

Dear reviewer,

Thank you for your thoughtful and constructive review of the manuscript. In the following paragraphs, we have summarized and responded to what we understand to be the main points of concern. We have also provided specific responses including plots to the individual minor comments.

Note: the color coding for the edit markings in the document are as follows:

Blue: directly addresses a comment from Reviewer #1

Green: directly addresses a comment from Reviewer #2

Red: additional update made by Authors

General comments/approach

It is evident from your comments that we are lacking clarity in the motivation of developing a DARE parameterization, which is key to the success of the paper. We appreciate this comment and acknowledge that the purpose was not made clear throughout the manuscript. The major advantage of developing the DARE parameterization is to encapsulate the transition from narrowband to broadband such that spectral aerosol and cloud properties are not required. The parameterizations require only one wavelength of input parameters and provides output of broadband DARE. Our goal was not to create an alternative to well-established radiative transfer formulae such as those presented in Coakley 1975, nor to replace full RT models such as libRadtran (with its solver “disort” created by Warren Wiscombe) [See our changes/additions on lines 99-101]. Rather, the difficulty concerns the *spectral* aerosol and cloud properties and their relationship to the *broadband* DARE. One cannot simply translate SSA, g , cloud albedo, etc. at a single wavelength into broadband DARE since their spectral dependencies are almost as important as the narrow-band single-wavelength values. Established formulae that are grounded in the physics (such as Coakley’s or other 2-stream approximations) still require spectral properties and subsequent broadband integration. Here, we replace the need for a full spectrum with narrow-band quantities at a wavelength in the mid-visible wavelength range where they are often available, and we also side-step the spectral integration. This, rather than the radiative transfer itself, is the essence of this work. More generally, we cast the collective radiative observations into an ORACLES-wide set of formulae and “resolve” the case-to-case DARE variability in terms of only two (or three) driving parameters. To express this better, we re-wrote the abstract of the paper (included in blue font below), and made changes in other places of the manuscript (e.g., lines 99-101).

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) and the albedo α of the underlying scene (either clouds or clear sky) by way of the first parameterization: $P(\text{AOD}, \alpha)$. 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 $P(\text{AOD}, \alpha)$. A second, extended, parameterization $PX(\text{AOD}, \alpha, \text{SSA})$ 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.

The *application* of the parameterizations has been made clearer in the revised manuscript, with the important addition of explicitly stating how this is already being used to obtain diurnally-integrated values of DARE for the study region (lines 514-519). We are actively working on a publication that uses the parameterization to establish diurnally-integrated DARE for the study region, based on satellite observations and aircraft observations of AOD and albedo. It is important to note, however, that the parameterizations were not developed for regions *beyond* that of ORACLES, since as you pointed out, the physics in other regions are most likely different and ours is an empirical formula that applies only to the data it is based on (we alluded to aerosol microphysics specifically in the revised abstract quoted above). While we recognize that the number of cases is small, the range of properties they cover is likely sufficiently representative of the conditions in the study region for developing the parameterization scheme. As more cases become available, even those falling outside of the envelope of, say, previously observed single scattering albedo, the *relationship between* mid-visible aerosol properties (AOD, SSA, asymmetry parameter), albedo on the one hand, and broadband DARE on the other would likely be similar as well. The advantage of our framework is that it can be continually updated. This will be done, for example, with the aforementioned paper that we are currently working on. Whereas the present paper only considers ORACLES 2016 and 2017, the follow-up paper extends the parameterization to ORACLES 2018, prior to the application of the parameterization to the calculation of campaign-averaged DARE. Data from other campaigns (e.g., from CLARIFY or even SAFARI) could similarly be included.

We understand that the two parts of the paper seem distinct from one another, and this is done purposefully. The manuscript is the second part to its companion paper, Cochrane et al., 2019, and they are meant to be read in succession. However, it was important that we include enough detail in the first part of the paper on the retrieval algorithm such that it can be understood at a surface level without reading the 2019 companion paper, as well as to make clear the changes made to the algorithm relative to the version described in the 2019 paper. The most important change of the 2020 version of the algorithm relative to its 2019 predecessor is that the data are automatically filtered and conditioned in a consistent manner. This allows ingesting of data without different treatment of cases and thereby biasing the results. In addition, part I of the current paper provides a data product on its own: the aerosol property retrievals of SSA and g . This database of these retrievals, though small, is important to include along with the parameterization of DARE as presented in part II.

To address your concerns, here is what we have done within the revised manuscript:

First, the abstract has been revised to better reflect the overall goals of the paper (see draft above in blue). Second, the two parts of the paper are now delineated at the end of the introduction (Lines 115-119). Regarding the body of the paper: The original version did not effectively capture the main objectives of the paper and potentially left the reader with an incorrect impression of what the paper is about. In the revised version, the benefit of a direct transition from narrowband to broadband through our parameterization has been highlighted in greater detail (lines 309-314). In addition, the application of our parameterizations has been more explicitly described (using the next paper as example; lines 514-519). We have made the separation between part I and part II of the paper even more apparent, making it possible to skip the aerosol property retrieval section and move directly into the DARE section (lines 115-119; section headings). However, we are hesitant to significantly reduce this section because of its value to the southeast Atlantic region research community.

Thank you again for your review of the paper and we hope that our changes have addressed your concerns satisfactorily.

With the above comments, we believe that we addressed the general (major) comments. Our response to sequential (minor) comments is below (in blue).

Minor comments:

Line 24: Does the “scene albedo” actually mean cloud albedo? If it really means “scene albedo” then what types of scenes (ocean, land, snow etc.) have been included?

ORACLES data was exclusively taken over the southeast Atlantic ocean. We use “scene albedo” rather than “cloud albedo” since we encountered scenes that included both cloudy and clear sky (partially cloudy or area contained cloud holes.) This is clarified in the revised manuscript at line 92 (and described in the newly drafted abstract quoted above).

Line 86: similarly, it should be clarified here if “scene albedo” actually just means “cloud albedo”. Note that the spectral signature of land reflectance/albedo is very different from cloud albedo.

Thank you for pointing out this ambiguity. For more clarity, line 92 has been 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.” A detailed discussion of the albedo has also been included in Appendix A.3.2.

Eq. (3) and (4): It should be pointed out explicitly if these equations are for instantaneous or diurnal averaged DARE. Also why is the dependence of DARE on solar zenith angle omitted in these equations? SZA is part of the parameterization (Table 4b), no?

Thank you, the sentence preceding eq 3 and eq 4 (line 85) has been 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:”

The dependence of DARE on SZA is not included in these equations, since the parameterization coefficients themselves do not depend on SZA. Rather, the parameterization coefficients are calculated separately for a range of SZAs.

To what extent is the DARE dependent on atmospheric profiles, such as water vapor profiles? There are some recent studies that suggest a correlation between the presence of above-cloud smoke and an enhanced water vapor in the ORACLE region. Should this correlation be considered in the parameterization?

This is an important question. The dependence of DARE on atmospheric profiles, specifically water vapor, is implicitly included within the parameterization. Each case that the parameterization is based upon has the measured water vapor profile included in its DARE calculations.

While we do not determine the extent to which DARE depends on water vapor, it should not depend too much on it since DARE is defined as $F_{\text{net}}(\text{with aerosol}) - F_{\text{net}}(\text{without aerosol})$. The water vapor profile for both terms was held fixed (as determined from observations for each of the cases), and therefore only has a bearing on DARE if it affects the incident radiation on top of the aerosol layer. Compared to the impact of the scene AOD, albedo, and SSA on the case-to-case DARE variability, the water vapor profile variability (as well as the aerosol microphysics variability) was negligible. However, the water vapor does have a bearing on the total heating rates, which is explored in a separate paper (in progress; if submitted before this paper is accepted, we will include a reference in the appropriate place)

Cochrane et al., 2019, has been cited many times in the paper, often in different formats. Please be consistent and also considering use abbreviation e.g., C19 to refer to Cochrane et al., 2019.

Thank you for catching this. The references to Cochrane et al., 2019 have been abbreviated to C19 (defined upon its first occurrence).

Line 148: Figure 3 should be Figure 2.

Thank you. This has been updated in line 156.

Eq. (5) and (6), why are the parameters a_{λ} and b_{λ} the same for upward and downward irradiances? What is the underlying physics?

Thank you for pointing this out, as our notation is ambiguous. $a_{\lambda}^{\uparrow}$ is not the same as $a_{\lambda}^{\downarrow}$ (similar for $b_{\lambda}^{\uparrow}$ and $b_{\lambda}^{\downarrow}$; equation 6 has been changed to $c_{\lambda}^{\uparrow}$ and $d_{\lambda}^{\downarrow}$ (line 171).

Again, Section 2.2.1 and 2.2.2 seem to be a replay of Cochrane et al., 2019. They should be put in the Appendix or substantially shortened.

As noted in the response to the general comments, we understand that the two parts of the paper seem distinct from one another, and this is done purposefully. The manuscript is the second part to its companion paper, Cochrane et al., 2019, and they are meant to be read in succession. However, it is important that we include enough detail in the first part of the paper on the retrieval algorithm such that it can be understood at a surface level without reading the 2019 companion paper, as well as to make clear the changes made to the algorithm relative to the version described in the 2019 paper. The most important change of the 2020 version of the algorithm relative to its 2019 predecessor is that the data are automatically filtered and conditioned in a consistent manner. This allows ingesting of data without different treatment of cases and thereby biasing the results. In addition, part I of the current paper provides a data product on its own: the aerosol property retrievals of SSA and g . This database of these retrievals, though small, is important to include along with the parameterization of DARE as presented in part II.

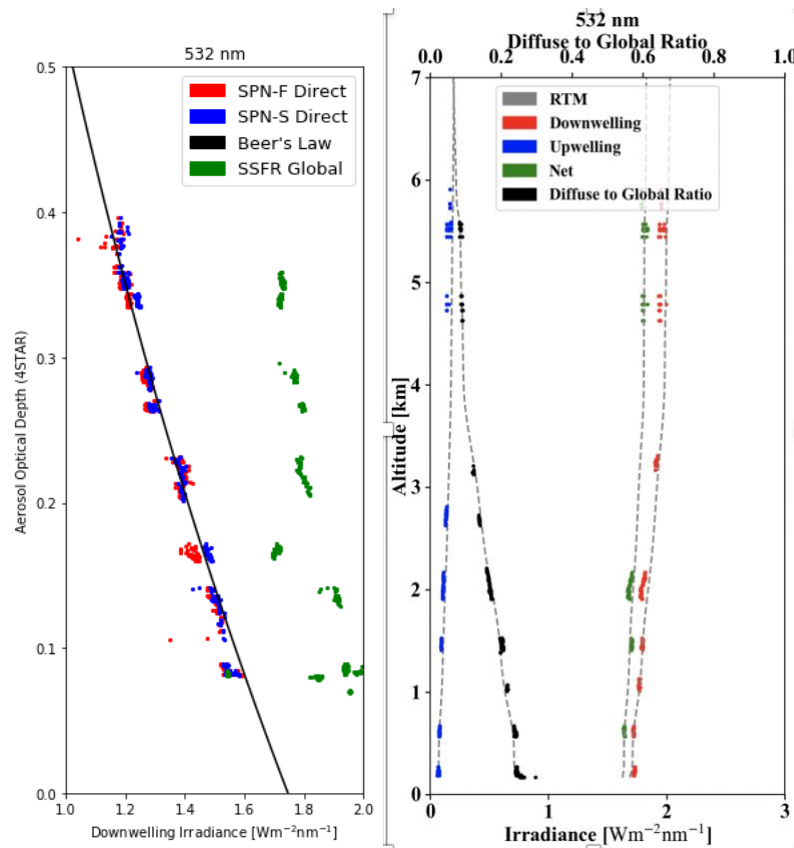
To address your concern, we have added additional text on lines 115-119 to better describe the two parts of the paper and have updated the section headings to reflect the separation (line 326).

Around line 150, this part is confusing and needs detail explanation. For example, “both upwelling (F_{up}) and downwelling (F_{dn}) irradiance profiles have an approximately linear relationship to AOD due to the absorption and scattering of the aerosol layer.” Shouldn’t the downwelling (F_{dn}) be exponential with AOD as a result of Beer’s law? “Any deviation from the linear relationship is attributed to changes in the underlying cloud” Why? Can’t the vertical variation of aerosol properties, e.g., SSA and/or g cause deviation from the linear relationship? How does cloud cause the deviation? These questions need to be clarified.

We did not intend to imply that the direct downwelling irradiance should not follow Beer’s law. However, in the SSFR measurements we cannot separate the global irradiance into its diffuse and direct components. We are using the linear assumption for the global downwelling as a simplification only for initial fitting for the subsequent filtering. We agree with the reviewer here because indeed, deviations from the linear relationship could be due to non-linearities as expected from Beer’s law, or vertical dependencies of aerosol parameters. However, we expect these to be negligible compared to changes in the underlying clouds, and therefore use deviations from a linear profile – just to filter our data. Otherwise, the results of the paper (i.e., the retrievals and the calculations) are not predicted on any linearity assumptions.

As can be seen from the figures below, the direct-beam downwelling (labeled SPN-f, SPN-S) *does* follow an exponential (these measurements were only available during 2018, and they are therefore not included in this manuscript). However, the global downwelling (SSFR global, green) can be approximated with a linear fit, at least for the range of AOD we encountered. We should add that the thin-layer approximation introduced by, e.g., Coakley et al. (1975) shows a

linear dependence of the global reflectance analytically (“In the thin atmosphere approximation, the reflectivity and transmissivity of the layer are linear functions of its optical depth”). The figure below on the right shows the irradiance components as a function of altitude.



While vertical variation of the aerosol properties could theoretically affect the linear relationship that we assumed, but changes in the cloud properties below outweigh aerosol-induced deviations from a linear profile by far, and can therefore be used as the basis for filtering.

To address this comment, we have added the following additional text on lines 159-163:

“This linear assumption for the global downwelling is a simplification only for initial fitting for the subsequent filtering, and deviations from the linear relationship could be due to non-linearities as expected from Beer’s law, or vertical dependencies of aerosol parameters. However, we expect these to be negligible compared to changes in the underlying clouds, and therefore use deviations from a linear profile to filter our data.”

Does the retrieval algorithm assume H-G phase function? What are the higher-order terms of the phase function expansion other than asymmetry factor, g ? What is the uncertainty associated with the phase function assumption?

Yes, it does assume an Henyey-Greenstein (HG) phase function with our retrieved asymmetry parameter (the first moment of the actual phase function) as input. The first moment of the phase function along with a parametric HG phase function is sufficient to calculate fluxes with a two-stream approximation. Higher moments are required when using higher-stream RT models (including disort) and can be generated from g (higher powers of it; for example, the second moment is simply g^2). 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 deviates from HG. However, $HG(g)$ represents the real phase function sufficiently well for our purposes. Using Coakley '75 again as an example: He does show that there are differences in the reflectance when using a two-stream formula relative to a multi-stream (in this case, doubling adding) RT. However, these differences arose because they were looking at directional reflectance. In our case, we retrieve g from *irradiance* observations, using disort, and then we use this same disort to calculate DARE. The retrieval uncertainty in g (provided in the manuscript) could propagate into higher moments, but since they are trivially derived from g for HG (g^2 , g^3 , etc.), this is not necessary here.

The following additional text is included at lines 253/254:

“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).”

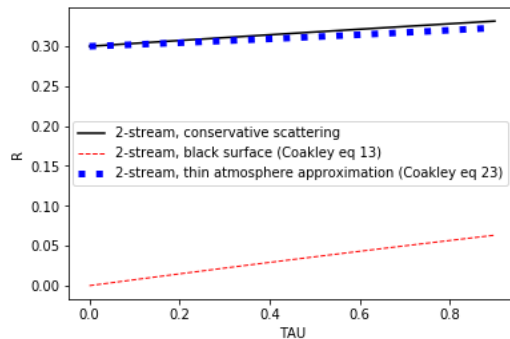
Line 286: Russel et al., 1997 should be Russell et al., 1997; deGraaf should be de Graaf

Thank you for pointing out these errors. We have removed this statement and the associated references due to a comment from another reviewer.

Eq. (12) and (13): The formula looks quite arbitrary. Is there any physics behind these polynomial parametrizations or are they only empirical? Note that there are some well-established 2-stream or 4-stream formula for layer reflection, e.g., Coakley (1975). Is it possible to draw some theoretical basis or physical meaning for the parameterization from these 2-stream or 4-stream approximations? Also, some previous studies have tried to use the concept of adding doubling to approximate the reflection of two layers (Lenoble 1985). Do you think these formulae might be helpful?

Thank you for this comment. Please see our general response above, in addition to our subsequent response. While the 2-stream (or 4-stream) formulae are appropriate for many applications, they are not suited for what we capture with our parameterization (see also general comments at the beginning of this response). What we have developed in this manuscript is not about the approximation, or creating a new approximation, but to directly represent the relationship between narrowband to broadband for which the spectral dependencies are implicit. The approximation formulas presented in Coakley (1975) would not work for our application because they would still require *spectral* properties which would need to be averaged or parameterized first, which is not as direct as our parameterization.

In addition, the approximations are not sufficiently accurate for our purposes since simplifications are required in these analytical formulae. For example, the thin layer approximation by Coakley '75, eq 23 (which does take absorption into account, for non-black surfaces) starts deviating even for fairly small optical thickness as can be seen in the figure below. Additionally, the non-linearity of DARE as a function of AOD (known from RTM calculations) would not be represented by the thin layer approximation.



To address your comment, we have added the following additional text on lines 99-101: “They are not meant to replace detailed or approximated radiative transfer calculations (e.g., Coakley 1975), which would require all these inputs, but rather to arrive at a broadband DARE with a minimum set of input parameters that drive its regional variability.”

Why is SZA dependence of DARE omitted in Eq. (12) and (13)?

The dependence of DARE on SZA is not included in these equations since the parameterization coefficients themselves do not depend on SZA. Rather, the parameterization coefficients are calculated separately for a range of SZAs.

Eq. (14) and (15): again, these parameterizations look arbitrary. Is there any underlying physics?

The underlying physics are only included insofar as that we parameterized broadband DARE as a functional dependence with respect to its driving parameters. The only way to transition from narrowband to broadband is through polynomial fitting; there is no direct way to include the physics of radiative transfer in aerosol layers above clouds, which has been well established since the mid-1970s. Of course, one could calculate the broadband DARE if all of the parameters were available, but that is not the point of this paper (see the other comments above). What we have developed in this manuscript is not about the approximation, or creating a new approximation, but to directly represent the relationship between narrowband to broadband for which the spectral dependencies are implicit. The analytical formulas which are based on physics do not work for our application because they would still require *spectral* properties which would need to be averaged or parameterized first, which is not as direct as our parameterization.

To make this more clear in the manuscript, additional text has been added on lines 99-101 and 309-314;

Eq. 21 – 24: I understand that dSSA term is introduced to make the parameterization scheme more general and more accurate. But as I mentioned above, a broadband RTM can easily compute the DARE given any type of SSA and g . Why bother developing such a complicated parameterization?

The parameterization removes the necessity of *spectral* SSA and g which are often difficult to obtain. We fully agree with the reviewer though: An RTM model would be more appropriate to compute the DARE *if* (!) spectral properties were known. However, most often spectral quantities are *not* known, unless they are tied to a fixed aerosol “type” that tabulates the spectral dependence (e.g., OPAC). More importantly, we show that *we do even not need to know these spectral details* as *explicit* inputs since just a few (2 or 3) mid-visible parameters explain the variability of DARE across the ORACLES cases we analyzed *to within the measurement uncertainty*. The spectral dependence of the parameters as measured/retrieved is thereby *implicitly* accounted for, and we can thus obtain broadband DARE with a minimum number of input parameters.

We have addressed this comment with revised abstract as well as the additional text on lines 99-101; 309-314; 522-528.

List of relevant changes:

- P.1-2, L. 21-39 (revised abstract)
- P.3, L. 91-92
- P.3, L. 99-101
- P.4, L. 110
- P.4, L. 115-119
- P.5, L. 157, 163, 164
- P.5, L. 158-162
- P.6, L. 170-173
- P.6, L. 177, 182
- P.7, L. 203, 211, 216
- P.8, L. 246
- P.8, L. 252-253
- P.9, L. 260
- P.10, L. 309-314
- P.11, L. 326
- P.17, L. 522-528

In addition to edits made according to the review comments, we have made the following updates:

- Added uncertainty estimates to table 4b.

- Updated the SSA extrapolation and description (figure A1a), requiring the RT calculations be re-calculated.
- Updated Figures 5/9/10/11/D1/D2 and tables 4a and 4b to reflect the new calculations
- Included supplementary material of code and coefficient files