02 August 2023

Dr. Zhanqing Li
Senior Editor, Atmospheric Chemistry and Physics

Dear Dr. Li,

I am herewith submitting our manuscript titled "A Remote Sensing Algorithm for the Quantifying Vertically-Resolved Cloud Condensation Nuclei Number Concentrations from Spaceborne Lidar Measurements" for consideration as a Research Article in the Atmospheric Chemistry and Physics.

This revised manuscript is a resubmission of a previously submitted paper to ACP (acp-2022-547). We received your decision and reviews on 16 December 2022. Just as a way of information, it has taken me a while to prepare the responses and the revised manuscript as I was dealing with persistent health issues this year and travelling a lot for the medical treatments. However, I and my coauthors are delighted to be submitting this revised manuscript for your consideration.

Our manuscript focuses on developing a remote sensing-based analytical algorithm (known as "ECLiAP") to retrieve cloud condensation nuclei (CCN) number concentration using spaceborne lidar measurements. This research work is motivated by the challenges identified by the Intergovernmental Panel on Climate Change (IPCC) in predicting climate change. The major challenge is the missing measurements of the vertically resolved CCN number concentration at a global scale for improving the understanding of processes associated with aerosol-cloud interactions (ACI).

We received comments from two anonymous reviewers and have addressed each and every comment in our point-wise response. We have provided multiple lines of evidence formulating the novelty of our algorithm and how that improves the retrieval of aerosol microphysical properties and CCN number concentrations compared to the existing techniques.

None of the material in the submitted manuscript has been published or is under consideration for publication elsewhere.

All data underlying our study are available in the public domain. We have provided links to the various data access portals in supplementary materials for the ground-based observations, airborne measurements, and satellite datasets used in the submitted manuscript.

Thank you for your consideration.

Sincerely,

Dr. Piyushkumar N. Patel

Remote Sensing Scientist (Consulting)
MethaneSAT LLC,
Austin, TX, USA
piyushether@gmail.com

Files uploaded: (i) manuscript file, (ii) point-wise response to reviewers, (iii) manuscript file with track changes to indicate where revisions occurred relative to the previously-submitted manuscript.

Comment on acp-2022-547
Anonymous Referee #2

General

The manuscript discusses a new lidar approach to estimate CCN concentrations. It is a useful contribution to the lidar literature. However, since a new lidar method is introduced and the manuscript deals with all details of this method including an uncertainty analysis, AMT would be more appropriate.

Minor revisions are necessary.

We thank the reviewer for taking the time to review our paper and providing constructive comments. We have addressed each of the reviewer's comments and included below a point-wise response, as well as included highlighted changes in the revised manuscript (main manuscript and supplementary material files). We hope our extensive comments in the initial review illustrate our genuine efforts to engage constructively with all review comments. The responses to the reviewer's comment are indicated in blue, while the modifications made in the revised manuscript are highlighted in red.

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.

Detailed comments:

p3, l97: Why do we need to know the particle size distribution? CCN is just the overalll particle number concentration (integral over all sizes)!
Authors: While it is true that CCN is often characterized by the overall particle number concentration, understanding the particle size distribution is crucial for several reasons:

1. **Activation Efficiency**: Activation efficiency refers to the likelihood of a particle serving as a CCN under certain supersaturation and participating in cloud formation. The activation efficiency of particles as CCN strongly depends on the interplay between the size of the particles and their chemical properties (*Farmer et al., 2015; Patel and Jiang, 2021*). The activation of aerosol particles occurs when the supersaturation within an air parcel exceeds a critical value, which is determined by the particle size and composition. This threshold refers to a critical radius. The critical radius is a fundamental concept in CCN studies. It represents the minimum size of a particle that can activate as a CCN and form a cloud droplet under a given supersaturation condition. Larger particles have a higher probability of activation

because they possess a larger surface area onto which water vapor can condense. As particle size decreases below the critical radius, activation becomes less probable, and smaller particles may require higher supersaturations to activate.

2. **Aerosol Composition**: The particle size distribution of aerosol is closely linked to their chemical composition. Certain chemical species and sources may preferentially contribute to specific size ranges. By investigating the particle size distribution, we can gain valuable information about the origins and properties of aerosol particles, enabling a better understanding of their CCN activity.

3. **Atmospheric Processes**: The behavior of aerosol particles, including their transport, transformation, and interactions with clouds, is influenced by their size. By understanding the particle size distribution, we can estimate the range of particle sizes present in the atmosphere and assess their activation potential. This knowledge is essential for predicting aerosol-CCN activity, cloud formation, understanding the aerosol indirect effect on climate, and improving cloud parameterizations in climate models.

This means that CCN number concentrations are not integral over all sizes of particles, but they are integral to the particle sizes ranging from the critical radius to the maximum. The same has been calculated using Eq. 10 in the manuscript. Therefore, the particle size distribution is a critical parameter for calculating the CCN number concentration. Moreover, the particle size distribution provides insights into the variability and heterogeneity of aerosol particles. It helps identify the dominant size ranges contributing to the CCN population, thereby enabling a better understanding of the sources, transport, and transformation processes affecting cloud properties. Therefore, considering the particle size distribution is crucial for a comprehensive understanding of CCN behavior, including the activation efficiency and the critical radius.

Regarding aerosol type, to my opinion the separation into dust and non-dust is sufficient. Size plays a strong role, but chemical composition a minor role…? Maybe discuss that a bit more.
Authors: In our study, we have categorized aerosols into dust and non-dust aerosols as a simplified approach to capturing the major sources of aerosol particles. This categorization allows us to assess the general influence of dust versus non-dust particles on cloud condensation nuclei (CCN) activity. However, we acknowledge the importance of both size and chemical composition in understanding aerosol behavior to act as CCN and their impacts on cloud processes.

While size plays a dominant role in determining aerosol properties, it is also essential to consider the influence of chemical composition. Chemical composition affects the hygroscopicity of aerosols, their ability to uptake water, and their CCN activity. Different chemical species exhibit varying degrees of water absorption and can undergo chemical transformations within the atmosphere, impacting their CCN potential. However, a study by *Dusek et al. (2006)* found that the cloud-nucleating ability of particles was primarily determined by their size distribution rather than their chemical composition. Larger particles with diameters between 100 and 200 nm were more effective at activating and forming cloud droplets than smaller particles with diameters less than 50 nm, even when the smaller particles had a more hygroscopic chemical composition. This is because larger particles can provide more surface area for water vapor to condense, irrespective of its hygroscopicity, and form cloud droplets. However, the effect of chemical composition on CCN activation is limited by the availability of hygroscopic material (i.e., organic matter or sulfate) in the aerosol particles, which are more effective at attracting water vapor and forming

cloud droplets. This suggests that the cloud-nucleating ability of particle are mainly controlled by the size of a particle, followed by the chemical composition.

It is important to note that the relative importance of particle size and chemical composition in CCN activation may vary depending on the specific environmental conditions and the types of aerosol particles present in the atmosphere. Nevertheless, a recent study by *Patel and Jiang et al. (2021)* analyzed the particle size distribution, chemical composition, and nucleating ability to act as CCN at various supersaturation for different airmass clusters using ground-based measurements. The study found that particle size was the most critical factor influencing the ability of aerosols to activate, whereas the effect of chemical composition was secondary. It also suggests that chemical composition and mixing state information are more crucial at lower supersaturation, whereas most particles become activated at higher supersaturation regardless of their chemical composition. Moreover, determining the accurate chemical composition of aerosol particles from space remains an unsolved challenge.

Therefore, in the present study, the particle size is the primary focus, followed by the chemical composition (described as a hygroscopicity) for the estimation of CCN number concentration. We recognize the importance of chemical composition in aerosol-cloud interactions, and while it is not extensively discussed in this study, the simplified categorization allows us to provide insights into the overall behavior of aerosols as CCN.

Thank you for raising this point, and we appreciate your valuable feedback.

What I learned from all these complex lidar inversion papers since Mueller et al. (1999) and Veseolovskii et al. (2002) is…. that it is impossible to retrieve the particle size distribution with good accuracy, even from a set of 3 backscatter and 3 extinction coefficients. Furthermore, the retrieved particle number concentration always shows the largest uncertainty (close to 100%). Now you use Look Up Tables. How can you have uncertainties < 50% when using a much more simple approach than these sophisticated inversion techniques? Please comment on that in the manuscript!
Authors: Thank you for your comment regarding the retrieval of particle size distribution and the associated uncertainties in lidar inversion techniques. We appreciate your insight into the challenges and limitations of these complex inversion methods.

Our study employed a Look Up Table (LUT) approach to estimate the particle size distribution. While LUT-based methods are more straightforward than sophisticated inversion techniques, they provide a practical and efficient means of evaluating particle properties, including size distribution, in real-time applications.

We acknowledge that the accuracy of particle size distribution retrieval from lidar measurements can be challenging, and previous studies have reported limitations and uncertainties in these retrievals (*Mueller et al., 1999; Veseolovskii et al., 2002*). The complexity arises from the non-uniqueness of the inverse problem (*Chemyakin et al., 2016*), where multiple combinations of particle size distributions can produce similar backscatter and extinction coefficients. By providing instances, *Chemyakin et al., (2016)* demonstrated that the particle size distribution from the inversion solution is not unique, and the LUT approach can reduce the uncertainty associated with non-uniqueness.

Our LUT approach addresses this challenge by pre-computing a comprehensive set of forward model simulations using a range of particle size distributions and associated optical properties. We can identify the particle size distribution that best matches the observations by comparing the measured backscatter and extinction coefficients with the LUT. The LUT approach offered in our study partially compensates for the difficulties of the inversion solution. While the LUT approach simplifies the inversion process, it still requires careful calibration and validation to ensure accurate results. To assess the uncertainties in our approach, we have performed rigorous exercises to quantify both systematic and random errors. Incorporating an additional measurement of the extinction coefficient at 1064 nm enhances the effectiveness of our algorithm in accurately determining the closest solution for the particle size distribution based on the provided lidar measurements. This further reduce the uncertainty arise due to the non-uniqueness solution of inversion. Therefore, we called it "$3\beta+3\alpha$ technique". The reduction in uncertainty is clearly reflected in the CCN estimation (as seen Figure 10).

In summary, while we recognize the complexities and limitations of particle size distribution retrieval from lidar measurements, our LUT approach offers a practical and efficient means of estimating these properties in real-time applications. We have included a discussion of the uncertainties associated with our retrieval method and the potential sources of error to provide a comprehensive understanding of the limitations of our study.

Thank you for raising this important point, and we appreciate the opportunity to clarify our approach and address the concerns regarding uncertainties in the manuscript.

P3, l101: Why do you mention the Rosenfeld et al (2016) method in this lidar paper?
Authors: The main objective of our study is to estimate the CCN number concentration from satellite measurements. While we focus on lidar-based retrieval of CCN, it is essential to acknowledge the broader context of CCN estimation from satellite observations.

The Rosenfeld et al. (2016) study is significant in the field of CCN research as it explores the potential of using passive satellite observations to retrieve CCN number concentration. Although their approach does not provide a vertically resolved picture of CCN, their work lays a foundation and demonstrates the feasibility of estimating CCN-related properties from satellite measurements. By mentioning the Rosenfeld et al. (2016) study, we aim to acknowledge the efforts and advancements made in CCN estimation from satellite data. We agree with your viewpoint that the Rosenfeld et al. (2016) method may seem tangential to a lidar-focused paper. However, it is important to highlight the collective progress in the field and the different approaches being explored for CCN estimation, including those from satellite observations. By referencing the Rosenfeld et al. (2016) study, we aim to emphasize the ongoing efforts in the field and the potential for future integration of lidar and satellite-based techniques in CCN research. Thank you for bringing this up, and we will ensure that the mention of the Rosenfeld et al. (2016) method is appropriately justified and positioned within the manuscript to provide clarity and relevance to our study objectives.

P4, l125: In addition to Burton et al. (2016) I think one should cite the original papers of Mueller et al., 1999, 2005 and of Veselovskii et al., 2002, 2004, 2012.

Authors: The suggested citation are added in the revised manuscripts.

The inversion solution using the combination of simultaneous measurements of backscatters at three wavelengths and extinction at two wavelengths, also called *3β+2α,* using lidar has been gaining prominence for aerosol microphysical (effective radius, total number, volume concentration, refractive index) retrieval (Burton et al., 2016; Müller et al., 1999, 2005, 2016; Veselovskii et al., 2002, 2004, 2012).

P4, l131: to my opinion, you have just a 2+2 lidar because the 1064 nm BSC is always a problem and the solutions are rather uncertain, because clear air calibration is very difficult, and a good calibration is only possible in the presence of ice clouds.

Authors: We appreciate the reviewer's concern regarding using the 1064 nm channel to retrieve aerosol properties and CCNs. However, we respectfully disagree that we have only 2+2 lidar observations.

From an airborne perspective, Advanced multiwavelength Raman lidars and High Spectral Resolution Lidar (HSRL2) have been increasingly used in recent years to retrieve aerosol and CCN properties *(Chemyakin et al., 2014; Müller et al., 1999, 2014; Burton et al., 2015; 2018)*. These lidar systems independently measure profiles of particle backscatter coefficients and particle depolarization ratios at 355, 532, and 1064 nm and particle volume extinction coefficients at 355 and 532 nm (*Müller et al., 2014; Burton et al., 2015; 2018*). However, the particle extinction coefficient at 1064 nm (α_{1064}) is derived using the backscatter coefficient and depolarization at 1064 nm using HSRL technique. Therefore, the *3β+2α* algorithm using lidar measurements has been gaining prominence for aerosol microphysical retrieval (*Müller et al., 1999, 2005, 2016; Veselovskii et al., 2002, 2004, 2012; Burton et al., 2016; Chemyakin et al., 2016; Lv et al., 2018; Tan et al., 2019*). While it is true that the existing three wavelengths (355, 532, and 1064 nm) lidar system (such as HSRL2) face challenges in clear air calibration of the 1064 nm due to atmospheric interference, and we acknowledge the calibration with ice clouds may not always be available. As stated in our paper, this can introduce limitations and uncertainties in our results. However, these issues can be mitigated with appropriate calibration and processing techniques. For instance, the HSRL2 system is designed to accurately measure aerosol backscatter and depolarization profiles at 532 nm wavelength, which can be combined with the 1064 nm signal to derive aerosol extinction profiles. Moreover, techniques such as range-dependent calibration and calibration using atmospheric model simulations have been shown to improve the accuracy of such lidar measurements (*Burton et al., 2012*).

In the context of the satellite perspective, it is important to note that our study focuses on estimating the particle size distribution and the subsequent estimation of cloud condensation nuclei (CCN) number concentration from satellite measurements. While we understand the challenges associated with the 1064 nm BSC measurements, our approach aims to leverage the available satellite data to derive valuable information about aerosol properties and their impact on cloud processes. Satellite lidar instruments typically operate at multiple wavelengths, including 1064 nm, to provide a more comprehensive characterization of the atmosphere. While the clear air calibration at 1064 nm can pose challenges due to the absence of strong scattering targets, such as aerosols or clouds, it is

worth noting that the calibration process for satellite lidar instruments is carefully designed and validated to ensure accurate measurements.

Nevertheless, the present study aims to leverage the available quality-assured measurements at 1064 nm to reduce the uncertainty due to the non-uniqueness problem and find the closest solution to particle size distribution. The inclusion of lidar measurements at 1064 nm provides important information on the vertical profile of aerosols and CCNs, particularly for coarse mode particles. In the present study, a comparative analysis between *3β+2α* and *3β+3α* (Figure 10) demonstrated that insertion of the α_{1064} signal improves the CCN estimation by ~15% in total and ~20% for the coarse mode-dominated aerosol subtypes (i.e., marine and dust aerosols) compared to *3β+2α*.

In addition, we have also validated our retrieved results against independent in-situ measurements from HSRL2-measured vertical profiles of aerosol optical properties from the NASA ORACLES airborne campaign. This provides additional confidence in our retrieval. Overall, we carefully evaluated the uncertainties associated with the use of the lidar measurements at 1064 nm and provided in section 3.1. However, we appreciate reviewer feedback and acknowledge that lidar calibration remains an active area of research and development.

We acknowledge the uncertainties and limitations associated with the 1064 nm measurements and the challenges of clear air calibration. These limitations are inherent to lidar measurements, and efforts are continually made to improve calibration techniques and minimize uncertainties.

P4, l147: Are you sure that the assumption of spheroidal dust particles is ok to obtain trustworthy dust lidar ratios at all three wavelengths? You may check the recent paper of Haarig et al. (2022).

Haarig et al., First triple-wavelength lidar observations of depolarization and extinction-to backscatter ratios of Saharan dust, Atmos. Chem. Phys., 22, 355–369, https://doi.org/10.5194/acp-22-355-2022, 2022.

Authors: Thank you for highlighting the limitation of assuming spheroidal dust particles in our study. We acknowledge that this assumption may introduce uncertainties in our dust-related retrievals. In light of this, we are committed to addressing this issue and improving the accuracy of our methodology. Our future research will focus on exploring alternative models that can efficiently capture the complex shapes of dust particles while remaining computationally efficient. By incorporating these advanced models, we aim to enhance the reliability and trustworthiness of our dust-related retrievals, ensuring that our results align with the latest understanding of dust particle properties. We appreciate your valuable input and will consider it in our ongoing efforts to refine and improve our methodology.

(ii) the non-spherical shape of dust particles. While this study considers the spheroidal shape of dust particles, 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 (Gasteiger et al., 2011; Saito et al., 2021) provide a more realistic representation of irregularly shaped dust particles, they are computationally expensive. We acknowledge this limitation and plan to explore alternative models in future studies.

P6-9, section 2.2: The main questions I had after reading section 2.2:

How do you handle all kinds of external mixtures, e.g., marine and dust, pollution and dust, smoke and dust, etc. The LUTs only include pure aerosol type information, right? So this is an open point that should be better explained in the manuscript.

Authors: We recognize that the characterization of internal mixtures can be complex and challenging to incorporate directly in to 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. 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 consider the dust mixtures (polluted dust and dusty marine). To avoid complications in the LUT approach, we exclude the mixture of dust and smoke due to the varying depolarization values of smoke, which can change as it ages from freshly emitted state.

The algorithm separates the optical properties of polluted dust into polluted continental and dust components, while for dusty marine aerosols, it distinguishes between dust and marine contributions. It utilizes a widely accepted technique for separating dust and non-dust components in the backscatter coefficient. This technique, as described by Tesche et al., (2009), relies on the particle depolarization ratio. By setting specific wavelength-dependent particle depolarization ratio, the algorithm distinguishes between pure dust and non-dust aerosol mixtures. The algorithm multiplies the resulting backscatter coefficients of dust and non-dust components by the corresponding lidar ratio for each aerosol subtype to determine the extinction coefficients for dust and non-dust aerosol components. The resulting separated aerosol optical properties combined with relative humidity and aerosol subtype information, is then utilized in the ECLiAP algorithm (as shown in Figure 2) to estimate CCN concentrations. Further details regarding the methodology to separate dust and non-dust components can be found in the revised manuscript. We added the more details in the revised manuscript.

Before applying hygroscopic correction, lidar-measured optical properties, particularly for dust mixtures (polluted dust and dusty marine), are separated into dust and non-dust components using the backscatter coefficients and particle depolarization ratio (Tesche et al., 2009). The methodology for separating dust mixture discussed initially in an Appendix A1, is now covered as a part of the methodology in the main manuscript. The section was revised by providing the additional information. The resulting dust and non-dust aerosol optical properties, along with aerosol subtype and relative humidity, is then utilized in the ECLiAP algorithm (as shown in Figure 2) to estimate CCN concentrations. Note that the direct inclusion of internal mixtures in our analysis and LUTs poses complexity and challenges. As a result, our approach primarily centers on studying and analyzing external mixtures of aerosol subtypes.

Thank you for pointing out the missing information. We added following details on methodology for separation of optical properties for dust mixture in Appendix A1.

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 S1. 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).

Relative humidity has a strong influence on all the retrievals. Are the growth or enhancement factors for all three wavelengths the same? Please provide more information. Just references are not sufficient. What about enhancement factors for internally mixed sulfate-BC-OC particles, or sulfate coated dust? What about growth factors for mixture of fine mode (urban haze) and coarse mode dust. The enhancement factor will then be clearly wavelength dependent, because 355 nm is very sensitive to hygroscopic small particles, and 1064 nm will be very sensitive to the hydrophobic dust particles. Is it possible to consider all these complex items?

Authors: We acknowledge that RH plays a significant role in aerosol characterization. In our study, we employed a wavelength-dependent hygroscopic enhancement factor account for the effects of RH on aerosol optical properties to a dry state.

We utilized a new physically based single-parameter representation approach proposed by Brock et al. (2016) was considered to estimate the hygroscopic enhancement factor. This approach showed better performance in describing light-scattering hygroscopic enhancement factors compared to the most frequently used gamma power-law approximation (Kasten, 1969). The formula of this scheme incorporates a dimensionless fitting parameter and the dry aerosol optical properties (as seen in Equation 6 in manuscript). To estimate the hygroscopic enhancement factor, we calculated aerosol optical properties (backscatter and extinction coefficients) at three different wavelengths (355 nm, 532 nm, and 1064 nm) over a range of RH values using Mie theory. Curve fitting was performed to determine using the derived aerosol optical properties at different RHs. We fixed the RH range to 60%-90% for the parameter fitting. The hygroscopic correction was only applied when RH was between 40% and 99%.
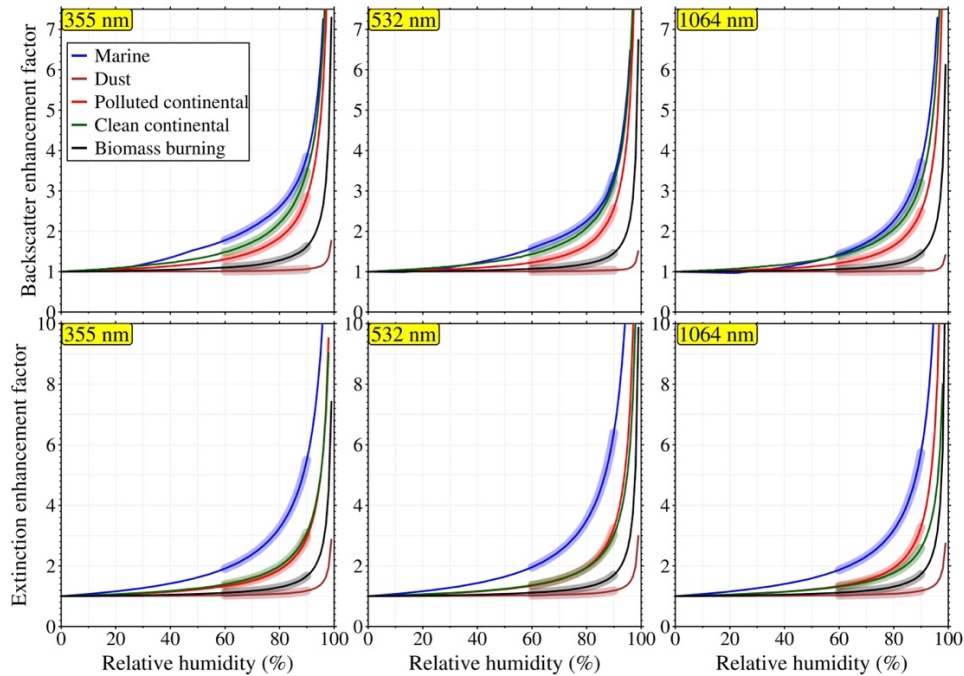
Figure 1 : Mean enhancement factor for backscatter and extinction coefficients at 355, 532 and 1064 nm are fitted using Eq (6) for five aerosol subtypes. This mean fitting curve is calculated with the set of PNSD and κ considered for the construction of LUTs. The thin line represents Mie model simulations, and the highlighted thick line (within RH range of 60-90%) are used to fit parameterization lines.

In our ECLiAP algorithm, individual values for each aerosol optical property and wavelength, along with the RH value, were used to calculate the dry aerosol optical properties separately for each aerosol subtype (see figure above). This approach allowed us to consider the wavelength dependence and the influence of RH on aerosol optical properties in the retrieval process. For more detailed information and graphical representations, please refer to Figure S1 and the detailed methodology provided in section 2.2.2 in the manuscript.

Finally, we acknowledge that the complexity of internally mixed aerosol types, such as sulfate-BC-OC particles or sulfate-coated dust, introduces additional challenges. However, due to the limitations in directly incorporating internal mixtures into our lookup tables (LUTs), our approach mainly focuses on external mixtures. Internal mixing is a complex topic requiring further investigation and the development of specific retrieval techniques.

Final point, RH values are at all obtained from models? Absolute uncertainties of plus-minus 20% have always to be kept in mind…. in many cases even 50% is my long-term experience. Are there papers that provide clear statements on modelled RH uncertainty?
Authors: We appreciate the reviewer's concern regarding RH uncertainties in CCN retrievals, and we have taken several steps to address and mitigate these uncertainties in our study.

In our validation process with airborne lidar observations, we utilized measured meteorological parameters from the airborne platform, which provides reliable in-situ measurements and reduces reliance on model-derived RH. This helps to ensure that the retrieved CCN values are validated using robust data sources. For CCN retrieval process using CALIPSO data, we relied on the RH values provided in the CALIPSO product, which are obtained from the GMAO modeling and assimilation office. While we made efforts to use the best available RH data, we acknowledge that model-derived RH values may have associated uncertainties.

To account for the potential impact of RH uncertainties, our algorithm incorporates a wavelength-dependent and aerosol-type dependent hygroscopic enhancement factor (as described in section 2.2.2). This factor allows us to correct the lidar-derived wet size distribution to the dry size distribution, ensuring accurate CCN calculations. By considering this hygroscopic enhancement factor, we aim to effectively account for the effects of RH on the aerosol optical properties and reduce the potential influence of RH uncertainties on CCN retrievals.

Furthermore, we conducted a sensitivity analysis to evaluate the influence of systematic and random errors in lidar-derived optical properties and auxiliary RH profiles on our CCN retrieval. This analysis allowed us to assess the overall performance of our retrieval algorithm and identify the main sources of errors. Through this comprehensive evaluation, we have considered and quantified the uncertainties associated with RH in our CCN retrievals.

While we acknowledge that uncertainties in model-derived RH values are inherent, our thorough error analysis and incorporation of the hygroscopic enhancement factor address these uncertainties to a significant extent. We have taken into account the changes in aerosol properties due to water uptake, which are influenced by RH, ensuring that our CCN retrievals are robust and reliable. Regarding the request for papers providing clear statements on modeled RH uncertainty, we have reviewed the literature extensively, and while there are studies discussing RH uncertainties, there is currently no consensus on a universal quantification of these uncertainties. The quantification of RH uncertainties depends on various factors, including the specific model used, the region of interest, and the time period considered.

In summary, we have implemented measures to address and mitigate RH uncertainties in our CCN retrievals. By incorporating a hygroscopic enhancement factor, conducting a sensitivity analysis, and considering the limitations of model-derived RH values, we have taken a comprehensive approach to ensure the reliability and accuracy of our results. We appreciate the reviewer's input, and we will continue to explore further improvements in addressing RH uncertainties in future research endeavors.

P9, l301: I am not sure, but is the critical radius not defined as the radius for which 50% of particles are activated….
Authors: The critical radius can be defined differently depending on the specific application and the experimental setup.

Traditionally, the critical radius has been defined as the size at which 50% of particles are activated, representing the point at which half of the aerosol population transitions to cloud

droplets under a given supersaturation condition. This definition is commonly used in cloud physics and aerosol activation studies (e.g., Twomey, 1959; Seinfeld and Pandis, 2016).

However, it is worth noting that the definition of critical radius can vary in different research contexts. In some studies, the critical radius may be defined as the minimum size required for activation under specific supersaturation conditions, corresponding to the smallest aerosol particles that can initiate cloud droplet formation. This definition focuses on the transition point from non-activated to activated particles rather than specifically targeting the 50% activation threshold.

In our manuscript, we have adopted the definition of critical radius as the minimum size of an aerosol particle that can activate into droplets for a given supersaturation ratio. This definition aligns with the concept of critical radius as the size threshold for activation without explicitly referring to the 50% activation criterion.

P9, l307: The critical radius can be as low as 25 nm for high supersaturations of 0.8 to 1%. The lidar backscatter and extinction is only sensitive to particles with radius of 50 nm and larger…. How can you then derive a critical radius of 25 nm? Please clarify and explain that in the manuscript!

Authors: The critical radius represents the size at which particles become activated as CCN. Indeed, lidar backscatter and extinction measurements are typically more sensitive to particles with larger sizes, generally around 50 nm and larger. However, it is essential to note that the estimation of the critical radius is not solely based on lidar measurements but involves additional considerations.

In our study, estimating the critical radius and subsequent CCN number concentration retrieval involves combining techniques, including lidar measurements and other ancillary data sources. While lidar measurements provide valuable information on the size distribution and extinction properties of particles, determining the critical radius relies on integrating various factors, such as aerosol size distribution, aerosol composition, supersaturation levels, and environmental conditions. The critical radius is derived by considering the Köhler theory or an equivalent approach, which considers the interplay between particle properties, such as size and composition, and the thermodynamic properties of the surrounding air parcel. By incorporating these factors, it is possible to estimate the critical radius even for particle sizes below the typical sensitivity range of lidar measurements.

We acknowledge that it is important to clearly explain the relationship between lidar sensitivity and the estimation of the critical radius in the manuscript. We revised the manuscript to include a detailed discussion of the methodology and considerations used to derive the critical radius, considering the limitations and sensitivity ranges of lidar measurements.

Thank you for raising this point, and we appreciate the opportunity to address it in the revised manuscript.

Probably you make use of Eq.(8), but that is then an assumption you use here… and causes an uncertainty. What about the impact of new particle formation on the actual Aitken mode (contributing to the CCN concentration)? This is an important uncertainty source, I could imagine!

Authors: In our retrieval methodology, we parameterize the particle size distribution within the range of 0.01 microns to 10 microns. This choice is based on the limitations of lidar measurements, which primarily provide sensitivity to particles with sizes larger than approximately 50 nanometers. The lidar backscatter and extinction signals are more sensitive to particles in the accumulation and coarse mode ranges, and the retrieval focuses on estimating aerosol properties within this range.

We acknowledge that this parameterization introduces uncertainties, particularly in capturing the contribution of particles smaller than 50 nanometers, such as those associated with new particle formation events. These particles can play a significant role in influencing the Aitken mode and CCN concentration.

Additionally, lidar measurements at the three wavelengths (355 nm, 532 nm, and 1064 nm) have different sensitivities to different aerosol types and sizes. The 355 nm wavelength is more sensitive to small particles, including hygroscopic species, while the 1064 nm wavelength is more sensitive to larger particles, such as dust. This wavelength dependence further adds to the complexity of accurately characterizing the aerosol population.

We included this as a limitation of lidar measurements, which subsequently appears in the estimation of CCN concentrations. We will also emphasize the need for further research to address the challenges associated with capturing the contribution of particles smaller than 50 nanometers and the impact of new particle formation on the CCN concentration.

Results:

p10, l317 and p11, l358: To my understanding, ECLiAP is not an inversion method. It is just an LUT approach. One should not mix that.

Authors: 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.

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 $3\alpha+2\beta$ technique typically used for inversion.

The relative humidity is a crucial input parameter. Uncertainties of 10-20% can never be excluded. So, resulting CCN retrieval uncertainties must be visualized up to RH plus minus 20%.

Authors: We appreciate the reviewer's concern regarding the impact of relative humidity (RH) on the resulting CCN retrievals. We acknowledge that uncertainties in RH can introduce variability in the retrieval outcomes, and we have given careful consideration to this aspect in our study. As indicated in the manuscript, we have accounted for systematic errors in RH by encompassing a range of RH uncertainties from -10% to 10%. This range was selected to cover typical variations in RH encountered in various atmospheric conditions. However, we understand the reviewer's interest in visualizing the impact of larger RH uncertainties up to plus minus 20%. We have indeed explored the effects of a 20% uncertainty in RH in our analysis, and the results demonstrate that this level of uncertainty introduces relatively larger errors in the NCCN estimation. Specifically, for negative RH errors.
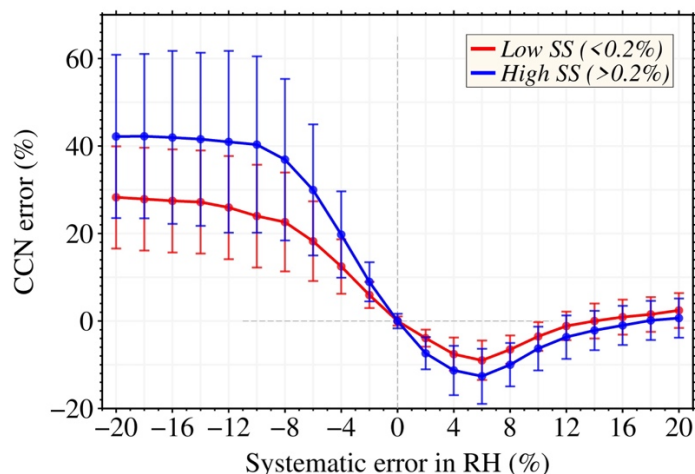
Figure: Systematic errors in retrieved N_{CCN}. This represents the errors in retrieved N_{CCN} as a function of systematic error in RH, combines for all aerosol subtypes, at low (≤0.2%) and high (>0.2%) supersaturations. The markers denote the mean value, and the error bars represent the standard deviation.

A 10% uncertainty in RH leads to approximately 20% underestimation in NCCN, and for negative RH errors, it results in around 40% underestimation. This finding serves to illustrate the significance of RH as a crucial parameter in the lidar-based retrieval. However, we should note that encountering a 20% RH uncertainty is infrequent in typical atmospheric conditions. The purpose of considering a 10% RH uncertainty was to demonstrate the sensitivity of our retrieval and its implications for typical RH variations observed in the atmosphere. Additionally, we have conducted an evaluation of the effects of random errors in RH, ranging from 5% to 20%, and have visualized the resulting NCCN retrieval errors under these different RH uncertainty scenarios. The results indicate that random errors in RH also contribute to the overall retrieval uncertainties, with the largest mean relative errors observed for coarse mode-dominated aerosol subtypes, consistent with their sensitivities to systematic errors.

We thank the reviewer for their valuable input, and we are confident that the current analysis, along with the comprehensive discussion in the manuscript, appropriately addresses the role of RH uncertainties in our CCN retrievals. Our aim is to demonstrate the significance of RH as a crucial

parameter in the lidar-based retrieval of NCCN and to provide valuable insights for the atmospheric science community.

P13, l461: The HSRL does not measure directly the 1064 nm extinction! ..is stated in line 461. This statement comes much too late. It must be clear from the beginning that HSRL is a typical 3+2 lidar instrument. You even do not know the 1064 nm lidar ratio. Please provide the lidar ratio! It is an important quantity! but not mentioned. May be you assumed 40 sr at 1064 nm, and in reality it is 80-100 sr, what are the consequences of such a bad assumption on the CCN retrieval? Please comment on that in the manuscript.

Authors: HSRL2 have been widely used to retrieve aerosol properties. These lidar systems independently measure the particle backscatter coefficients and depolarization ratios at three wavelengths (355, 532 and 1064 nm), and particle volume extinction coefficients at two wavelengths (355 and 532 nm) (*Müller et al., 2014; Burton et al., 2015; 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).

It is important to note that our approach does not involve a direct retrieval of aerosol properties. Instead, 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. We appreciate your feedback and have made the necessary revisions to better understand our methodology and the implications of incorporating the inferred lidar ratio at 1064 nm.

P13, l471: That is my basic question: How do you handle mixtures: dust, marine, and smoke…. in your retrieval…
50100620403426

Authors: We appreciate your continued interest in our study. As mentioned in the earlier response, our study primarily focuses on external mixtures of aerosol subtypes, as the direct incorporation of internal mixtures into the Look-Up Tables (LUTs) used in our analysis can be complex. To account for mixtures, we separated the optical properties of polluted dust and dusty marine aerosols and utilized a technique based on particle depolarization ratio to distinguish between dust and non-dust components. The resulting separated aerosol properties, along with relative humidity and aerosol subtype information, were used in the ECLiAP algorithm to estimate CCN concentrations. Additional details on the methodology can be found in Appendix A1 of the revised manuscript.

P15, l506-511: I have no idea what you mean here…. because CALIOP is a 2+0 lidar, or even a 1+0 lidar. Please explain better …!
Authors: 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.

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.

P15, l512-526: Again,I have no idea what you did? Did you apply the depolarization-ratio based method to separate dust and non-dust components? And then what did you do in the next steps? Must be clear in the manuscript.
Authors: Yes, we applied a wavelength-dependent depolarization ratio method to separate dust and non-dust components. As mentioned above, the resulting separated aerosol properties, along with relative humidity and aerosol subtype information, were used in the ECLiAP algorithm to estimate CCN concentrations (as seen in Figure-2). Additional details on the methodology can be found in Appendix A1 of the revised manuscript.

P16, l548: Again, there is a mixture of dust and continental pollution. What are the different steps of the retrieval. Please explain in detail!
Authors: Details on the methodology can be found in Appendix A1 of the revised manuscript.

P16, l573: What does that mean… a more realistic LUT-based approach using the 3+3 wavelength technique? You do not have any good information about the 1064 nm lidar ratio!
Authors: 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.

In the summary and conclusion section one could discuss: How large is the chance that there will be an airborne or spaceborne 3+3 HSRL in the near future (within the next 10 years).
Authors: We appreciate the reviewer's suggestion. The NASA Langley Research Center airborne lidar instrument HSRL-2 measures aerosol backscatter and depolarization at three wavelengths (355 nm, 532 nm, and 1064 nm) and aerosol extinction at 355 nm and 532 nm using the HSRL technique (Burton et al., 2018; Shipley et al., 1983). At 1064 nm, extinction is derived from the product of aerosol backscatter at 1064nm and an inferred lidar ratio at 1064 nm. So, technically it is 3+2 but not the 3+3. 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 "$3\beta+3\alpha$ technique".

P20, l684, the depol value of 0.31 holds for 532 nm only!
Authors: Thank you for pointing out the missing information. Detailed information on the values of particle depolarization ratio is added in the Appendix A1. We utilized a reported wavelength-dependent particle depolarization ratio from various campaigns (listed in Table S1). For this investigation, we employ mean values of particle depolarization ratio 0.24, 0.31, and 0.06 for dust and 0.03, 0.05, and 0.02 for non-dust at 355 nm, 532 nm, and 1064 nm, correspondingly.

Tables 1 and 2: If the numbers are so small, do we really have to show this?
Authors: We understand reviewer's concern regarding the small numbers presented in Table 2 and Table 3 (they were Table 1 and Table 2 in the original version of manuscript). While the values may appear small, they still hold important information for the analysis.

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.

By including these tables, we ensure transparency and provide comprehensive insights into the performance and limitations of our methodology. This strengthens the scientific rigor of our study and enables other researchers to reproduce and validate our findings effectively.

Figure 6 triggered my basic question: How are aerosol mixtures handled in the entire retrieval procedure?
Authors: Details on the methodology can be found in Appendix A1 of the revised manuscript.

Figure 8: I have no idea how you got all the shown information and how you could estimate CCN at the end. And who can evaluate the quality of the products? We have just to believe.
Authors: In Figure-8, we used a single CALIPSO overpass to showcase the capability of ECLiAP in retrieving N_{CCN} from space-borne lidar observations. The aerosol extinction coefficient at 532 nm and aerosol subtypes information illustrated in Figure 8(a & b) are already taken directly form the data product, whereas particle concentrations and CCN concentration in Figure 8 (c & d) are derived using ECLiAP.

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.

Burton, S. P., Hair, J. W., Kahnert, M., Ferrare, R. A., Hostetler, C. A., Cook, A. L., Harper, D. B., Berkoff, T. A., Seaman, S. T., Collins, J. E., Fenn, M. A., & Rogers, R. R. (2015). Observations of the spectral dependence of linear particle depolarization ratio of aerosols using NASA Langley airborne High Spectral Resolution Lidar. *Atmospheric Chemistry and Physics*, *15*(23), 13453–13473. https://doi.org/10.5194/ACP-15-13453-2015

Burton, S. P., Hostetler, C. A., Cook, A. L., Hair, J. W., Seaman, S. T., Scola, S., Harper, D. B., Smith, J. A., Fenn, M. A., Ferrare, R. A., Saide, P. E., Chemyakin, E. V., & Müller, D. (2018). Calibration of a high spectral resolution lidar using a Michelson interferometer, with data examples from ORACLES. *Applied Optics*, *57*(21), 6061. https://doi.org/10.1364/AO.57.006061

Chemyakin, E., Müller, D., Burton, S., Kolgotin, A., Hostetler, C., & Ferrare, R. (2014). Arrange and average algorithm for the retrieval of aerosol parameters from multiwavelength high-spectral-resolution lidar/Raman lidar data. *Applied Optics*, *53*(31), 7252. https://doi.org/10.1364/AO.53.007252

Chemyakin, E., Burton, S., Kolgotin, A., Müller, D., Hostetler, C., & Ferrare, R. (2016). Retrieval of aerosol parameters from multiwavelength lidar: investigation of the underlying inverse mathematical problem. *Applied Optics*, *55*(9), 2188. https://doi.org/10.1364/AO.55.002188

Lv, M., Wang, Z., Li, Z., Luo, T., Ferrare, R., Liu, D., Wu, D., Mao, J., Wan, B., Zhang, F., & Wang, Y. (2018). Retrieval of Cloud Condensation Nuclei Number Concentration Profiles From Lidar Extinction and Backscatter Data. *Journal of Geophysical Research: Atmospheres*, *123*(11), 6082–6098. https://doi.org/10.1029/2017JD028102

Müller, D., Wandinger, U., & Ansmann, A. (1999). Microphysical particle parameters from extinction and backscatter lidar data by inversion with regularization: theory. *Applied Optics*, *38*(12), 2346. https://doi.org/10.1364/AO.38.002346

Müller, D., Mattis, I., Wandinger, U., Ansmann, A., Althausen, D., & Stohl, A. (2005). Raman lidar observations of aged Siberian and Canadian forest fire smoke in the free troposphere over Germany in 2003: Microphysical particle characterization. In *Journal of Geophysical Research D: Atmospheres* (Vol. 110, Issue 17). https://doi.org/10.1029/2004JD005756

Müller, D., Hostetler, C. A., Ferrare, R. A., Burton, S. P., Chemyakin, E., Kolgotin, A., Hair, J. W., Cook, A. L., Harper, D. B., Rogers, R. R., Hare, R. W., Cleckner, C. S., Obland, M. D., Tomlinson, J., Berg, L. K., & Schmid, B. (2014). Airborne multiwavelength High Spectral Resolution Lidar (HSRL-2) observations during TCAP 2012: Vertical profiles of optical and microphysical properties of a smoke/urban haze plume over the northeastern coast of the US. *Atmospheric Measurement Techniques*, *7*(10), 3487–3496. https://doi.org/10.5194/AMT-7-3487-2014

Müller, D., Böckmann, C., Kolgotin, A., Schneidenbach, L., Chemyakin, E., Rosemann, J., Znak, P., & Romanov, A. (2016). Microphysical particle properties derived from inversion algorithms developed in the framework of EARLINET. *Atmospheric Measurement Techniques*, *9*(10), 5007–5035. https://doi.org/10.5194/AMT-9-5007-2016

Tan, W., Zhao, G., Yu, Y., Li, C., Li, J., Kang, L., Zhu, T., & Zhao, C. (2019). Method to retrieve cloud condensation nuclei number concentrations using lidar measurements. *Atmospheric Measurement Techniques*, *12*(7), 3825–3839. https://doi.org/10.5194/AMT-12-3825-2019

Veselovskii, I., Dubovik, O., Kolgotin, A., Korenskiy, M., Whiteman, D. N., Allakhverdiev, K., & Huseyinoglu, F. (2012). Linear estimation of particle bulk parameters from multi-wavelength lidar measurements. *Atmospheric Measurement Techniques*, *5*(5), 1135–1145. https://doi.org/10.5194/AMT-5-1135-2012

Veselovskii, I., Kolgotin, A., Griaznov, V., Müller, D., Franke, K., & Whiteman, D. N. (2004). Inversion of multiwavelength Raman lidar data for retrieval of bimodal aerosol size distribution. *Applied Optics*, *43*(5), 1180–1195. https://doi.org/10.1364/AO.43.001180

Veselovskii, I., Kolgotin, A., Griaznov, V., Müller, D., Wandinger, U., & Whiteman, D. N. (2002). Inversion with regularization for the retrieval of tropospheric aerosol parameters from

multiwavelength lidar sounding. *Applied Optics*, *41*(18), 3685. https://doi.org/10.1364/AO.41.003685

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., & Andreae, M. O. (2006). Size matters more than chemistry for cloud-nucleating ability of aerosol particles. *Science*. https://doi.org/10.1126/science.1125261

Farmer, D. K., Cappa, C. D., and Kreidenweis, S. M.: Atmospheric Processes and Their Controlling Influence on Cloud Condensation Nuclei Activity, Chem. Rev., 115, 4199–4217, https://doi.org/10.1021/cr5006292, 2015.

Patel, P. N., & Jiang, J. H. (2021). Cloud condensation nuclei characteristics at the Southern Great Plains site: role of particle size distribution and aerosol hygroscopicity. *Environmental Research Communications*, *3*(7), 075002. https://doi.org/10.1088/2515-7620/AC0E0B

Kasten, F. (1969). Visibility forecast in the phase of pre-condensation. *Tellus*, *21*(5), 631–635. https://doi.org/10.3402/TELLUSA.V21I5.10112

Brock, C. A., Wagner, N. L., Anderson, B. E., Attwood, A. R., Beyersdorf, A., Campuzano-Jost, P., Carlton, A. G., Day, D. A., Diskin, G. S., Gordon, T. D., Jimenez, J. L., Lack, D. A., Liao, J., Markovic, M. Z., Middlebrook, A. M., Ng, N. L., Perring, A. E., Richardson, M. S., Schwarz, J. P., … Murphy, D. M. (2016). Aerosol optical properties in the southeastern United States in summer - Part 1: Hygroscopic growth. *Atmospheric Chemistry and Physics*, *16*(8), 4987–5007. https://doi.org/10.5194/ACP-16-4987-2016

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 $\varepsilon_0 = 1.4$, the transition occurs at approximately $x_c \approx 125$, while for prolate spheroids with $\varepsilon_0 = 3.0$, 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 "$3\beta+3\alpha$ 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.

Burton, S. P., Hostetler, C. A., Cook, A. L., Hair, J. W., Seaman, S. T., Scola, S., Harper, D. B., Smith, J. A., Fenn, M. A., Ferrare, R. A., Saide, P. E., Chemyakin, E. V., & Müller, D. (2018). Calibration of a high spectral resolution lidar using a Michelson interferometer, with data examples from ORACLES. *Applied Optics*, *57*(21), 6061. https://doi.org/10.1364/AO.57.006061

Dubovik, O. (2002). Variability of absorption and optical properties of key aerosol types observed in worldwide locations. *J. Atmos. Sci.*, *59*, 590–608. https://doi.org/10.1175/1520-0469(2002)059<0590:voaaop>2.0.co

Giannakaki, E., Van Zyl, P. G., Müller, D., Balis, D., & Komppula, M. (2016). Optical and microphysical characterization of aerosol layers over South Africa by means of multi-wavelength depolarization and Raman lidar measurements. *Atmospheric Chemistry and Physics*, *16*(13), 8109–8123. https://doi.org/10.5194/ACP-16-8109-2016

Hänel, A., Baars, H., Althausen, D., Ansmann, A., Engelmann, R., & Sun, J. Y. (2012). One-year aerosol profiling with EUCAARI Raman lidar at Shangdianzi GAW station: Beijing plume and seasonal variations. *Journal of Geophysical Research Atmospheres*, *117*(13). https://doi.org/10.1029/2012JD017577

Kim, M. H., Omar, A. H., Tackett, J. L., Vaughan, M. A., Winker, D. M., Trepte, C. R., Hu, Y., Liu, Z., Poole, L. R., Pitts, M. C., Kar, J., & Magill, B. E. (2018). The CALIPSO version 4 automated aerosol classification and lidar ratio selection algorithm. *Atmospheric Measurement Techniques*, *11*(11), 6107–6135. https://doi.org/10.5194/AMT-11-6107-2018

Komppula, M., Mielonen, T., Arola, A., Korhonen, K., Lihavainen, H., Hyvärinen, A. P., Baars, H., Engelmann, R., Althausen, D., Ansmann, A., Müller, D., Panwar, T. S., Hooda, R. K., Sharma, V. P., Kerminen, V. M., Lehtinen, K. E. J., & Viisanen, Y. (2012). Technical Note: One year of Raman-lidar measurements in Gual Pahari EUCAARI site close to New Delhi in India-Seasonal characteristics of the aerosol vertical structure. *Atmospheric Chemistry and Physics*, *12*(10), 4513–4524. https://doi.org/10.5194/ACP-12-4513-2012

Lv, M., Wang, Z., Li, Z., Luo, T., Ferrare, R., Liu, D., Wu, D., Mao, J., Wan, B., Zhang, F., & Wang, Y. (2018). Retrieval of Cloud Condensation Nuclei Number Concentration Profiles From Lidar Extinction and Backscatter Data. *Journal of Geophysical Research: Atmospheres*, *123*(11), 6082–6098. https://doi.org/10.1029/2017JD028102

Mamouri, R. E., & Ansmann, A. (2016). Potential of polarization lidar to provide profiles of CCN-and INP-relevant aerosol parameters. *Atmospheric Chemistry and Physics*, *16*(9), 5905–5931. https://doi.org/10.5194/acp-16-5905-2016

Müller, D., Ansmann, A., Mattis, I., Tesche, M., Wandinger, U., Althausen, D., & Pisani, G. (2007). Aerosol-type-dependent lidar ratios observed with Raman lidar. *Journal of Geophysical Research Atmospheres*, *112*(16). https://doi.org/10.1029/2006JD008292

Pérez-Ramírez, D., Whiteman, D. N., Veselovskii, I., Kolgotin, A., Korenskiy, M., & Alados-Arboledas, L. (2013). Effects of systematic and random errors on the retrieval of particle microphysical properties from multiwavelength lidar measurements using inversion with regularization. *Atmospheric Measurement Techniques*, *6*(11), 3039–3054. https://doi.org/10.5194/AMT-6-3039-2013

Tan, W., Zhao, G., Yu, Y., Li, C., Li, J., Kang, L., Zhu, T., & Zhao, C. (2019). Method to retrieve cloud condensation nuclei number concentrations using lidar measurements. *Atmospheric Measurement Techniques*, *12*(7), 3825–3839. https://doi.org/10.5194/AMT-12-3825-2019

Torres, B., Dubovik, O., Fuertes, D., Schuster, G., Eugenia Cachorro, V., Lapyonok, T., Goloub, P., Blarel, L., Barreto, A., Mallet, M., Toledano, C., & Tanré, D. (2017). Advanced characterisation of aerosol size properties from measurements of spectral optical depth using the GRASP algorithm. *Atmospheric Measurement Techniques*, *10*(10), 3743–3781. https://doi.org/10.5194/AMT-10-3743-2017

Veselovskii, I., Kolgotin, A., Griaznov, V., Müller, D., Franke, K., & Whiteman, D. N. (2004). Inversion of multiwavelength Raman lidar data for retrieval of bimodal aerosol size distribution. *Applied Optics*, *43*(5), 1180–1195. https://doi.org/10.1364/AO.43.001180