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Comment on acp-2022-547

Anonymous Referee #3

Referee comment on "Resolving Vertical Profile of Cloud Condensation Nuclei Concentrations from Spaceborne Lidar Measurements" by Piyushkumar Patel et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-547-RC2, 2022

The paper by Patel et al. presents a methodology to infer the concentration of cloud condensation nuclei (N_{CCN}) 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).

- **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).
- **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.
  - 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.

- 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 reviewers knows that the likelihood for such instruments becoming a common occurrence is negligible. But readers might not and, thus, should be informed about this.

- **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.
  - 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
  - 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.
  - 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.
  - 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 (\sqrt(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.
- **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.

- 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.

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