## acp-2019-892 (Painemal et al.)

### Responses to Reviewer 1

We appreciate the reviewer's insightful comments and suggestions. His/her comments and recommendations are addressed below (in blue).

The paper starts off by mentioning the ACI construct, but then doesn't seem to refer to the ACI again. One very interesting potential consequence of this work is the impact on the ACI of the biases identified here. Is the error in the ACI using AOD random (in which case the mean ACI and RFaci are relatively unchanged) or is it a systematic bias, which would affect RFaci quantification. This is not a large addition to the manuscript, but it would be a potentially significant result that might not otherwise stand as a paper on its own.

The reviewer raises an important point. We decided to focus on the implicit assumption of the ACI construct, that is, the linearity of the aerosol-cloud relationship (in logarithmic scale). While we agree with the reviewer that the ACI quantification is relevant, this is left for future work, as robust estimates will require the use of the full CALIPSO data record. Nonetheless, following the reviewer's recommendation, we provide insight into this point: Based on the AOD-$\sigma_{BC}$ depicted in Fig. 3b, AOD can be empirically expressed as:

$AOD = \alpha \cdot \sigma_{BC}^{\beta}$ (1)

Eq. (1) can be used to recast $ACI_{AOD}$ in terms of $\sigma_{BC}$ as:

$$ACI_{AOD} = \frac{\partial lnAOD}{\partial lnN_d} \approx \beta \cdot \frac{\partial ln\sigma_{BC}}{\partial lnN_d}$$

Therefore ACI expressed in terms of AOD and $\sigma_{BC}$ can be related via $\beta$ as:

$$ACI_{AOD} \approx \beta \cdot ACI_{\sigma}$$

It is clear from Fig. 3b that the linear pattern between AOD and $\sigma_{BC}$ is only valid for specific regimes of variability, typically for $\sigma_{BC}$ and AOD less than or greater than 0.1 ($km^{-1}$ for extinction). Thus, biases in ACI expressed in terms of AOD will depend on the observed range of AOD. For relatively pristine conditions (AOD<0.1), $ACI_{AOD} \approx 0.15 \cdot ACI_{\sigma}$, and for AOD>0.1 the relationship is $ACI_{AOD} \approx 0.85 \cdot ACI_{\sigma}$. That is, the fractional underestimation relative to $ACI_{\sigma}$ would encompass the range [0.15 0.85]. This analysis is now included in the revised manuscript.

Secondly, there appears to be a reliance on spatial correlations across the globe. Given that cloud properties vary across the globe and that these variations might be expected to generate variations in the CCN-$N_d$ relationship (e.g. through updraft variations), is this not slightly dangerous? One factor that might be interesting is global maps of the correlations (or ACI), which might help to identify any regional effects.

We agree with the reviewer that updraft velocity is an important factor to be taken into account for understanding the spatial distribution of aerosols that ultimately module the cloud droplet number concentration (e.g. Sullivan et al. 2016). Similarly, precipitation and entrainment are likely contributors to the Nd and CCN budget. However, despite the complexity of the cloud-aerosol microphysical processes, observational studies have shown concomitant westward gradients in aerosol concentration and cloud droplet number concentration over the eastern Pacific and northeast Atlantic (e.g. Bretherton et al., 2010;

Painemal et al., 2015; Bretherton et al., 2019; Wood et al., 2019), where low aerosol concentrations are associated with low $N_d$ (e.g. Wood et al., 2019, Painemal et al., 2015). So spatial co-variability between aerosol properties and $N_d$ should be expected in subtropical boundary layer clouds. In the revised manuscript, we caution the reader that the spatial correlation approach is an oversimplification of the processes that control aerosol activation, yet the spatial correlation has been corroborated in in-situ observations over the eastern Pacific and northeast Atlantic.

As discussed in our response to Comment 1, the ACI mapping will be included in a standalone manuscript. Regarding regional correlations, they are reported for the eastern Atlantic and Pacific only. Given the sparse CALIOP's spatial coverage, we will need a much larger dataset in order to report regional correlations for the entire globe (which we hope to include in the standalone manuscript).

References:

Sullivan SC, Lee D, Oreopoulos L, Nenes A (2016) Role of updraft velocity in temporal variability of global cloud hydrometeor number. Proc Natl Acad Sci USA 113:5791–5796.

Bretherton, C. S., Wood, R., George, R. C., Leon, D., Allen, G., and Zheng, X.: Southeast Pacific stratocumulus clouds, precipitation and boundary layer structure sampled along 20° S during VOCALS-REx, Atmos. Chem. Phys., 10, 10639–10654, https://doi.org/10.5194/acp-10-10639-2010, 2010.

Bretherton, C.S., I.L. McCoy, J. Mohrmann, R. Wood, V. Ghate, A. Gettelman, C.G. Bardeen, B.A. Albrecht, and P. Zuidema, 2019: Cloud, Aerosol, and Boundary Layer Structure across the Northeast Pacific Stratocumulus–Cumulus Transition as Observed during CSET. *Mon. Wea. Rev.,* **147**, 2083–2103.

Painemal, D., Minnis, P., and Nordeen, M. ( 2015), Aerosol variability, synoptic-scale processes, and their link to the cloud microphysics over the northeast Pacific during MAGIC. *J. Geophys. Res. Atmos.*, 120, 5122– 5139. doi: 10.1002/2015JD023175.

Wood, R., Stemmler, J. D., Rémillard, J., and Jefferson, A.( 2017), Low-CCN concentration air masses over the eastern North Atlantic: Seasonality, meteorology, and drivers, *J. Geophys. Res. Atmos.*, 122, 1203– 1223, doi:10.1002/2016JD025557.

L34 - product of

Modified, thanks.

L45 - Stier (2016) noted that vertical mismatches are important to get a high correlation, but results using other aerosol-climate models (Gryspeerdt et al, PNAS, 2017) suggested that this does not have a large impact on the inferred RFaci (as long as the PD-PI aerosol product is appropriate). This paper focuses on the aerosol-$N_d$ correlation, which is one step further removed from the RFaci. As the initial justification of this work is based around the RFaci, it would be good to mention how these results are linked to it/might affect it.

We appreciate the reviewer's suggestions. We have included the following sentence in the revised Discussion:

"Lastly, in light of the results presented here, it would be informative to assess the extent of which aerosol extinction coefficient can be combined with climate models to quantify the ACI radiative forcing from the pre-industrial time as in Gryspeerdt et al. (2017) but expressing ACI in terms of $\sigma_{BC}$ instead of MODIS AI."

Going beyond this discussion is difficult as Stier (2016) and Gryspeerdt et al. (2017) rely on climate models for their aerosol assessment. Keeping in mind the limitations of climate models in simulating the aerosol vertical structure (e.g. Koffi et al., 2016), it is unclear whether the simulated relationships between different aerosols proxies are valid representation of those observed in nature. In contrast, our work present an observationally based perspective of Stier (2016).

Reference:

Koffi, B., et al. ( 2016), Evaluation of the aerosol vertical distribution in global aerosol models through comparison against CALIOP measurements: AeroCom phase II results, *J. Geophys. Res. Atmos.*, 121, 7254– 7283, doi:10.1002/2015JD024639.

L117 - CALIPSO with CloudSat's

Section 2.2 - It sounds like the MODIS pixels are only paired with CALIOP pixels across the track, rather than along the track. This would mean that some closer pixels are ignored, as they might be in the along-track direction.

The reviewer is correct. This configuration was designed for minimizing clear-sky contamination in MODIS, and aerosol swelling near the cloud edges in CALIOP extinctions. By only considering MODIS samples at least 1 km east/west from the CALIOP track, with the filtering discussed in section 2, we are minimizing the potential swelling effect on aerosol extinction by removing aerosol embedded in regions with extensive cloud cover. Recall that simultaneous retrievals of cloud and aerosol in time and space are not possible, and therefore any satellite-based method has to rely on matching neighboring cloud and aerosol pixels.

L142 - Zhang and Platnick (JGR, 2011 - their Fig. 14) suggest that the 3.7um channel re is (slightly) negatively biased in in-homogeneous cases. how does this fit with the reasoning here? Also, is the 'cloudy-sky' retrieval mostly limited to high CF scenes (and might this explain the weaker $N_d$-CF relationship)?

Zhang and Platnick (2011) and our research (e.g. Painemal et al., 2013) show that 3.7-um cloud effective radius (r_e) is smaller than its 2.1-um counterpart; however, this should not interpreted as a bias in 3.7-um r_e. Instead, the differences mentioned by the reviewer are the consequence of the higher sensitivity of the 2.1-um channel to spatial inhomogeneity. In contrast, the 3.7-um is less sensitive to sub-pixel inhomogeneity and 3D radiative transfer effect. The advantages of the 3.9-um channel for retrieving r_e were briefly discussed in L96-L98.

Regarding the second point: cloudy $N_d$ refers to $N_d$ averaged over 5kmx5km grid with cloud fraction>0.9 (90%). These 5kmx5km averaged $N_d$'s are then further averaged in 25-km segments along the CALIPSO track.

L149 - I am unclear on how this binning procedure works. Not much needs to be included here, but it should be specified. What CF range is used?

CF was averaged using 50 bins with each of them containing the same number of samples. This binning is better described in the revised version.

L161 - I was probably just being slow, but it was only once I got here that I realized the below and above cloud top σ values are separate. Perhaps just make the in L124 a plural (σs) to make is clearer?

We appreciate the reviewer's suggestions. His/her comment was addressed accordingly.

L168 - This has been noted in previous studies (e.g. Ma et al, Nature Communications, 2018)

We appreciate the reviewer's comment. This is indeed a relevant work that is now discussed in the revised manuscript.

L176 - Is this unexpected? Several studies in the past have hinted at a saturation of aerosol effects at high AOD.

The reviewer is correct in that aerosol effect saturation at high AOD has been previously observed (e.g. Breon et al., 2002). Nevertheless, our analysis based on $\sigma_{BC}$ suggests the AOD-Nd saturation is possibly an artifact rather than explained by thermodynamical processes described in Feingold et al. (2001).

References:

Bréon, F.-M., Tanré, D., and Generoso, S.: Aerosol effect on cloud droplet size monitored from satellite, Science, 295, 834–838, 2002.

Feingold, G., Remer, L. A., Ramaprasad, J., and Kaufman, Y. J. ( 2001), Analysis of smoke impact on clouds in Brazilian biomass burning regions: An extension of Twomey's approach, *J. Geophys. Res.*, 106( D19), 22907– 22922, doi:10.1029/2001JD000732.

L179 - It is stated here that there is a narrower range of $N_d$ for each value of $\sigma_{SFC}$, but that it has a lower correlation coefficient with $N_d$ that $\sigma_{BC}$. How does these observations fit together?

The sentence refers to the $N_d$ range relative to the range of $\sigma_{SFC}$, rather than individual values of extinction. Therefore, the narrow range of variability in $N_d$ combined with a more scattered relationship give rise to lower correlations with respect to $N_d$-$\sigma_{BC}$. In the revised manuscript, we rephrased the sentence to read:

"In general, it is found a narrower range of the binned $N_d$ as a function $\sigma_{SFC}$ than that for $\sigma_{BC}$."

L182 - Give that the relationship is non-linear, would a measure such as Spearman's rank be more appropriate that an correlation coefficient?

We did calculate the Spearman's rank correlation in a previous version of the manuscript, and the differences were modest, which is why we did not include them in the manuscript. The standard Pearson correlation is more appealing as it allows inferences about variance.

L198 - It is difficult to compare the contours with the MODIS $N_d$ image. I am not sure that this needs an extra panel, but I can't see a clear alternative. It could even just be stated I think (as it is not a vital part of this work)

In the revised the manuscript, we use dots and circles to represent regions with free tropospheric aerosols: 0-0.015, 0.015-0.03, 0.03-0.45 $km^{-1}$. The manuscript was updated with the new figure (see below)

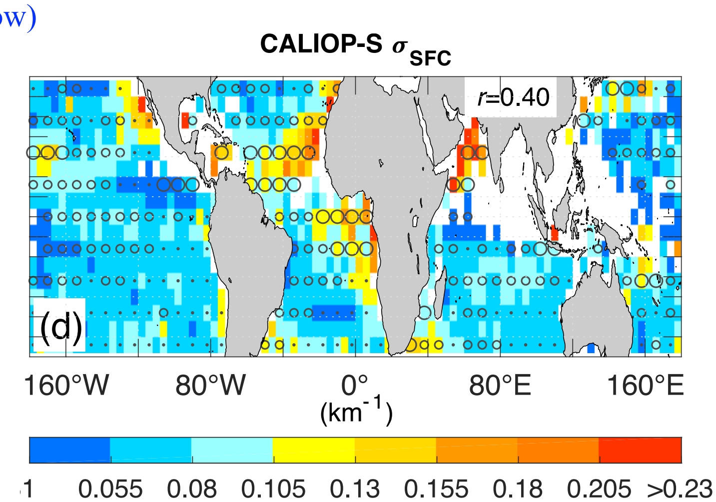

Figure 1: New surface and free tropospheric extinction coefficient in Fig. 4.

Fig 5 and 7 - Could these be included in Fig. 4? Especially as they are mainly here to compare to 4a

While we agree that the inclusion of Fig 5 and 7 into Fig. 4, we prefer to keep them as separate figures, as they are only used to support the results derived from CALIOP-S.