"Pollution slightly enhances atmospheric cooling by low-level clouds in tropical West Africa", submitted to ACP by Valerian Hahn et al., 2022

We thank Referee #1 for his valuable comments and the effort that has been put in reviewing the submitted manuscript on the pollution effects and atmospheric cooling by low-level clouds in tropical West Africa.

General Comment 1:
A. Introduction: I don't understand the primary motivation for this study. There are lots of nice details about prior work but no clear story about what it all adds up to and where important knowledge gaps remain. Why is this new work needed? How does it relate to the previous work?
Answer General Comment 1:
We thank Referee#1 for the indication to emphasize the motivation of this study. To get a better understand of the basic idea for the analysis of the microphysics of low-level clouds in West Africa, one has to see the underlying scientific questions tied to the design of the DACCIWA campaign. As Knippertz et al. (2015) outline tropical West Africa faces a major population growth and urbanization. The effect of an increase in anthropogenic emissions is not only of concern in view of health aspects, but poses an uncertainty for future local climate. Knippertz et al. (2015) suggest that clouds above West Africa could be 'highly susceptible to increases in anthropogenic pollution'.
Previous studies found discrepancies in local weather models, also associated with a misrepresentation of multilayered cloud structures in this region.
The contribution of low-level clouds to the energy budget during the monsoon onset phase in West Africa was a significant scientific question to be answered by the DACCIWA campaign. This is where a knowledge gap was identified by the DACCIWA consortium und our study aims at contributing to fill this gap.
Our study can be very well embedded in the body of existing studies and expands their current state of knowledge within the scope of the DACCIWA campaign, such as Haslett et al. (2019), who analyzed the aerosol provenance, van der Linden et al. (2015) and Hill et al. (2018), who investigated the occurrence of cloud structures and patterns from remote sensing retrievals and latter calculated an overall radiation budget. Whereas Taylor et al. (2019) focus on a statistical analysis of in-situ cloud data and distinguish between local and continental sources (inland) as well as transported maritime air masses, our study makes a distinction between polluted and less polluted clouds, based on CO mixing ratios, which we can reference to accumulation mode aerosol. Furthermore, Taylor et al. (2019) explained with a sensitivity study using a parcel model the observed difference between maritime and inland low-level clouds.
What is left unanswered to this point is the role of polluted and less polluted low-level clouds within the regional radiation budget. Our study extends the current body of literature by adding radiative transfer calculations fed with a derived representation of measured low-level clouds under consideration of the degree of pollution involved in air associated with the genesis of and/or entrainment in low-level clouds. We look at how the Twomey effect influences the instantaneous radiative forcing and instantaneous heating rates. Although we cannot simulate and evaluate overall cloud adjustments with our RT model, in order to come up with an effective radiative forcing, this still is a crucial step towards classifying the microphysical and radiative influence of increased aerosol emissions on low-level clouds during the monsoon onset period in tropical West Africa.
A greater emphasize on the significance of our study and its aim to answer a major scientific question of the DACCIWA campaign will be included in the manuscript.

General Comment 2:
B. Cloud adjustments and radiative forcing: It would be helpful to be more precise in the discussion here. What you are calculating is the instantaneous radiative forcing due to the Twomey effect alone (effect of greater aerosol leading to greater cloud droplet number concentration and smaller effective radius for the same amount of cloud water). It neglects both cloud adjustments to the Twomey effect and other atmospheric adjustments to the changes in heating profiles and thus is not the effective radiative forcing, which is a distinction worth pointing out. It also may be worth thinking about the effect of aerosols on the clear-sky fluxes, as the "clean" and "polluted" cases would presumably also have different AOD values.
Answer General Comment 2:
The distinction between an instantaneous radiative forcing and the effective radiative forcing as Referee#1 emphasizes is an important one to be made. Surely, this point should be underlined when discussing the RT model results. We will review the manuscript in order to make sure to use the terms instantaneous net radiative forcing and instantaneous heating rates throughout the entire manuscript correctly. The investigation of cloud and atmospheric adjustments according to the demonstrated aerosol-cloud interaction cannot be simulated in the discrete RT-model libRadtran and lies outside the scope of our study.
Analyses of adjustment mechanisms with data from the DACCIWA period were analyzed e.g. by Pante et al. (2021), who find a likely cause of reduced precipitation caused by enhanced aerosol levels, but a cloud lifetime effect, such as shown by Christensen et al. (2020) has not been analyzed. Nevertheless, cloud and ongoing atmospheric adjustments are already implicitly included in the measured data set. Just like Douglas and L'Ecuyer (2020) a determination and quantification of cloud adjustment processes need a more holistic approach and might feature a synopsis of additional data sources and measurements to in-situ data alone, such as remote sensing and microphysical models.
The question of a varying AOD is interesting in the context of determining effective radiative forcing when quantifying of the influence of local sources versus aerosol sources from long-range transport. However, our study specifically examines the influence of a ubiquitous aerosol background and its influence on cloud microphysics and the mere aerosol-cloud effect on instantaneous radiative forcing.
This study investigates a significant open research question posed by the DACCIWA consortium prior to the campaign, and extends other studies by performing radiative transfer calculations.
Our findings provide an excellent starting point for further model studies, e.g. to determine an effective radiative forcing of the overall atmospheric system in the region.

General Comment 3:
C. Cloud specification in RT model: In general, does the ~1 km cloud make sense? Is that how thick the clouds typically are? It would be helpful to perhaps discuss distributions of cloud top and base heights for the polluted and clean cases. This would also be relevant for thinking about how to interpret the vertical profiles in Figure 6 (see comment below).
Answer General Comment 3:
We thank the Referee for the suggestion to include a discussion of the distribution of cloud top and cloud base heights in the manuscript.
The determination of the vertical cloud structure is a representation of a height averaged profile over all campaign flights with the DLR-Falcon, including cloud base height (CBH) at an average of 666.29 ±82.82 m and cloud top height (CTH) around 1909.40 ±440.12 m.
While interpreting the vertical profile, one has to consider the underlying measuring and flight principles inherent in aircraft-based in-situ measurements: Contrary to genuine vertical profiles e.g. from balloon soundings, aircraft measurements incorporate a much larger horizontal component. An aircraft based vertical atmospheric survey only features measurements from altitudes along the flight trajectory. Unless the flight plan was designed to allow for multiple transects of a single cloud, including flights at CBH and CTH, no statement towards individual cloud dimensions can be made. Additionally, the commencement of a flight has to comply with local flight rules i.e. terrain clearance. Hence, we derived a vertical profile from all individual cloud transects of low-level clouds as a

proxy. Instead of speaking of a single continuous compact cloud layer, one must rather use the notion of different cloud encounter along a multitude of low-level clouds that were compiled in a single vertical profile.
Nevertheless, the used vertical low-level cloud statistics can be considered as representative for low clouds in the considered period.

To highlight this, a comparison between an example of a single day of measured low-level clouds to the statistical vertical profile support the aptness of the derived vertical profiles and their input in the RT model.
The vertical profile of the cloud measurements of July 5 suggest the occurrence of low-level clouds between 750 m and 1750 m between 11:30 to 13:50 UTC, with maximum droplet number concentrations of 1200 cm^{-3}. The flight was intended as a comparison flight from Lome to Savé and a turn over the ground station called KITcube. A comparison with the ceilometer at station, a model CHM15k NIMBUS (https://www.lufft.com/de-de/produkte/wolken-schneehoehensensoren-306/ceilometer-chm-15k-nimbus-2093/), supports the flight measurements in terms of occurrence of low-level clouds. The ceilometer retrieval (provided by N. Kalthoff) shows the presence of a nocturnal stratiform cloud cover as a ~200 m thick band around 300 m agl during night and the early morning hours, resulting from an interaction of the African easterly jet and the nocturnal low-level jet (Zouzoua et al.,2021). As the sun rises, the CBH lifts and the thickness of the cloud cover increases with a rise in CTH. Around noon, the stratiform clouds break up and clouds are found in the entire range between 700 m agl and 1700 m agl. These results fit to the inflight measurements aboard the DLR Falcon-20 on this specific day, as well as to the vertical profile in the low-level cloud statistics from all research flights.

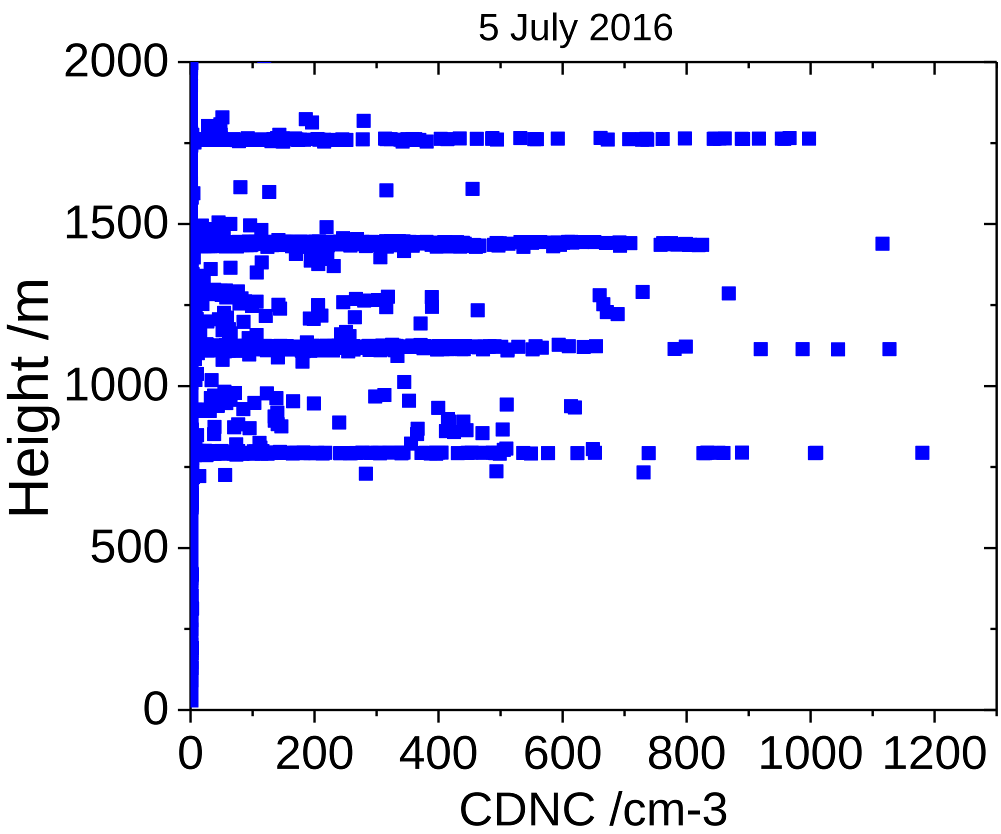

Figure 1: Representation of a vertical profile of cloud droplet number concentration measured aboard the DLR Falcon 20 research aircraft of low-level clouds from 5 July 2016 on a comparison flight from Lome, Togo to Savé Benin where a ground station was overflown. The representation of the vertical profile shows cloud encounter between 750 m agl and 1750 m agl, with higher concentrations on four distinct altitudes, which correlate to the main cruising altitudes.

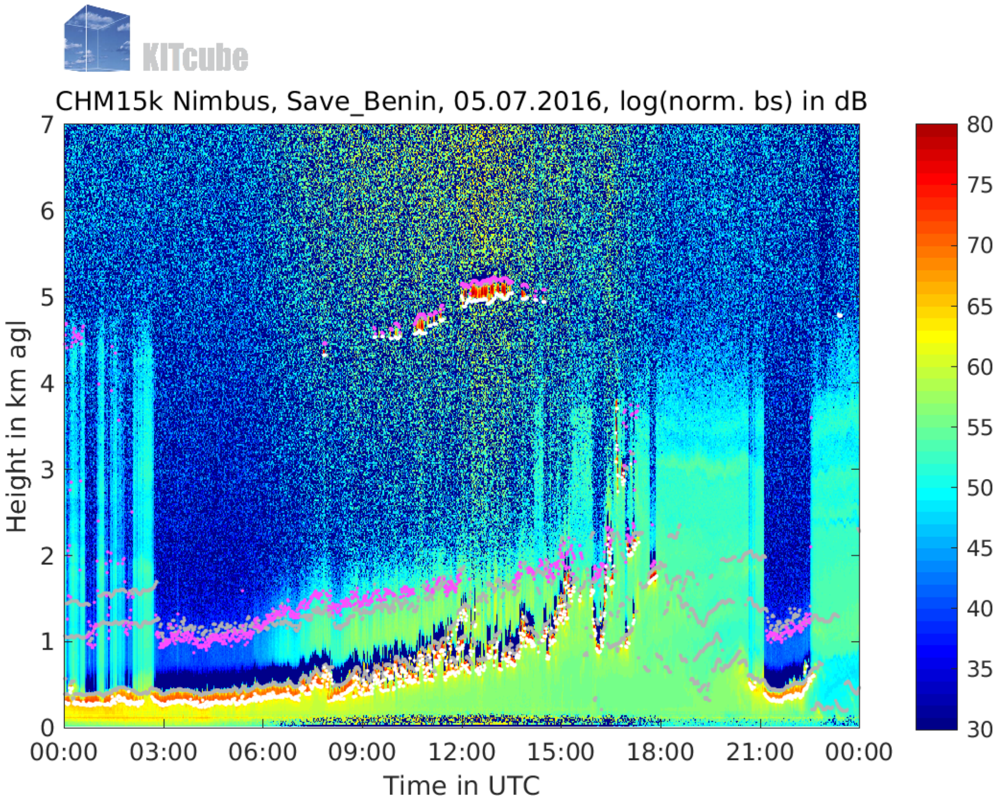

Figure 2: Ceilometer retrieval from the KITcube station in Savé, Benin from 5 July 2016. The retrieval shows the evolution of low-level clouds, starting from a nightly stratiform cloud deck that lifts after sunrise, broadens and breaks up around noon. Similarly to the insitu cloud measurements from the aircraft clouds are found on similar altitudes. Credits: N. Kalthoff

To give an idea about the radiative influence of a standard deviation modified CBH and CTH, sensitivity studies were performed with the DISORT solver in accordance with the conditions published in the study for the profile of a polluted and less polluted cloud.

As a starting point of the of the sensitivity study, the corresponding measurement profiles from the study were used with a solar zenith angle of 17 November at 12:00 local solar time. Assuming a CBH of 0.8 km and a CTH of 1.85 km, the instantaneous delta net radiative forcing is -17.14 Wm^{-2} at the top of atmosphere and -14.58 Wm^{-2} at the ground.

Based on this case, the entire cloud was raised by 450 m, which is approximately the standard deviation of the CTH (as well as lowered by 100 m, which corresponds to the CBH). This results in delta net radiative forcing at TOA of -17.40 Wm^{-2}(-17.04 Wm^{-2}) and -14.42 Wm^{-2} (-14.61 Wm^{-2}) at BOA. Hereby it is evident that raising (lowering) the vertical profile derived from the measurements by the standard deviation does not significantly change the instantaneous net radiative forcings caused by the Twomey effect.

Only for the case that the total cloud is stretched by 450 m, so that the CBH is held at 0.8 km but the CTH is now at 2.3 km, results in a delta net radiative forcing at TOA of -12.98 Wm^{-2} and -9.51 Wm^{-2} at BOA. Only with a change of the total optical thickness of the cloud the delta in the instantaneous radiative forcing increases. This, however, means that its completely different cloud, which no longer corresponds to the measurements from the DACCIWA flight campaign.

Specific Comments:

Comment 1:
Line 71: What is meant by "large mode"? A large fraction of the accumulation mode?
Answer 1:
Yes, exactly. This statement refers to the observation by Haslett et al. (2019) that in all probed airmasses during the DACCIWA aircraft campaign, classified as upwind marine, continental background and urban outflow the aerosol particle number concentration of accumulation aerosol seems to be somewhat comparable (Fig. 1). This finding, including an assessment of the chemical composition of the probed air masses, led them to the conclusion that the entire region is already influenced by advected biomass burning aerosol from remote sources, likely from agricultural fires in southern and central Africa.
For a clearer understanding the wording of this sentence will be modified accordingly in the revised version of the manuscript.

They find a large contribution of accumulation mode aerosol from biomass burning ~~aerosol~~ transported from the southern hemisphere ~~on the large mode of~~ in the background aerosol distribution in West Africa, which acts as cloud condensation nuclei.

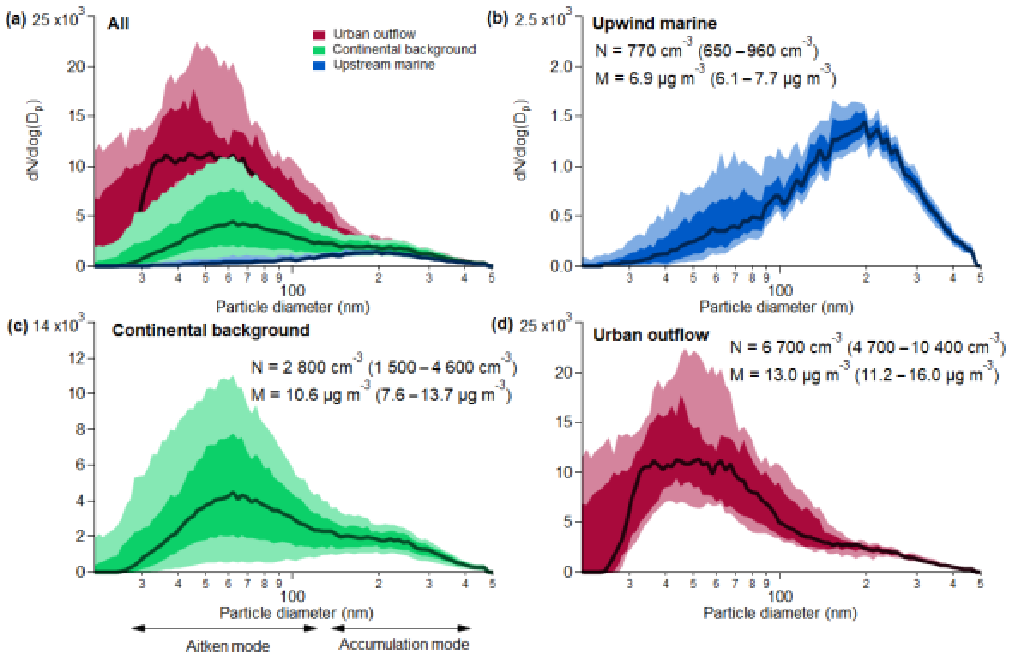

Figure 3. Size distributions of aerosol in the urban outflow, continental background and upstream marine regimes, measured by the SMPS on board the ATR aircraft. For each regime, the median size distribution is shown by the dark line, the dark shading contains 50 % of the data, and the light shading contains 80 % of the data. The comparison of all three plots in panel **(a)** shows a stable accumulation mode that exists in all three regimes, centred at around 200 nm, while the smaller Aitken mode is much more variable. In panels **(b)–(d)**, N shows the median total number concentration summed across the whole distribution, with the lower and upper quartiles shown in brackets; M shows the calculated aerosol mass, assuming an aerosol density of 1.6 g cm^{-3} (Haslett et al., 2019), with the interquartile range again shown in brackets. The Aitken and accumulation modes are labelled in panel **(c)**.

Figure 3: Aerosol modes from Haslett et al. (2019) divided in upwind marine, continental background and urban outflow, all having in common a similar accumulation mode, which supports the assumption of ubiquitous aged biomass burning aerosol from central and southern Africa.

Comment 2:
Lines 72-73: There are many more studies than just Painemal et al. (2014) that study the effect of smoke CCN on low level clouds! Is there a particular point you want to make here about that paper?
Answer 2:
There Is no specific intent in mentioning just Painemal et al. (2014). Further citations can be drawn from Kaufman and Fraser, 1997; Kaufman et al., 2005; Ramanathan et al., 2001;

Douglas and L'Ecuyer, 2020; Christensen et al., 2020; Menut et al., 2018, just to mention a few. These will be featured in the revised version of the manuscript.

Comment 3:
Line 72: CCN has not yet been defined.
Answer 3:
The term cloud condensation nuclei abbreviated as CCN has been introduced in the updated draft.

Comment 4:
Line 108: Instantaneous observations are unable to directly measure the "influence of aerosol loading on microphysical properties".
Answer 4:
This wording is inaccurate or at least imprecise. We will modify the sentence as follows:
~~The influence of aerosol loading on m~~Microphysical properties of low-level clouds ~~is derived from~~ are measured in-situ ~~measurements of clouds~~ with the Cloud and Aerosol Spectrometer CAS installed at a wing station of the Falcon 20 research aircraft.

Comment 5:
Line 152: Wouldn't CO also trace biomass burning plumes?
Answer 5:
This is true. However, the dilution of air masses from central and southern Africa, influenced by biomass burning during the long-range transport, provides the CO background mixing ratio. Additional contributions from local sources such as urban emission plumes, as well as local biomass burning sites, add up to an enhancement of the local CO mixing ratio above the background.
In the revised version of the manuscript, local sources of biomass combustion are considered.
Since data coverage of accumulation mode aerosol measurements with the SkyOPC is insufficient within clouds, CO concentrations have been used to derive location and dilution of local sources such as urban emission plumes as well as local biomass burning sites, factored by a vast amount of combustion products of organic matter within the plumes (Haslett et al., 2019).

Comment 6:
Line 201: Why not give the details of when these clouds were observed here?
Answer 6:
The used proxy for a mid-level cloud was observed on 6 July 2016, before 13 UTC. These details will be given in the modified version.

Comment 7:
Lines 218-219: Where is the AOD assumed to reside vertically?
Answer 7:
The assumed aerosol profile fed into the RT simulations is a standard profile using the background option with urban in the lower 2 km (Mayer and Kylling, 2005). While taking an AOT of 0.381 from measurements of the Aeronet data from the Region (e.g. Savé, Benin) and campaign period, we assume a representative aerosol profile.
Nevertheless, we performed a sensitivity evaluation to see the dependence of top of atmosphere net fluxes (SW and LW) on variations in AOT. For a clear sky case at noon a variation of ±20 % of the assumed AOT lead to a change of ±4.9 Wm-2. A doubling of the AOT still reduces the TOA net flux by 21.4 Wm-2.
Since the focus of our study was drawn on low-level cloud microphysics and resulting radiative properties, an in-depth analysis and variation of the aerosol background was not our upmost concern. Still, we are convinced that the combination of a measured Aeronet AOT coming from the same region and time period in combination with the selected profile

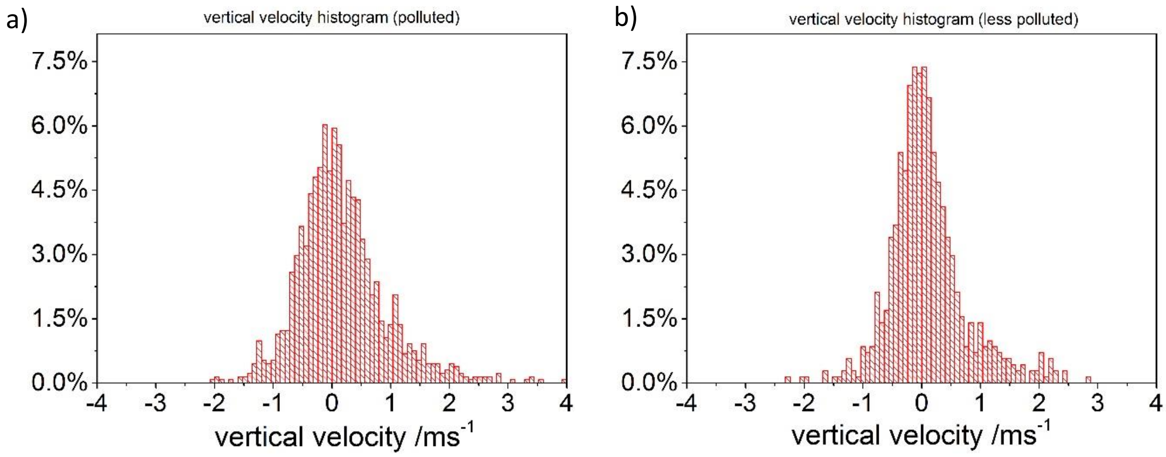

Figure 4: Histograms of vertical velocities for the polluted cases a) and less polluted cases b) show an overall similar distribution.

from libRadtran yields a close to realistic representation of aerosol distribution for our low-level cloud RT study.

Comment 8:
Line 292: Is the CLARIFY value referring to median CO in the boundary layer? CO in the free tropospheric biomass burning plumes observed during CLARIFY and ORACLES were substantially higher than this value. Also, the median value between clean air and heavily polluted plumes might not be a particularly meaningful metric.
Answer 8:
This objection is correct: Compared to measurements from the CLARIFY campaign that have been performed directly within heavily polluted biomass burning plumes coming from central and southern Africa are in three orders of magnitude higher than what has been measured during the DACCIWA or ACRIDICON campaign. This will be clarified in the updated version of the manuscript. Still, we deem the classification of a polluted versus a less polluted airmass or cloud according to its CO mixing ratio which we can correlate to accumulation mode aerosol as a valid method.
In light of median CO mixing ratios of 75 pp~~b~~mv above the South Atlantic Ocean measured directly in biomass burning plumes~~, as measured on~~ during the CLARIFY-2017 campaign (Haywood et al., 2021), the measured CO average mixing ratio during DACCIWA is about three orders of magnitude smaller, but can still be used to qualify between polluted and less polluted air parcels ~~has to be regarded as moderately polluted~~.

Comment 9:
Line 303: Are these measurements only around noon? This should be clarified in the methods section.
Answer 9:
Although it was no strict strategic flight planning measurement flights were conducted somewhat between 10 o'clock am and 2 o'clock pm. With earliest to latest measurements ranging between 9 o'clock am and 3 o'clock pm. Eventually all flights covered noon. No measurements have been taken during late afternoon, evening, night and early morning hours, thus the term noon-time low-level clouds. This will be elaborated in the modified version.
The mean effective diameter of the ~~noon-time~~ low-level less polluted clouds is 14.8 µm for 90 % of measurements ranging between 10 o'clock am until 2 o'clock pm.

Comment 10:
Lines 323-326: Why is this not shown?
Answer 10:

Vertical velocity measurements from the basic instrumentation system aboard the Falcon research aircraft revealed mean updrafts of 0.20 ms^{-1} within clouds, among the discussed data set of polluted and less polluted low-level clouds in West Africa.
Both histograms of observed updraft speeds in low-level clouds for the polluted, as well as the less polluted cases reveal a quite similar behavior, except a slightly broader distribution for the former.
Taylor et al. (2019) assume in their packet model the 3rd quartile as estimate for morning updraft speeds. Following their example in also considering the 75th percentile as updraft speeds we yield a value of $w = 0.453$ ms^{-1}, which fits to Taylor et al. (2019). Analyzing the 75th percentile of the updraft speeds of each of the low-level cloud cases individually reveal a deviation of approximately 10 %.
These assumptions subsequently fit quite well to the modelled (observed) CDNC increase of 31 % (26 %) between polluted and less polluted low-level clouds, as also calculated by Taylor et al. (2019).

Comment 11:
Figure 6: The droplet size results are potentially convolving cloud vertical structure and microphysical differences. An adiabatic cloud should have increasing droplet size with height. Are the distributions of cloud tops and bases similar between the more and less polluted cases? If not, that would influence the comparison here.
Answer 11:
Cloud bases and cloud tops of both cloud types are comparable, as the clouds emerging from the stratus cover were formed under the same conditions. Observations show that typically for this time of year, deep stratus clouds form overnight in the region, as a result of the transport of cool and humid maritime air inland. The Surface warming throughout the morning causes the nocturnal stratiform clouds to rise and thicken, until their break up during noon. Only towards the afternoon the influence of a convective component becomes more influential (van der Linden et al., 2015; Kalthoff et al., 2018). Figure 3 from Taylor et al. (2019) shows the diurnal cycle of low -level cloud fraction from Satellite observations from SEVIRI during the DACCIWA aircraft campaign, with high cloud fractions between 8 am and 2 pm local solar time over land. Figure 4 shows an inflight image from the DLR Falcon research aircraft of gaps in the former stratiform cloud cover during afternoon break up.

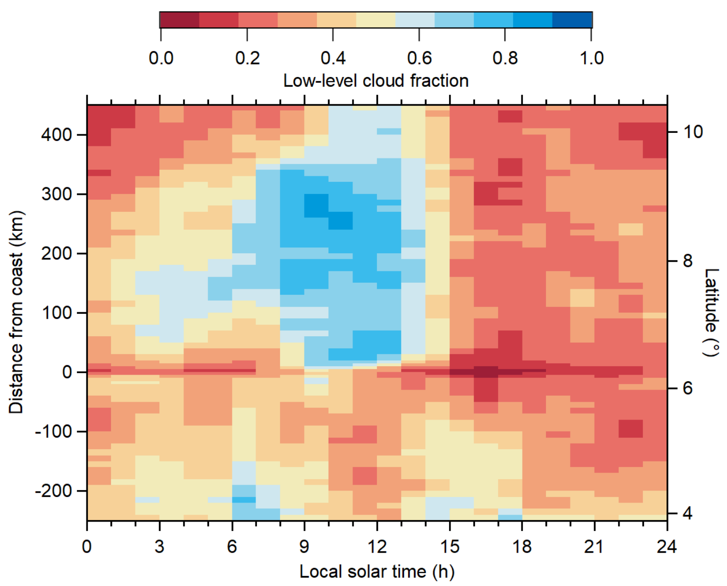

Figure 5: Cloud fraction of low-level clouds according to solar time and distance from coast in Wet Africa. High cloud fractions between 6 and 14 local solar time are found from the coast line up to 400 km inland. From van der Linden et al. (2015).

Figure 6: Representative Inflight image from the Falcon research aircraft during DACCIWA campaign in tropical West Africa from 12 July 2016 around 10:30 am a) above clouds b) below clouds.

Comment 12:

Lines 344-347: This is a somewhat confusing and oversimplified discussion of indirect aerosol effects. Cloud adjustments to the Twomey effect (holding LWC constant, greater aerosol leads to greater CDNC/lower ED) can be large in magnitude and substantially enhance or counteract the radiative forcing from the Twomey effect alone. Your study only addresses the Twomey effect, but the neglect of adjustments should be mentioned as a source of uncertainty.

Answer 12:

Adjective "instantaneous" has been added in line 223 as well as in line 349.

Comment 13:

Line 361: Twomey effect only, not "pollution effect," which could encompass direct, semidirect, and indirect effects.

Answer 13:

Changed accordingly in the modified manuscript.

Comment 14:

Lines 379-381: I don't follow where this discussion is coming from.
Answer 14:
Taking the net radiative forcing for the low-level cloud case, integrated over 24 hours yields $RF_{net} = -3.9$ W m^{-2}. Under consideration of an additional medium level cloud with a COT=3.1 averaged over 24h yields a net forcing at top of atmosphere of $RF_{net} = -4.0$ W m^{-2}. Increasing the COT of the medium-level cloud, as done in our sensitivity study, increases the 24h averaged net forcing of the two-cloud-layer case, thus has a greater impact on the net forcing at TOA by our low-level clouds alone. Vice versa a smaller coverage (small COT) of the medium- level cloud over the homogeneous boundary layer cloud has less impact on the net forcing at TOA as exerted by low level-clouds alone.
Formulations have been changed and the last sentence "…; conversely, a homogeneous medium level cloud …" has been deleted because this has not been studied and corresponding numbers from radiative transfer calculations are not available.

Comment 15:
Lines 398-399: The SW rate isn't converging to the LW values, the total is converging to LW, right? Wouldn't it just be easier to say the SW rate approaches zero?
Answer 15:
Sentence has accordingly been changed

Comment 16:
Line 413: Lower than? Not "smaller."
Answer 16:
Changed accordingly in the modified manuscript.

Comment 17:
Line 427: "For an entire day" is ambiguous here, as the net forcing is positive at night. Integrated over an entire day?
Answer 17:
Thank you for pointing this out. The objection is correct. Meant here is the integration over the entire day and not "for an entire day". Changed accordingly in the modified manuscript to "integrated over an entire day".

Comment 18:
Line 446: I'm not sure how you're using "climate sensitivity" in this sentence.
Answer 18:
Climate sensitivity is used to refer to the atmospheric cooling brought on by low-level clouds. As our study demonstrates that this effect does not increase linearly with population growth and ongoing urbanization in tropical West Africa, but is attenuated in the presence of ubiquitous background aerosol.
We agree that discussing about "climate sensitivity" might be far-fetched, when we only regard the Twomey effect alone during the monsoon onset season and discussing net radiative forcings and derived instantaneous heating rates (without regarding a cloud adjustment), without having drawn our conclusions from the effective radiative forcing.
The new manuscript will find a more accurate classification. The last two sentences of the "Discussion" have been reworded to make the statements clearer and correct.

Comment 19:
Forcing values aren't accounting for any change in AOD
Answer 19:
This is correct. Although AOD measurements from the Aeronet measurement network were taken from July 2016, for the radiative transfer calculations there has been no variations of AOD involved in this study, which solely focused on the contribution of low-level clouds.

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