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Reply on RC2

Goutam Choudhury and Matthias Tesche

Author comment on "Estimating cloud condensation nuclei concentrations from CALIPSO lidar measurements" by Goutam Choudhury and Matthias Tesche, Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2021-358-AC2>, 2021

We thank the reviewer for the time and effort in reviewing our manuscript. We found the comments to be very helpful in enhancing the quality of our article. Below are our point-by-point replies to his comments.

- As mentioned in the general comment, after L48, it should be added another paragraph to state the motivation of proposing this new method for estimating CCN concentrations, which is different from the widely used POLIPHON method. To my point of view, the potential advantage for the proposed method is not necessary for calculating those region-varied conversion factors to obtain $n_{j,dry}$ (Ansmann et al. 2019AMT, He et al., 2021AMT), which cannot be obtained in the regions without sun photometer observations. Instead, CAMel and MOPSMAP package can solve this problem, making the global estimation of CCN concentrations possible. If so, add a new paragraph to state that.

We appreciate the reviewer's comment. It is crucial to note that the size distributions and refractive indices included in the CALIPSO aerosol model are derived from cluster analysis of AERONET measurements and from independent measurement campaigns. In either case, the site-specific measurements limit our coverage of different regions of our globe. However, the cluster analysis of long-term AERONET measurements already has provided us with deeper insights into the major global aerosol types and their consistent microphysical properties across different measurement sites (Omar et al., 2005). Having said that, we still believe this consistency will be disrupted in real-atmosphere conditions of complex aerosol mixtures. This circumstance motivated us to perform a sensitivity analysis and to quantify the effect of variations of the size distributions in our algorithm on the derived CCN concentrations. Also, the POLIPHON method has not yet been applied to long-term CALIPSO data for studying aerosol-cloud interactions. This is our long-term goal and to achieve it, we wanted to develop a new methodology that is self-consistent within the CALIPSO framework. To emphasize this circumstance, we have added the following sentences to the Introduction:

"The approach for retrieving cloud-relevant aerosol microphysical properties has not yet been implemented for spaceborne lidar measurements. This study, therefore, presents a new methodology for obtaining height-resolved aerosol number concentrations from CALIPSO measurements within the CALIPSO framework, i.e. without relying on externally

inferred conversion factors."

- The basic information of the data set used in subsection 4.1 and 4.2 are absent, such as the location (global or a few sites) and period (a few months or year) of the selected data. This may confuse the readers, especially considering the large differences of conversion factors in the POLIPHON method. It is suggested to show some cases (after figure 5) that compare the CCN concentration profiles for five aerosol types with POLIPHON and OMCAM algorithms.

We are sorry for the confusion. The sensitivity tests (Section 4.1) and theoretical comparison between OMCAM and POLIPHON methods (Section 4.2) are performed solely based on synthetic data. As stated in lines 239, 269, and 270 of the revised manuscript, while performing the sensitivity experiments, we use a preset extinction coefficient value of 0.1 km⁻¹ and zero relative humidity, and vary the input size distribution parameters (mean radii, standard deviations, and volume fraction) of our algorithm. Coming to Section 4.2, we preferred the theoretical comparison of both the algorithms because it is hard to find real world scenarios with only one aerosol subtype and thus using real data can skew the results. We have added the following sentences (lines 307-310) for further clarification in the revised paper.

"In this section, we present a theoretical comparison of the CCN concentrations estimated using the OMCAM and POLIPHON methods (Mamouri and Ansmann, 2016). Both algorithms' primary input is the aerosol type-specific extinction coefficient. Hence, we consider a range of extinction coefficients and compute the corresponding theoretical CCN concentrations with both algorithms. To estimate CCN concentrations with POLIPHON, we use the extinction-to-CCN conversions given in Eq. (1)."

- The difference (or important improvement) between this method and Georgoulias et al. (2020) should also be stated.

Thank you for pointing this out. We have added the following sentences in our revised manuscript (lines 331-338):

"When it comes to ease of application, the POLIPHON method with its simple extinction-to-CCN conversion is more straightforward while the OMCAM algorithm – at present stage – is more complex and computationally expensive. Despite of the complexities, OMCAM incorporates a hygroscopicity correction methodology which is essential for a CALIPSO-based CCN retrieval (Georgoulias et al., 2020). Furthermore, the computation time in the OMCAM algorithm can be drastically reduced by either (i) parameterizing the output CCN concentrations in terms of the type-specific extinction coefficient and RH values or (ii) creating a look-up table of CCN concentrations at different extinction coefficients and RH values for different aerosol subtypes. However, such developments are not within the scope of the present work which focuses on the theoretical description of the OMCAM algorithm."

- L26-27: "Quaas et al., 2008; Quaas et al., 2020" -> 'Quaas et al., 2008, 2020'

Thank you for identifying this mistake. We have rectified it in the updated manuscript.

- L40: n_{280} has been modified to n_{250} in the following papers (Mamouri and Ansmann, 2016ACP; Ansmann et al., 2019ACP).

Thank you for pointing this out. We have changed n_{280} to n_{250} in the updated manuscript.

- L76: volume or particle depolarization ratio? It should be clearly stated.

Thank you for pointing this out. We use the particle depolarization ratio and have stated it in the updated manuscript.

- L92-93: What if CALIPSO level-2 aerosol subtype is misclassified (Ansmann et al., 2021 FENVS)? It should be mentioned and added some sentences to evaluate the related impact.

Thank you for your suggestion. We have added the following sentence (lines 299-300) to our manuscript along with the reference. *"Furthermore, aerosol misclassification in the CALIPSO aerosol-typing scheme (Ansmann et al., 2021a) may introduce errors in the OMCAM algorithm."*

- L103: 'spheroids' -> 'non-spheroids'? please confirm.

We apologize for the confusion. The MOPSMAP package uses combination of the T-matrix method and improved geometric optics method for spheroid aerosols.

- L116: define the α in equation (1) here. x is aerosol extinction exponent. The unit should be given for each parameter in equation (1). Besides, you should also cite when giving equation (1) (Shinozuka et al., 2015ACP).

Thank you for pointing it out. The parameter " α " was already defined before Eq. (1) in line 124 of the updated manuscript. We have added units to all parameters and now cite the article in our updated manuscript.

- L124: Conversion factors vary from region to region. Therefore, as suggested before, it is better to choose a specific site (maybe the site in Limassol, where many target aerosol types can be observed) and give CCN profile comparison between POLIPHON and OMCAM algorithms.

As explained in the response to the second comment, we have performed a theoretical comparison between OMACM and POLIPHON retrievals which is much more universal than the Referee's suggestion. As the POLIPHON conversion factors may vary with regions, we use their averaged values suggested by Ansmann et al. (2019, 2021) for the application to CALIPSO data (already stated in lines 135-136).

- L127-134: This paragraph is logically confusing and should be rephrased to introduce the general content of section 3.

We apologize for the confusion. We have modified the paragraph in the revised manuscript as:

"This section describes the algorithm used in the present work to derive CCN concentrations from the CALIPSO profiles of extinction coefficient, backscatter coefficient, depolarization ratio, and aerosol subtype information. We begin with the scaling procedure of the normalized size distributions from the CAMel to obtain the actual aerosol size distribution. After that, we explain the hygroscopicity correction followed by the CCN parametrization adopted in our algorithm. Finally, we discuss the application of the CCN retrieval algorithm to CALIPSO level 2 aerosol profile data."

- L232: How can the conclusion 'fine mode as the primary contributor to the output aerosol number concentration' be drawn from figure 2?

As discussed in the same paragraph, from Figure 2, we see that the fine mode size distribution parameters such as mean radius and standard deviations have the maximum impact on the output $n_{j,dry}$ concentrations of up to 600% and 100%, respectively.

- L312: Rephrase this sentence to more clearly state that conversion factors for marine aerosols are estimated from the Barbados AERONET site?

We have rephrased the sentence in the revised manuscript as:

"The POLIPHON conversion factor for marine aerosol is estimated from 7.5 years of measurements between 2007 and 2015 at the Barbados AERONET site (Mamouri and Ansmann 2016)."

- L135 and Figure 5: There is a systematical factor of around 3-5 for marine aerosol types between the CCN concentration from two different methods. It would be better to try to give a more specific discussion, especially considering the proposed method is expected to extend to a global scale.

We understand reviewer's point here. We have addressed the possible causes behind the differences in lines 323-331 of the revised manuscript as given below. In short, we believe that this may be a result of different approaches and sample sizes used to derive the size distributions in both the methods. Furthermore, we stress the need for a detailed validation study to identify which algorithm is more accurate in estimating CCN concentrations for marine aerosols.

"This may be due to the different approaches followed and sample sizes considered to derive the size distributions used in the two algorithms. The POLIPHON conversion factor for marine aerosol is estimated from 7.5 years of measurements between 2007 and 2015 at the Barbados AERONET site (Mamouri and Ansmann, 2016). In contrast, the marine model used in OMCAM is derived from in-situ measurements of sea-salt size distributions produced from breaking waves, taken during the SEAS experiment at Bellows Air Force Station, Oahu, Hawaii between 21 and 30 April 2000. Studies found that the AERONET size distributions can be significantly different from the in-situ measurements – especially under high relative humidity conditions (Chauvigne et al., 2016; Schafer et al., 2019). Further studies involving type-specific comparisons of both the aerosol number concentrations and the CCN concentrations with in-situ measurements are required to test the reliability of both algorithms (Mamali et al., 2018)."

- Figure 6: The trajectory of the sub-satellite point should be added.

Thank you for your suggestion. It is aimed at better placing the location of the measurements. We have tried adding an inset of the satellite track but were not satisfied with the results. Therefore, we kept the figure as is and revised the caption for a better description of the location of the measurement to *"for a CALIPSO overpass over the Thessaloniki region of northern Greece on 9 September 2011."*

- Figure 7: It is great to see that CCN concentration profiles from POLIPHON and OMCAM are consistent with each other after hygroscopicity correction very well. It is expected to conduct more comparison with in-situ measurement in the future as reported in (Mamali et al., 2019AMT).

We have added the suggested reference in the revised manuscript.

