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

Dear Referees,

thank you for your thoughtful comments on the draft, which once again helped to further improve the scientific quality and the clarity of our manuscript.

General Comment:
The authors have addressed many, but not all, of my concerns with their revisions. Specific comments are below, but I would highlight a) that while it should not preclude publication, I would strongly encourage the authors to update their introduction to better motivate the importance of their study, and more importantly b) the discussion of adjustments to the Twomey effect is still lacking.
Reply to General Comment:
We thank the Referee for this comment and revised the introduction and included a discussion on the adjustments to the Twomey effect.

Comment 1:
1. Line 35: It's not clear that this is the minimum value, not the diurnally-averaged value. I would suggest that the diurnally-averaged value is the more appropriate one to report and is easier to square with your assertion that the effect is "non-negligible" yet still small.
Reply 1:
Thank you for the suggestion to rather present the diurnally-averaged values for instantaneous net radiative forcing and -heating rates. For a better understanding, we now list both, the diurnally-averaged and the noon values in the revision and apply the following changes:

Lines 34-37 (revised ms): Radiative transfer simulations show a non-negligible influence of higher droplet number concentrations and smaller particle sizes on the diurnally-averaged (noon) net radiative forcing at the top of atmosphere of -3.9 W m^{-2} (-16.3 W m^{-2}) of ~~the~~polluted with respect to ~~the~~less polluted clouds and lead to a change in instantaneous heating rates of -22.8 K day^{-1} (-17.7~~8~~ K day^{-1}) at the top of ~~the~~ clouds ~~at noon~~.

Lines 407-409(revised ms): Differences in net heating rates between both cases range between -17.5 K day-1 at noon and -25.3 K day-1 at 6 UTC. Averaging the net heating rate differences at TOC over 24 hours gives a value of -22.8 K day^{-1}.

Comment 2:
2. Introduction: There were minimal changes to the introduction, which still reads more like a laundry list of previous findings without strong connective tissue. Why should a reader continue on with this paper? "It was the next thing to be done" isn't very compelling.
Reply 2:
In order to sharpen the purpose of our study we rewrote the introduction as follows:

Lines 45-89 (revised ms):

The sensitivity of climate change to the properties of clouds is yet ambiguous and as such limits the confidence in model simulations of projected climate change from global climate models (Eyring et al., 2020). In-situ measurements in rarely studied locations, such as tropical West Africa, are required to improve and constrain the cloud droplet number concentration (CDNC) and effective droplet diameter (ED) in global climate models (Righi et al., 2020). Cloud formation processes and their dependence on updraft and cloud condensation nuclei concentrations, although being studied for quite some time, are still subject to scientific research with in different regions of the globe (e.g. Kirschler et al., 2022; Braga et al., 2017a, b and 2022; 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). Thus, the role of widespread occurrence of low-level clouds (below 800 hPa, ~1800 m) during the West African summer monsoon, for the local atmospheric energy budget is yet to be better understood (Knippertz et al., 2017; Hill et al., 2018; van der Linden et al., 2015; Flamant et al., 2018a).

With an annual 6 % increase of its gross domestic product (Knippertz et al., 2017), sub-Saharan Africa's economy and socio-economic system is undergoing major changes. The ongoing population growth, urbanisation and industrialisation leads to strong increases in emissions from industry and the transport sector in particular in the major cities, but also from domestic fires in urban and more rural areas (Liousse et al., 2014). 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. Hence, a scientific goal of the international DACCIWA project (Dynamics-Aerosol-Chemistry-Clouds Interactions in West Africa) was to quantify and better understand the effects of increased pollution levels within the boundary layer on low-level clouds over tropical West Africa and on the corresponding radiation budget (Knippertz et al., 2015).

This project combined large scale satellite observations and local ground-based measurements with detailed trace gas, aerosol, cloud and meteorology observations from three airborne platforms: the German Deutsches Zentrum für Luft- und Raumfahrt (DLR) Falcon 20, the French Service des Avions Français Instrumentés pour la Recherche en Environnement (SAFIRE) ATR 42, and the British Antarctic Survey (BAS) Twin Otter (Flamant et al., 2018b). The deployment of the research aircraft to Lomé, Togo took place from 29 June to 16 July 2016 and involved survey flights over Togo, Benin, Ghana and Côte d'Ivoire.

In their comprehensive analysis of the aerosol composition Haslett et al. (2019) find a large contribution of accumulation mode aerosol from biomass burning transported from the southern hemisphere in the background aerosol distribution in West Africa, which acts as cloud condensation nuclei.

While Deroubaix et al. (2022) use a modelling approach, with a regional meteorology chemistry model (Baklanov et al., 2014; Menut et al., 2019), to study the sensitivity of low-level clouds and precipitation, Taylor et al. (2019) present a comprehensive statistical analysis of cloud properties from the three research aircraft and assess the cloud droplet activation by a variation of local aerosol sources. Hill et al. (2018) statistically analyse the radiative effects from 12 distinct cloud types over south West Africa during the summer period from June to September using satellite data and calculate a surface cooling of all indicated cloud types including only a small radiative cooling of the atmosphere attributed to low-level clouds. They identified the frequent obscuring of low-level clouds by higher cloud layers (Figure 1) as an obstacle in satellite data, limiting the information on low level cloud properties.

This study complements the satellite observations and the radiative transfer calculations of Hill et al. (2018) with an in-situ perspective as input to sensitivity studies of radiative transfer modelling. In addition, it extends the work of Taylor et al. (2019) by using the in-situ cloud measurements as an input and a base to investigate the effect of pollution on low-level clouds, radiative transfer and climate in West Africa. We investigate the radiative impact of increased anthropogenic pollution on low-level during the monsoon onset phase

in West Africa by calculating instantaneous radiative forcings and heating rates. The basis is a comprehensive data set of in-situ cloud observations from the 12 measurement flights of the Falcon 20 research aircraft during the DACCIWA airborne campaign.
An overview of the instruments and the methodology is presented in Section 2. Sect. 3 and 4 describe the microphysical quantities of low-level clouds, that serve as input to radiative transfer model simulations. Model results and sensitivity of the clouds radiative impact to aerosol, meteorology and diurnal impact are presented in Sect. 5.

Comment 3:
3. Line 292: The "ppmv" must be a typo in Haywood et al. (2021), CO values were nowhere near that high even in the densest plumes. From the Haywood paper, my interpretation is that Figure 17 (showing the ~75 ppbv mode of CO) is from the boundary layer, measured at Ascension Island. Values of several hundred ppbv were more common in free tropospheric plumes during ORACLES and CLARIFY. Perhaps it would be good to take a step back and reconsider what the purpose of this CLARIFY comparison is in the first place?
Reply 3:
We also consider the "ppmv" mentioned by Haywood et al. (2021) in Figure 17 to be a typo, so we have revised the classification of the individual CO mixing ratio measurements in the PBL between DACCIWA and CLARIFY as follows:

Lines 263-266 (revised ms):
~~In light of median~~CO mixing ratios between 60 ppbv and 160 ppbv~~of 75 ppbv above the South Atlantic Ocean~~ within the Southern Atlantic maritime boundary layer as measured on Ascension Island during the CLARIFY-2017 campaign (Haywood et al. 2021),~~ the measured CO average mixing ratio during~~ show comparable CO enhancement as the DACCIWA measurements, for periods within biomass burning plumes advected from Central Africa.~~has to be regarded as moderately polluted.~~

Comment 4:
4. Lines 354-363: The new additions really aren't responsive to my main concern, which is that adjustments in terms of liquid water path and cloud fraction can substantially offset or enhance the Twomey effect. To just give a few recent high profile examples, Toll et al. (2019) found that liquid water path adjustments tend to offset ~1/3 of the Twomey effect in pollution tracks whereas Manshausen et al. (2022) found that liquid water path increases greatly enhance cooling under weaker inversions. Meanwhile, Chen et al. (2022) found that cloud response to an effusive volcanic eruption was dominated by cloud fraction increases. Wall et al. (2022) also found that the cloud fraction effect rivals or exceeds the Twomey effect (technically Twomey effect combined with liquid water path response) globally when accounting for meteorology.

To be honest, the "negative Twomey effect" literature is unpersuasive to me (you really need very high aerosol concentration for a decrease in the absolute number activated as opposed to decreased sensitivity of increases) and I'm not sure how it's relevant to your paper, as you measured increases in drop number/decreases in droplet size for a relatively moderate aerosol enhancement.

Reply 4:
We thank the referee for pointing out the relevant studies and want to include the discussion of adjustment mechanisms in the revised manuscript. We removed the discussion of an anti-Twomey effect and discuss the saturation effect as described by Wang et al. (2015).

Lines 332-349 (revised ms):

The origin and long-range transport of aerosol, as is the case in West Africa could play a role for the relationship between aerosol number concentration and the ED of clouds (e.g. Panicker et al., 2010). A saturation of the Twomey effect has been observed at AOTs of 0.4 to 0.5 and above by Wang et al. (2015). A mean AOT of 0.38 from the Aeronet data in the campaign region might explain the comparatively small difference in effective diameters between polluted and less polluted low-level clouds in our study.

Toll et al. (2019) found in pollution tracks in various regions around the globe that aerosols generally increase cloud brightness, mainly due to the Twomey effect. An increase of the liquid water path (LWP) as a result of cloud adjustment would be cancelled out by an entrainment effect, which leads to an overall reduced sensitivity of LWP towards anthropogenic emissions. This study shows that a decrease of the LWP can offset the Twomey effect by 23 %.

Wall et al. (2022) suggest that LWP is reduced by increases in sulfate aerosol, accompanied by a delay or suppression of precipitation. Also, Pante et al. (2021) find a correlation between increased anthropogenic aerosol emissions and reduced rainfall during the DACCIWA campaign.

Another study by Manshausen et al. (2022) found a significant negative forcing as a result of an increase of the LWP in ship tracks. The presence of a strong inversion opposes this effect, leading to the assumption that a deepening of clouds is necessary for an LWP amplification.

The pollution effect identified in our study, without being able to distinguish between various contributions to cloud adjustments, is used as a basis to derive the instantaneous cloud radiative forcing and the instantaneous heating rates based on greater CDNC and smaller ED.

Further changes:

Line 68, 226: Changed Ivory Coast to Côte d’Ivoire

Lines 260-263 (revised ms):

Figure 4: Correlation of accumulation mode aerosol and CO mixing ratio from all~~flights~~ measurements <1800 m of the DLR Falcon with a linear best fit of $N_{cacc\text{-}aerosol} = -248.05 \pm 2.65 + 2.83 \pm 0.02 \cdot CO$ [ppbv]. The CO mixing ratio hence is used as a proxy for the degree of pollution and the abundance of activated cloud condensation nuclei within low-level clouds. Henceforth clouds with CO levels ≤135 ppbv (22nd percentile) and ≥155 ppbv (79th percentile) are characterised as less polluted or substantially polluted, respectively.