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

Reply Referee#2

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:

The manuscript presents a report of cloud measurements conducted during DACCIWA, which are classified as either clean or polluted clouds, and then used as inputs to radiative transfer calculations to estimate TOA radiative forcing across the cases. The results are consistent with the first indirect effect -- there is more TOA net radiative cooling for the polluted cloud cases than for the clean cloud cases (as estimated by CO). While this is a nice summary of some of the campaign measurement results with extension to radiative forcing via simple calculations, the scientific depth of analysis is rather shallow. What are we to conclude from this campaign with regard to aerosol-cloud interactions, and would we expect that the results of this interesting set of cases from June-July, 2016, would be regionally representative or consistent with other seasons or years? A good first step would be to add a conclusions section to the manuscript that sums up the impact and implications of the campaign results. Overall, the manuscript is well written and the methods and results sections are appropriately detailed. It is appropriate for ACP. I'd recommend the manuscript be revised to bolster the conclusions and to address the minor comments below:

Reply to the General Comment:

We thank the Referee for the suggestion and completely agree to add a conclusion section to the manuscript, which outlines the impact and implications of this study.

This includes the discussion of the representativity of the results concerning season and location.

This analysis comprises a data set from the DACCIWA aircraft campaign segment in June and July 2016. In light of a predicted tripling of anthropogenic emissions in tropical West Africa between 2000 and 2030 alongside an increasing urbanization and demographic growth, the overarching goal of the DACCIWA aircraft campaign, as part of a multidisciplinary project, was to quantify the consequences for the local climate. The underlying motivation was to better understand interactions between emissions, clouds, radiation, precipitation, and regional circulations, with an emphasis on a better understanding of the two-way cloud and aerosol impacts on the radiation and energy budgets from the cloud scale to the scale of the West African monsoon circulation with a certain attention to low-level clouds (Knippertz et al., 2013).

With our study we directly address one essential campaign objective by analysing the radiative impact of polluted versus less polluted low-level clouds, which before the campaign were suspected of being highly susceptible towards local emissions (Knippertz

et al., 2015). Knippertz et al. (2017) describe the meteorology and chemistry corresponding to this monsoon onset season, for which the data set at hand is representative.

Closely correlated to this study is the ubiquitous background aerosol, transported from central and south Africa into the measurement region. This is correlated to agricultural land use in southern and central Africa where each year slash-and-burn methods are used for land cultivation. Outside this period (including a certain transport delay) background biomass burning aerosol from these sources vanish.

Nevertheless, this instance allows us to draw conclusions for similar cases, where additional urban emissions are released into an already polluted background environment. Take cities in South America for instance, here, slash-and-burn methods are used in the Amazon Rainforest. A blending with urban emissions from densely populated conurbations likely has comparable implications. The same holds for regions in south east Asia, either in terms of biomass burning from agriculture or in the agglomeration regions of megacities, where there is no seasonality. These Implications will be added as in a conclusion section to bolster the significance of our study.

Changes in accordance to the general comment:

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 456-460 (revised ms): Although a high biomass burning aerosol background entrained from central Africa reduces the delta between both classes and reduces the susceptibility to additional local emissions, an aerosol-cloud effect is still visible in cloud microphysical properties, showing effective droplet diameter between 12.3 μ m in the polluted case, compared to an ED of 14.8 μ m in the less polluted case and CDNCs that almost double (median) in the polluted case.

Lines 497-500 (revised ms): 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-andburn 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.

Comment 1:

Line 25: is accumulation aerosol on a number concentration basis?

Reply 1:

Accumulation aerosol has been used in terms of ambient particle number concentrations. The new draft has been modified accordingly. Line 22-25 (revised ms): Clouds below 1800 meter altitude, identified as boundary layer clouds, were classified according to their carbon monoxide (CO) pollution level into pristine and less polluted clouds (CO < 135 ppbv) and polluted low-level clouds (CO > 155 ppbv) as confirmed by the linear CO to accumulation aerosol number concentration correlation.

Comment 2:

Line 31: Add close parenthesis

Reply 2:

A parenthesis has been included at this spot in the revised version in line 31 (new ms).

Comment 3:

Line 36: I don't understand what is being said in the sentence: "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". Are the authors making the case that the indirect cooling from polluted clouds is similar to the direct cooling from pollution aerosols in the absence of clouds? Please clarify.

Reply 3:

The phrasing of this sentence might be inconclusive. We have changed it in the revised version to:

Lines 39-41 (revised ms): 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 exerted atmospheric cooling by low-level clouds only increases ever so slightly in light of their formation in an environment with a substantial increase of accumulation mode aerosol on top of an already elevated background aerosol concentration.

Comment 4:

Line 234: Should this be $a\Delta\lambda$?

Reply 4:

This has been accounted for in the revised version (line 246 new ms).

Comment 5:

Line 266: Were the OPC size distributions fitted (say to a lognormal function) in order to account for the accumulation mode contribution below 250 nm?

Reply 5:

The particle number concentration described as accumulation mode aerosol used in this study comes entirely from the OPC Instruments with a cut off at 250 nm. No subsequent fitting to a density function has been performed. Although this lower measurement cut off does not consider the entire accumulation mode, this estimate was regarded as sufficient to be correlated to CO as a pollution tracer.

Comment 6:

Lines 270-272: Were aerosol measurements made within the vicinity of the cloud? Is the accumulation mode aerosol just below cloud well correlated with the CO-correlation-based estimate?

Reply 6:

The aerosol measurements for the CO-correlation were entirely outside, but constantly in the vicinity of clouds. The flight strategy, in order to accommodate all campaign goals, included only few instances where we probed along a prescribed flight track on various altitudes. Unfortunately, a precise analysis of accumulation mode aerosol measurements just below individual clouds is not possible.

Due to the break-up of the shallow stratiform cloud deck during the late morning and noon, flying below 1800 m altitude necessarily involved sampling in the immediate vicinity of low-level clouds.

Comment 7:

Figure 5: what is the lowest droplet diameter shown on the x-axis?

Reply 7:

We thank the Referee #2 for this hint. The x-axis was readjusted according to the low droplet size threshold of 3 μ m (Figure 5 new MS) and a remark was made in the figure description in Line 325 (new MS).

References:

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