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Comment on acp-2022-154

Charles Brock (Referee)

Referee comment on "Evaluation of aerosol number concentrations from CALIPSO with ATom airborne in situ measurements" by Goutam Choudhury et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-154-RC1, 2022

This manuscript provides a good evaluation of algorithms to derive aerosol number concentration from space-based lidar measurements against independent, in-situ observations made during a recent, global-scale airborne campaign, the Atmospheric Tomography mission (ATom). The authors use trajectory simulations to match the locations of the aircraft measurements with the CALIOP lidar measurements, and exclude data taken above 5 km, where the lidar signal is not sufficient. They also exclude data where dry extinction values disagree by more than 50%. To these matched lidar observations they apply two algorithms to estimate the number concentration of particles with diameters >0.1 and >0.5 μm. They compare these number concentrations with those directly measured by the airborne observations. One of the algorithms, OMCAM, did not match the marine observations well, while the other, POLIPHON, performed acceptably. The authors propose a modification to OMCAM based on a different marine aerosol model that improves agreement with the observations.

The manuscript is well-written, clear, and focused. The analysis is fine, with some relatively minor changes suggested below. The manuscript provides a useful independent (from the training set) test of the ultimate outcome of the POLIPHON and OMCAM algorithms when applied to the CALIOP backscatter measurements, and as such is informative regarding the level of confidence with which these algorithms can be applied.

That said, I would suggest future analysis that would involve more direct comparison of the ATom dataset to the algorithms to calculate aerosol number concentration from the CALIOP measurements. I was the PI for the ATom aerosol microphysics measurements, and have recently produced a dataset of calculated optical and microphysical properties that could be profitably compared to the CALIOP measurements (Brock et al., 2021). I feel it would be more straightforward to directly use these derived optical properties from the ATom dataset, such as aerosol optical depth, ambient extinction, or backscatter, and perform the regressions to determine the coefficients found in the POLIPHON parameterization. By comparing these derived regression coefficients, one would bypass the need for the backtrajectories and matching of the air masses that produces substantial scatter and uncertainty in the comparison presented here. Further, this expanded ATom dataset could be used to examine the horizontal and vertical variability of these coefficients (i.e., build statistics on their variability), which would help bound the uncertainties in the estimation of particle number concentration from the lidar measurements.

For the case of the OMCAM parameterization, the ambient size distributions derived from the in situ ATom measurements, which include lognormal fits to the nucleation, Aitken, accumulation, and coarse modes, could be used to validate the CALIPSO aerosol model size distributions that were selected as best representative of the ambient aerosol extinction coefficient. They could also be compared with the marine aerosol size distribution that was applied to derive new coefficients when poor agreement was found with the in situ measurements. There is clearly sensitivity of the OMCAM algorithm to these inputs, so understanding their variability is important to evaluate their general applicability globally.

The new ATom dataset in Brock et al. (2021) also provides hygroscopic growth factors calculated from the composition measurements, CCN concentrations at different supersaturations, and vertically integrated AOD values. These all have the potential to be compared with the CALIOP measurements and derived quantities. More importantly, they can be used to develop and constrain the parameterizations, often based on a small number of observations over a limited region, that provide the inputs to the algorithms that are applied to remote sensing data. Better understanding of the spatial variability of these aerosol characteristics can only improve our ability to apply space-borne sensors to derive aerosol quantities that are not directly measurable remotely.

Minor comments:

1) Lines 15-18. Correlation coefficients are used to evaluate "good agreement". Correlation is correlation, not quantitative agreement, which is better estimated by RMSE and bias.

2) Section 2.3.2 (OMCAM): The relationship given by Eq. 3 must depend very sensitively on the choices for the size distribution modes described in Eq. 2, especially for sizes in the CCN-active region ($r>0.025$ µm) where the size distribution is often very steeply sloped. It would be useful for this manuscript to provide more information, in the form of a table, on the choices for the lognormal parameters for the different aerosol types that were used. Some discussion on their variability would also be useful.

3) Lines 171-173: I'm not sure what is meant by "using the microphysical properties by Sayer et al. (2012) in the OMCAM algorithm and estimate the ANC separately". The OMCAM algorithm is used to derive the ANC via Eq. 3; how can you estimate the ANC "separately"? Perhaps it's better to explain that a different marine model is applied in Sect. 3.2.1 upon finding that the Sayer et al. model produced significant biases when

compared with the in situ data. At least that's clear, if perhaps providing a bit too much foreshadowing of results.

4) Summary: One of the take-home messages for me was that, while these algorithms do surprisingly well for the r>50 aerosol number concentration, they are pretty sensitive to assumptions about the size distribution model in the training set. Clearly there needs to be a better understanding of the variability in space and time of these parameters. So comparison with measurements such as Schmale et al. (2017) would be useful, but I would think a more comprehensive evaluation of the range of size distribution parameters in different airmass types would be an important long-term objective. There are many potential datasets out there; for example, for the marine aerosol, there is an extensive ship-borne global dataset from Quinn et al. (2017), as well as an airborne datasets by Clarke and Kapustin (2002) and Clarke et al. (2010). A more comprehensive survey may be in order to thoroughly bound uncertainties in the space-borne retrieval of ANC and CCN. Certainly beyond the scope of this work, but perhaps a target for the future.

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