

Dear Authors - Thank you for submitting your manuscript to ACP. I have reviewed the referees' comments and your responses to them, and have a few additional comments I would like you to consider before this manuscript goes to publication. The manuscript will not go back to the referees again. The page and line numbers of the comments are based on the Author's Response file you provided. It doesn't look to me that that shows the tracked changes to the original manuscript, as requested by the journal, but I can live with that. You may follow suit with your next revision but I would like to see your responses indicated point-by-point to the comments below.

Sincerely, Paquita Zuidema

Abstract, line 18: "this has yet to be quantified" is a bit strong. Radiative heating profiles have been calculated before, but not specifically for the air masses you are targeting. Please consider revising this sentence.

Abstract: My understanding is that acronyms should not be used or defined within the abstract unless substantially used. I only see SW and RRTM referred to twice, and my perception is that all of the acronyms can be removed from the abstract without loss of meaning.

Introduction, p. 25, first sentence: this sounds a little strange "...tend to underestimate the cloud fraction and cloud optical depth due to underestimates in cloud albedo and liquid water path..." as this is not really a cause and effect? One could leave off the phrase beginning with 'due to' with no loss of meaning.

p. 25 line 20-21: actually, close to the coast, the ORACLES campaign also highlights the presence of BBA in the boundary layer during August, e.g., Kacarab et al., 2020, ACP Special Issue. It might be good to qualify this sentence further, either with "initial space-based observations" or, "in September-October".

p. 26, line 25: the Lagrangian approach within Diamond et al 2018 was applied to the cloud layer, whereas in the current study, the aerosol layer is followed. These air flows are quite different.

Section 2.1: how are the authors treating orographic effects upon the clouds? And are MFRSR AODs, which come from the elevated AMF1 site, treated the same as the AERONET values taken at the airport? These will have a systematic bias.

p. 28: line 11 mentions MERRA2 thermodynamic profiles are used within the RT calculations, while line 27 indicates the INTERPSONDE profiles are used. Please clarify.

P. 29 top paragraph: the first sentence implies one SSA value was used while line 8 states 3 different SSA values were used. Please clarify.

P. 30 line 14-15: "Brown carbon tends to be more absorbing than organic carbon". Doesn't this depend on how brown carbon is defined? I should also note that the SSA values in Zuidema 2018 correspond to an RH of 45-65%, and to aerosols smaller than 1 micron in aerodynamic diameter, which will exclude the larger sea salt particles possessing a larger SSA.

Section 3 p. 30: Fig. 2: it seems strange to me that BC and OC are not collocated better, e.g., 6 Sept.. What do you make of this? Do BC and OC relate differently to RH within MERRA2 (though this is not the explanation I don't think) ?

Section 3 p. 30 Fig. 4: What's also clear about this is that back trajectories associated with higher AODs experience stronger winds. This is why their back trajectories extend further into Africa. Some of this is likely explained by the AEJ-S.

Section 4, Fig. 5: MERRA2 does not seem to capture the boundary layer decoupling at all. It would be worth mentioning how many levels MERRA2 is resolving within the boundary layer, either here or in Section 2.2

p. 31 line 23: are you sure the LASIC radiosondes have been assimilated into MERRA-2? That is quite valuable to know.

p. 31 line 24: it would be helpful to indicate contours for high MERRA-2 values of either BC or BrC (or some combination) on top of the moisture plot.

p. 32 line 14: "exhibit," -> "August 28-29, exhibit"

p. 32 line 18: "-1" needs to be superscripted.

p. 32 line 27: needs to be bolded and enlarged?

Section 5.1, here and in Fig. 9, caption: make clear there is no conditional sampling based on an AOD threshold applied. At least, I believe that is the case.

p. 33 line 17: "-1" needs to be superscripted. This occurs twice.

p. 33 lines 18-19: it would be good to report the RH here, as one would not expect aerosol swelling until a fairly high RH.

p. 33 line 26: the Shinozuka et al. 2020 model-observational comparison paper would be good to cite here as well, as it indicates a wide range of SSAs in use across models.

Section 5;2: several instances of '-2' that need to be superscripted.

p.35 line 24: Does this sentence need to be modified to indicate that it is small aerosols such as BBA that don't impact LW radiation, in contrast to large aerosol such as dust?

Section 5.2, Fig. 12: this indicates DARE is only a cooling, even when cloud is present. I am surprised by this. Does this mean the cloud optical depth or cloud albedo is so small, that a positive DARE can never be actualized? Is DARE being calculated relative to the cloud albedo? Please comment, an additional plot showing the TOA DARE as a function of the underlying albedo might even be a nice addition. If DARE is indeed always a cooling for the MICROBASE cloud properties, that is worth including in the abstract.

p.37 line 21 and line 25: superscript the '-1'

p. 38 line 24: the heating could also help drive an anomalous ascent. Something in fact must happen because the observed temperature profiles don't deviate from each other by say more than 2K, less from day to day when the synoptics are more similar.

p. 38 line 27-28: You could check with Art Sedlacek but I don't think there is evidence for tar balls over the southeast Atlantic.

p. 39, line 19-20: I'm not quite following is, is LW cooling at cloud top reduced because the warmer free-tropospheric temperatures emit more LW, for the same water vapor amount -the effect shown in Fig. 13? Or is it because of the increasing free-tropospheric opacity associated with the moisture colocated with the aerosol, as noted in the correspondence between Fig 2 and 6?

Table 3 caption: superscript '-1'

Overall: latitude and longitude indicators throughout need degree signs.

Check abstract again after you have finished to be sure you feel it captures your most salient findings.