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

Reply Referee# 1

Dear Reviewer,

Your feedback is greatly appreciated and was helpful in improving the quality of this research. We value the constructive criticism and thoughtful comments, which have helped to identify areas that require further clarification and refinement.

We carefully considered your suggestions and incorporated them into the revised manuscript, as specified below.

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?

Reply General Comment 1:
We thank Referee#1 for the indication to emphasize the motivation of this study. 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 leads to uncertainties for future regional climate. Knippertz et al. (2015) raise the question on the susceptibility of clouds to increases in anthropogenic pollution in West Africa.
Previous studies found discrepancies in local weather models, also associated with a misrepresentation of multilayered cloud structures in this region (van der Linden et al., 2015; Birch et al., 2014).
Also, the contribution of low-level clouds to the energy budget of the atmosphere during the monsoon onset phase in West Africa was investigated for which detailed in-situ cloud measurements were required.
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 abundance in western Africa, 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. While 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 link to accumulation mode aerosol. Also, Taylor et al. (2019) use a parcel model to explain the formation and the observed difference between maritime and inland low-level clouds.
Still, the role of polluted and less polluted low-level clouds for the regional radiation budget is not answered. Here we use radiative transfer calculations with the measured low-level clouds to investigate the impact of pollution on clouds and the radiation budget. We derive instantaneous radiative forcing and heating rates, as a first step to quantify the effect of increased aerosol emissions on low-level clouds in tropical West Africa.

We changed the manuscript as follows:

Abstract lines 34-40 (revised MS):
Radiative transfer simulations show a non-negligible influence of higher droplet number concentrations and smaller particle sizes on the net radiative forcing at the top of atmosphere of -16.3 W/m² of ~~the~~ polluted with respect to ~~the~~ less polluted clouds and lead to a change in instantaneous heating rates of -18 K day^{-1} at the top of ~~the~~ clouds at noon. ~~It was found that the net radiative forcing at top of atmosphere accounts for only 2.6 % of the net forcing of the cloud-free reference case. Thus, polluted low-level clouds add only a relatively small contribution on top of the already exerted cooling by low-level clouds in view of a background atmosphere with elevated aerosol loading.~~ Thus, the atmospheric cooling by low-level clouds increases only slightly in the polluted case due to the already elevated background aerosol concentrations.

Lines 105-113 (revised ms): ~~In our study we calculate the radiative impact of inland continental low-level clouds in West Africa based on a comprehensive 110 data set of in-situ observations from the 12 measurement flights of the Falcon 20 research aircraft during the DACCIWA airborne campaign. Additionally, we simulate how the increased anthropogenic pollution of low-level clouds affects the radiation budget and in which direction such effects could change the local climate.~~ Here we investigate the impact of low-level clouds to the energy budget during the monsoon onset phase in West Africa. We calculate the instantaneous radiative impact of inland continental low-level clouds in West Africa based on a comprehensive data set of in-situ observations from the 12 measurement flights of the Falcon 20 research aircraft during the DACCIWA airborne campaign. Based on the measurements we simulate the impact of increased anthropogenic pollution on low-level clouds, the atmospheric radiation budget and heating rates.

Lines 446-448 (revised ms): This data set contributes to fill the gap of scarce cloud measurements in ~~this region at~~ the Gulf of Guinea ranging ~~as far as~~ from Benin to the east and Côte d'Ivoire in the west. We also investigate effects of pollution on low-level clouds and on the radiation budget in this region.

Lines 493-500 (revised ms): The growing economy and ongoing urbanisation of the West African suggest further increases in aerosol emissions and related changes in cloud condensation nuclei concentrations in future. Still our study suggests that clouds have a lower susceptibility to aerosol in a regime with high background aerosol concentrations and that related cloud radiative forcing are small and might be damped by a medium-high and high clouds. Results of this study are representative for the monsoon onset period in West Africa, associated with long-range transport of biomass burning aerosol related to agricultural land use in southern and central Africa where each year slash-and-burn methods are used for land cultivation. It remains to be investigated whether the results from this study can be transferred to other regions in the world with higher pollution levels.

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.
Reply General Comment 2:

The distinction between an instantaneous radiative forcing and the effective radiative forcing as pointed out by Referee#1 is important. We have reviewed 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 cloud 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.

We changed the manuscript as follows:

(revised ms): Throughout the ms we use the term instantaneous heating rates, instantaneous radiative effects and instantaneous net radiative forcing.

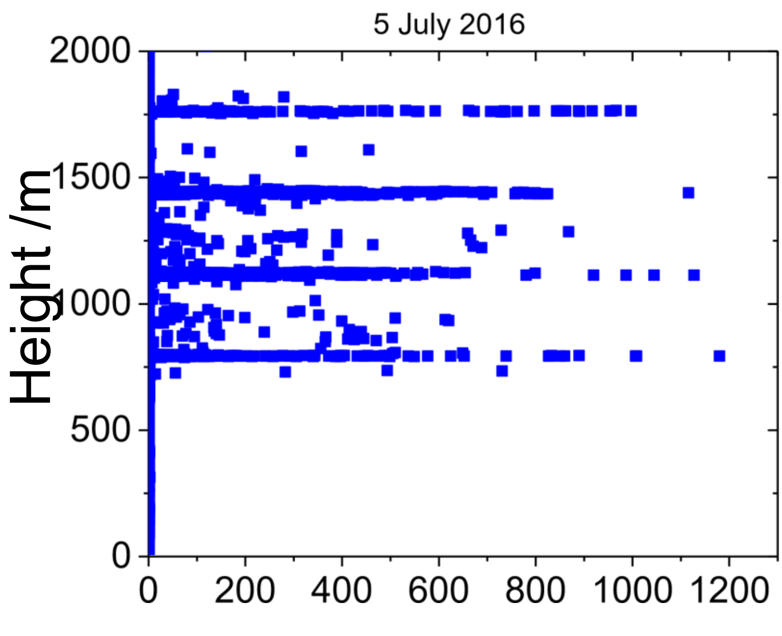
Lines 74-77 (revised ms): Interactions of cloud condensation nuclei (CCN) from biomass burning aerosol and low-level clouds have also been studied by previous studies (Painemal et al., ~~(~~2014~~)~~; Douglas and L'Ecuyer, 2020; Christensen et al., 2020; Menut et al., 2018; Kaufman and Fraser, 1997; Kaufman et al., 2005; Ramanathan et al., 2001)

Lines 365-377 (revised ms):
An aerosol effect on clouds and its implications for the radiation budget (Twomey, 1991) could lead to an increase in cloud droplet number concentration, at the same time reducing the effective radius, given that the liquid water content is similar (since LWC is basically given by the amount of condensable water and thus by the updraft speed).
As Panicker et al. (2010) describe, the origin and long-range transport of aerosol, as is the case in West Africa during the campaign, could play a role in the formation of a positive or negative relationship between aerosol number and ED. A saturation and eventual reversal of the Twomey effect is observed in Wang et al. (2015) at AOTs of 0.4 to 0.5 and above. A mean AOT of 0.38 from the Aeronet data might explain the comparatively small difference in effective diameters between polluted and less polluted low-level clouds in our study. Qiu et al. (2017) suggest that the precipitable water vapor and increasing collision-coalescence moderates the relationship between AOT and ED, which may lead to an anti-Twomey effect. Radiative effects resulting from increased aerosol loading can also lead to a reversal of the relationship through entrainment and coalescence (Kathri et al., 2022).
The Twomey effect identified in our study is used as a basis to derive the instantaneous cloud radiative forcing and the instantaneous heating rates based on both, greater CDNC and smaller ED, while neglecting any other adjustment effects.

Lines 483-487 (revised ms): In order to determine an effective radiative forcing based on instantaneous heating rates, further studies of cloud and atmospheric adjustment are needed using a thermodynamic model that also accounts for adjustment of cloud microphysics. The adjustments according to the demonstrated aerosol-cloud interaction and corresponding radiative response cannot be simulated by use of a one-dimensional RT-model like the UVSPEC/DISORT routine from libRadtran and the calculation is outthe scope of our study.

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).
Reply 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 performed around noon. They show a 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.

The comparison between ceilometer measurements and airborne in-situ measurements above and in the vicinity of the ground station from 5 July validates the extent of the derived vertical cloud profiles. 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.

CDNC /cm-3

Figure 1: Vertical profile of cloud droplet number concentrations measured aboard the DLR Falcon 20 research aircraft on 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 encounters between 750 m agl and 1750 m agl, with higher concentrations on four distinct altitudes, which correlate to the main cruising altitudes.

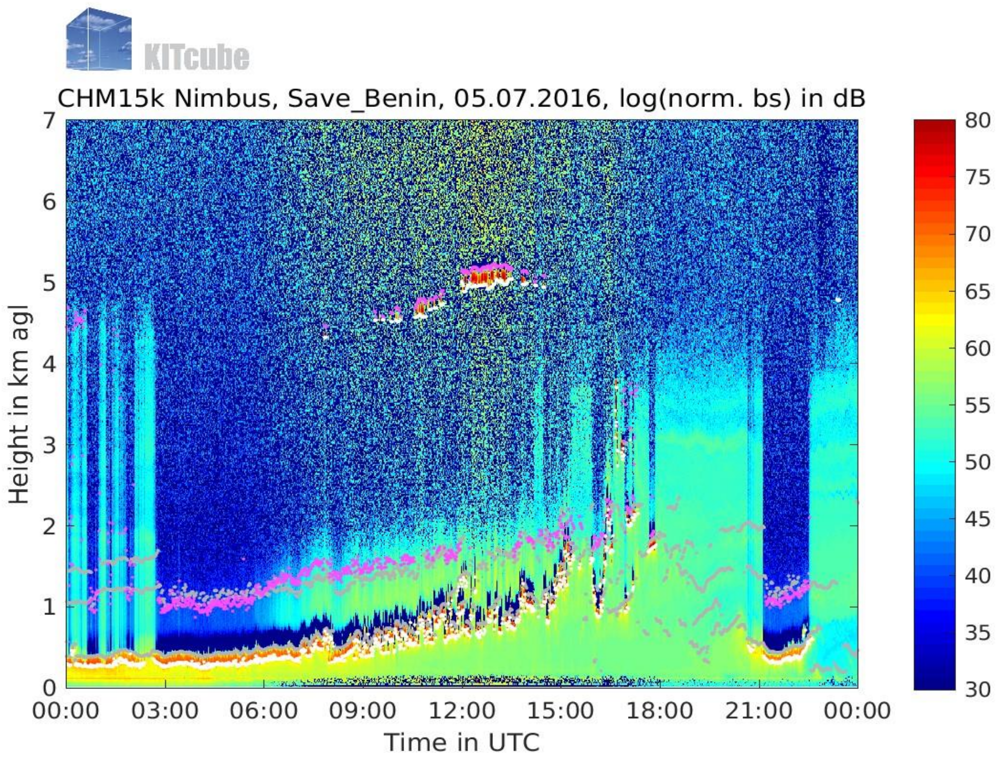


Figure 2: Ceilometer retrieval from the KITcube station in Savé, Benin from 5 July 2016. The retrieval shows the evolution of low-level clouds, starting from a nightly stratiform cloud deck that lifts after sunrise, broadens and breaks up around noon. Credits: N. Kalthoff-DACCIWA presentation 2017.

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.

We changed the manuscript as follows:
Lines 206-207 (revised ms): The low-level clouds measure around noon are compiled into a one-layer surrogate, which is embedded between 790 m and 1870 m and discretizised into 60 model layers.

Lines 345-348 (revised ms):
The determination of the vertical cloud structure is a representation of a height-bin averaged profile over all campaign flights with the DLR-Falcon performed around noon. They show a cloud base height (CBH) at an average of 666 ±83 m and cloud top height (CTH) around 1909 ±440 m. Instead of a single continuous compact cloud layer, different cloud encounters of different low-level clouds were compiled into a single vertical profile.

Specific Comments:

Comment 1:
Line 71 (old ms): What is meant by "large mode"? A large fraction of the accumulation mode?
Reply 1:
Yes, exactly. This statement refers to the observation by Haslett et al. (2019, their figure 3) 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.

Lines 72-74 (revised ms): 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.

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?
Reply 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. The references have been included in the revised version of the manuscript.

Lines 74-77 (revised ms): Interactions of cloud condensation nuclei (CCN) from biomass burning aerosol and low-level clouds have also been ~~studied~~ investigated by previous studies (Painemal et al., ~~(~~2014~~)~~; Douglas and L'Ecuyer, 2020; Christensen et al., 2020; Menut et al., 2018; Kaufman and Fraser, 1997; Kaufman et al., 2005; Ramanathan et al., 2001).

Comment 3:
Line 72: CCN has not yet been defined.
Reply 3:
We now define cloud condensation nuclei (CCN) in line 75 of the revised ms.

Comment 4:
Line 108: Instantaneous observations are unable to directly measure the "influence of aerosol loading on microphysical properties".
Reply 4:
This wording is inaccurate or at least imprecise. We have modified the sentence as follows: Lines 118-120 (revised ms): ~~The influence of aerosol loading on m~~Microphysical properties of low-level clouds ~~is derived from are~~ were 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?
Reply 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.
Lines 161-164 (revised ms):
Since ~~data coverage of~~ accumulation mode aerosol measurements with the SkyOPC are affected by the presence of~~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?
Reply 6:
We used vertical cloud sequences between 4770 m and 4840 m and 9100 m and 9700 m measured on 6 July 2016 during a climb as input for the mid-level cloud in the RT-simulation. These details are now given in the revised version of the manuscript.

Lines 212-214 (revised ms): LWC/IWC and ED profiles for these clouds have been taken from measurements from 6 July 2016 ~~stem from single measurements sometime during the campaign~~ and are then gradually adjusted for sensitivity studies (section 5.2).

Comment 7:
Lines 218-219: Where is the AOD assumed to reside vertically?
Reply 7:
We use a standard urban background profile in the lower 2 km taken from Mayer and Kylling (2005) and scale it to the AOT of 0.381 measured by AERONET stations in the region (KITcube Savé, Benin, and Koforidua, Ghana) and campaign period.
Also, we performed a sensitivity study to evaluate 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 is on low-level cloud microphysics and resulting radiative properties, an in-depth analysis and variation of the aerosol background was not intended. 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 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.
Reply 8:
Measurements from the CLARIFY campaign that have been performed directly within heavily polluted biomass burning plumes from central and southern Africa are significantly higher compared to DACCIWA or ACRIDICON (Wendisch et al.,2016) data. Still, the classification of polluted versus less polluted airmasses or clouds according to the CO mixing ratio which we correlated to accumulation mode aerosol is a valid method.

Line 300-303 (revised ms):
With 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 significantly lower, but can still be used to distinguish 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.
Reply 9:
As reported by Taylor et al, (2019) three aircraft were operated during Dacciwa and often the Falcon went second during the day and as a consequence around Falcon measurements were performed around noon between 10 am and 2 pm with earliest to latest Falcon measurements between 9 am and 3 pm. Eventually all flights covered noon.
Line 316-317 (revised ms)
The mean effective diameter of the ~~noon-time~~ low-level less polluted clouds is 14.8 µm and most of the measurements were performed between 10 am and 2 pm.

Comment 10:
Lines 323-326: Why is this not shown?
Reply 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 polluted case.
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).
We now include Figure xx in the revised manuscript and added the following text:
Changes in the revised ms:

Lines 333-342 (revised ms): ~~The mean updraft of 22 cm/s indicates low updraft velocities in the noontime within low-level clouds. There is no statistically significant difference in updraft speed distributions for the high and less polluted clouds. For our study, differences in updraft speed distributions are small for the polluted and less polluted case and have a minor influence on changes in cloud particle size distributions.~~
As shown in Figure 5, the difference between the two vertical velocity distributions is minor, as peak relative frequency of occurrence differs by 1.5%. The median vertical velocities of 0.01 m s^{-1} (-0.09 m s^{-1}) in polluted (less polluted) low-level clouds indicate no discernible overall trend in vertical motion during measurements, with no statistically significant difference in updraft speed distributions for high and less polluted clouds. In both cases, the standard deviations are 0.69 m s^{-1} (0.62 m s^{-1}), with maximum vertical velocities reaching 4 m s^{-1} (3 m s^{-1}). Since differences in updraft speed distributions are small for the polluted and less polluted cases in our study, we assume that it has only a minor influence on changes in cloud particle size distributions.

We included Figure 5 in the revised ms.

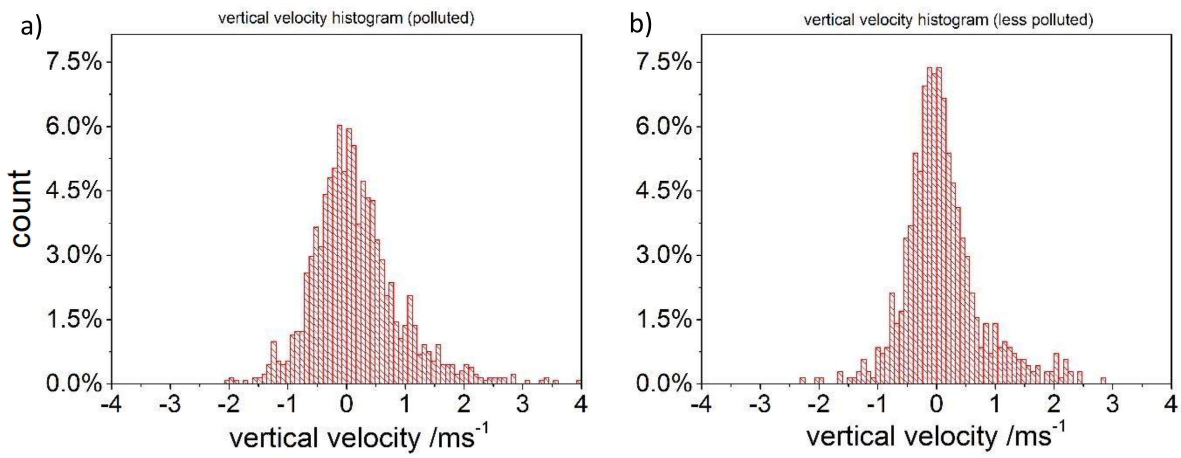


Figure 5: Histograms of vertical velocities for the polluted a) and less polluted b) low-level clouds show an overall similar distribution, with median updraft speeds close to 0 ms-1 and standard deviations lower than 0.69 ms-1 in both cases.

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.
Reply 11:
In Figure 7 (revised ms) we also show the vertical profile of cloud properties and the expected increase in ED. The distributions of cloud tops and bases have been discussed in General Comment 3 and they are similar for more and less polluted clouds as the Falcon measurements were made around noon. There is a diurnal evolution of the clouds as discussed by Taylor et al., (2019).

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.
Reply 12:
We thank the Referee for this valuable hint. An improved discussion on the indirect aerosol effect the is included in the revised version of the ms.

Lines 365-377(revised ms):
An aerosol effect on clouds and its implications for the radiation budget (Twomey, 1991) could lead to an increase in cloud droplet number concentration, at the same time reducing the effective radius, given that the liquid water content is similar (since LWC is basically given by the amount of condensable water and thus by the updraft speed).
As Panicker et al. (2010) describe, the origin and long-range transport of aerosol, as is the case in West Africa during the campaign, could play a role in the formation of a positive or negative relationship between aerosol number and ED. A saturation and eventual reversal of the Twomey effect is observed in Wang et al. (2015) at AOTs of 0.4 to 0.5 and above. A mean AOT of 0.38 from the Aeronet data might explain the comparatively small difference in effective diameters between polluted and less polluted low-level clouds in our study. Qiu et al. (2017) suggest that the precipitable water vapor and increasing collision-coalescence moderates the relationship between AOT and ED, which may lead to an anti-Twomey effect. Radiative effects resulting from increased aerosol loading can also lead to a reversal of the relationship through entrainment and coalescence (Kathri et al., 2022). The Twomey effect identified in our study is used as a basis to derive the instantaneous cloud radiative forcing and the instantaneous heating rates based on both, greater CDNC and smaller ED, while neglecting any other adjustment effects.

Comment 13:
Line 361: Twomey effect only, not "pollution effect," which could encompass direct, semidirect, and indirect effects.
Reply 13:
Changed accordingly in the modified manuscript.

Line 394-395 (revised ms): The net forcing, which, as defined in this study, is based on the ~~pollution~~ Twomey effect only, decreases with decreasing cloud cover.

Comment 14:
Lines 379-381: I don't follow where this discussion is coming from.
Reply 14:
Taking the net radiative forcing for the low-level cloud case, integrated over 24 hours yields RFnet = -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 RFnet = -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.

Lines 413-417 (revised MS): These numbers are valid for a horizontally homogeneous medium-high cloud with 100 % coverage. Note, a~~A~~ smaller coverage of ~~the~~ medium-high~~level~~ clouds over the homogeneous boundary layer cloud would have ~~has~~ less dampening impact on the net forcing at TOA due to a reduced effective optical thickness.~~; conversely, a homogeneous medium level cloud over broken boundary layer clouds will reduce the net forcing values at TOA.~~

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?
Reply 15:
Thank you, the sentence has been changed.

Line 429-430 (revised ms): With increasing SZA, the SW warming decreases approaching to zero ~~converging to the value of~~, at SZA ≤ 0 only ~~the~~ LW cooling is effective ~~rate~~.

Comment 16:
Line 413: Lower than? Not "smaller."
Reply 16:
Thanks, this has been changed in the modified manuscript.

Line 448-450 (revised ms): The characterisation of low-level clouds (~~smaller~~below 1800 m altitude) over~~across~~ this region, between 29 June and 14 July 2016 shows median cloud droplet number concentrations around 270 cm^{-3}, as measured from the CAS underwing probe.

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

Lines 462-464 (revised ms): Seeding the simulation with the microphysical properties from the in-situ measurements shows a negative radiative net forcing at TOA ($F_{net,\ SW+LW,\ CO\geq155\ ppbv}$ minus $F_{net,\ SW+LW,\ CO\leq135\ ppbv}$ ~~CO ≥155 ppbv minus CO ≤135 ppbv~~) integrated over ~~for~~ the day for the~~a~~ low-level boundary layer cloud.

Comment 18:
Line 446: I'm not sure how you're using "climate sensitivity" in this sentence.
Reply 18:
Climate sensitivity is not the correct wording here. We changed the sentence to:

Lines 490-496 (revised ms): ~~This study confirms that the climate sensitivity towards a projected steady increase in aerosol emissions and such cloud condensation nuclei concentrations in the boundary layer, due to proceeding urbanisation and growth of the West African economies might be damped, as the background aerosol loading, at least at the time of measurement, is already significantly enhanced.~~
The growing economy and ongoing urbanisation in West Africa suggest further increases in aerosol emissions and related changes in cloud condensation nuclei concentrations in

future. Still, our study suggests that clouds have a lower susceptibility to aerosol in a regime with high background aerosol concentrations and that related instantaneous cloud radiative forcing changes are small and might be damped by a medium-high and high clouds.

Comment 19:
Forcing values aren't accounting for any change in AOD
Reply 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.

References:

Birch, C. E., D. J. Parker, J. H. Marsham,D. Copsey, and L. Garcia-Carreras (2014),A seamless assessment of the role ofconvection in the water cycle of theWest African Monsoon,J. Geophys. Res.Atmos.,119, 2890–2912, doi:10.1002/2013JD020887.

Christensen, Matthew W., Jones, William K., Stier, Philip: Aerosols enhance cloud lifetime and brightness along the stratus-to-cumulus transition, PNAS Vol. 117, No. 130, https://doi.org/10.1073/pnas.1921231117, 2020.

Douglas, A. and L'Ecuyer, T.: Quantifying cloud adjustments and the radiative forcing due to aerosol– cloud interactions in satellite observations of warm marine clouds, Atmos. Chem. Phys., 20, 6225– 6241, https://doi.org/10.5194/acp-20-6225-2020, 2020.

Haslett, S. L., Taylor, J. W., Evans, M., Morris, E., Vogel, B., Dajuma, A., Brito, J., Batenburg, A. M., Borrmann, S., Schneider, J., Schulz, C., Denjean, C., Bourrianne, T., Knippertz, P., Dupuy, R., Schwarzenböck, A., Sauer, D., Flamant, C., Dorsey, J., Crawford, I., and Coe, H.: Remote biomass burning dominates southern West African air pollution during the monsoon, Atmospheric Chemistry and Physics, 19, 15 217–15 234, https://doi.org/10.5194/acp-19-15217-2019, 2019.

Kalthoff, N., Lohou, F., Brooks, B., Jegede, G., Adler, B., Babić, K., Dione, C., Ajao, A., Amekudzi, L. K., Aryee, J. N. A., Ayoola, M., Bessardon, G., Danuor, S. K., Handwerker, J., Kohler, M., Lothon, M., Pedruzo-Bagazgoitia, X., Smith, V., Sunmonu, L., Wieser, A., Fink, A. H., and Knippertz, P.: An overview of the diurnal cycle of the atmospheric boundary layer during the West African monsoon season: results from the 2016 observational campaign, Atmos. Chem. Phys., 18, 2913–2928, https://doi.org/10.5194/acp-18-2913-2018, 2018.

Khatri, P., Hayasaka, T., Holben, B.N. et al. Increased aerosols can reverse Twomey effect in water clouds through radiative pathway. Sci Rep 12, 20666 (2022). https://doi.org/10.1038/s41598-022-25241-y

Kaufman, Y. J. and Fraser, R. S.: The effect of smoke particles on clouds and climate forcing, Science, 277, 1636–1639, https://doi.org/10.1126/science.277.5332.1636, 1997.

Kaufman, Y. J., Koren, I., Remer, L. A., Rosenfeld, D., and Rudich, Y.: The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic Ocean, P. Natl. Acad. Sci. USA, 102, 11207–11212, https://doi.org/10.1073/pnas.0505191102, 2005.

Knippertz, Peter: Dynamics-aerosol-chemistry-cloud interactions in West Africa-DACCIWA Fact Sheet, https://www.imk-tro.kit.edu/download/Dacciwa-FactSheet-eng.pdf, 2013.

Knippertz, P., Coe, H., Chiu, J. C., Evans, M. J., Fink, A. H., Kalthoff, N., Liousse, C., Mari, C.,Allan, R. P., Brooks, B., Danour, S., Flamant, C., Jegede, O. O., Lohou, F., and Marsham, J. H.: The DACCIWA project: Dynamics-aerosol-chemistry-cloud interactions in West Africa, B. Am. Meteorol. Soc., 96, 1451–1460, https://doi.org/10.1175/BAMS-D-14-00108.1, 2015.

Knippertz, P., Fink, A. H., Deroubaix, A., Morris, E., Tocquer, F., Evans, M. J., Flamant, C.,

Mayer, B. and Kylling, A.: Technical note: The libRadtran software package for radiative transfer calculations – description and examples of use, Atmos. Chem. Phys., 5, 1855-1877, 2005. DOI:
10.5194/acp-5-1855-2005.

Menut, L., Flamant, C., Turquety, S., Deroubaix, A., Chazette, P., and Meynadier, R.: Impact of biomass burning on pollutant surface concentrations in megacities of the Gulf of Guinea, Atmospheric Chemistry and Physics, 18, 2687–2707, https://doi.org/10.5194/acp-18-2687-2018, https://www.atmos-chem-phys.net/18/2687/2018/, 2018.

Painemal, D., Kato, S., and Minnis, P.: Boundary layer regulation in the southeast Atlantic cloud microphysics during the biomass burning season as seen by the A-train satellite constellation, Journal of Geophysical Research, 119,11,288–11,302, https://doi.org/10.1002/2014JD022182, http://doi.wiley.com/10.1002/2014JD022182, 2014.

Panicker, A.S., Pandithurai, G., Dipu, S.: Aerosol indirect effect during successive contrasting monsoon seasons over Indian subcontinent using MODIS data, Atmospheric Environment, Volume 44, Issue 15, 2010, Pages 1937-1943, ISSN 1352-2310,https://doi.org/10.1016/j.atmosenv.2010.02.015.

Pante, G., Knippertz, P., Fink, A. H., and Kniffka, A.: The potential of increasing man-made air pollution to reduce rainfall over southern West Africa, Atmos. Chem. Phys., 21, 35–55, https://doi.org/10.5194/acp-21-35-2021, 2021.

Qiu, Y., Zhao, C., Guo, J. & Li, J. 8-Year ground-based observational analysis about the seasonal variation of the aerosol-cloud droplet effective radius relationship at SGP site. Atmos. Environ. 164, 139–146. https://doi.org/10.1016/j.atmosenv.2017.06.002,(2017)

Ramanathan, V., Crutzen, P. J., Kiehl, J. T., and Rosenfeld, D.: Aerosols, climate, and the hydrological cycle, Science, 294, 2119–2124, https://doi.org/10.1126/science.1064034, 2001.

van der Linden, R., A. H. Fink, and R. Redl(2015), Satellite-based climatology of low-level continental clouds in southern West Africa during the summer monsoon season, J. Geophys. Res. Atmos.,120,1186–1201, doi:10.1002/2014JD022614.

Wang, Fu, Guo, J., Zhang, J., Huang, J., Min, M., Chen, T., Liu, H., Deng, M., Li, X.: Multi-sensor quantification of aerosol-induced variability in warm clouds over eastern China, Atmospheric Environment, Volume 113, 2015, Pages 1-9, ISSN 1352-2310, https://doi.org/10.1016/j.atmosenv.2015.04.063.

Wendisch, M., Pöschl, U., Andreae, M. O., Machado, L. A., Albrecht, R., Schlager, H., et al. (2016). ACRIDICON–CHUVA campaign:Studying tropical deep convective clouds and precipitation over Amazonia using the new German research aircraft HALO.Bulletin of theAmerican Meteorological Society,97(10), 1885–1908. https://doi.org/10.1175/BAMS-D-14-0025.1, 2016.

Zouzoua, M., Lohou, F., Assamoi, P., Lothon, M., Yoboue, V., Dione, C., Kalthoff, N., Adler, B., Babić, K., Pedruzo-Bagazgoitia, X., and Derrien, S.: Breakup of nocturnal low-level stratiform clouds during the southern West African monsoon season, Atmos. Chem. Phys., 21, 2027–2051, https://doi.org/10.5194/acp-21-2027-2021, 2021.