Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2022-547

Anonymous Referee #2

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-RC1, 2022

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.

Detailed comments:

p3, l97: Why do we need to know the particle size distribution? CCN is just the overall particle number concentration (integral over all sizes)!

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.

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!

P3, l101: Why do you mention the Rosenfeld et al (2016) method in this lidar paper?

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.

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.

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.

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.

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?

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?

P9, l301: I am not sure, but is the critical radius not defined as the radius for which 50% of particles are activated….

P9, l307: The critical radius can be as low as 25 nm for high super saturations 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!

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!

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.

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%.

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.

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

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 …!

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.

P16, l548: Again, there is a mixture of dust and continental pollution. What are the different steps of the retrieval. Please explain in detail!

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!

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).

P20, l684, the depol value of 0.31 holds for 532 nm only!

Tables 1 and 2: If the numbers are so small, do we really have to show this?

Figure 6 triggered my basic question: How are aerosol mixtures handled in the entire retrieval procedure?

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.