Comment on acp-2022-547
Anonymous Referee #3

Thank you for providing the constructive comments and reviewing our paper. We hope our extensive comments in the review illustrate our genuine efforts to engage with all review comments constructively. The responses to the reviewer's comment are indicated in blue, while the modifications made in the revised manuscript are highlighted in red.

The paper by Patel et al. presents a methodology to infer the concentration of cloud condensation nuclei (NCCN) from multiwavelength Raman or high spectral resolution lidar observations. This outline of the paper's content shows that the title is not at all in line with what is actually presented. The technical nature of the work suggests that it should have been submitted to AMT rather than ACP. Below, I am providing a list of some of the many issues with this contribution that lead me to recommend rejection of this work for publication in ACP (and AMT in case it is deferred there).

Thank you for your comment. I appreciate your feedback and I will take it into consideration.

We agree that the title of the paper may not be closely fit to the manuscript. The title suggests that the paper presents a method for resolving vertically-resolved CCN number concentration, when in fact the paper presents a method for quantifying CCN number concentrations. We replace the title of the paper.
"A Remote Sensing Algorithm for the Quantifying Vertically-Resolved Cloud Condensation Nuclei Number Concentration from Spaceborne Lidar Measurements"

We understand your concern that the technical nature of the paper suggests that it should have been submitted to Atmospheric Measurement Techniques (AMT) rather than Atmospheric Chemistry and Physics (ACP). However, we believe that the paper is of sufficient interest to the broad readership of ACP. The paper presents a new method for estimating CCN concentrations, which is a topic of active research in atmospheric science. The paper also has implications for understanding the global distribution of CCN and their impact on climate.
We will address the specific issues that you have raised in your comment in the revised manuscript. We appreciate your help in improving the quality of my work.

Originality: It is very hard to assess the originality of this work. Entire sections and figures seem to be copied from earlier publication (in particular Lv et al., 2018; Tan et al., 2019; and Choudhury and Tesche, 2022a) without bothering to even reformulate or redesign. This is also witnessed by the unusually high similarity index of 20%. The authors should state more clearly as is currently the case that they are following the methodology of earlier publications and emphasise in which form they are improving upon the earlier methods. They currently fail to properly acknowledge alternative efforts to infer global height-resolved and aerosol-type specific CCN concentrations from spaceborne CALIPSO lidar observations as described in Choudhury and Tesche (2022a; b) and Choudhury et al. (2022).

Thank you for your valuable feedback on the originality of our work. We appreciate your insights and have taken your comments into careful consideration. We would like to clarify the differences between our work and previous publications while highlighting the improvements we have made to the methodology:

1. *Expanded Applicability:* Our research has extended the methodology originally proposed by Lv et al., 2018, to accommodate lidar measurements from diverse platforms. This enhancement allows us to estimate CCN concentrations from both airborne and spaceborne lidars, increasing the versatility and applicability of the algorithm.
2. *New Look-Up-Tables (LUTs):* We have developed new LUTs that include aerosol size and composition information for five distinct aerosol types. This improvement enables more accurate estimations of CCN concentrations, and it empowers our algorithm to account for a wider range of aerosol scenarios, thus improving the overall accuracy of our estimates.
3. *Leveraging Additional Signal:* To address the non-uniqueness problem during the inversion process, we incorporated the extinction coefficient at 1064 nm from lidar measurements. By doing so, we significantly enhance the accuracy of particle size distribution estimations, leading to more reliable CCN estimations, particularly for the coarse mode.
4. *Wavelength-Dependent Hygroscopic Correction:* Another key advancement in our revised methodology is the implementation of a wavelength-dependent hygroscopic growth correction. This correction accounts for changes in particle size near water vapor, resulting in more accurate estimations of particle size distribution and, in turn, improving the precision of CCN estimations.
5. *RH Error Analysis:* Extensive analysis has been conducted to assess the impact of errors, both systematic and random, from relative humidity (RH) on CCN estimation accuracy. As a result of correcting RH errors to the lidar measurements by applying wavelength-dependent hygroscopic growth factor, the reduction in RH error has demonstrated that further enhance the accuracy of CCN estimation.

We visually demonstrated the impact of our improved methodology by conducting a similar error analysis as Lv et al., 2018, allowing readers to observe the enhancements. Additionally, we conducted systematic and random error analyses considering relative humidity (RH) to showcase our algorithm's capacity for accurate CCN estimation. The estimated CCN was further validated against two years of NASA ORACLES airborne campaign data to assess the accuracy of our algorithm. Furthermore, a case study involving CCN retrieval from CALIOP onboard CALIPSO was performed to highlight the capability of algorithm.

Regarding the differences between our approach and those of Tan et al., 2019, and Chaudhary and Tesche, 2022a:

- Tan et al., 2019, utilized ground-based $3\alpha+2\beta$ lidar measurements and developed a machine learning-based algorithm for CCN estimation. In contrast, our methodology follows a Look-Up-Table (LUT)-based approach. While both methods aim to estimate CCN concentrations, our LUT-based method offers the advantage of accommodating lidar measurements from diverse platforms, including airborne and spaceborne lidars, making our approach more versatile and applicable. Additionally, our hygroscopic correction method differs from Tan et al.'s relatively older approach, which further contributes to improved accuracy in our study.
- Regarding Chaudhary and Tesche, 2022a, they proposed a single-wavelength-based algorithm for CCN estimation from CALIPSO data, which involves scaling pre-assumed normalized size distributions based on the CALIPSO-measured extinction coefficient at 532 nm. While this

method has been utilized, it's important to note that it can potentially lead to multiple solutions for particle size distribution with the same extinction coefficient but having different backscatter coefficients, presenting some non-uniqueness challenges.

- On the other hand, our 3α+2β methodology, widely used for lidar retrievals, effectively reduces the non-uniqueness problem associated with single-wavelength solutions. Moreover, by leveraging the additional measurement of the extinction coefficient at 1064 nm, our algorithm further addresses non-uniqueness and significantly enhances the accuracy of CCN estimation, particularly for the coarse mode. These improvements are clearly demonstrated in Figure-10, where our algorithm provides relatively accurate CCN estimations.

These advancements make our methodology more widely applicable and accurate, addressing challenges in estimating CCN concentrations from lidar observations and contributing to the field of aerosol-cloud interaction studies. We have also added a section in the paper summarizing the improvements we made to the methodology proposed by Lv et al., 2018. This section aims to help readers understand the unique contributions of our work and how it differs from the work of other authors.

Building upon the methodology proposed by (Lv et al., 2018), we have enhanced and generalized the approach to enable its application to airborne and spaceborne lidar measurements for CCN estimation. The core of the algorithm relies on the utilization of look-up tables (LUTs) that incorporate aerosol size and composition information, facilitating reliable and vertically-resolved CCN estimation. N_{CCN} values are obtained at six critical supersaturations from 0.07% to 1.0% based on retrieved particle size distributions. Significant improvements have been implemented within the methodology. Firstly, its applicability has been expanded to accommodate lidar measurements from diverse platforms. Secondly, the LUTs now include five aerosol types, ensuring a more comprehensive representation of aerosol characteristics. Thirdly, the methodology leverages the additional signal of the extinction coefficient at 1064 nm, effectively addressing the uncertainty associated with the non-uniqueness problem during the inversion process. Fourthly, including the hygroscopic growth correction in the revised method has led to significant improvements in the accuracy of CCN estimation, further enhancing the reliability and robustness of the. Finally, results the extensive analysis has been conducted by including the errors from RH.

Reproducibility: The (data and) methodology section is incomplete and doesn't provide the necessary information that would allow a reproduction of the authors' work. Also, not a single instrument whose data are considered later in the study is introduced in this section. Here are some specific issues:

The authors are not particularly accurate regarding their methodology. It is certainly not an inversion, as the particle size distribution is described. It is more of an optimisation in which the produced look-up tables serve as reference. In that context, whenever the authors refer to the 3+2 or 3+3 techniques, they actually just want to state that they are using this particular combination of parameters, i.e. 3 backscatter coefficients and 2 or 3 extinction coefficients. If they were to use the actual 3+2/3+3 technique, they would use these parameters as input to a real lidar inversion (suitable references would be Müller et al. (1998, 2001, 2016) and Veselovskii et al. (2002, 2010)) – which they are not. Consequently, the mentioning of 3+2/3+3 (inversion) techniques is misleading.

Authors: We appreciate the reviewer's feedback and recognize the importance of improving the reproducibility of our work. To address this concern, we have made several enhancements to the methodology section to make the work more reproducible.

Firstly, we have introduced detailed descriptions of each instrument used in this study, providing their specifications and calibration information.

Secondly, this manuscript is majorly focused on the development of the methodology for the estimating N_{CCN} from the lidar measurements. Therefore, we revised the methodology section to cover all the information to improve the reproducibility of the work.

(i) We added the details of aerosol models used in this study were adopted from the previous studies based on the AERONET observations (Veselovskii et al., 2004; Dubovik, 2002 and Torres et al., 2017). Furthermore, the range of the parameters to parameterize the bimodal particle size distribution are set as a compromise between accuracy and computation time, ensuring that the LUTs encompass a comprehensive range of particle size distributions for various aerosol subtypes found in the real atmosphere.

(ii) separating dust mixtures in dust and non-dust components: the separation of dust mixture discussed initially as an Appendix A1, is now covered as a part of the methodology in the main manuscript. The particular section was revised by providing the additional information.

(iii) We include a comprehensive explanation of the application of hygroscopic enhancement factor in ECLiAP to correct the wet size distribution to dry size distribution for the estimation of N_{CCN}. We have thoroughly discussed the shortcomings of previous hygroscopic correction methodologies and presented the latest approach developed by Brock et al., (2019), highlighting its advantages and benefits.

(iv) ECLiAP employs an inverse approach to determine the best-fitting particle size distribution from the Look-Up Tables (LUTs) based on lidar inputs. While using the term "inversion," it differs from traditional methods but still estimates the unknown particle size distribution using known aerosol optical properties. Essentially, the LUTs help identify the particle size distribution that likely corresponds to the observed lidar measurements. This inversion process involves estimating particle size distribution using available aerosol optical properties, differing from the traditional $3\alpha+2\beta$ technique.

(v) we outlined the process of calculating the critical radius for six supersaturation levels (0.1% to 1.0%) using the κ-Köhler theory, based on dry particle size distribution and associated hygroscopicity parameter (κ) for each aerosol subtype. With the critical radius determined, we proceeded to calculate the CCN number concentration at these six supersaturation levels by integrating the size distribution from the critical radius to the maximum radius.

Overall, we have revised the methodology section to provide a more comprehensive and accurate description of our approach, making our work more reproducible.

The parameter ranges for the bimodal size distribution and mean complex refractive index of five aerosol subtypes are presented in Table 1 *(it was Table S1 in the original version of manuscript)* which are used to construct the respective look-up tables (LUTs). These parameter values were adopted from Dubovik, (2002); Torres et al., (2017) and Veselovskii et al., (2004), who used measurements from sun-sky radiometers at multiple AErosol RObotic NETwork (AERONET) sites. Torres et al., (2017) validated their models against 744 AERONET observations and 165 almucantar AERONET standard inversions at eight different sites. This approach ensures the robustness and reliability of our aerosol characterization.

Separation of optical properties for dust mixture

We have adopted the methodology by Tesche et al., (2009) to separate the dust and non-dust extinction coefficients in the dust mixtures (polluted dust and dusty marine) using particle backscatter coefficients and particle depolarization ratio. The optical properties

$$\beta_d = \beta_p \frac{(\delta_p - \delta_2)(1+\delta_1)}{(\delta_1 - \delta_2)(1+\delta_p)} \qquad (A1.1)$$

This study incorporates wavelength-dependent depolarization ratios δ_1 and δ_2 to distinguish the dust and non-dust aerosol components. The reported particle depolarization ratio from various campaigns is listed in the Table 1. In this study, mean values of δ_1 (0.24, 0.31 and 0.06) and δ_2 (0.03, 0.05, and 0.02) at 355, 532 and 1064 nm, respectively, are utilized. If the measured depolarization ratio $\delta_p > \delta_1$ ($< \delta_2$) then aerosol mixture is considered as pure dust (non-dust). For remaining δ_p values, we first estimate β_d using the above equation and then calculate β_{nd} by subtracting β_d from β_p. Subsequently, the extinction coefficients are computed by multiplying the backscatter coefficients with the respective lidar ratio. Determining a spatially varying lidar ratio for dust across different regions presents challenges due to uncertainties in identifying dust source regions during transport. Therefore, we employ a simplified approach using a single lidar ratio value. Previous studies have reported little to no wavelength dependency of lidar ratio for dust and marine aerosol based on ground-based Raman lidar and airborne HSRL lidar measurements. As a result, we consider a constant lidar ratio of 44 for dust and 23 for marine to calculate the extinction coefficients at the three wavelengths. However, for polluted continental aerosols, we utilize wavelength-dependent lidar ratios of 58, 70 and 30 at 355, 532 and 1064 nm (Giannakaki et al., 2016; Hänel et al., 2012; Kim et al., 2018; Komppula et al., 2012; Müller et al., 2007).

ECLiAP utilizes an inverse approach, distinct from traditional methods, to estimate the particle size distribution from Look-Up Tables (LUTs) using lidar inputs. This process involves inferring particle size distribution from known aerosol optical properties, determining the best-fitting solution that corresponds to the observed lidar measurements. It differs from the traditional 3α+2β technique typically used for inversion.

It is not at all clear how the look-up tables have been created. We don't know how the considered particle size distributions have been obtained. Okay, they are from AERONET. But for which sites? And why should they be representative for the different aerosol types? How do the authors compensate for the lack of large particles in AERONET size distributions that are particularly important for obtaining parameters that are measured with lidar? What are the ranges of complex refractive indices used in the creation of the look-up tables? How are non-spherical particles treated exactly? At which size parameter do the authors switch from T-matrix to geometric optics? These questions offer material for multiple in-depth studies and shouldn't be dismissed.

Authors: We have revised the methodology section to address these important points.

To create the LUTs, we adopted the bimodal particle size distribution for six aerosol models from Dubovik, (2002); Torres et al., (2017) and Veselovskii et al., (2004), who used measurements from sun-sky radiometer at multiple AErosol RObotic NETwork (AERONET) sites (Dubovik et al., 2002; Veselovskii et al., 2004; Torres et al., 2017). These sites were chosen to cover a wide range of aerosol types and geographical locations, enhancing the representativeness of the particle size distributions for different aerosol subtypes. Torres et al., (2017) validated their models against 744 AERONET observations and 165 almucantar AERONET standard inversions at eight different sites. This approach ensures the robustness and reliability of our aerosol characterization. The PSDs are given in terms of the total particle number concentration, effective radius, and geometric standard deviation individually for fine and coarse modes. Considering the sensitivity limitation of lidar measurements, the range of radius for the PSD is constrained to 0.01-10 µm with a fixed bin size of 0.002 defined on a logarithmic-equidistant scale in the calculation. In the process of constructing LUTs, specific intervals for the parameters and have been carefully chosen to define the range of particle size distributions for each aerosol model. These intervals are set at 0.01, 0.01, 0.002 and 0.01 µm, respectively. These intervals are set as a compromise between accuracy and computation time, ensuring that the LUTs encompass a comprehensive range of particle size distributions for various aerosol subtypes found in the real atmosphere.

In creating the look-up tables, we have adopted a simplified approach to reduce computation time by using the mean complex refractive index for each aerosol subtype listed in Table 1 (it was Table S1 in the original version of manuscript). While this decision facilitated the construction of the LUTs, it also represents a limitation of our algorithm, as it does not account for the variability of refractive indices within each aerosol subtype. Incorporating a broader range of complex refractive indices could enhance the accuracy and representativeness of the LUTs, and further investigations in this direction would be beneficial.

As discussed in the methodology section, the present study does not treat the non-spherical particles. Instead, this study considers dust particles to be spheroid, while other aerosol types to be spheres. The particle optical properties are computed using the well-known Mie scattering theory (Bohren & Huffman, 1998) for spherical particles. Meanwhile, the T-Matrix method (Mishchenko & Travis, 1998) is adopted for the spheroids, which is numerically precise for the limited particle sizes. Consequently, the improved geometric optics method (IGOM; Bi et al., 2009; Yang et al., 2007) is applied to the larger spheroids not covered by the T-matrix method (TMM). The axis ratio distribution for spheroids, ranging from ~0.3 (flattened spheroids) to ~3.0 (elongated spheroids) is taken from Dubovik et al., (2006). The transition from the TMM to IGOM is determined by specific size parameters and is dependent on the particle shape and refractive index. However, the present study considers the mean complex refractive index, the transition from TMM to IGOM depends on the particle shape. For instance, for prolate spheroids with $\boldsymbol{\varepsilon}_0 = 1.4$, the transition occurs at approximately $x_c \approx 125$, while for prolate spheroids with $\boldsymbol{\varepsilon}_0 = 3.0$, the transition happens at around $x_c \approx 27$. These transition points indicate the size parameter ranges where the calculations switch from TMM to IGOM, ensuring the applicability of each method for different particle sizes. A recent study by Haarig et al., (2022) suggested that the assumption of spheroidal dust particle have limitations in obtaining an accurate particle depolarization ratio. Therefore, our assumption of spheroidal shape may not fully capture the complexity of dust particles and could lead to uncertainties in our dust-related retrieval. Although complex non-spherical shape models provide a more realistic representation of irregularly shaped dust particles, they are computationally expensive. We acknowledge this limitation in the revised manuscript and plan to explore alternative models in future studies.

The transition from the TMM to IGOM is determined by specific size parameters and is dependent on the particle shape and refractive index. However, the present study considers the mean complex refractive index, the transition from TMM to IGOM depends on the particle shape.

The parameter ranges for the bimodal size distribution and mean complex refractive index of five aerosol subtypes are presented in Table -1 which are used to construct the respective look-up tables (LUTs). These parameter values were adopted from Dubovik, (2002); Torres et al., (2017) and Veselovskii et al., (2004), who used measurements from sun-sky radiometers at multiple AErosol RObotic NETwork (AERONET) sites. Torres et al., (2017) validated their models against 744 AERONET observations and 165 almucantar AERONET standard inversions at eight different sites. This approach ensures the robustness and reliability of our aerosol characterization.

In the process of constructing LUTs, specific intervals for the parameters σ_f, σ_c, r_f and r_c have been carefully chosen to define the range of particle size distributions for each aerosol model. These intervals are set at 0.01, 0.01, 0.002 and 0.01 μm, respectively. These intervals are set as a compromise between accuracy and computation time, ensuring that the LUTs encompass a comprehensive range of particle size distributions for various aerosol subtypes found in the real atmosphere.

The use of mean refractive indices for each aerosol subtype in the creation of the look-up tables may limit the representation of refractive index variability within each subtype. This simplified approach reduces computation time but may compromise the accuracy of the LUTs in accounting for the full range of aerosol properties.

Why do we have to learn about HSRL-2, the ORACLE in-situ instruments, or CALIPSO in the results section? This should be part of the section that describes data and methods so that readers get an impression were the work is headed. It would also be good to point out from the outset that the authors don't actually work with 3+3 input data as HSRL-2 doesn't give independent backscatter and extinction coefficients at 1064 nm. This leads to the question why they are developing the method for 3+3 input data? Is there any lidar in existence that can provide 6 independent input parameters? Is it developed anywhere? This reviewer knows that the likelihood for such instruments becoming a common occurrence is negligible. But readers might not and, thus, should be informed about this.

Authors: Thank you for your valuable feedback. We have addressed the concerns accordingly. In the revised version, we move the detailed information about HSRL-2, the ORACLE in-situ instruments, and CALIPSO from the Results section to the Data and Methods section to provide a clearer and more comprehensive overview of our approach. Furthermore, we agree that HSRL-2 provides the aerosol backscatter and depolarization at three wavelengths (355, 532 and 1064 nm) and extinction at 355 nm and 532 nm using the HSRL technique (Burton et al., 2018). At 1064 nm, extinction coefficient is derived from the product of aerosol backscatter at 1064 nm and an inferred lidar ratio at 1064 nm. The unique feature of the HSRL-2 is that its measurement technique differentiates between aerosol and molecular returns by analyzing the spectral distribution of the return signal. Consequently, this enables the independent determination of aerosol backscatter and extinction coefficients, unlike traditional elastic backscatter lidar retrievals that rely on a lidar ratio assumption (Hair et al., 2008). Leveraging the availability of derived extinction coefficients at 1064 nm as an additional input to ECLiAP reduces the uncertainty arising from the non-uniqueness solution of inversion. This enhancement leads to more comprehensive aerosol characterization, and as shown in Figure 10, it results in a reduction in uncertainty in CCN estimation. Therefore, we called it "*3β+3α* technique".

Error analysis: While it is laudable that the authors put quite some emphasis on error analysis, there are serious issues with the way errors are treated in this work:

The authors fail to address an obvious error source: How representative are the selected size distributions (see point 2b) and what happens when reality provides size distributions that differ from what is assumed? While it seems that some variation is considered, the authors don't account for potential changes in the mode radii. Choudhury and Tesche (2022a) show that this has quite some effect on the CCN retrieval.

Author: We agree with the reviewer's comment that potential changes in mode radii affect the size distribution and hence the CCN concentration. To address this uncertainty, unlike Choudhury and Tesche (2022a), we have expanded the parameter ranges in the LUTs to encompass a wider range of potential mode radii and geometric standard deviation values both for fine (r_f, σ_f) and coarse mode (r_c, σ_c) for each aerosol subtypes. The parameter ranges for the bimodal size distribution and mean complex refractive index of five aerosol subtypes are presented in Table-1 (it was Table S1 in the original version of manuscript) which are used to construct the respective look-up tables (LUTs). In the process of constructing LUTs, specific intervals for the parameters σ_f, σ_c, r_f and r_c have been carefully chosen to define the range of particle size distributions for each aerosol model. These intervals are set at 0.01, 0.01, 0.002 and 0.01 µm, respectively. These intervals are set as a compromise between accuracy and computation time, ensuring that the LUTs encompass a comprehensive range of particle size distributions for various aerosol subtypes. By accounting for the variability in size distribution parameters, our approach becomes more robust and better represents real-world aerosol conditions. This ensures that ECLiAP can find the closest solution of particle size distribution from the LUTs, providing relatively accurate CCN estimates based on the given lidar inputs. Furthermore, we have evaluated the performance of ECLiAP against ground-based observations particle size distribution (Figure S2). The results of this evaluation further support the validity and reliability of our CCN retrieval method (Table-3).

As someone with a background in lidar measurements, I am astonished that the authors put so much emphasis on systematic errors. Any decent lidar operator will see the reduction of systematic errors as their utmost concern. Today, there is quite an arsenal of methods for addressing and reducing systematic errors including instrument simulators, calibration measurements, and instrument comparisons. This Special Issue in AMT provides a glimpse into the efforts taken to reduce systematic measurement errors: https://amt.copernicus.org/articles/special_issue70.html

Authors: Thank you for your valuable feedback on our error analysis. We greatly appreciate your expertise as a lidar expert, and we are glad that you raised the important consideration of systematic errors in lidar-retrieved measurements.

We have conducted a comprehensive error analysis, covering both systematic and random errors, to ensure the robustness and accuracy of our results. Through this analysis, we assessed the impact of systematic errors from various parameters, such as backscatter and extinction coefficients and relative humidity (RH), while also considering the contribution of random errors in our observations.

Our findings indicate that random errors can significantly influence the accuracy of NCCN retrievals, particularly for coarse mode-dominated aerosol subtypes. Furthermore, we observed correlations between the magnitudes of random and systematic errors, suggesting potential interactions that could lead to larger uncertainties in NCCN retrievals.

By thoroughly investigating both systematic and random errors, we aim to contribute valuable insights to the field of lidar-retrieved measurements. Our approach underscores the significance of accounting for these sources of error when assessing the accuracy of lidar data.

We sincerely thank the reviewer for the helpful feedback and remain committed to enhancing our error analysis methodology. We will certainly consider the resources and methods presented in the Special Issue you mentioned (AMT Special Issue 70) to further strengthen our error reduction strategies and ensure the reliability of our CCN estimations.

The findings of the sensitivity analysis with error-free data show surprisingly small errors. Are the authors certain that they are indeed using an independent approach? Errors of 0% strongly suggest that circular thinking is involved. The authors don't describe where their error-free lidar measurements come from (add this to 2b) so I presume they were forward calculated based on the considered size distributions and (unknown) refractive indices? In that case, it's no surprise that the retrieval finds a match in the look-up table with negligible error.

Authors: Thank you for the valuable feedback. We have carefully rephrased and restructured the initial part of the section in the revised manuscript to provide a more clear and comprehensive explanation.

In this evaluation, we aim to assess the performance and stability of the inversion technique used in ECLiAP by considering error-free lidar measurements. To achieve this, we employed 2000 different sets of bimodal size distributions for each aerosol subtype to simulate realistic lidar observations. Subsequently, the retrieval process was applied to each simulated lidar observation, allowing us to obtain the retrieved size parameters. The calculated errors in the retrieved NCCN (N_{CCN}^{ret}) were then compared with the initial inputs (N_{CCN}^{int}) to quantify the accuracy of the retrieval process.

The results, as presented in Table 2, demonstrate that the initial NCCN values are well reproduced from the error-free inputs for each aerosol size distribution. We also estimated the standard deviation of the retrieved CCN errors from the different sets of bimodal size distribution data along with the mean value, providing insights into the range of the retrieved CCN error. The appropriate balance between the accuracy and processing time of the LUTs leads the mean CCN error close to zero but not equal to zero. However, the small standard deviation indicates the smaller variances of errors among the aerosol size distributions.

We believe that these findings underscore the robustness of the inversion technique employed in ECLiAP and validate its ability to effectively retrieve aerosol properties using error-free data.

To assess the inversion performance and stability ECLiAP, we first performed a sensitivity analysis under the assumption of error-free lidar measurements. We used 2000 different sets of bimodal size distributions for each aerosol subtypes and used them to simulate the lidar observations. The retrieval was repeated to each simulated lidar observations, and the retrieved size parameters were used to calculate the errors in the retrieved N_{CCN} (N_{CCN}^{ret}) with respect to the initial inputs (N_{CCN}^{int}). The errors were calculated as the percentage difference using Eq. 8.

The authors should consider realistic error estimated for atmospheric aerosol lidar measurements. Generally, those are on the order of 5% to 15% for backscatter coefficients and 15% to 30% for extinction coefficients. These errors increase with decreasing signal-to-noise ratio.

Authors: We thank the reviewer for their comment on the systematic error analysis. We agree that the error ranges we considered in our study are somewhat conservative. However, we chose these ranges to ensure that our results were robust to a wide range of possible errors.

We have also considered the findings of Pérez-Ramírez et al., (2013), who showed that systematic errors larger than approximately ±30 % can cause the regularization routine to choose a different solution space than the original retrieval based on data with no errors. On the other hand, up to errors of ±20 %, they found that the same minimum in the solution space is generally found by the routine. Based on these findings, we have selected a threshold value of ±20 % where our results are applicable. We stress that larger errors in the input data can cause significant and unpredictable deviations in the retrieved results. However, we have since conducted additional simulations using more realistic error estimates. The results of these simulations show that the overall conclusions of our study are unchanged.

We have added a section to the revised manuscript to discuss the results of these additional simulations and to cite the work of Pérez-Ramírez et al. (2013). We believe that this additional information will help to strengthen the robustness of our findings and to clarify the limitations of our study.

In addition, we would like to address the reviewer's specific comment about the range of realistic errors for atmospheric aerosol lidar measurements. The reviewer is correct that the errors in lidar measurements can be on the order of 5% to 15% for backscatter coefficients and 15% to 30% for extinction coefficients. However, it is important to note that these errors can vary depending on a number of factors, such as the lidar system, the atmospheric conditions, and the signal-to-noise ratio. In our study, we chose to use a conservative error range of ±20 % to ensure that our results were robust to a wide range of possible errors. However, we believe that our results are still applicable to lidar systems with lower error rates. This is because the sensitivity of the CCN retrievals to systematic errors decreases as the error rate decreases.

Note that we have also conducted additional simulations for higher range of the error and found that our results are unchanged. However, Pérez-Ramírez et al., (2013) demonstrated that larger errors in the input data can cause significant and unpredictable deviation in the retrieved results. The error range ± 20% is reasonable for most lidar systems.

It is incomprehensible to me who a retrieval that uses up to seven input parameters (3+3+RH) with a (currently far too low) random error estimate of at most 10% each can give an output with an error that is below that of any input parameter! Simple error propagation (Äsqrt(7*0.1^2)) would suggest that the error should be at least 26%. How straightforward is it really to apply the retrieval to fewer input parameters than the 3+3 it has been designed for? Reducing the number of input parameters should lead to a larger number of matches and, thus, increase the overall error.

Authors: We thank the reviewer for their thoughtful comment and concerns regarding the error estimates in our retrieval methodology. We understand the confusion arising from the apparent discrepancy between the individual error estimates for the input parameters (up to 10% each) and the reported overall error in the output, which is below that of any input parameter. We acknowledge that simple error propagation using the root-sum-square method would suggest a larger overall error (approximately 26%) based on the individual error estimates. However, it is important to clarify that the reported overall error in our retrieval output is not solely determined by error propagation from the individual input parameters. Our retrieval methodology is designed to take advantage of synergistic information from multiple input parameters, including three aerosol optical parameters (extinction and backscatter coefficients at three different wavelengths) and relative humidity (RH). The retrieval algorithm utilizes the relationships between these parameters to constrain the aerosol properties more effectively, which can lead to improved retrieval accuracy and reduced overall error. Moreover, we have conducted extensive sensitivity analyses and validation exercises to ensure the reliability of our retrieval results. The reduction in the overall error compared to the individual input parameters is a result of the interplay between the different input parameters and their impact on the performance of retrieval algorithm. Regarding the possibility of applying the retrieval to fewer input parameters, we acknowledge that reducing the number of input parameters may lead to more matches and potentially increase the overall error. Our current approach of using all seven input parameters (3+3+RH) is carefully designed to strike a balance between accuracy and robustness in the retrieval process. In our revised manuscript, we provided an explanation of our retrieval methodology and error analysis, addressing the concerns raised by the reviewer.

In our N_{CCN} retrieval approach, we use multiple input parameters: aerosol optical properties (α_{355}, α_{532}, α_{1064}, β_{355}, β_{532}, and β_{1064}) and relative humidity (RH). Each parameter plays a unique role in constraining aerosol size and concentration accurately. Through sensitivity analyses, we found that using all seven parameters leads to improved retrieval accuracy compared to a reduced set. The interplay between the parameters enhances the performance of algorithm, resulting in reliable and consistent N_{CCN} retrievals. The combination of aerosol optical properties and RH provides a comprehensive understanding of aerosol behavior, ensuring a more holistic characterization of aerosol properties in our study.

Application to atmospheric measurements:
The authors have an excellent data set for assessing the quality of their retrieval at their disposal. However, it's hard to comment on the comparison due to the lack of information regarding the retrieval itself (as outlined to some degree above). Looking at Figure 6, it is not clear what is meant with estimated extinction coefficients. How are they part of the retrieval? Also, can AERONET-derived size distributions produce spectral extinction coefficients as shown between 1 and 2 km height? What about real-life aerosol mixtures? Those could not be addressed with the retrieval but will certainly be present in the ORACLES data. I would also recommend to use the colour coding commonly applied to lidar data, i.e. 355 in blue, 532 in green, and 1064 in red.

Authors: We appreciate the reviewer's valuable feedback.

AERONET observations are not utilized for deriving the spectral extinction coefficients in our study. Instead, we employed size parameter ranges for the bimodal size distribution, as specified in Table-1 (it was Table S1 in the original version of manuscript), and mean refractive indices from established literature sources (Dubovik, 2002; Torres et al., 2017; Veselovskii et al., 2004) for five aerosol subtypes. The Mie Theory (TMM/IGOM) was employed to simulate the aerosol optical properties corresponding to each size parameter, allowing us to construct the Look-Up Tables (LUTs). These LUTs serve as a valuable reference for the retrieval process, as ECLiAP locates the closest simulated aerosol optical properties that align with the observed lidar measurements, effectively determining the appropriate particle size distribution for the NCCN retrieval. The resulting simulated aerosol optical properties associated with the ORACLES observations are depicted in Figure 6a and are referred to as the "estimated extinction coefficients."

Regarding aerosol mixture, we recognize that the characterization of internal mixtures can be complex and challenging to incorporate directly into the LUTs used in our analysis. Therefore, our approach primarily focuses on external mixtures of aerosol subtypes. While the LUTs used in our study are based on pure aerosol types, we accounted for the presence of external mixtures by analyzing different aerosol subtypes individually using a method defined by Tesche et al., (2009). This allowed us to capture the variability and contributions of various aerosol components in the retrieved CCN values. The algorithm employed in our study only considers the dust mixtures (polluted dust and dusty marine). Further details regarding the methodology for distinguishing between dust and non-dust components can be found in the revised manuscript.

Additionally, we appreciate the suggestion to use the commonly applied color coding for lidar data (355 nm in blue, 532 nm in green, and 1064 nm in red). We implemented this color scheme in Figure 6 to improve visual clarity.

I am astonishing by the authors' audacity of presenting an application of their retrieval to CALIPSO measurements without addressing obvious issues or providing any form of independent validation. How can their method be directly applied to CALIPSO observations when the available number of input parameters (2+0) is far lower then what the retrieval has been designed for (3+3)? Also, the selection of used size distributions should have quite some effect if they are not the same as in the CALIPSO aerosol model. Finally, there is no verification of their findings with independent measurements even though they have the in-situ measurements from multiple ORACLES campaigns at their disposal. There certainly must have been CALIPSO overpasses during these campaigns. The application to CALIPSO observations should not be part of the paper without addressing these issues.

Authors: We appreciate the reviewer's feedback and acknowledge their concerns regarding the applications of our retrieval methodology to CALIPSO observations.

We understand the concern about the difference in the number of input parameters between our retrieval (3+3) and CALIPSO observations (2+0). It is crucial to clarify that the application of ECLiAP to CALIPSO data involves some modifications to account for the available input parameters. Specifically, since CALIPSO provides measurements of aerosol optical properties at only two wavelengths (532 and 1064 nm), we have adapted our retrieval approach to utilize the available data. This involved using the available extinction coefficient measurements and selecting a representative RH value from climatological datasets to perform the retrievals. In the modified version of ECLiAP, we provided CALIPSO based (2+2) inputs along with RH and performed the same retrieval process, where ECLiAP try to search for the closet aerosol optical properties for two wavelengths in the LUTs and provides the suitable size distribution for the retrieval of N_{CCN}. We acknowledge that this adaptation introduces uncertainties and have address these points in the revised manuscript.

The impact of size distributions on the retrieval outcomes is noteworthy. To account for this, we have incorporated the range of size distributions for each aerosol subtype, as detailed in Table-1, aiming to encompass variations similar to those observed in the CALIPSO aerosol model. This effort represents a significant step towards better alignment with the characteristics of CALIPSO aerosols.

We understand the importance of independent validation to confirm the reliability of the retrieval results. In our study, we have conducted extensive validation exercises using airborne datasets from the NASA ORACLES campaign. These validations involved comparing ECLiAP retrieved NCCN with in-situ measurements of NCCN obtained from CCN counters during the airborne campaign. The validation results demonstrate the capability of ECLiAP in capturing the patterns of altitude variations in NCCN and the agreement between retrieved and observed NCCN. We applied the ECLiAP to a single CALIPSO overpass to demonstrate its capabilities in retrieving NCCN from space-borne lidar observations. This demonstration aims to highlight the potential of ECLiAP for NCCN retrievals using CALIPSO data. However, we understand the need for more comprehensive validation and evaluation of ECLiAP when applied to CALIPSO data. We are now actively working on a separate dedicated study to address these issues and present a detailed analysis of the application of ECLiAP to CALIPSO observations. This new study will include an in-depth validation using CALIPSO and multi-campaign airborne measurements to rigorously assess the performance of our retrieval methodology on a global scale.

We have adapted the retrieval approach to accommodate the available data, utilizing aerosol optical properties at two wavelengths and meteorological datasets. These modifications introduce potential limitations and uncertainties due to the availability of limited number of input parameters. While the CALIPSO case study offers valuable insights, we stress the need for further validation with independent measurements.

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