Dear Jim Haywood,

Thank you for your positive review of our manuscript. We appreciate the comments and suggestions you provided and we believe they will greatly improve the clarity and scientific robustness of the manuscript. We would like to take this opportunity to provide you with our plan to address your main concerns of the manuscript *prior to submitting a revised manuscript* to allow for a continued discussion as necessary. Below we have provided responses (in boldface) to each of the major and minor comments.

Review of Cochrane et al, AMT

This paper builds on the previous work of Cochrane et al. (2019) and examines the direct radiative effect of biomass burning aerosols over cloudy and clear-sky conditions during the ORACLES 2016 and 2017 measurement campaigns. There are three main aspects: 1) assessing the aerosol optical parameters that give consistent radiative closure, 2) developing a basic and extended algorithm that parameterizes the DARE as a function of the scene albedo and the optical depth (at 550 nm) and 3) adding an additional dependency on SSA.

I was very interested and impressed by the SSA retrievals when compared to some of the more accurate assessments of SSA that are now possible using more advanced instrumentation (e.g. airborne CRD and PAS measurements that are not subject to the artefacts associated with missing scattering (nephelometers) or with scattering/absorption artefacts (filter based absorption measurements). I have included some additional references to these measurements as they were not included by the authors – I do think that these complementary studies provide excellent additional supporting information for the validity of the approach.

The second part of the paper which documents the performance of the P_DARE and PX_DARE could certainly be of use when compared to satellite data, but one would have to have an estimate of the above cloud/above surface AOD and the scene albedo. The most obvious place where this could have applicability might therefore be a combination of e.g. space borne lidar and broadband scene albedo from e.g. GERB; it might be worth explicitly stating this as a future possibility.

I deal with the comments as more major and minor below.

More Major:

Point 1: L325 onwards: It is worth emphasizing that many of the in-situ retrievals of SSA have, in the past and in the ORACLES measurements, relied on filter based measurements for absorption and nephelometers for scattering (authors' Figure 4). These instruments have relatively large uncertainties associated with them because of corrections needed for scattering and absorbing artefacts (e.g. Cappa et al., 2008). Much more accurate measurement systems such as Cavity Ring Down (extinction) and Photo Acoustic Spectrometry have been developed.

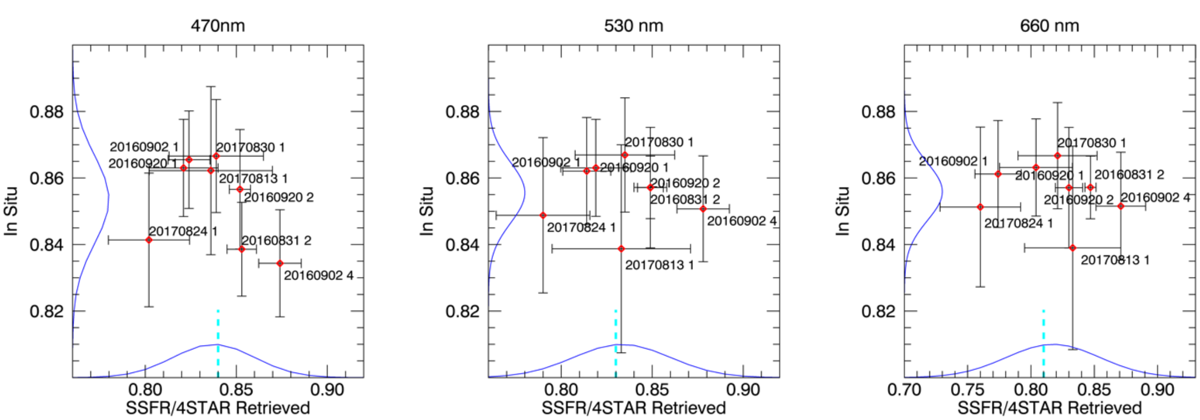
Cappa, C. D., Lack, D. A., Burkholder, J. B., and Ravishankara, A. R.: Bias in filter-based aerosol light absorption measurements due to organic aerosol loading: Evidence from laboratory measurements, Aerosol Sci. Technol., 42, 1022–1032, https://doi.org/10.1080/02786820802389285, 2008.

I know that it is difficult to keep up with the contemporary literature, particularly with the concentrated efforts over the SE Atlantic region, but there is some recent work from the CLARIFY- 2017 team that very much supports the values of the SSA (and the wavelength dependence). I would suggest adding something like this:-

“New, more accurate, cavity ring down and photo acoustic spectrometry instrumentation has recently been deployed to the SE Atlantic during the CLARIFY-2017 deployment. Davies *et al*. (2019) performed an analysis of the SSA of aerosol dominated by biomass burning aerosol using such instrumentation and found mean SSA values of 0.84, 0.83 and 0.81 at interpolated wavelengths of 467, 528, and 652 nm respectively. Wu *et al*. (2020a) extended this analysis by examining the BBA in the free troposphere, finding a mean and variability in BBA SSA of 0.85 ± 0.02 (1stdev) and 0.82±0.04 (1stdev) at 405 and 658 nm with evidence that the BBA at higher altitudes in the free troposphere is less absorbing. These results appear entirely consistent with those derived here.”

The authors should consider including a combination of the Davies et al. (2019)/Wu et al. (2020) paper on their Figure 4 as the agreement is so good.....Of course, you would have to caveat this with the fact that there are different temporal and geographical sampling regions etc.

Thank you for this comment and we apologize for our unintentional exclusion of the other SEA campaigns beyond ORACLES. We have included the suggested text on line 360. In figure 4, we will add an additional indicator of the Davies et al., 2019 results. We choose not to include the Wu et al., 2020 results since the wavelengths are not as similar.



Point 2: Line 234. The RT calculations themselves have a degree of uncertainty associated with them. For example, I note that the aerosol is characterized by the asymmetry factor. Does this mean that the higher order moments of the phase function are not accounted for? Is the RT code 2-stream? Is delta-Eddington rescaling applied? How is the surface reflectance modelled? A few more details would be appropriate here as would some acknowledgement that radiative transfer models that treat aerosols have their own inherent uncertainty (e.g. Boucher et al., 1999).

Thank you for pointing out the lack of detail surrounding the RT calculations within the manuscript. The RT calculations are performed with a multi stream RT model, disort, run with 6 streams. There is no delta-Eddington rescaling. The model assumes a Henyey-Greenstein (HG) phase function with our retrieved asymmetry parameter as input for the DARE calculations. We do not at all claim here that HG is, in fact, the actual phase function. On the contrary, the real phase function most likely significantly deviates from HG. However, since we retrieve g from *irradiance* observations, using disort, and then we use this same disort to calculate DARE, HG(g) *represents* the real phase function sufficiently well for our purposes. As you know, the first moment of the phase function along with a parametric HG phase function is sufficient to calculate fluxes with a two-stream approximation, whereas higher moments are required when using higher-stream RT models (including disort). The higher moments of the phase function can be generated from g (higher powers of it. For example, the second moment is simply g^2). The retrieval uncertainty in g (provided in the manuscript) could be propagated into higher moments, but since they are derived from g for HG, this is not necessary here.

For the aerosol retrievals and DARE RT calculations, we simply set the "surface" to the level of the cloud and define the albedo as the spectral cloud albedo. Therefore, we do not model the surface in the main calculations within the manuscript. However, for the DARE parameterization, the spectral cloud albedo "grid" at each SZA was obtained through cloud optical thickness and effective radius retrievals.

The translation from the mid-visible albedo to the spectral albedo (described in Appendix A.3.2) starts with the actually measured albedo spectrum. From that, COD and REF are retrieved, from which the spectrum is calculated. For the other albedos in the "grid", the COD is simply varied such that the albedo at the mid-visible wavelength changes as needed (while changing all of the other wavelengths as well).The COT/REF retrieval uses two wavelengths (860 and 1630 nm) to construct the lookup table, with no aerosol since the retrieval is done on the below aerosol, above cloud measurements. In these calculations, the surface for the cloud retrievals is standard Lambertian with an albedo value of 0.03. The COT range begins at 0, and this translates to a 0 "surface" albedo for the parameterization.

To address this comment in the manuscript, we will add the following information to this section in the paper:

Line 242: The RTM is run with 6 streams, assumes a Henyey Greenstein Phase function, and no delta-Eddington scaling is applied, all of which contribute to the inherent uncertainty within the RTM (Boucher et al., 1999).

Line 252: the RTM ingests the spectral cloud top albedo from SSFR (set at the altitude of the above-cloud leg, around 2km)

We should add that these simplifications were made to combine as many legs (cases) as possible in a common framework. In terms of radiative transfer, an opaque cloud effectively replaces the surface as boundary condition. However, for partially transparent clouds, the surface contributes to the reflectance. On the other hand, DARE is the *difference* between the fluxes with/without the aerosol above the altitude at which the albedo is prescribed in the model, and the approximations we used should therefore be insignificant compared to other uncertainties. Finally, it is acknowledged that clouds do not exhibit a Lambertian albedo. However, for irradiance calculations, the cloud albedo (non-Lambertian) can be substituted with a Lambertian albedo. We will add a brief clarification on these subtleties in the revised manuscript, provided our explanations here will be acceptable as path forward.

Point 3: The authors chose a SZA of 20degrees to demonstrate the RT calculations and the parameterization fits. As demonstration purposes, this is OK, but there should be an acknowledgement that, when comparing to models, the DRE is typically calculated over the full range of SZAs that are experienced in the region and then diurnally averaged.

Thank you for bringing this up. To make it clear that our parameterizations are instantaneous, the sentence preceding eq 3 and eq 4 will be changed from "ORACLES measurements are used collectively to develop two parameterizations in the form of:" to "ORACLES measurements are used collectively to develop two parameterizations of instantaneous DARE in the form of:" In addition, we will add the caveat to figure 5: "It should be noted that a 20° SZA is not representative of the mean in the region." We will further add that in the first application of our parameterization, the diurnal integration will be done (see also our response regarding applications below).

Minor typos/clarifications

L24: spanned -> determined

The abstract has been rewritten (below, in blue) to better reflect the main goals of the paper. This wording no longer remains.

Revised Abstract: In this manuscript, we use observations from the NASA ORACLES (ObseRvations of CLouds above Aerosols and their intEractionS) aircraft campaign to develop a framework by way of two parameterizations that establishes regionally representative relationships between aerosol-cloud properties and their radiative effects. These relationships rely on new spectral aerosol property retrievals of the single scattering albedo (SSA) and asymmetry parameter (ASY). The retrievals capture the natural variability of the study region as sampled, and both were found to be fairly narrowly constrained (SSA: 0.83 ± 0.03 in the mid-visible, 532 nm; ASY: 0.54 ± 0.06 at 532 nm). The spectral retrievals are well suited to calculate the direct aerosol radiative effect (DARE) since SSA and ASY are tied directly to the irradiance measured in presence of aerosols – one of the inputs to the spectral DARE.

The framework allows for entire campaigns to be generalized into a set of parameterizations. For a range of solar zenith angles, it links the broadband DARE to the mid-visible aerosol optical thickness (AOD) τ and the albedo α of the underlying scene (either clouds or clear sky) by way of the first parameterization: $P(\tau, \alpha)$. For ORACLES, the majority of the case-to-case variability of the broadband DARE is attributable to the dependence on the two driving parameters of $P(\tau, \alpha)$. A second, extended, parameterization $PX(\tau, \alpha, \varpi)$ explains even more of the case-to-case variability by introducing the mid-visible SSA ϖ as third parameter. These parameterizations establish a direct link from two or three mid-visible (narrowband) parameters to the broadband DARE, implicitly accounting for the underlying spectral dependencies of its drivers. They circumvent some of the assumptions when calculating DARE from satellite products, or in a modeling context. For example, the DARE dependence on aerosol microphysical properties is not explicit in P or PX because the asymmetry parameter varies too little from case to case to translate into appreciable DARE variability. While these particular DARE parameterizations only represent the ORACLES data, they raise the prospect of generalizing the framework to other regions.

L38: just off -> off (as the Sc extends 1000km....)

This will be adjusted.

L48-49: "In a region like the southeast Atlantic, this makes determining DARE challenging since the cloud fields change rapidly". I would suggest adding some idea of why the cloud changes rapidly - not only because of cloud dynamics, but because the cloud field advection is dominated by the flow in the MBL while the aerosol advection is dominated by the flow in the residual continental marine boundary layer which is frequently in the opposite direction.

Thank you, additional details will be included here.

L52: Chand was not the first to coin the phrase critical surface albedo. I'd suggest adding Haywood and Shine, 1995 reference here (Haywood, J.M., and Shine, K.P., 1995. The effect of anthropogenic sulfate and soot aerosol on the clear sky planetary radiation budget. Geophys. Res. Letts., 22, 5, 603- 606; see their Fig 1).

Reference will be added. Thank you for pointing out our oversight.

L75 (and probably other instances) aerosol optical depth -> AOD as you've already defined it

Thank you, this will be updated.

I like Figure 1. It captures the essence of the filters and the criteria.

Thank you.

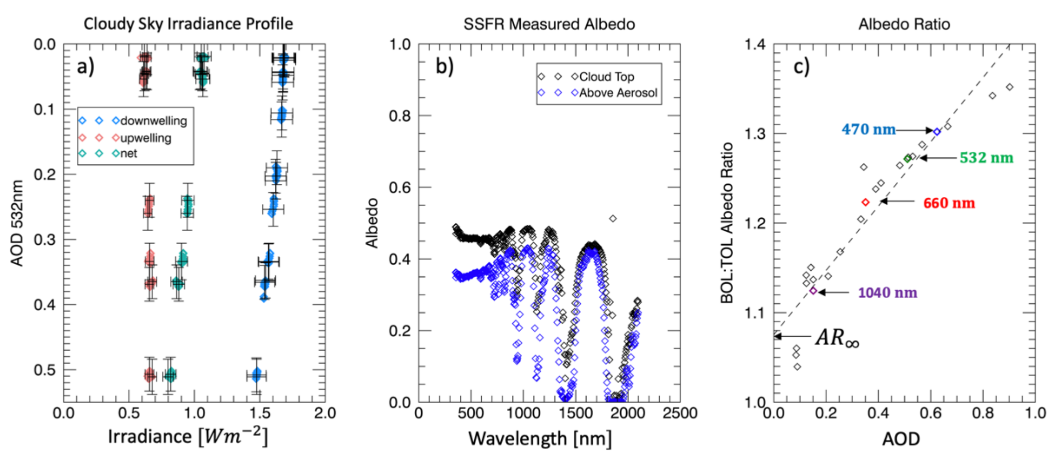
L148: Figure 3a -> Figure 2a.

Figure reference will be updated.

Caption for Figure 2. “c) The ratio between the BOL and TOL albedo spectra shown against the BOL AOD spectrum.” I think that this needs a little more explanation. Presumably the AOD is 532nm? What about the albedo – is this the broadband albedo (i.e. weighted by the solar flux)?

Figure 2c shows the ratio between the two albedo spectra from 2b (BOL and TOL SSFR spectral albedo) as a function of the 4STAR AOD spectrum at the BOL. To make this more clear, the figure caption will be updated to: “c) The ratio between the BOL and TOL albedo spectra (taken from Fig 2b) shown against the BOL AOD spectrum at the 4STAR wavelengths. The intercept of the fit line is criteria 3 (AR_∞); if the intercept deviates largely from 1.0, the case cannot be used for an aerosol retrieval. Select wavelengths are labelled to highlight the spectral importance of this method.”

In addition, we will update the figure to highlight a few wavelengths so it is clear that the data shown is spectral. Below is the proposed figure:



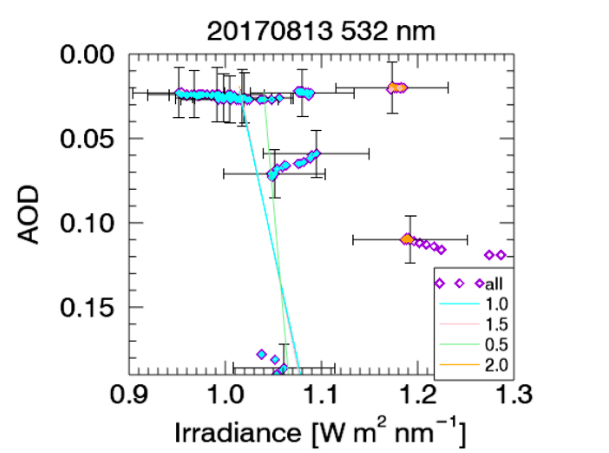
L163-L164: “The filter, which is applied to the upwelling profile, retains only those data within the 68% confidence interval (1 sigma) of the linear fit line”. This is fine if the error distribution is Gaussian, but is it? It would be worth checking that this is the case. It seems as though the two cases where the original filtering method is retained may not be Gaussian which might give you a reason for applying a different filtering method.

Technically, the measurement data should be Gaussian, since Gaussian is essentially measurement repeatability with some variability. For the stationary condition, if we are measuring over the same surface the measurements should be Gaussian distributed, or if noise is present, that should be Gaussian distributed. However, if the cloud changes within the measurement period, one no longer measures the same thing, and that will be reflected in the data. The idea is that through the filtering, we get rid of anything that falls outside of that Gaussian envelope since the data become too unlikely.

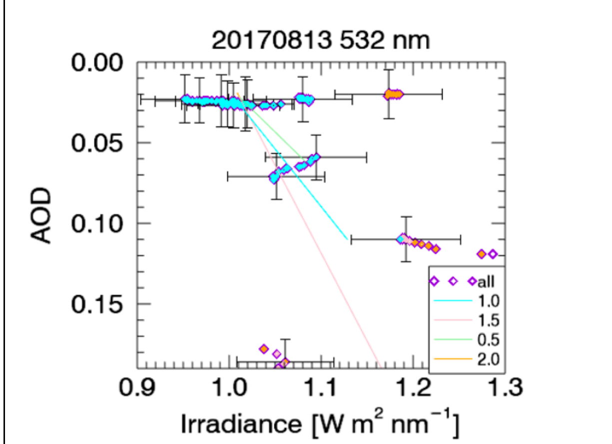
In the original filtering method (from Cochrane et al., 2019), the 1-sigma standard deviation limit is based on the mean value of the profile. The updated filtering method uses the 1-sigma standard deviation based on the *linear fit* of the profile. In most cases, the new filtering provided better outcomes for the quality criteria (C1, C2, C3 in Fig 1) that follow the filtering, indicating that the new filter was eliminating more of the outlying data and retaining the quality data. However, in the exception cases, we found the opposite and the new filter resulted in poorer values of the quality criteria. In these cases, we reverted back to the old filter method.

Below are the comparisons between the new and old filtering below (shown for the two exception cases that pass the criteria for a retrieval and one for a standard case that uses the new filtering). The cyan color is data within the 1 sigma fit and therefore retained; the lighter colors (pink, orange, purple) are data that do not pass the filter and not retained. In the first case, the new filter eliminates the highest AOD data. In the second case, the new filter retains too much variability at the highest AOD (lowest altitude).

Case 1:

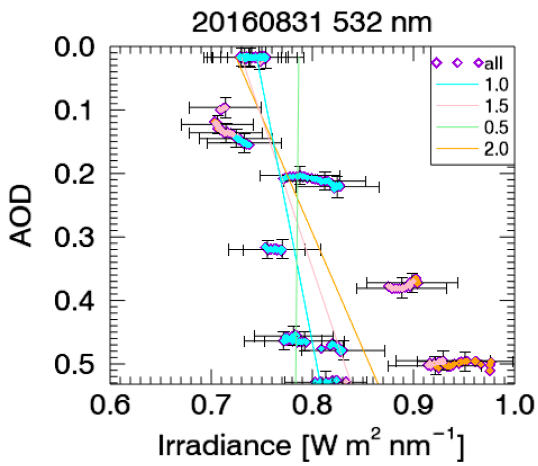


Original fit

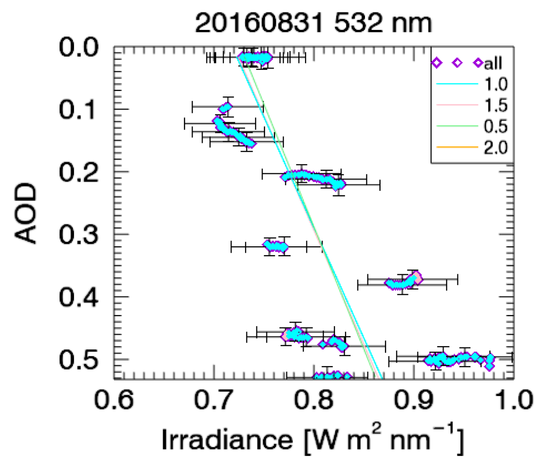


New fit

Case 2:

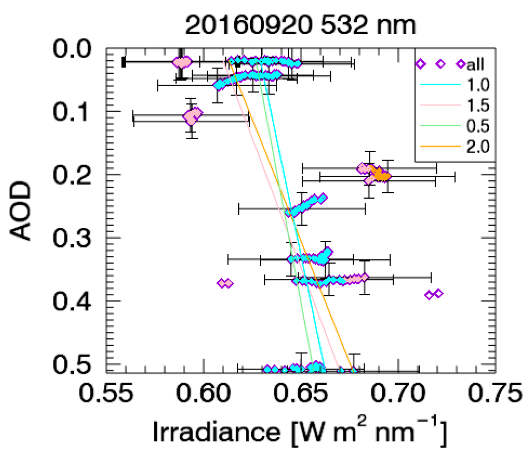


Original fit

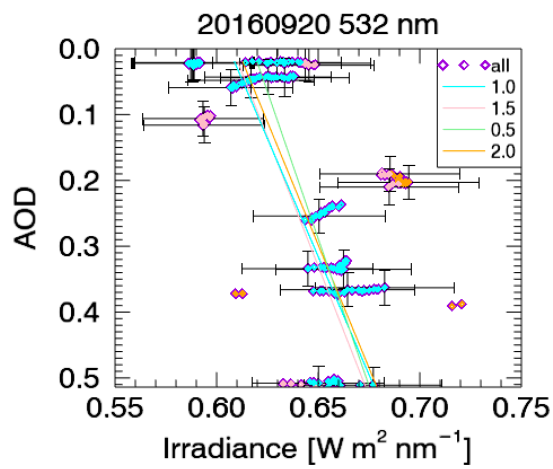


New fit

In most cases, the difference between the two filtering methods is very small. Below is an example of a standard case for which the new fit is used.



Original fit



New fit

Line 255. I was initially concerned that the retrieval algorithms rely on a mean solar zenith angle. The spirals typically take 30mins to achieve. This means that the SZA could be 15minutes out at the TOL and BOL. I reckon that 15minutes is approximately equivalent to 3.75degrees error is SZA given that the sza changes by around 15degrees per hour. So assuming a mean SZA of 30degrees (Table 2) could give an error of around 4% in the fluxes, or around 40Wm-2 assuming that you'd get around 1000Wm- 2 from the product of the solar constant and atmospheric transmission. However, I note from an earlier section (line 152) that the fluxes are corrected according to Equation 3 of Cochrane et al. (2019); which is quite easy to miss on a first read. This therefore really needs to be reiterated here so emphasize that the observations are corrected and the observations and the modelling are therefore consistent.

We will amend this to: "As mentioned in section 2.2.1, the irradiances are corrected to the SZA at the midpoint of the spiral to account for the changing solar position during the spiral. For consistency, the SZA within the RTM is set to the same SZA of the spiral midpoint."

L269: "We focus on the TOL calculations since radiative effects can be directly related to radiative balance at the TOL (Matus et al., 2015)." I think that it's better to say that the TOL calculations will resemble those calculated at the tropopause which is used as a metric for the cooling/warming impact of aerosols (e.g. Forster et al., 2007 which you already reference).

Thank you for pointing this out. Indeed, our TOL results should certainly resemble the top of the troposphere more than the entire column. This will be updated.

L281: is DARE -> is the DARE

This will be updated.

L286: Russel -> Russell; deGraaf -> de Graaf

Thank you for catching this error, it will be corrected.

L288: "no studies that we are aware of have generalized RFE to account for these complexities in a quantitative framework." Be careful here. General circulation modelling studies can (and do) turn out these numbers on a regular basis simply by taking the all sky DARE and dividing it by the AOD, which implicitly has all of the detailed RT calculations implicit within it. It is quoted numerous times for different aerosol types for a multitude of e.g. AEROCOM simulations. You could also argue that above cloud satellite retrieval estimates have also implicitly accounted for this in their look-up-tables (e.g. de Graaf et al., 2012, which you already cite). What you mean is that "no detailed observational based studies"......

Based on your comment, it is clear that our intended meaning is not expressed through this statement and we therefore have decided to remove it from the manuscript. Rather than the RFE itself, this statement was intended to pertain to the parameterization, going from a single-parameter concept (RFE, used with AOD) to multiple parameters (as the ones driving P/PX, i.e., AOD, albedo, SSA).

L295: Again, some care is needed here: "has the significant advantage that the complexities of transitioning from narrowband to broadband for many parameters are incorporated into the parameterization coefficients, allowing for use across large spatial scales since minimal information is required". If you have a different type of aerosol, or your aerosol is mixed with mineral dust (as it frequently is in west Africa), then your algorithm will fail because the above cloud AOD and the DARE will differ when compared to BBA alone. This note of caution needs to be included I think – easiest way is to tone down the "large" spatial scales, which is semi-quantitative to "regional".

We agree that caution is needed in the application of the parameterization to larger regions, since with a different aerosol type the coefficients will no longer be valid. We will make this more clear by changing the text to: "has the significant advantage that the complexities of transitioning from narrowband to broadband for many parameters are incorporated into the parameterization coefficients, allowing for use across regional spatial scales for biomass burning aerosol since minimal information is required as input. Of course, the parameterization is only applicable for the region where the measurements were taken. It also cannot be generalized to apply for a different aerosol type."

L330: Russel -> Russell

This will be updated.

L356: Might want to include reference to the Wu et al. (2020) paper here as that suggests that there is a variation in the vertical profile of SSA.

Reference will be added, thank you.

L360: (scene or cloud albedo). I'm a little confused – is the scene albedo, the albedo with the aerosol and the cloud in it, while the cloud albedo is the albedo of the cloud layer alone? The two must differ otherwise the aerosol is having no effect. A few words of clarification would be appropriate.

When referring to scene albedo, we mean that as the scene *below* the aerosol layer. This work, especially in relation to the parameterizations, does not focus on the albedo measured from above the aerosol layer. For more clarity, line 86 in the paper will be changed from "The 550 nm albedo is the albedo of the scene below the aerosol layer and the SSA is a measure of aerosol absorption." to "The 550 nm albedo is the albedo of the scene below the aerosol layer (open ocean and/or cloudy scene), and the SSA is a measure of aerosol absorption."

We differentiate between scene and cloud in this instance because the parameterizations include albedo values down to 0. Again for more clarity, we will amend this phrase to: (scene or cloud albedo below the aerosol layer). See also our response above how cloud-free scenes are handled in the radiative transfer.

L382 and Figure 5. The authors have tended to slip into the terminology of "surface albedo" which tends to mean the physical reflectance of the surface rather than the "effective underlying albedo" (i.e. the albedo of the combined Rayleigh scattering, cloud, MBL aerosol and surface) which is I think what the authors mean. Again, this should be clarified. Is there an explicit assumption that the underlying albedo is Lambertian? While this might be a decent approximation for heterogeneous thick cloud, is it sufficient for the sea-surface reflectance? What impact would this have?

There are several figures where the albedo goes down to zero, but Figure 10 suggested a minimum surface albedo of around 3.5%, which is similar to a 'real' ocean surface – some more explanation is warranted I think.

This is a very good point. All mentions of surface albedo in the manuscript will be changed to underlying albedo.

All of the albedo spectra in the paper are at cloud level, around 2 km, so they do include Rayleigh scattering. We realize Rayleigh scattering will change with altitude, but within the parameterization that change will be small (since the altitude variation between cases is small) and only would matter for the albedo translation. Within the radiative transfer calculations (as described under response to main point #2), the 'clear sky' 0 albedo is the albedo in the limit of 0 cloud optical thickness, and not a true clear sky with a 0 surface albedo. There is still a cloud with an optical thickness of 0 and Lambertian assumption.

Also, it is acknowledged that a sea surface is even less of a Lambertian reflector than a cloud (one only needs to think of sun glint). However, this is precisely the simplification that we made to fit both cloudy and cloud-free skies into a common framework. In a sense, one could call our albedo an "effective" albedo, which represents the true surface reflectance assuming a Lambertian reflectance distribution. We will add a statement in the manuscript about this point.

Finally, since we are looking at DARE (the difference of fluxes) rather than the fluxes themselves, our simplifications should lead to only negligible effects relative to the contributing measurement uncertainties.

L385-386. Fig 7 is referred to before Figure 6. Easy to sort out by swapping the order of this sentence.

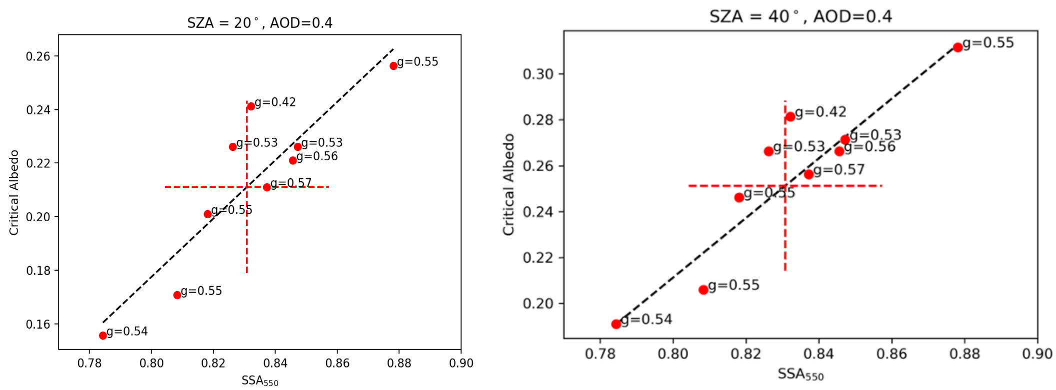
The sentence order will be switched.

L405: "and rather strongly on the SZA (not shown; for example, it can attain 0.6 at low Sun elevations)." I think it would be appropriate to include a reference here – the classical reference for the dependence of the DRE on the radiative forcing is Boucher et al (1999) although this is for sulfate aerosol.

Thank you, reference will be added.

One of the problems with presenting the critical SSA for a solar zenith angle of 20degrees is this rather strong solar zenith angle dependence. The values at 20degrees (close to local noon) is not likely to be representative of the mean SZA that is experienced in the region. A caveat to in this regard would be appropriate.

Thank you, we will add in this caveat into the caption of figure 5 (since this is the first figure shown at 20 degree SZA.) For your reference, below are the critical SSA figures for SZA=20 and SZA=40:



L 490: “We cannot judge whether our approach will be useful for predictive models, ... “. I agree that there are issues with whether the community will take up the parameterized approaches the RT calculations are a fundamental necessity. You might want to more explicitly suggest that a combination of lidar derived AODs and scene albedos from e.g. the geostationary GERB instrument or similar for future assessments of biomass burning DRE.

Thank you for this suggestion. In fact, we are working on an application paper that does just what you are suggesting here! We will include the additional statement: “However, current work implementing our approach using albedo data from the geostationary Spinning Enhanced Visible and Infrared Imager (SEVIRI) in combination with ORACLES AOD data from HSRL-2 and 4STAR is already underway (Chen et al., 2020 in preparation).”

L494: “At the very least, the SSA and asymmetry parameter retrievals coming out of our and other ORACLES studies will constrain the aerosol optical properties in a range of models”. Again I agree – the text and references that document the very encouraging agreement between the absolute values of the SSA and the spectral dependence of the SSA and those from higher accuracy CRD and PAS measurement systems coming out of CLARIFY-2017 should again be noted here I think. I would also suggest changing “ORACLES studies” to “ORACLES/LASIC/CLARIFY-2017/AEROCLO-Sa studies (Zuidema et al., 2016)”.

We agree that this should be highlighted. The text will be updated to: “At the very least, the agreement between the absolute values and spectral dependence of the SSA and asymmetry parameter retrievals coming out of our and other ORACLES/LASIC/CLARIFY-2017/AEROCLO-Sa studies (Zuidema et al., 2016) such as Davies et al. (2019) and Wu et al. (2020) will provide robust and consistent constraints of the aerosol optical properties in a range of models.”

References :

Boucher, O., et al., 1998. Intercomparison of models representing direct shortwave radiative forcing by sulphate aerosols. J. Geophys. Res., 103, 16979-16998.

Davies, N.W., C. Fox, K. Szpek, M.I. Cotterell, J.W. Taylor, J.D. Allan, P.I. Williams, J. Trembath, J.M. Haywood, and J.M. Langridge, Evaluating biases in filter-based aerosol absorption measurements using photoacoustic spectroscopy, Atmos. Meas. Tech., 12, 3417-3434, DOI: 10.5194/amt-12-3417- 2019, 2019.

Wu, H., J.W. Taylor, K. Szpek, J. Langridge, P.I. Williams, M. Flynn, J.D. Allan, S.J. Abel, J. Pitt, M.I. Cotterell, C. Fox, N.W. Davies, J. Haywood, H. Coe, Vertical and temporal variability of the properties of transported biomass burning aerosol over the southeast Atlantic during CLARIFY-2017, doi:10.5194/acp-2020-197, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-197, 2020.