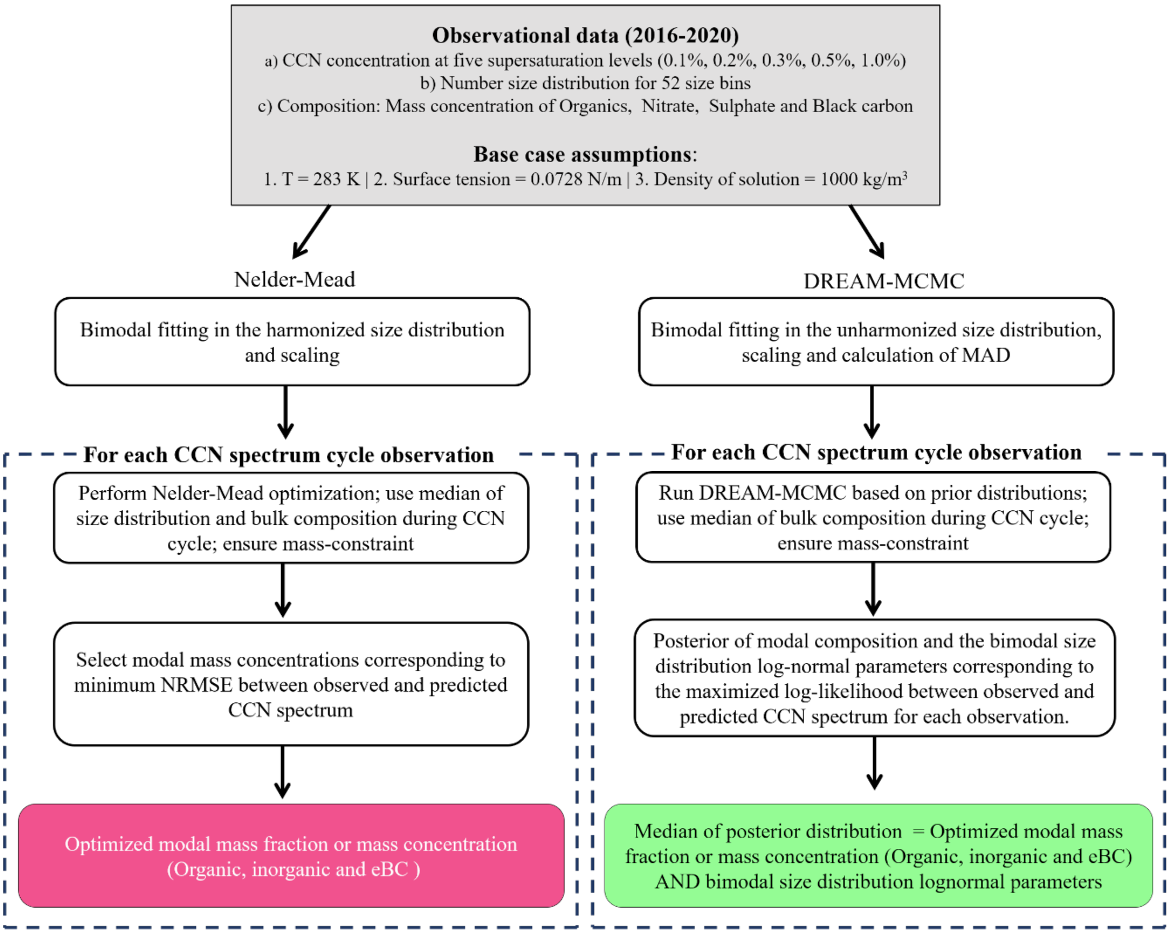

**Figure R1.** Workflow of the two inverse closure methods: the Nelder–Mead algorithm (left) and the DREAM-MCMC (right) approach. Bimodal fitting - representation of the aerosol size distribution as two lognormal modes. Harmonized size distribution - size distribution data harmonized to CCN data; data 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.*

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. We have added a brief discussion about it to the revised manuscript in the conclusion section as:

“If modal or size-resolved $\kappa$ (in addition to just having bulk chemical composition) were available, our approach could be extended to derive more detailed size-dependent chemical composition, while also helping to constrain $\kappa$ values by identifying those that best match 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.*

We agree that the methodology, assumptions, simplifications, and limitations should be described more clearly to enhance transparency and reproducibility. 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. We agree that the methods section was at parts unclear and lacked some important details. To account for comments from other reviewers as well, we have now revised the text to improve clarity and add more details. For example, we have included new sections where the Nelder–Mead inverse closure method is explained in detail, as well as a section which describes the added DREAM-MCMC approach. We have also added a new flowchart (Fig. R1) to give a clearer overview of the workflow with the inverse closure methods.

***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.*

Thank you for the suggestion. We have written a new abstract that better represents the main methods and results:

“With reductions in anthropogenic emissions, natural aerosols from boreal forests are expected to play a crucial role in total aerosol loadings. Understanding their cloud-forming potential is therefore crucial. Observational data on aerosol particle number size distribution and chemical composition is required for predicting cloud condensation nuclei (CCN) concentrations using Köhler theory. However, long-term online measurements of bulk chemical composition typically provide data on total sub-micron particulate mass, which only represents the larger end of the number size distribution. Previous studies have shown significant differences in the hygroscopicity of Aitken and accumulation mode particles in boreal environments. Neglecting this size-dependence can substantially overestimate cloud condensation nuclei (CCN) concentrations — particularly at supersaturations (*SS*) where Aitken particles activate. We applied $\kappa$-Köhler theory to a multi-year dataset (2016–2020) from Hyytiälä, Finland, to evaluate different representations of aerosol chemical composition for CCN prediction. Forward closure tests using either bulk chemical composition or a constant $\kappa$ value of 0.18 (Sihto et al., 2011) overpredicted CCN, with geometric mean bias (GMB) highest at *SS* = 0.1% (1.56 and 1.35) and also notable at *SS* = 1.0% (1.34 and 1.26). To mitigate this bias, we performed inverse closure by optimizing size-resolved composition using: (1) Nelder–Mead method with the size distribution fixed to its median during each 2-hour CCN spectrum cycle, and (2) MCMC accounting also for the variability in the particle number size distribution during the CCN measurement cycle. Both methods improved closure at *SS* = 1.0% (GMB = 1.20 and 0.99), with moderate improvement at 0.1%. The optimized results revealed organic enrichment in the Aitken mode in many occasions (as compared with the overall bulk chemical composition): the Aitken mode was enriched in organics in 77% of cases using method (1) and 46% using method (2) – with typical $\kappa$ values around 0.1 and 0.3 for Aitken and accumulation modes, respectively. The results are generally in line with what is known about size-dependent chemical composition in Hyytiälä, and suggest that most of the variability of aerosol hygroscopicity in Hyytiälä is due to variability in Aitken mode composition. The results also highlight the important role of the highly-variable Aitken mode size distribution in influencing the overall CCN variability at the site. Our results demonstrate the potential of inverse CCN closure methods for obtaining valuable information of the size-dependent chemical composition beyond the reach of bulk chemical composition measurements."

*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.*

Thank you for pointing it out. $SS_{max}$ is now defined appropriately when mentioned for the first time 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.*

Thanks for this suggestion. We agree and have now included more details in the manuscript:

"Importantly, some organic aerosol properties beyond hygroscopicity – such as solubility or surface film formation which reduces surface tension – 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.*

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.

*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.*

Thank you for mentioning this. We have corrected it in the new version of the 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.*

Thank you for this helpful suggestion. We agree that explicitly mentioning the hydrophobic nature of black carbon adds clarity. Accordingly, we have revised the sentence in the manuscript according to your suggestion to better reflect this point.

*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.*

Thanks for mentioning a lack of clarity here. We have added the sentence "The further steps required in the inverse closure are described in more detail below." To the end of the paragraph, which hopefully clarifies that the bimodal fitting is indeed only the first step in the inverse closure approach. As mentioned above, we have generally added more detail on the inverse closure methodology to the revised manuscript.

*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.*

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. 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. We have added a reference to the work by Liwendahl (2023) to the revised manuscript.

*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.*

Thanks for suggestion, which we have accounted for in the revised manuscript. We have also included a description of the measurement cycle to obtain the complete CCN spectrum.

*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.*

Thanks for suggestions. We have made appropriate changes in the manuscript.

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

Thanks for pointing it out. We have corrected it in the 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.*

Thank you for the comment – please see our response to comment #8.

*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).*

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.

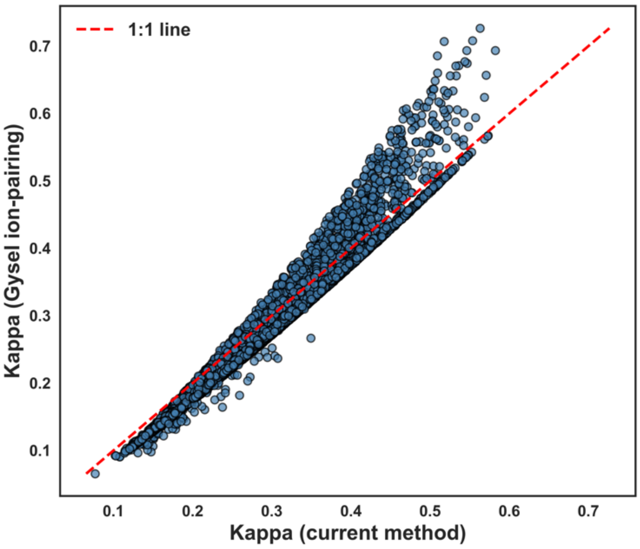

**Figure R2.** Comparison of $\kappa$ values calculated using two different methods: (i) a simplified full neutralization approach assuming complete conversion of $SO_4^{2-}$ and $NO_3^-$ to ammonium sulfate and ammonium nitrate, and (ii) a more chemically detailed ion-pairing scheme based on Gysel et 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.*

Thank you for this valuable suggestion. We agree, and have added a great deal of more details on the inverse modelling approach to both, the main manuscript and the supplement (see also Fig. R1). For instance, the descriptions of the two inverse modelling approaches and the related supplementary notes now read as below:

### "2.2.3.1 Nelder-Mead

The Nelder–Mead simplex algorithm (Gao and Han, 2012) is suitable for both one-dimensional and multidimensional optimization problems and is relatively fast in our application. In our case, we need to optimize only one variable (the fraction of total organic mass in Aitken mode, $M_{\mathrm{org1}}$) and the remaining masses can be derived from it through mass closure constraints. For each time step, the optimization begins with an initial simplex of three trial values of $M_{\mathrm{org1}}$, and the NRMSE is evaluated at each point. The worst-performing value is reflected across the midpoint of the better two to explore whether a more accurate estimate can be found in the opposite direction. If this improves the fit, the algorithm attempts an expansion, pushing further in the same direction. If reflection does not improve the result, a contraction step is taken to move closer to the midpoint. If neither reflection nor contraction improves the outcome, the simplex undergoes shrinkage, tightening around the best-performing solution to focus the search locally. This process continues until the optimization converges, resulting in an estimate of $M_{\mathrm{org1}}$ that minimizes the NRMSE between modeled and observed CCN concentrations. Note that Nelder–Mead works well for simple, low-dimensional problems like optimizing just one parameter (e.g., $m_{\mathrm{orgl}}$), but it starts to struggle as the number of variables increases and have a tendency for converging to local minima. Hence, in the inverse closure calculations using the Nelder-Mead algorithm we assumed the particle number size distribution and chemical composition to stay constant throughout the CCNc measurement cycle.

### 2.2.3.2 DREAM-MCMC

In order to assess the importance of the variability of the bimodal size distribution parameters within each CCN cycle, we conduct a second inverse-closure experiment with the number concentration and mean diameter for both modes as additional optimization parameters (simultaneously with $M_{\mathrm{orgl}}$ ). Since optimizing both size distribution parameters and composition introduces a more complex and higher-dimensional parameter space, and we are interested parameter uncertainty, we use a Bayesian inference approach to estimate the parameter posterior distributions. Specifically, we chose the DiffeRential Evolution Adaptive Metropolis Markov Chain Monte Carlo (DREAM-MCMC) algorithm (Vrugt et al. 2009), which has been previously used for inverse CCN-closure studies in idealized cases (Partridge et al. 2012) and is available in the Python PINTS library (Clerx et al. 2019). DREAM-MCMC is an efficient MCMC method (Metropolis et al. 1953, Gelfand et al. 1990) that evaluates multiple Markov chains in parallel and automatically adapts its proposal strategy during sampling, making it particularly efficient for correlated, multi-modal problems such as aerosol-cloud microphysical interactions. To know more about MCMC and Bayesian inference, please see Supplementary note R1.

We initialize the MCMC optimization with Cauchy priors for each parameter, centered on the median values of the fitted bimodal size distributions for each CCN cycle, specifically, the number concentration and geometric mean diameter. For chemical composition we use the median of the ACSM observations during each CCN spectrum cycle. The scale value is the smaller of either 1 (resulting in a Student-$t$ distribution) or the median absolute deviation (MAD) of the observations within the given CCN cycle. We constrain the total aerosol mass in each mode to remain within ±10% of the total mass observed by the ACSM and aethalometer.

The priors are truncated to positive values only. We use a heteroskedastic Gaussian likelihood function, such that the highest likelihood is typically where the parameters provide the least squares fit to the CCN observations. The likelihood is defined as

$$L(\theta|\mathrm{Y}) = \prod_{i=1}^{n} \frac{1}{\sqrt{2\pi s_i^2}} exp\left[-\frac{1}{2} s_i^{-2}\left(y_i - \phi(X_i, \theta)\right)^2\right]$$

where $s$ is standard deviation of the noise on each output $i$, which we assume is 10% of the CCN observations at each supersaturation value. We perform the optimization in a log-transformed parameter space, which improves sampler efficiency by normalizing scale differences between parameters. For each CCN observation window, we run five chains with 40,000 iterations per chain, of which the first 15,000 are used as burn-in/adaptation. Up to two chains are discarded if they deviate strongly in central tendency after burn-in, and the last 20,000 steps of all accepted chains are then used to calculate posterior statistics. Convergence is assessed with the $\hat{R}$-statistic (Gelman and Rubin, 1992), using a relaxed threshold of $\hat{R} < 2.5$ for all five parameters to retain a window in the analysis. The $\hat{R}$-statistic compares the variance within chains to the variance between chains; values close to 1 indicate well-mixed, converged chains. We use a relaxed threshold because the $\hat{R}$-statistic is quite conservative and because our problem has high correlation between parameters and the potential for multi-modality if there are multiple distinction aerosol populations within one window, which is penalized by the $\hat{R}$-statistic but realistic in this case. Overall, 19% of windows are discarded due to high $\hat{R}$-statistic values. Even with the relaxed threshold, some windows are excluded where the MCMC identifies reasonable parameter values and CCN spectra but the chains fail to mix well and we cannot guarantee the posterior is well explored.

### 2.2.2 Metrics for assessing variability of lognormal size distribution parameters during CCN cycle

Unlike the Nelder–Mead optimization method, which uses the median of the size distribution during the CCN cycle period, the DREAM-MCMC setup requires the variability of the size distribution as input. To account for this, we calculate the median absolute deviation (MAD) of each lognormal

parameter for every CCN cycle observation. The overall distribution of MAD values for the full 5-year dataset is presented in Supplementary note R2 and Fig. R3. MAD for individual CCN cycle period is calculated as follow:
Let $I_c$= [ $t_c^{start}$, $t_c^{end}$) be the time window for CCN cycle $c$; For a given lognormal parameter, $k$ (among geometric mean diameter (GMD), geometric standard deviation (SD) and number concentration in each mode; so total 6 parameters), collect the samples inside this window as $\{x_k\ (t): t \in I_c\} = \{x_{k,1}, x_{k,2}, \ldots, x_{k,n_c}\}$.
Median in the interval is $m_k(c)$:

$$\text{median}\{x_{k,1}, x_{k,2}, \ldots, x_{k,n_c}\}$$

Median Absolute Deviation (MAD) in interval $c$**:**

$$\text{median}|x_{k,i} - m_k(c)|, \text{ where } i \text{ varies from 1 to } n_c$$

.”

“**Supplementary note R1**
In this section we give a short summary of Bayesian inference and MCMC, then describe the detailed setup of the DREAM-MCMC algorithm used in this study and provide some summary statistics. For a comprehensive review of Bayesian methods, see Gelman et al. (2013). Bayesian inference is a rigorous method for quantifying uncertainty in model parameters, using probability statements. Unknown parameters are treated as random variables with some joint posterior probability distribution, which can be written using Bayes law as:

$$p(\theta|Y) = \frac{p(\theta)L(Y|\theta)}{p(Y)}$$

where $p(\theta)$ is the prior distribution which encompasses what is known about the parameters prior to observing any data, $L(Y|\theta)$ is the likelihood function which measures how well the model fits observed data, and

$$p(Y) = \int p(\theta)p(Y|\theta)\mathrm{d}\theta$$

is the marginal distribution of Y, which represents the probability of observing Y given all possible parameter values $\theta$. The result of conditioning the prior distribution with some observations is the posterior probability distribution $p(\theta|Y)$, which represents the updated probability of the model parameters. Bayesian inference is therefore a process of creating a probability model and iteratively updating that model based on some observations, resulting in a best estimate of the parameters and knowledge about their uncertainty, sensitivity, and correlation (in the case where $\theta$ is vector-valued).

Monte Carlo Markov Chain (MCMC) simulations are a methodology for sampling from posterior distributions. Generally, they involve repeatedly and sequentially sampling $\theta$ such that each new draw depends only on the previous sample and therefore forms a Markov chain, and correcting those draws so that the chain converges to the target distribution. Many different algorithms have been proposed for generating and correcting chain samples. Here we use the DiffeRential Evolution Adaptive Metropolis Markov Chain Monte Carlo (DREAM-MCMC) algorithm (Vrugt et al. 2009). This algorithm runs multiple chains simultaneously and adaptively updates the proposal distribution using a randomized subset of the chains’ joint history. It also supports large proposal jumps and outlier rejection during the initial burn-in phase, which accelerate convergence. This type of self-adaptive evolutionary strategy is particularly well suited to heavy-tailed or multi-modal posteriors, such as in this study where different combinations of aerosol chemical composition and size distributions parameters could result in similar CCN spectra. ”

“**Supplementary note R2**

Figure R3 shows the probability density of median absolute deviation, MAD values fitted with chi-squared distributions for both the Aitken and accumulation modes. The results indicate that the variability is generally small, with distributions strongly centered close to zero and narrow tails. The fitted chi-squared parameters suggest that fluctuations in diameter and sigma are low, whereas number concentrations show comparatively larger spread. Overall, this analysis confirms that the derived parameters remain fairly stable within CCN cycle period, with occasional variability in particle number concentration.

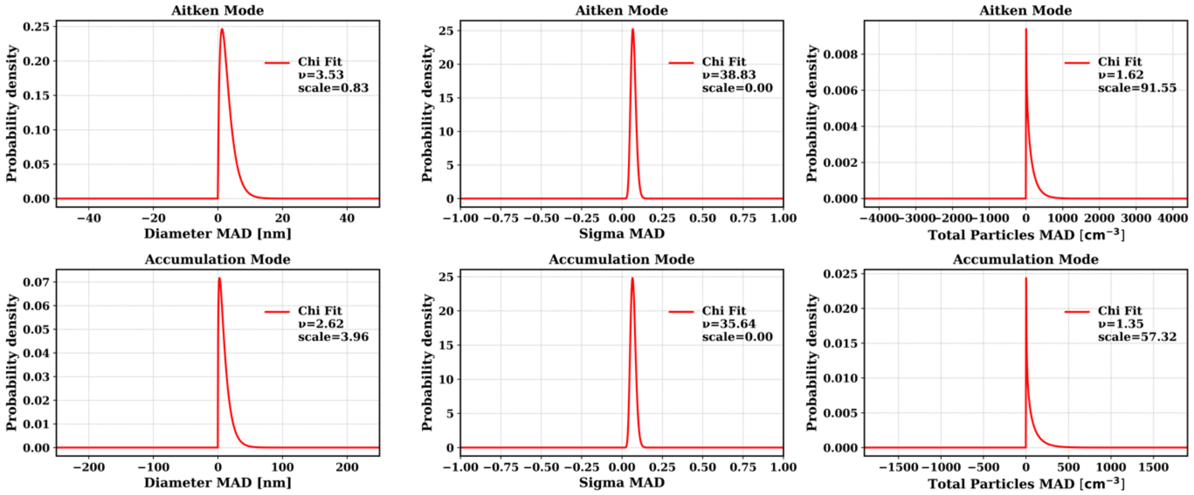

**Figure R3.** Chi-squared probability density functions (PDFs) fitted to the median absolute deviation (MAD) values of aerosol size distribution parameters calculated with respect to median of corresponding parameters during CCN cycle. The data represent entire 5 years period between 2016 to 2020. Panels show Aitken mode (top row) and accumulation mode (bottom row) MADs for mode diameter, geometric standard deviation ($\sigma$), and particle number concentration. The fitted parameters (degrees of freedom, $v$, and scale) are reported in the legends. The fits are constrained to non-negative values to reflect the definition of MAD.

*i) Lines 412 – 413: —This makes $\kappa_{opt}$ variable in time as well as a function of particle size.*

*Was $\kappa_{opt}$ variable as a function of particle size, or two $\kappa_{opt}$ values were assigned (i.e., one for the Aitken and one for the accumulation modes)? This needs to be clarified.*

Indeed, the sentence should have read "two $\kappa_{\mathrm{opt}}$ values were assigned (i.e., one for the Aitken and one for the accumulation modes)". This is corrected in the revised manuscript.

*ii) The equation(s) relating $\kappa_{opt}$ to the measured CCN activation spectra is missing. The authors in section 2.2.1 provide the generalized equation of the κ-Köhler theory only (see equation 2 in the manuscript). Did they use this equation, during the inverted closure? If yes, what was the value(s) assigned in Dp,wet?*

Thank you for pointing this out. Yes, we did use the $\kappa$-Köhler theory (as shown in Equation 2) during the inverse-closure analysis. In this process, the $\kappa$-Köhler equation is applied repeatedly for different combinations of Aitken and accumulation mode chemical compositions. These combinations are explored using the Nelder–Mead optimization algorithm. The key difference between the forward and inverse closure is that in the forward closure, we use a single set of Aitken and accumulation mode mass fractions — both taken from the bulk composition —and apply the $\kappa$-Köhler equation once to compute the predicted CCN spectrum. In contrast, during the inverse closure, the $\kappa$-Köhler equation is run multiple times across a wide range of combinations of Aitken and accumulation mode chemical compositions. For each combination, we calculate a predicted CCN spectrum and then evaluate the normalized root-mean-square error (NRMSE) between the predicted and observed CCN spectra. The composition that results in the lowest NRMSE is selected as the best-fit solution. Regarding the wet diameter used in the $\kappa$-Köhler calculations, in both forward and inverse closures we use the value that corresponds to the assigned composition and dry diameter in any given size bin (defined by the size grid of the DMPS data).

*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.*

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 of exploring 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).*

We agree, and hope that the inclusion of Fig. R1 addresses this valuable 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.*

Here we meant to say that in every season, the $D_{act}$ is greater than previously reported. We agree that more information is required for better clarity, and have modified the revised manuscript accordingly. We have also added the quantitative comparison requested.

*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%).*

Thank for pointing it out, we have revised the manuscript accordingly.

*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.*

Many thanks for this suggestion. We acknowledge that there has to be correction in the description of this figure. In the latest version of the manuscript, we have incorporated the changes. The inclusion of the MCMC inverse closure method, we have also gained further insights into the supersaturation-dependence of the goodness of the closure – particularly for 0.1% – which have also been added to the revised manuscript. The relevant paragraph now reads:

"While the seasonal variation in median activation diameters $D_{\mathrm{act}}$ is not pronounced across all *SS*, more detailed inspection (Fig. S3) reveals a decrease in $D_{\mathrm{act}}$ at the lowest supersaturation (0.1%) during the transition from autumn into winter (November to April). This trend is consistent with a reduced contribution of organic aerosols and a higher relative abundance of inorganic components during winter, as also indicated by the bulk chemical composition (Fig. S9). Since the activation diameters at 0.1% *SS* fall within the accumulation mode, the size range where ACSM measurements are most representative, the observed seasonal variation in $D_{\mathrm{act}}$ at this *SS* level can be directly linked to changes in aerosol composition. Overall, across all supersaturations, an increase in $D_{\mathrm{act}}$ is generally observed during the transition from spring to summer which is more pronounced at 0.1%, 0.2%, and 1.0% *SS*, while being relatively weak at 0.5% *SS*."

*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.*

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 — 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 will add a description of these results to the revised manuscript.

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.

*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.*

Thank you for this comment. We have modified the Abstract and Conclusions of the revised manuscript to include more clear and quantitative take-home messages (see response to specific comment #1). The relevant paragraph in the Conclusions now reads:

"CCN concentrations at Hyytiälä exhibit clear seasonal variations, peaking in summer and reaching their lowest in winter, reflecting overall particle number trends. Our closure calculations generally agree reasonably well with observed CCN concentrations, with Pearson correlations exceeding 0.8. However, all of the applied methods tend to overpredict CCN concentrations to varying degrees. As expected, the inverse closure methods perform the best, especially at higher supersaturations (0.3%, 0.5% and 1.0%), where both accumulation and Aitken mode particles can activate, highlighting the importance of accounting for the size-dependent nature of aerosol composition for more accurate CCN predictions. Overall, the GMB remains well below 1.3 for $\kappa_{\mathrm{MCMC}}$, $\kappa_{\mathrm{opt}}$ and $\kappa_{0.18}$ across all supersaturations (see Table S1 in supplementary), except at 0.1%. The best agreement is observed at 0.2% and 0.3% supersaturations, where the GMB is around 1.1 for all methods, except for $\kappa_{\mathrm{MCMC}}$, for which the best agreement occurs at 0.5% and 1.0%, suggesting that most of the overprediction at higher supersaturations, where the Aitken mode activates can be reduced if variability in the lognormal parameters of the size distribution is also considered. However, at a supersaturation of 0.1%, the use of size-dependent composition i.e. $\kappa_{\mathrm{opt}}$ and $\kappa_{\mathrm{MCMC}}$ don't significantly reduce the error. This suggests that the primary source of the error at this supersaturation arises from another factor — most likely, the substantial measurement uncertainty of the CCN counter at low supersaturation, as previously discussed (see Sect. 3.2)."

*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.*

Great point. The sentence now reads "The Aitken mode has the lowest $\kappa$ values in winter, while summer features higher Aitken mode hygroscopicity (lowest accumulation mode $\kappa$) possibly due to decreasing BC content." We have also modified the conclusions and the comparison between Aitken and accumulation mode hygroscopicities obtained with the two inverse closure methods to the revised manuscript.

*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).*

We thank the reviewer for this comment. Our results now indicate that – except in some cases – the 2-hour resolution of the CCN cycle is often enough to get a reasonable idea of the modal composition of the aerosol population. We have added extensive explanations to clarify this in the revised manuscript, and removed the confusing sentence that the reviewer refers to.

*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?*

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. The manuscript has been revised extensively to clearly illustrate this point.

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