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.

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).

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.

Minor typos/clarifications

L24: spanned -> determined

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

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.

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).

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

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

L148: Figure 3a -> Figure 2a.

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)?

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.

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.

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).

L281: is DARE -> is the DARE

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

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"……

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".

L330: Russel -> Russell

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.

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.

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.

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

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.

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.

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.

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)".

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.

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