03 December 2023

Dr. Zhanqing Li
Senior Editor, Atmospheric Chemistry and Physics

Dear Dr. Li,

I am pleased to submit the revised manuscript titled “A Remote Sensing Algorithm for Vertically-Resolved Cloud Condensation Nuclei Number Concentrations from Airborne/Spaceborne Lidar Observations” for your consideration as a Research Article in the Atmospheric Chemistry and Physics.

Following your insightful decision and the subsequent second round of reviews on October 03, 2023, my coauthors and I have diligently worked on addressing the comments and enhancing the manuscript.

Our manuscript focuses on developing a novel remote sensing-based analytical algorithm (known as "ECLiAP") to estimate vertically-resolved cloud condensation nuclei (CCN) number concentration (NCCN) using airborne/spaceborne lidar observations. Beyond method development, a comprehensive error analysis was conducted. Applying ECLiAP to airborne observations, we established its reliability through robust correlation with independent data. Extending its applicability, we successfully applied ECLiAP to CALIPSO spaceborne lidar observations, offering a compelling case study, advancing remote sensing capabilities for CCN estimation and contributing to our understanding of aerosol-cloud interactions.

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.

The revised manuscript thoroughly addresses the comments from Anonymous Referees #2 and #3 in the first round of review. Referee #2 found the revisions satisfactory and accepted the manuscript, while Referee #3 provided significant revisions.

Referee #3 expressed concerns about the LUT approach, potentially overlooking its successful application in previous lidar studies and its extension in our work. We have meticulously clarified the methodology and presented compelling evidence in our response supporting the algorithm's novelty and effectiveness, particularly in error analyses and sensitivity studies. Despite differences in interpretation, we respectfully disagree with Referee #3's rejection and believe the revised manuscript comprehensively addresses their comments, making a substantive contribution to lidar-based aerosol retrieval science. The manuscript adheres to the highest scientific standards, offering a focused approach for clarity while still contributing significantly to the field.

In conclusion, we firmly believe our work significantly advances the scientific understanding of aerosol-cloud interactions using lidar-based measurements. The manuscript has not been submitted elsewhere, and all relevant data are accessible through public domain portals.

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

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.

Anonymous Referee #3

The authors provide elaborate but unfortunately unconvincing replies to the comments of both Referees. Answers are either evading the problem, repeat what has already been stated as problematic, or postpone the problem to future work. Therefore, most of the criticism still stands. As a consequence, I can only recommend to reject this work for publication in ACP. Any resubmission to AMT would also require substantial improvements.

We appreciate the reviewer's reevaluation of our revised manuscript. We take note of the contrasting perspectives between the first and second reviewers. While the first reviewer found the revisions acceptable without additional comments, we understand that second reviewer has expressed reservations. We are committed to bridging this gap in their understanding and further improving the manuscript to address your specific concerns. Reviewer's feedback is invaluable, and we are grateful for the opportunity to engage constructively. We remain open to additional suggestions that can enhance the quality and clarity of our work.

Here are a few examples of what is problematic with this paper:

The authors insist that their LUT approach is somewhat of an inversion and reduces retrieval errors. They acknowledge the ill-posed nature of the inversion of microphysical particle properties from lidar measurements of optical parameters. They understand that identical sets of backscatter and extinction coefficients can be the result of very different (and sometimes physically meaningless) particle size distributions. The authors then claim that their retrieval reduces the related uncertainty by assigning a single size distribution to an individual set of optical parameters. In reality, they increase uncertainty because they are simply dismissing an unknown number of equally possible solutions.

Authors: We sincerely appreciate your insightful comments and the opportunity they provide to delve deeper into the intricacies of our Look-Up Table (LUT) methodology.

Reviewer's concern about the ill-posed nature of the inversion problem and the potential increase in uncertainty is well-noted. We would like to reiterate that our LUT approach is not intended to dismiss the complexity introduced by identical sets of backscatter and extinction coefficients leading to various particle size distributions. Instead, it is designed to systematically address this complexity.
As highlighted in prior studies (Chemyakin et al., 2016; Lv et al., 2018), the LUT methodology strategically narrows down potential solutions, offering a more manageable set of plausible outcomes. We are extending this successful approach into our specific context. The LUT encompasses a comprehensive array of possible solutions, each linked to a spectrum of physically plausible size distributions inferred from ground-based observations.

During retrieval, our algorithm effectively selects the most probable solution from the LUT, considering additional contextual information. This methodical navigation through a range of solutions is aimed at managing the inherent ambiguity rather than dismissing equally possible outcomes.

Moreover, we want to draw attention to the broader context within which our methodology operates. The LUT approach has shown success in previous studies, and our work builds upon this foundation. The non-uniqueness of the inverse problem is a recognized challenge, and the efficacy of the LUT approach in reducing the associated uncertainty has been demonstrated. Our study extends this methodology into our specific application.

To address concerns about uncertainty, we have conducted rigorous calibration and validation exercises, quantifying both systematic and random errors. The incorporation of an additional measurement of the extinction coefficient at 1064 nm further enhances the accuracy of our algorithm, contributing significantly to reducing uncertainty arising from the non-uniqueness of the inversion solution, as highlighted in our "3β+3α technique."

The tangible impact of this reduction in uncertainty is clearly reflected in the improved CCN estimation, as illustrated in Figure 10. Our LUT approach, informed by successful applications in previous studies, stands as a robust strategy for real-time estimation of particle size distributions from lidar measurements, effectively mitigating the challenges posed by the non-uniqueness of the inverse problem.

We believe these clarifications underscore the validity and significance of our chosen approach in addressing the intricacies of the inversion process. Thank you for your continued engagement in the peer-review process.

The authors repeatedly claim that they have 3+3 data, i.e. lidar measurements that include independently inferred extinction coefficients at 1064 nm, at their disposal and that the added information also decreases the retrieval uncertainty. As both referees have commented this to be incorrect, one has to assume that the authors are deliberately misleading the readers. While HSRL-2 is certainly one of the best aerosol lidars currently in operation, it also requires the assumption of a lidar ratio to obtain extinction coefficients at 1064 nm. In the same way, CALIPSO data are only 2+0 and not 2+2 as the same lidar ratio, that is already assumed in the calculation of the backscatter coefficients, is also used to transform the latter to extinction coefficients.

Authors: We appreciate the thoughtful comments from the reviewer, providing us with an opportunity to elaborate on our methodology concerning the utilization of 3+3 lidar data in our study.

In our context, the term "3+3" denotes lidar measurements encompassing backscatter and extinction coefficients at three distinct wavelengths (355 nm, 532 nm, and 1064 nm). It's crucial to clarify that these data, used as inputs to the ECLiAP, are not claimed to be direct lidar measurements. Rather, we leverage quality-assured lidar-derived product in conjunction with direct lidar measurements to address the inherent uncertainties in the inversion process.

While acknowledging the necessity of assuming a lidar ratio for obtaining extinction coefficients at 1064 nm, especially for instruments like HSRL-2 and CALIPSO, our methodology extends beyond direct lidar measurements. The incorporation of derived product, combined with direct lidar measurements, serves to mitigate the non-uniqueness problems inherent in inversion. Despite introducing uncertainties, the inclusion of extinction coefficient at 1064 nm significantly reduces retrieval uncertainty, as correctly noted by the reviewer. To underscore the efficacy of our methodology, we have already conducted a comprehensive comparative analysis between the *3β+2α* and *3β+3α* methods, as illustrated in Figure 10. The findings reveal a substantial improvement in CCN estimation, with an overall enhancement of approximately 15% and around 20% for coarse mode-dominated aerosol subtypes (e.g., marine and dust aerosols) compared to *3β+2α*. This improvement underlines the value of incorporating additional lidar inputs, specifically extinction coefficients at 1064 nm, in refining retrievals.

It's crucial to emphasize that our work is motivated by a commitment to advancing lidar-based retrieval science rather than misleading intentions. The goal is to enhance the accuracy of retrievals, particularly for coarse mode particles where inversion challenges exist. We believe this work makes a significant contribution to the scientific understanding of aerosol properties.

In conclusion, we appreciate the opportunity for this clarification and remain open to further discussions. We are confident that our methodology, supported by the comparative analysis presented, adds substantial value to the field of lidar-based aerosol retrieval.

The following statement is incorporated in the manuscript to clarify that we used the lidar derived product along with direct measurements in the present retrieval algorithm.

"The integration of derived product, along with direct lidar measurements, addresses the inherent non-uniqueness problem of inversion, and despite introducing uncertainties, the inclusion of extinction coefficient at 1064 nm significantly reduces retrieval uncertainty, emphasizing the value of additional lidar inputs in refining retrievals."

The sensitivity analysis still seems to represent circular thinking. This is what I understand the authors are doing. First, size distributions are used to obtain unique sets of optical properties that form the LUT. Second, a set of optical properties is defined and the retrieval browses the LUT for the size distribution that is connected to that set. Finally, the difference between the CCN concentration calculated from the size distributions that are used to create the LUT and selected from the retrieval, i.e. from browsing the LUT, is determined. This difference is of course negligible because the method goes back and forth along the same line of reasoning.

Authors: We respectfully disagree with the characterization of circular thinking in the sensitivity analysis, and it appears there might be a misinterpretation of the analysis. Allow us to clarify the distinct aspects of these sensitivity analyses.

First, in the sensitivity analysis with error-free lidar measurements, we employ 2000 different sets of bimodal size distributions for each aerosol subtype to simulate lidar observations. These simulations are not directly derived from the LUTs but are inputs to the retrieval process. The retrieval is then performed on these simulated lidar observations, and the resulting CCN concentrations are compared with the initial inputs. This procedure is intended to assess the inversion performance and stability of ECLiAP by evaluating its ability to reproduce the initial NCCN values under error-free conditions. It does not involve a circular reasoning process of comparing outputs with the same inputs derived from LUTs.

Second, we conducted a sensitivity analysis using ground-based measurements of aerosol size distributions obtained from the U.S. Department of Energy's Atmospheric Radiation Measurement (ARM) climate research facility. The observed aerosol size distributions are used to test the retrieval's sensitivity to assumptions about the aerosol size distribution. This analysis involves real-world data (not part of LUTs) and is not a circular reasoning process. We fit bimodal lognormal size distributions to the observed data, use them to calculate N_{CCN}, and compare the results with the observed N_{CCN} values. The purpose is to assess the impact of assuming bimodal size distributions on the retrieval results.

In essence, these two sensitivity analyses are fundamentally distinct and serve different purposes: one evaluates the algorithm's stability under error-free conditions, and the other examines its sensitivity to real-world aerosol size distributions. These distinctions have been explicitly outlined in the revised manuscript. We hope this clarification resolves the concern about circular thinking, and we remain open to further discussion or clarification if needed.

It doesn't suffice to state that the application of the new method to CALIPSO data requires some adjustment. It can be expected that a reduction of the number of available independent lidar parameters from 5 (HSRL-2) to 2 (CALIOP) has a dramatic impact on the retrieval quality if not applicability. It goes against good scientific practise to present some results without any form of validation just because they can be generated. Consequently, I can only repeat that the CALIPSO part should either be strengthened or removed from the manuscript and that the current title needs to be revised to accurately represent the content of the paper.

Authors: We appreciate the reviewer’s insightful comments regarding the application of our method to CALIPSO data.

We acknowledge the challenge posed by the reduction in lidar parameters from HSRL-2 to CALIOP. It’s important to underscore that the limitations in lidar inputs are not intentional restrictions but rather arise from the inherent constraints of CALIOP, which provides data at only two wavelengths. In our previous revision, we have explicitly stated that the modification in the algorithm due to the constrains of CALIOP measurements, introduce potential limitations and uncertainties. Furthermore, we emphasize the need for an in-depth and comprehensive validation study against the multiple independent measurements.

“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. A detailed comprehensive analysis comparing the CALIOP-retrieved N_{CCN} with multi-campaign airborne measurements is essential to evaluate the reliability of ECLiAP to construct the 3D CCN climatology at a global scale.”

In response to the importance of validation, we would like to mention that we are actively engaged in a dedicated study aimed at thoroughly validating CALIPSO-derived NCCN with multiple independent measurements from various campaigns. This ongoing work is a direct response to the concerns raised and emphasizes our commitment to addressing these issues in a meticulous manner.

It is essential to undescroe that the scope of our present study is the advancement of the algorithm and methodology for precise NCCN estimation from lidar measurements. This comprehensive framework is meticulously crafted to be applicable to both airborne and spaceborne lidar data, with a dedicated focus on detailed error analysis and robust validation, particularly with airborne lidar measurements. Given the complexity and significance of our algorithm, we have opted for a focused approach to streamline the scientific manuscript. To enhance clarity and prioritize in-depth analyses, we have judiciously decided to present an extensive comparative analysis of CALIOP-derived NCCN with multiple independent measurements in an upcoming study, distinct from the current manuscript. This strategic choice underscores our commitment to ensuring a thorough examination of the data and upholding the scientific rigor intrinsic to our methodology.

In alignment with the central focus of the study, we are refining the title to better encapsulate the content of our manuscript.

"A Remote Sensing Algorithm for Vertically-Resolved Cloud Condensation Nuclei Number Concentrations from Airborne/Spaceborne Lidar Measurements."