Reviewer #1

The authors use measurements from multiple aircraft to construct a large dataset of cloud properties. The authors show differences in these cloud properties based on location, and provide explanations for their differences based on pollution tracers and the understanding of the local dynamics and meteorological phenomenon. The authors conclude that significant increases in population in south west Africa and resulting enhanced pollution will not greatly affect cloud properties in the region, because a considerable fraction of aerosol particles are high concentrations of biomass burning smoke transported from the southern hemisphere. I have some concerns about the evidence being used to make this claim. I think the use of SMPS measurements would make a stronger case than OPC measurements alone. Furthermore, it is understandable that mixing state can be difficult to determine, but the expected hygroscopicities of different particles are very different and simply assuming an internal mixture weakens the argument (aged biomass, fresh emissions, sea salt).

We thank the reviewer for their useful comments and will address them individually below.

Major concerns:

A considerable amount of the introduction is attributed to explaining the effect of the low level jet on the inland clouds. While it is necessary for the reader to understand the role of the LLJ, I suggest limiting the explanation some since it is not the focus of the results.

We thank the reviewer for the suggestion. We have shortened the paragraph about the role of the LLJ.

I would suggest performing a few parcel model simulations to identify how sensitive the simulation results are to mixing state, even if they are speculative. Proving to the reader that this (arguably bad) assumption cannot account for major changes in CDNC would be helpful to your results.

We have added to section 4.2

"We performed some additional simulations to explore some possible sensitivities in our assumptions. Firstly we considered the presence of a coarse sea salt aerosol mode, which we isolated from the GRIMM size distributions. The size distribution of this mode is listed in Table 2, and the composition was assumed to be NaCl. As the concentration in the sea salt mode is so low compared to the accumulation mode, this addition of a coarse sea salt mode made less than a 1% difference to the derived CDNC. We also performed some illustrative runs on the inland aerosol- one possible scenario is that the local Aitken mode aerosol were from primary combustion. We assumed them to be 100% organic, and proportioned the composition of the local accumulation mode to match the total measured organic/inorganic ratio. Compared to the base case, this increased the inorganic

fraction in the local accumulation mode from 31% to 41%, and the resultant CDNCs showed around a 12% increase. These simulations show that there are some differences that are attributable to assumptions in aerosol composition and mixing state, but they are likely to be smaller than the differences we show related to the total amount of aerosol and updraft velocity."

Section 3.4 analysis with CDNC and Reff seems a little pointless, and possibly misleading. It's nice and simple to group up all your data, show the statistics and interpret them, but you're mixing together very different cases. You pointed out the fact that your profiles often contain multiple layers of clouds.

We think it's worth providing these statistics to provide a basis of comparison for models. If a model is not producing average properties like these then it is not accurately reproducing the microphysical properties of the clouds. We have added the caveat to the caption to Figure 5

"The cloud profiles show the average properties of the cloud field, which often contained multiple discreet layers."

In the text relating to Figure 5c (the Reff profile) we already explicitly discuss the presence of multiple cloud layers.

And you pointed out that offshore measurements of CDNC are only for a day or so, but you still make plenty of comparisons despite this and I don't see the point. These offshore cloud measurements are certainly not enough to suggest they are representative of the regions offshore clouds and they are distracting from your real results/objectives.

For the offshore cloud measurements it seems strange to omit them entirely, given we rely on the offshore aerosol measurements for the basis of our analysis. Reviewer #3 has also explicitly said they think we should not remove them. In every point in the results where they are discussed, we have already noted that they are limited in scope. They do also support the big picture argument we are trying to make, that the polluted background aerosol is having an impact on the clouds. We have made significant changes to the discussion in response to this and Reviewer#3's significant comments on this issue

Did you have a CPC? Figure 6 would benefit from such measurements. They include particles that are too small to be CCN, but N250 certainly misses a lot of CCN. CPC measurements would also provide some insight on the pollution levels.

Haslett et al (2019) provide CPC measurements for the offshore/inland data. In the urban outflow regions, large numbers of Aitken mode particles are present, that are too small to act as CCN. We thought would confuse the issue given we are interested in CCN, and the accumulation mode aerosols show a very different trend. Also see below regarding the comparison to the SMPS.

Beginning of Page 14: Often the majority of the accumulation mode particles (and CCN) are smaller than 250 nm so based on the OPC data alone it's not entirely convincing that the offshore particles account for the majority of the accumulation mode. I understand there are limitations in the SMPS data, but for an analysis where you are averaging all concentration measurements over the land, it should not be a problem. Furthermore, I believe the next paragraph, where you compare CDNC between off-shore and inland clouds to counter this conclusion with the inland CDNC often being more than double the offshore CDNC. Though again, there aren't quite enough off-shore clouds measured, but stating that inland clouds have 85-175% more droplets makes your argument less convincing. It's likely the different dynamics on and offshore may play a role here, so comparing the concentration of particles below cloud (from a CPC and SMPS) would be more useful.

Regarding SMPS data, we have added this to section 3.5

"The average inland enhancement is in excellent agreement with Haslett et al. (2019), who also found a 40% enhancement in accumulation mode aerosol in continental background areas compared to upwind marine, using SMPS measurements averaged over the whole campaign. The day-to-day averages of the accumulation mode aerosol number concentration from the SMPS showed good correlation with the GRIMM, with $R 2 = 0.7$, but data were only available for 9 days inland and 2 days offshore. The GRIMM data therefore show a similar trend as the SMPS, but with greater coverage."

We have also added to the to the discussion

"Our measurements showed an average ~40% increase in regional accumulation mode aerosol inland compared to offshore. The accumulation mode is the most important for CCN, as particles smaller than this are too small to activate. Haslett et al. (2019) also found a 40% enhancement in the inland accumulation mode aerosol concentrations compared to offshore using SMPS data, which is in excellent agreement with our GRIMM measurements of N_{250} . In localised city plumes, highly concentrated emissions had a noticeable increase in CDNC, but on a regional scale the majority of accumulation mode aerosol were due to long-range transport."

Generally aerosol size distributions should not be averaged together unless they look similar (similar peak locations). Can you add all the distributions averaged in Figure 8 with light lines? It would also give the reader a good picture of the distribution variability.

This is a good suggestion to show the variability in the size distributions, and this plot can be found in Haslett et al (2019). We have added a clarification to section 4.1

"While the Aitken mode was variable inland due to highly localised anthropogenic emissions, the accumulation mode showed minimal variability as the majority of particles in this size range were due to long-range transport (Haslett et al., 2019).

This averaging approach is therefore reasonable to inform cloud activation modelling, as the accumulation mode is the source of CCN"

Page 18 first paragraph: The argument on entrainment is a bit week. Sure you had a lot of layered clouds, but clouds with greater than 1 g mˆ-3 of LWC were often likely convective, and therefore, higher updraft velocities. At the very least I would include standard deviations for these CDNCs and the number of cases.

We have removed this part of the paragraph, which we agree was a bit speculative. We have replaced it with

"The values of modelled CDNC are between the 75th and 90th percentiles of measured CDNC shown in Fig. 7a, meaning the difference between the average model and measured values is smaller than the variability in the measured data. ACPIM is an idealised simulation and we have used it here primarily to explore the sensitivity to local aerosol emissions, rather than arbitrarily try to tune factors such as aerosol mixing state, more realistic dynamics, or entrainment (of which we have limited or no measurements) to make the CDNC agree perfectly."

Minor points:

Page 8 line 15 change 'an' to 'and'.

Done

Page 13 "This is"

Done

Page 15 line 19. Do you mean scanning time?

Done

Reviewer #2

The presents and summarises cloud and aerosol observations in low-level clouds over the coast of southern West Africa made during the DACCIWA campaign. Further, an analysis of cloud cover in the wider area is presented based on satellite data and the impact of local aerosol emssions on cloud properties is investigated using a parcel model. The authors draw the conclusions that there is discernible impact of local emissions on the cloud properties and that changes in local emission strength would only have a small impact on CDNC and cloud properties. In general, the paper is well written, the methods are mostly suitable, and the results are for the most part presented well and discussed logically. My main concerns regarding the main conclusions is a lacking discussion of whether the area proportion affected by local aerosol emissions is reflected roughly correctly in the aircraft data.

Also in several places, it is not clear how the authors arrive at certain statements (s.comments below). If the necessary clarifications are included, I recommend to publish the paper.

We thank the reviewer for their useful comments, which we will address individually below.

1 Specific comments

1. p. 2, l. 4 ff: I believe the authors summarise the typical diurnal cycle during the campaign. This should be stated somewhere.

The abstract first paragraph now reads

 "This novel dataset allows us to assess spatial and temporal , diurnal, and day-to-day variation in the properties of these clouds over the region."

2. p.5, l. 18 & Fig. 1: The text states Fig. 1 is showing the flight tracks of the aircraft, while the caption says it shows the locations of cloud observations. Please clarify this inconsistency.

We've clarified this caption, it's the parts of the flight track where cloud measurements were made

3. p. 5, l. 26: It would be helpful to state the actual number of days, for which observations over the sea are available here. Both for aerosol observations and cloud observations. Also can you clarify, how and why you arrive at the conclusion that the number of aerosol observations is sufficient to estimate offshore aerosol variability?

The number of days is shown in figures 5 and 6- in Figure 6 it is easy to see. We've clarified in the text, changing "several days" to *"11 days"*. It seemed reasonable that this would give us some understanding of the variability throughout the campaign, as we have >50% coverage and it is fairly evenly spread, as shown in Figure 6.

4. p. 6, l. 14 ff: For a better understanding of the relevance / meaning of the statistical comparisons, it would be good to include at least a short statement on the sampling times (hours of day), locations and strategies of the different aircraft.

We have taken effort to minimise the influence of any differences in sampling locations/strategies by limiting this comparison to a small geographical area and altitude <1km. This is already stated in the text.

5. p. 6, l. 21 ff: Can you also provide a comparison of the stastics of R_{eff} other than the mean?

This now says

"For R_{Eff} , average vertical profiles between 0 and 100 km inland showed the median values agreed within ~1 μm, which is within the uncertainties of the instruments, and the interquartile ranges were similar for each platform."

6. p. 7, l. 25: Is this the percentage difference in cloud fraction or the absolute difference in cloud fraction?

We've added that it's absolute difference

7. p. 8, l. 17 ff: The limitations of the modelling scheme are described. However, a discussion on how these assumption could impact the results is not included.

Could you at least speculate on the impact of the limitation on the results (maybe in the discussion section)?

We have addressed this is our addition to section 4.2 with the extra model runs, in response to reviewer #1.

8. p. 9, l. 15: It maybe would be interesting to have a figure similar to Fig. 2 with the standard deviation (or variance) in cloud fraction. This would allow a better judgement on, how robust the analysis of the mean field is.

We have added this as a part (b) to figure 2, and added the following text to section 3.2

"The areas of high mean LLC are the most robust features, with the lowest standard deviation, meaning patches of continuous cloud were present on almost all days during the campaign. Areas with lower average LLC fraction had higher standard deviation and were therefore more variable; the cloud cover in these regions varied day-to-day but was lower on average."

9. p. 10, l. 5ff: Can you elaborate on how you determine whether there is or is nota clear effect on the cloud cover?

We have added to section 3.2

"Pollution plumes are expected to extend for hundreds of kilometres downwind of the major cities (Deroubaix et al., 2019). Figure 3 does not show any features extending

downwind of the major cities that would represent a signature for anthropogenic influence on cloud cover on a regional scale"

10. p. 11, l. 13: Is there some means of quantifying this fraction? Is it over- or underrepresented in the observational data? Do analysis of cloud microphysical profiles in the polluted region only?

We have added that the CO threshold of 160ppbv is the 93rd percentile from our measurements, and also added

"Our measurements in this polluted tail were generally in and around the major cities, which were the one of the research themes of the DACCIWA campaign. On the regional scale these are likely to be overrepresented compared to the more sparsely populated regions such as Ghana and Côte d'Ivoire, but may be underrepresented compared to the more polluted regions of coastal Nigeria."

11. p. 12, l. 27ff: I am a bit lost with the argumentation here. You say CDNC differences are "larger than differences in aerosol", "correlate with differences in the aerosol vertical profile" and are not "directly related to differences in accumulation mode aerosol"?

This was a typo- "did correlate with differences in aerosol" should have been "did not correlate with differences in aerosol". Essentially we see differences in CDNC but they are larger in magnitude than the differences in aerosol and don't correlate with them, so it's not obvious that the variation in CDNC is due to aerosol. This is addressed in the updated discussion.

12. p. 13, l. 8ff: Are clouds over the ocean and the land multi-layered. If so the arguments regarding the connection between CDNC and R_eff need to be considered more carefully, as with observations at the same altitude and varying cloud base altitudes this connection is not robust.

Please see our response to Reviewer #1's comment starting "Section 3.4"

13. p. 13, l. 16 / 23: What is the basis for the claim of "little systematic spatial variability" and the statement of "fairly homogeneous" clouds inland?

If you look at Figure 5, the medians for both aerosol and cloud properties for most alititude bins look almost the same for all 3 inland traces. Systematic spatial variability would be something like decreasing CDNC further inland, which the data do not show. The reviewer's "fairly homogeneous" statement here is a bit unfair- the full quote is *"Although the properties of individual clouds were variable, the average inland clouds below 1 km were fairly homogeneous regardless of the distance inland"*, which is true based on Figure 5 and the above.

14. p. 14, l. 5: What do you mean with a "degree of correlation"?

We've added that there was $R^2 = 0.72$ between the medians for CDNC and N_{250} for the time period in question. Admittedly it is difficult to see from eyeballing the plot as the CDNC was so variable locally, we have added

"On a cloud-to-cloud basis this correlation was dwarfed by the inherent local variability in CDNC."

15. p. 14, l. 13: If this is solely based on Fig. 6b, I do not find the claim of an impact of the aerosol concentration on the dailz CDNC very convincing. Just from looking at the plot, I do not think there is any correlation in this data.

See the previous comment

16. p. 15, l. 4: Can you provide any information regarding the position of the flight data relative to cloud base?

See the next comment

17. p. 18, l. 7: What part of the observed vertical velocity distribution are you using for this comparison? How does the observed vertical velocity relate to the cloud base vertical velocity?

For this and the previous comment, we have added in to section 3.6

"Vertical velocity does not vary strongly with altitude in stratocumulus clouds (Wood, 2012), so although our measurements are not just from cloud base, they are still suitable to use in our simulations."

We've also clarified in section 4.2 that we were referring to the simulations *"using the representative morning and afternoon updrafts"*

18. section 3.1 & 3.2: It should be mentioned somewhere explicitly that the area anlaysed here is significantly larger than that covered by the aircraft observations. Orographic effects etc. discussed here have only a limited impact on the detailed cloud data.

This is a good point to make- we have added near the start of section 3.3

"The limited spatial area of the aircraft sampling means that regional scale orographic effects discussed in Sect. 3.2 are not likely to have a large effect on in situ cloud properties."

19. Fig. 9: What does the blue shaded region around the inland line show?

That's the variability in CDNC that you get from varying the offshore aerosol modes by +- 1 stdev, as labelled in the figure key.

2 Technical corrections

• p. 5, l. 23: " ... included measuring emissions ..." (?)

Done

• p. 8, l. 15: "... updraft velocities and aerosol number ... "

Done

• p. 12, l. 14: What is "it" refering to?

Clarified in the paper as *"N_{250} does not show…."*

• p. 13, l. 5: I belief there is something missing from the sentence starting with "In the free troposphere ..."

We've clarified it's R_{Eff} we're talking about

• p. 13, l. 6: "This is due ..."

Done

• p. 13, l. 8: Do you mean cloud observations at higher altitude were not necessarily higher above the cloud base than observations at lower altitude?

That's a good way of phrasing it and we've added that in

• p. 13, l. 9: "... which is consistent with ..."

Done

• p. 14, l. 24: " ... indicating a greater cloud ..."

Done

• p. 16, l. 28: "... updraft than in aerosol ..."

Done

• p. 18, l. 5: "... measurements of aerosol composition ..."

Done

Reviewer #3

The work describes insights on the low level cloud deck which forms during the Monsoon season over South West Africa. These were derived from aircraft measurements of number concentrations of both cloud droplets and aerosol particles in combination with satellite data and some modeling efforts.

It is generally well written, but it seems that data shown in figures does not always support the conclusions drawn. My major concern is about the conclusion that changes in aerosol particle number concentrations (APNC) do not effect cloud droplet number concentrations (CDNC). And I will not argue that there is a large effect, but the presented data also does not support that there is no effect.

I should say upfront that I started reading this paper not having any agenda of my own on this topic. While writing this report, time and again I saw hints in the manuscript which support an influence of aerosol particle number concentrations (APNC) on cloud droplet number concentrations (CDNC). But a contrasting interpretation was given, arguing that the amount of data for the "low concentrations cases" (i.e., offshore) is too small for interpretations. In the following, I will comment on a number of related text passages and on data shown in your figures which seem to indicate a different result. Your work needs to account for that.

Nevertheless, the topic is timely and a revised version may merit publication in ACP.

We thank the reviewer for their useful comments. We will address them individually below.

Major concerns:

As said above, this major concern deals with a possible effect of APNC on CDNC. I understand that you try not to over-interpret the (comparably small number of) data you got offshore. But still, I see the same there, over and over again: Cloud droplet number concentrations (CDNC) are lower offshore than inland, and so are aerosol particle number concentrations (APNC, in your case particles larger than 250nm (N_250). And also the aerosol particle size distribution (APSD) you derived for the modeling is clearly lower in number and different in shape offshore, compared to inland.

If I understood it correctly, you argue that even offshore the air masses are always polluted, originating from biomass burning aerosol that prevails in this region during this time of the year (e.g., page 21, line 24-25). This is a bit confusing, as you also describe that you went > 20km offshore "to unambiguously remove any possibility of local terrestrial emissions affecting" (page 10, line 25-26). So this would mean that the pollution from biomass burning aerosol should be ubiquitous in the whole region, also >

20km offshore. But the size distribution you show as representative for > 20km offshore (Fig. 8) does not indicate that biomass burning pollution is prominently present (for that a pronounced mode of particles < 100nm would be expected). So I somewhat wonder how "polluted" air masses in the offshore region really are.

Generally, you omit the lower concentrations offshore and if you refer to them at all, you say that these data are not to be trusted, due to their scarcity. It is true that they are scarce, but if you really do not trust them, you should not use them in the first place. This is NOT what I would recommend. But I would still recommend that you spend some thoughts on the idea that there might be a real increase in APNC as the air masses go over land, and related to that an increase in CDCN. And I also recommend that you adjust your interpretations in the text.

We thank the reviewer for their in-depth and detailed comments. The preceding set of paragraphs cover several aspects and are difficult to divide up, so we will try to address them as a group. Firstly, the reviewers comment about the biomass burning size distribution does not apply to biomass burning that has undergone any sort of ageing, and literature shows aged biomass burning is primarily in the accumulation mode and consistent with our offshore measurements (e.g. Haywood et al., 2003; Sakamoto et al., 2014) . It sounds like the reviewer is confused about the source of the biomass burning- it is long-range transported from fires in central Africa- we have said

"Haslett et al. (2019) recently described the aerosol properties measured during the DACCIWA aircraft campaign, and showed that a large background of transported biomass burning pollution from the southern hemisphere was ubiquitous in the West African boundary layer"

The idea of going >20km offshore was to unambiguously remove any local component to demonstrate clearly that there was indeed a ubiquitous background of long-range transported pollution across the whole offshore region.

The final point refers to the offshore CDNC measurements. It is not clear what the reviewer is referring to by saying "you omit the lower concentrations offshore" as they are included in many plots and are discussed in numerous places throughout the manuscript. Reviewer #1 has said we should remove them entirely as there's not enough data, but this reviewer has suggested they are either very important or we could remove them if we don't trust them. We do trust them, however we acknowledge they are limited in number.

The main issue is whether the local aerosols are having a large effect on the cloud properties. We have shown that there is no large effect on the broad-scale cloud field downwind of major cities (Section 3.2), and the thick city plumes are a small fraction of the dataset (Section 3.3). With the limited offshore measurements the best way to assess the average effect of local aerosols (as a whole, not just in the city plumes) is by the modelling approach in Section 4.

Having said that, the reviewer's comment has highlighted that it would be useful to state more clearly why the modelling section is there and why this is the best way to address this problem. We have made several changes in response to this comment and ones further down.

In the abstract:

"To assess the relative importance of local and transported aerosol on the cloud field, we isolated the local contribution to the aerosol population by comparing inland and offshore size and composition measurements"

To the start of section 4:

"The limited offshore dataset and the possibility of differing dynamics inland makes it difficult to assess the impact of local aerosol emissions on cloud properties based on measurements alone."

To the conclusions:

"In order to assess the impacts of local urban emissions and transported background pollution on cloud properties, we performed a sensitivity analysis with a parcel model…"

We have also made rewritten several paragraphs of the discussion- the main changed parts are quoted here:

"In DACCIWA, the limited measurements of offshore clouds were split into one day with CDNC around $300 - 700\ cm^{-3}$, which was ~35% higher than the corresponding inland measurements, and four days with CDNC around $100 - 200\ cm^{-3}$, $45 - 65\%$ lower than inland. None of the offshore measurements were representative of clean clouds; offshore clouds were universally more polluted, with higher CDNC and lower R_{Eff}, than those in the clean regions of the south Atlantic. The ubiquitous presence aged biomass burning smoke transported from fires in western Central Africa towards SWA provides a source of CCN in greater concentrations than may be found in clean, remote regions.

One of the aims of this paper was to assess the relative impacts of local and transported aerosols on the microphysics of continental clouds over SWA. None of the offshore differences in CDNC, either between the different offshore CDNC on different days, or the different offshore/inland ratios, corresponded with differences in the measured aerosol concentrations. Therefore other factors must have been at play to cause the variability in the offshore cloud measurements, such as different dynamics, thermodynamics, or simply random biases due to our limited set of observations. Our measurements of offshore clouds were sporadic rather than systematic, and it is difficult to assess the statistical significance of the variability within these offshore clouds, or the differences observed between the clouds measured offshore and inland. Figure 2 shows that in our measurement region, the low-level cloud fraction offshore was much lower than inland, and most of the inland cloud formed over the continent, rather than forming offshore before moving inland. The complex dynamic and thermodynamic structure of the marine and continental boundary layers (Wood, 2012), and the diurnal cycle of these differences, mean that the offshore and inland clouds are not directly comparable to assess the impact of local aerosol emissions.

In contrast, the offshore aerosol measurements are much more robust. There were aerosol measurements from a larger number of days, with a much longer sampling time on each day, and aerosol concentrations are much less susceptible to changes in boundary layer thermodynamics compared to clouds. Our measurements showed an average ~40% increase in regional accumulation mode aerosol inland compared to offshore. The accumulation mode is the most important for CCN, as particles smaller than this are too small to activate. Haslett et al. (2019) also found a 40% enhancement in the inland accumulation mode aerosol concentrations compared to offshore using SMPS data, which is in excellent agreement with our GRIMM measurements of N_{250}. In localised city plumes, highly concentrated emissions had a noticeable increase in CDNC, but on a regional scale the majority of accumulation mode aerosol were due to long-range transport.

Our model simulations allow us to isolate the effects of specific aerosol modes and dynamics. The increase in CDNC inland due to local aerosols is strongly sensitive to the updraft velocity used in the simulation, but for our representative morning and afternoon updrafts local aerosols increased inland CDNC by and 44 and 65%. As these numbers are larger than our measured increase in accumulation mode aerosol, some of the particles in the Aitken mode fits must be having an influence. Regardless, local aerosols constitute less than half of the regional CCN over SWA. Any future increases in the local anthropogenic emissions during the summer monsoon will be on top of the transported background, and this reduces the sensitivity of CDNC to local emissions, which form less of the total."

Below I collected the passages in your manuscript I stumbled upon, which all more or less deal with this issue:

On page 10, line 4-5, referring to Fig. 3, you say "Despite the presence of urban emissions, anthropogenic aerosols had no clear effect on cloud cover downwind of the major cities,...". This seems vague - by simply looking at Fig. 3, one region with the highest LLC fraction (in the middle of the figure, longitude 3-4◦, latitude 8-9◦) is north of the biggest city you indicated (LA) and also close to the highest "density" of cities on the map (IB, OG, IL). And then there is another high LLC fraction north of AJ, another larger city. - At the same time, there are mountain slopes, so you might be correct, but judging from the figure you cannot say that anthropogenic aerosol did not have a clear effect. (I wrote this remark given above before reading on, and later found moreon this:) You do show later on that there is an effect (Fig. 4d) on CDNC (which is, on average, increased to~1.5 times higher values, page 12, line 2), and you again show this in your modelling – so there might not be a large effect, but still, there is one, so your remark on page 10, line 4-5 should be tuned down, maybe already pointing towards these later results, or deleted at all. And I think it is all the more interesting that you see this increase in the CDND in Fig. 4d ALTHOUGH "However, these highly polluted plumes represent less than 10% of the inland data measured over the region."(as you say on page 12, lin 2-3).

The reviewer does not seem to be clear on the wind direction. We have added this to Figure 3 to show the range at this time of day. Looking at the area the reviewer says is downwind of Lagos (LA), much of it is actually not downwind of Lagos. It is arguably far downwind of Lome (LO) and Cotonou (CO), but if we imagine a plume travelling downwind from these cities, the cloud cover only increases when you reach the hills. Most of the area they say is downwind of Abidjan (AJ) is actually not downwind of Abidjan. Bearing the wind direction in mind, the reviewer's argument doesn't really stand up to scrutiny. The part about CDNC being 1.5x higher inland refers only to the thickest city plumes, and was not widespread. This is already stated in the manuscript. Please also see our response to Rev#2 starting "We have added to section 3.2"

page 11, line 10: Already upon my first reading, I made the following note here, and this got only more relevant: The most striking difference between offshore and inland data, as I see it, is the larger average CDNC for the inland data. You do not discuss this at all. Would there be generally larger aerosol

concentrations over land (also from natural sources)? Or any other idea where that could come from?

Please see our response to R3 Major concerns, and our changes to the discussion

page 12, line 14-15: Here you clearly describe that the ground acts as an aerosol source: "Inland, accumulation mode aerosol concentrations were highest closer to the ground, and decreased with altitude up to around 2 – 2.5 km." There is a factor of roughly 2.5 between values of N_250 on ground and above 2km.

Agreed, this is a good insight. We have added to Sect. 3.4

"Together, these aerosol profiles suggest local terrestrial sources adding to the background aerosol on a regional scale."

page 13, line 1-2: When looking at Fig. 5, it struck me that up to 2.5km mostly N_250 and CDNC are roughly similar for the offshore cases, while for the inland cases CDNC are roughly double the amount of N_250. That would imply that there must be more particles < 250nm in the air inland (compared to offshore) which can be activated to cloud droplets, otherwise the CDNC are difficult to explain.

We agree it is difficult to explain using aerosol alone. We performed an extensive set of model runs to investigate the relative impacts of the offshore and local aerosol in clouds with different dynamics. Please see the updated discussion, which addresses this issue. We also agree that there are differences in the aerosols smaller than 250nm, as we have shown in Fig. 8. This is discussed further by Haslett et al. (2019).

page 13, line 14ff: You say "There are differences in aerosol and CDNC between inland and offshore clouds, but not in R_Eff, : : : " – below 1.5km, the "bump" that shows up in CDNC for the offshore case (towards lower values at 0.75 km and 1.25 km) is reflected in larger R_Eff (comparing _ 8-9 micrometer offshore to _ 6-7 micrometer inland). So this sentence needs a reformulation. Already Fig. 4b showed a totally different cloud droplet size distribution for offshore and inland conditions.

In the preceding sentence, we have already said

"Between 0.5 and 1.5 km, the clouds offshore tended to have higher R_{Eff} than those measured inland at the same altitude…."

Figure 4b does not show a cloud droplet size distribution, but we have already addressed the different PDFs of CDNC, that it does show, multiple times in the results and discussion.

page 14, line 2-3: There is a factor of _ 1.5 between N_250 that you give for offshore and inland, again with concentrations inland being larger. This, together with my comment concerning page 13, line 1-2, I am not sure if it can really be said that "aerosol imported from offshore comprised the majority of accumulation mode aerosol inland".

There would need to be a factor of >2 increase inland in order for the majority of aerosol to not be from offshore. The local contribution would have to be larger than the transported pollution, which our measurements show it is not.

page 15, line 8-10: Increase in mean updraft and in CDNC do not really mirror each other. While CDNC increase from 7 to 13 LST and then stay quite stable, the vertical velocity, implying a possibly stronger role of dynamics, is quite stable and has a distinct peak at 17 LST (followed by data ta 15 and 19 LST). So it is not so clear to me that dynamics explain the observations.

We have changed this to say

"….suggest that dynamics played a role in the diurnal cycle of CDNC, but cannot fully explain the changes."

page 15, line 23-25: It would be good to know how the inland size distribution exactly was derived (measured when and where)? But in any case, here it can clearly be seen that inland and offshore particle number size distributions differ massively, with much higher concentrations for particle sizes _ < 200 nm for the inland aerosol.

Please see our response to Rev #1's comment starting "Generally aerosol size distributions…"

page 19, line 21-25: As said above, looking at your data gave me an impression that is deviating from your interpretation. Instead, and increase in particle number concentrations from offshore to inland conditions might indeed play a role.

Our modelling simulations agree that the local pollution played a role, and we have said as much in the abstract, results, discussion and conclusions.

page 19, line 21-23: "It may be that our poor statistics of offshore data present a misleading distribution in Fig. 4b, or that there was a systematic and ubiquitous increase in CDNC due to some other factor such as stronger updrafts inland." Again, the "poor statistics" offshore is certainly better constrained than stronger updrafts inland. Admittedly, an increase by a factor of 1.5 for CDNC might not be huge, but at least there seems to be some effect, there.

Please see our earlier comment regarding changes to the discussion

page 20, line 23-24: In light of all the above, this sentence should be revised.

This refers to the broad-scale cloud field, please refer to our comment starting "The reviewer does not seem to be clear on the wind direction…"

page 21, line 1-3: And again, there is evidence in your data that APNC increased as soon as air masses were transported over land, although they might have been already "mildly polluted". (The particle number size distributions offshore and inland are VERY different!) And it is true that polluted air masses are less susceptible to a further increase in APNC, but you do see an increase, nevertheless. So again, this sentence should be revised.

We have changed "limited susceptibility" to "reduced susceptibility". They do have reduced susceptibility because doubling the local aerosol only causes a 13% increase in CDNC, not a ~100% increase. We have replaced similar terms like "limited effect" with "reduced effect" throughout the paper

page 21, line 24-25: Again, the "high aerosol background from transported smoke" was somewhat lower than what that sentence suggests.

We have changed it to just "the aerosol background"

Minor and technical comments:

page 9, line 11-13: You write "A further region of higher cloud cover was seen just inland : : : ". If I get the color scale right, then closer to the coast the coverage is 70%, drops to 60% and then again increases to 70%. The description here sounds a bit different. To me this area does not necessarily look like two separate regions, and the clouds closer to the sea do not really dissipate as they move further inland. A reformulation is needed.

The inland region starts forming at 3AM, but the sea breeze clouds start forming around 9AM, so they are different. There's not necessarily a full gap between them at midday in terms of total cloud cover, but the two bands show different daily cycles.

page 13, line 6: "is" is missing between "This due".

Done

page 13, line 12: The sentence is more general than what you report. Following "This implies that offshore clouds" insert "at altitudes above 1.5 km" and in the following line change "that clouds offshore" to "that these offshore clouds".

Done

page 14, line 18-19: The CDNC does not really "peak around 15:00 LST". There is a clear increase from before to after local noon, but after that, looking at all mean and percentiles, it seems to rather reach a plateau, until after 18:00.

This now says

"A trend is apparent in the data, as CDNC increased inland throughout the morning and remained higher through the afternoon"

page 17, line 1-2: You say ": : : for the average updrafts the main sensitivity of CDNC is not so clear-cut." What do you mean by "is not so clear-cut"? For the average updrafts, there is a difference, as you state in the text above, so I am confused by this statement here.

We have replaced this with

"…the clouds are in a regime where they are sensitive to both changes in updraft and aerosol"

page 17, line 3-4: You say that "A large fraction of the day-to-day variability : : : derives from the day-to-day variability in the offshore aerosol brought inland." But isn't this indicated by the bluish color band? The variability of the inland aerosol (greyish color band) is much larger. This confused me a little (I thought you might have mixed up the colors, but maybe you didn't?), and it would be fair to at least mention the fact that the inland aerosols' variability is even larger.

We have added a sentence to clarify

"At higher updrafts, the variability from offshore aerosol causes ~50% of this inland variability, but at lower updrafts this fraction increases to 100%."

page 17, line 4: Following the above mentioned sentence, you mention "these average updrafts", and maybe I was still confused from that "color issue"

above, but I first didn't get which updrafts you mean. Maybe it would be better to relate to them as "the average morning and afternoon updrafts as defined above", or something like this.

Done

page 17, line 18-19: The change in CDNC you report for omitting local emissions in your model also nicely reflects the change in CDNC between offshore and inland that you showed above. You should mention this.

We have addressed this issue in the updated discussion

page 17, line 20ff: "The effect of varying local aerosol emissions does not produce a linear relationship between CDNC and total aerosol concentrations." One additional reason for this, which you do not mention explicitly, yet, is that there is this rivalling effect where the maximum super-saturation at a given updraft is lower when there are higher concentrations of CCN, due to the water vapor being consumed faster, hence the critical size above which particles activate to droplets becomes larger.

We have added

"This nonlinearity may be related to a reduction in supersaturation for a given updraft velocity, as CCN number concentration increases and water vapour condenses more quickly."

page 18, line 21: Is "flat" a good way to describe an ocean surface? Maybe it is (I'm not a native speaker), but I would think there might be a better word, like "calm" or something else?

We have clarified *"flat (compared to the land surface)"*

page 22, line 22: There are too many quotation marks around "borderline".

Fixed

References not in the main text

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Aerosol influences on low-level clouds in the West African monsoon

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Abstract. Low-level clouds (LLC) cover a wide area of southern West Africa (SWA) during the summer monsoon months, and have an important cooling effect on the regional climate. Previous studies of these clouds have focused on modelling and remote sensing via satellite. We present the first comprehensive set of in situ measurements of cloud microphysics from the region, taken during June – July 2016, as part of the DACCIWA (Dynamics-Aerosol-Chemistry-Clouds Interactions in

5 West Africa) campaign. This novel dataset allows us to assess spatial ~~and temporal~~ , diurnal, and day-to-day variation in the properties of these clouds over the region.

LLC developed overnight and mean cloud cover peaked a few hundred kilometres inland around 10:00 local solar time (LST), before clouds began to dissipate and convection intensified in the afternoon. ~~Additional sea breeze clouds developed near the coast in the late morning, reaching a maximum extent around 12:00 LST.~~ Regional variation in LLC cover was largely

10 orographic, and~~determined by the modulation of the cool maritime inflow by the local orography, with peaks on the upwind side of hills and minima on the leeward sides. In the broad-scale cloud field,~~ no lasting impacts in cloud cover related to ~~anthropogenic aerosol~~pollution plumes were observed downwind of major population centres.

The boundary layer cloud drop number concentration (CDNC) was locally variable inland, ranging from 200 to 840 cm^{-3} (10th and 90th percentiles at standard temperature and pressure), but showed no systematic regional variations. Enhancements were seen in pollution plumes from the coastal cities, but were not statistically significant across the region. The majority of accumulation mode aerosols, and therefore cloud condensation nuclei, were from ubiquitous biomass burning smoke trans-

5   ported from the southern hemisphere. ~~Consequently, all clouds measured (inland and offshore) had significantly higher CDNC and lower effective radius than clouds over the remote south Atlantic from literature.~~

To assess the relative importance of local and transported aerosol on the cloud field, we isolated the local contribution to the aerosol population by comparing inland and offshore size and composition measurements. A parcel model sensitivity analysis showed that doubling or halving local emissions only changed the calculated CDNC by $13 - 22\%$, as the high background

10   meant local emissions were a small fraction of total aerosol. As the population of SWA grows, local emissions are expected to rise. Biomass burning smoke transported from the southern hemisphere is likely to dampen any effect of these increased local emissions on cloud-aerosol interactions. An integrative analysis between local pollution and Central African biomass burning emissions must be considered when predicting anthropogenic impacts on the regional cloud field during the West African summer monsoon.

15   **1   Introduction**

During the summer monsoon in June – September, large areas of southern West Africa (SWA) are covered by low-level clouds (LLC), which form overnight and thicken in the morning, before breaking up in the early afternoon (van der Linden et al., 2015). By presenting a high albedo surface close to the ground, these clouds generate a strong surface cooling. Many climate models struggle to accurately represent LLC (Hannak et al., 2017), and measurements of LLC (and their radiative

20   interactions with higher-level cloud layers) are a key uncertainty in the quantification of the overall cloud radiative effect in SWA (Hill et al., 2018). Most of the population centres in SWA are located near the coast, and plumes of local anthropogenic pollution are transported inland (Deroubaix et al., 2018), potentially being entrained into cloud base. Over the coming decades, the population of SWA is expected to undergo large increases (United Nations, 2017), leading to corresponding increases in emissions of anthropogenic pollution (Liousse et al., 2014). Such increases may affect dynamics and cloud microphysics in

25   the region, and it is therefore of interest to determine any impact on the regional climate such as changes in cloud cover and precipitation (Knippertz et al., 2015b).

The DACCIWA (Dynamics-Aerosol-Chemistry-Clouds Interactions in West Africa) project (Knippertz et al., 2015a) was developed to provide a comprehensive overview of cloud-aerosol-precipitation interactions in the region. The program studied different scales, from local emission measurements near source, to regional sampling using aircraft, remote sensing and model

30   analyses. This study focuses mostly on in situ cloud measurements made during the DACCIWA aircraft campaign (Flamant et al., 2018b), which took place between 29 June and 16 July 2016. Three research aircraft, each equipped with a suite of atmospheric measurement probes, were based out of Lomé, Togo, and conducted 50 research sorties flying over Togo, Benin, Ghana and Côte d'Ivoire, of which 33 included in-situ sampling of cloud properties.

The flying campaign took place in typical, post-onset West African monsoon conditions (Knippertz et al., 2017). In the daytime continental boundary layer, the southwesterly monsoon flow dominates the wind field, particularly in the lower kilometre of the atmosphere. Above around 1.5 – 2 km, the wind direction shifts to easterly, and some easterly waves and vortices passed through the region during the study period (Knippertz et al., 2017). Near the coast, winds are slowed by boundary layer

5   turbulence, generating sea breeze clouds along a convergence front that moves up to a few tens of kilometres inland during the day (Adler et al., 2017; Deetz et al., 2018b; Flamant et al., 2018a). As the turbulence subsides in the evening, the monsoon flow strengthens to bring this front inland as the nocturnal low-level jet (NLLJ), a strong flow of cool and humid maritime air, often penetrating over 180 km inland (e.g. Kalthoff et al., 2018). We follow the convention of Adler et al. (2019) in referring to the various flows bringing marine air inland as the Gulf of Guinea maritime inflow, or simply maritime inflow for short.

10   The NLLJ appears to be a key factor for initiating the formation of nocturnal LLCs, and previous studies have suggested several factors may be at play, including the transport of moisture and cold air inland and the impact this has on factor such as turbulence and the radiation budget. Recent studies by Babić et al. (2019) and Adler et al. (2019) suggest the main factor may be the cooling effect of a current of cold air moving inland. ~~The jet is strongest a few hundred metres above ground level Kalthoff et al. (2018), and Schrage and Fink (2012) suggested shear-driven turbulence below this maximum draws moisture~~

15   ~~upwards from near the surface. Schuster et al. (2013) concluded that cloud formation was the result of a subtle balance of several factors including shear-driven turbulence, orographic lift, and cooling by advection of cool air in the maritime inflow. Adler et al. (2019) recently quantified the boundary layer heat budget from 11 case studies using observations from a ground station near Savé during the DACCIWA campaign. They calculated that 50% of the cooling prior to LLC formation was due to advection of cold maritime air, with roughly 20% each from radiative and sensible heat flux divergence. A similar analysis by~~

20   ~~Babic et al. (2019) on a single case study found advection of cool air was responsible for 72% of boundary layer cooling.~~ The importance of the maritime inflow in cooling a particular location means that orography plays a significant role in determining geographical variation in the cloud field. Higher LLC coverage is found on the leeward side of slopes, where the maritime flow can reach more easily, and where this flow is also forced to rise orographically (van der Linden et al., 2015).

Deetz et al. (2018b) modelled the effects of changing aerosol emissions on the cloud fields in SWA. They suggested that

25   the reduction of land-sea temperature gradient from increasingly hazy conditions led to a weakening of the monsoon flow and nocturnal low-level jet, and a delay in stratocumulus to cumulus transition and cloud breakup. Haslett et al. (2019) recently described the aerosol properties measured during the DACCIWA aircraft campaign, and showed that a large background of transported biomass burning pollution from the southern hemisphere was ubiquitous in the West African boundary layer. Although city emissions resulted in aerosol load increase directly downwind of urban conglomerates (Brito et al., 2018), the

30   injection of aerosols prone to activate as cloud nuclei (> 0.1 μm) are thought to have limited impact on an already elevated background. This transported biomass burning background was reproducible in a modelling study by Menut et al. (2018). Painemal et al. (2014) showed that cloud-aerosol interactions with biomass burning smoke were the main factor governing cloud microphysical properties in clouds north of 5°S over the South Atlantic, but so far no study has measured the microphysical properties of clouds over SWA to quantify the relative influences of transported pollution and local anthropogenic aerosol

35   emissions.

The objectives of this paper are to:

1. Provide a statistical overview of in situ cloud properties measured during the DACCIWA aircraft campaign.

2. Compare the broad-scale cloud field during the DACCIWA aircraft campaign to previous overviews of the region, to assess the representativeness of the study region and period.

3. Assess the impacts of local urban emissions and transported background pollution on cloud properties in the region.

The conclusions we draw may be used in process and regional modelling studies, to improve assessments of the impacts of increasing urban emissions on regional cloud, and consequently precipitation and climate, under different pollution scenarios.

2 The DACCIWA aircraft project

The flying campaign took place from 29 June – 16 July 2016, and was a collaboration between science teams flying on three aircraft: the British Twin Otter operated by British Antarctic Survey, the French ATR42 (hereafter referred to as the ATR) operated by SAFIRE (Service des Avions Francais Instrumentés pour la Recherche en Environnement), and the German Falcon aircraft operated by DLR (Deutsches Zentrum für Luft- und Raumfahrt). The project has been described in detail (Flamant et al., 2018b); here we provide a brief description of the flights and relevant instrumentation. Research sorties were conducted in daylight hours from Lomé airport (6.17°N, 1.25°E), though the ATR refuelled twice in Abidjan (5.26°N, 3.93°W). Figure 1 shows flight tracks from all three aircraft below 1 km. The local time in Togo, Ghana and Côte D'Ivoire is UTC, though Benin uses UTC+1.

Many sorties were flown along a SW-NE axis from Lomé to Savé (as shown in Fig. 1), close to the average direction of the low-level monsoon flow. These flights included profiles up and down through cloud, as well as straight and level runs above, in, and below cloud to provide statistical mapping of clouds, aerosol and radiation. Other flight objectives included measuring emissions from different sources (such as cities, oil rigs, and ships), and mapping clouds and pollution further west over Ghana and Côte d'Ivoire. The large majority of measurements were taken over the African continent. Flights over the sea were carried out on ~~several~~eleven days during the campaign, and these are sufficient to gauge the variability of aerosol offshore. Cloud penetrations on these offshore flights were relatively few, and it is difficult to gauge how statistically robust the offshore cloud measurements were. In this analysis we use cloud data from all flights of the campaign, and mostly only consider data from the lowest 1 km to investigate the effects of aerosol on boundary layer clouds over the African continent.

2.1 In situ instrumentation

Each aircraft was equipped with a suite of instrumentation to measure basic meteorological variables such as temperature, humidity, pressure, and winds. Further details are provided by Flamant et al. (2018b). Cloud drops 3 – 50 μm in diameter were measured using a cloud droplet probe (CDP) on the Twin Otter, CDP and/or fast CDP on the ATR, and a cloud-aerosol spectrometer (CAS) on the Falcon (Baumgardner et al., 2001; Voigt et al., 2017). Both CDPs had modified pinholes to reduce

coincidence (Lance, 2012), however the CAS did not, and the CAS CDNC measurements were corrected for coincidence at high concentrations as described by Kleine et al. (2018). Each instrument had its sample area measured using a droplet gun prior to the campaign, and sizing was calibrated in the field using glass beads of known size and refractive index. To ensure comparability between CDNC measurements on the different platforms, we performed a statistical analysis of all CDNC measurements made between 0 and 100 km inland over Togo/Benin at altitudes below 1 km. CDNC measured on the different platforms showed excellent agreement, with the medians and quartiles all agreeing within 5%.

We also consider cloud drop effective radius, defined as

$$R_{\text{Eff}} = \frac{\int_0^\infty R^3 n(R) \, dR}{\int_0^\infty R^2 n(R) dR},$$

where $n(R)$ is the number concentration of drop with radius R.

For R_{Eff}, average vertical profiles between 0 and 100 km inland showed the ~~cloud probes~~median values agreed within ~1 μm, which is within the uncertainties of the instruments, and the interquartile ranges were similar for each platform.

Data were considered in-cloud when the measured liquid water content (LWC) was greater than 0.1 g m^{-3}. This relatively high LWC threshold minimises the effects of diffuse cloud edges on our measurements, and removes swollen aerosol layers (see Deetz et al., 2018a; Haslett et al., 2018). Measurements of CDNC are reported in number per cubic centimetre (cm^{-3}), corrected to standard temperature and pressure (STP, 273.15 K and 1013.25 hPa).

Aerosol composition was measured by compact time-of-flight aerosol mass spectrometers (AMS, Drewnick et al., 2005). Two AMS instruments were mounted on the ATR and Twin Otter, which were each calibrated using nebulised ammonium nitrate and ammonium sulfate to determine absolute and relative ionization efficiency. We also consider aerosol size distributions, which were measured using GRIMM optical particle counters (model 1.109 on the ATR and 1.129 on the Twin Otter and Falcon), and a scanning mobility particle sizer (SMPS) on board the ATR. Further details on the AMS and SMPS are provided by Brito et al. (2018) and Haslett et al. (2019). From the GRIMM measurements we consider only the total concentration of particles larger than 250 nm (N_{250}), and from the SMPS we use aerosol size distributions in the range 20 – 500 nm. The strength of the GRIMM dataset is that all three aircraft had instruments running on every flight, making it a useful dataset for measuring changes in accumulation mode aerosol concentrations. The SMPS data coverage is more limited, and is restricted to straight and level runs, but provides a more detailed measurement of the aerosol size distribution. Aerosol data were screened for cloud using a threshold for LWC of 0.01 g m^{-3} and CDNC of 10 cm^{-3}.

Carbon monoxide (CO) concentrations were measured using infrared absorption spectrometry, by an Aerolaser AL5002 on the Twin Otter, SPectrometre InfraRouge In situ Toute altitude (SPIRIT, Catoire et al., 2017) on the Falcon, and a Picarro Analyzer G2401-m on the ATR. We also consider vertical velocity, which was measured using wind probes on the ATR and Falcon.

2.1.1 Cloud satellite measurements

Using satellite measurements allows us to view the cloud field over a large region. We used the optimal cloud analysis (OCA, Watts et al., 2011) product taken from the Meteosat Spinning Enhanced Visible and InfraRed Imager (SEVIRI) spectrometer.

This product provides cloud top pressure (CTP) and cloud optical thickness (COT) for the top two cloud layers (looking from above), for scans every 15 minutes. We use this product to derive the LLC fraction (for pressures above 680 hPa), as described in appendix A. Comparison with the LLC fraction using ceilometers, based at the DACCIWA supersites near Savé and Kumasi (Kalthoff et al., 2018), showed that this satellite-derived product agreed with ground measurements within ~10% absolute cloud fraction, capturing both the absolute values and also the diurnal cycle, particularly during daylight hours. Further details are provided in appendix A.

2.1.2 Parcel modelling

Parcel model simulations of aerosol activation were carried out using the Aerosol-Cloud and Precipitation Interactions Model (ACPIM) (Connolly et al., 2009). Multiple aerosol modes of defined dry size distribution and composition can be initialised, and the simulated air parcel rises at a prescribed updraft velocity. ACPIM uses bin microphysics and thermodynamics to evaluate the rate of condensation of water in each size bin numerically using the diffusional drop growth equation and Köhler theory (see Topping et al., 2013), so does not rely on bulk parametrizations of cloud drop activation. The number of particles in each aerosol mode growing above the critical diameter is then evaluated to determine the CDNC.

ACPIM was initialised starting at 95% humidity, 296 K, and 960 hPa, which are typical of conditions just below cloud base over Togo. Aerosol size distributions from SMPS and relative compositions from AMS were averaged over regions at various distances offshore or inland over SWA, and these were used to initialise the model for several runs at different updraft velocities. The hygroscopicity parameter (κ) values of inorganic and organic aerosols were assumed to be the same as ammonium sulfate and fulvic acid, which is chemically similar to highly aged organic aerosol (Jimenez et al., 2009). By using fixed, representative thermodynamic starting conditions, varying the updraft velocities and aerosol number, size, and composition, allows us to determine the relative sensitivities of CDNC to both variables.

Our simple modelling scheme does not include factors such as entrainment, collision-coalescence, or an investigation into the impacts of aerosol mixing state, which can have a large effect on calculated CCN (Ren et al., 2018). We have also only considered a limited number of aerosol species, and other components such as black carbon, sea salt, and mineral dust will have been present. This is not intended to be an exact simulation, but a sensitivity analysis using physically reasonable approximations. A detailed study of aerosol activation is beyond the scope of this analysis and is non-trivial (e.g. Sanchez et al., 2017). Additionally, we have no measurements with which to constrain factors such as the size-dependence of chemical composition and mixing state, which strongly affect the accuracy of CCN closure calculations (Moore et al., 2013), so such an investigation would be purely speculative.

3 Results

3.1 Diurnal cycle of low-level cloud cover

Figure 2 shows the average diurnal cycle of LLC fraction over Togo and Benin, where the aircraft operated, plotted versus distance North of the coast and approximate latitude. LLC peaked around 10:00, and fell to a minimum around 18:00. The initial surface warming in the few hours after sunrise causes the nocturnal clouds to rise and thicken, before eventually breaking up (van der Linden et al., 2015; Kalthoff et al., 2018). Cloud cover was generally greater inland than offshore, except for a period in the late afternoon / early evening. A region of more extensive cloud cover was seen developing overnight, around 50 – 150 km inland. This is the region the maritime inflow reaches on a typical evening, though it is interesting that cloud cover did not increase overnight nearer the coast. The highest LLC fraction was around 250 km inland, in a region that began to develop overnight but became particularly pronounced during daylight hours. A further region of higher cloud cover was seen just inland of the coast. These are most likely sea breeze clouds developing from mid-morning to the early afternoon, and moving up to ~50 km inland before dissipating. Lower levels of cloud cover (average 0.24) were seen over all inland areas between 16:00 and 00:00. By taking the mean over several weeks we lose the extremes of these values- on some evenings cloud cover was zero and on some mornings it reached 100%. This averaging allows us to assess which features are statistically robust, and minimises transient features in the cloud field. Figure 2 bears a striking similarity to similar diagrams of boundary layer temperature and relative humidity presented by Deetz et al. (2018a), suggesting transport of cool, humid air inland by the maritime inflow is a key factor in determining LLC cover.

3.2 Regional variation in low-level cloud cover

Figure 3 shows the orography of West Africa, and a map of LLC fraction at its 10:00 UTC peak. The LLC fraction decreases dramatically in the drier regions north of ~10 – 11°N. South of this latitude, the largest LLC fractions were seen on the upwind side of slopes, and the lowest on the leeward sides, for a southwesterly monsoon flow. Additionally, a patch of lower LLC fraction is seen just offshore of Ghana, Togo and Benin, which is related to the colder waters there due to the coastal upwelling system (see Flamant et al. (2018a)). The areas of high mean LLC are the most robust features, with the lowest standard deviation, meaning patches of continuous cloud were present on almost all days during the campaign. Areas with lower average LLC fraction had higher standard deviation and were therefore more variable; the cloud cover in these regions varied day-to-day but was lower on average. The features in our mean LLC fraction over the region are broadly similar to those presented in multi-year satellite observations by van der Linden et al. (2015), meaning the measurement period is likely to be broadly representative of a typical monsoon season. The absolute values we present are more in line with the synoptic observations than the satellite measurements in the previous work, as the LLC fraction product here takes into account times when LLC would not be visible due to higher cloud obscuring the view.

Figure 3 also shows the major population centres in the region. Pollution plumes are expected to extend for hundreds of kilometres downwind of the major cities (Deroubaix et al., 2019). Figure 3 does not show any features extending downwind of the major cities that would represent a signature for anthropogenic influence on cloud cover on a regional scale. on Despite

~~the presence of urban emissions, anthropogenic aerosols had no clear effect on cloud cover downwind of the major cities, and~~
This was also the case in plots similar to Fig. 3 at all times of day.

3.3 Regional variation in cloud microphysics

To investigate the effect of local emissions on cloud microphysical properties, we collated cloud measurements below 1 km in altitude from all three aircraft and compared them with distance to the coast (and the population centres nearby) on a north/south axis, as shown in Fig. 4a. Figure 4b shows corresponding normalised histograms in the different regions. The limited spatial area of the aircraft sampling means that regional scale orographic effects discussed in Sect. 3.2 are not likely to have a large effect on in situ cloud properties. The heterogeneous nature of the dataset means that some areas and some days have large numbers of data points recorded, while others have relatively few. In particular, cloud measurements over the sea were sparse compared to inland. To give an indication of how statistically representative our data are, the number of data points and number of individual days the data are from, are also listed in Fig. 4a.

Offshore, we split the data up into two bins. The aim of the offshore analysis is to consider clouds and pollution conditions representative of the air upwind of the DACCIWA region, that is then brought inshore by the wind. Close to the coast, several factors may cause terrestrial pollution to be transported to an area offshore. Firstly, some areas of the coast are more prominent than others, for example the area located just offshore near Lomé may be downwind of Accra if the wind was blowing from a west-south-westerly direction. Secondly, sea breeze circulations may act to recirculate pollution (Flamant et al., 2018a). Additionally, a shipping corridor near to the coast presents a local source of CO and aerosol. In Fig. 4a there is no apparent effect of any pollution near to the coast, but plumes of CO and aerosol were observed up to 20 km offshore that would affect the analysis in Sections 3.6 and 4. For the remainder of this analysis, when considering "'offshore"' data, we therefore only consider measurement made at least 20 km south of the coast, to unambiguously remove any possibility of local terrestrial emissions affecting our offshore measurements.

In the offshore bin, there were CDNC measurements from 5 individual days, and the distribution of measured CDNC in Fig. 4b appears bimodal, with a main mode centred around $100 - 200 \ \mathrm{cm}^{-3}$, and a smaller mode centred around $500 \ \mathrm{cm}^{-3}$. Looking at Fig. 4a, this bimodality is the result of sampling different populations of clouds. This may be due to variable pollution conditions offshore, or some other factor. Bennartz (2007) studied CDNC in the global marine boundary layers, and found average CDNC in the pristine south Atlantic was $67 \ \mathrm{cm}^{-3}$, reported in ambient temperature and pressure. For stratocumulus topping the south Atlantic marine boundary layer at 800 hPa and 280 K, this number is roughly equivalent to $87 \ \mathrm{cm}^{-3}$ when corrected to STP, which lies around the 10th percentile of the CDNC measured in offshore clouds in the DACCIWA region, meaning the offshore measurements presented here were representative of moderately polluted clouds.

Moving inland, several differences are apparent. The inland CDNC distributions in Fig. 4b are approximately Gaussian, centred around ~400 cm^{-3}, but with a long tail extending up to over 1000 cm^{-3}, which was measured most frequently near the urban centres near the coast, and diminished further ~~from the coast~~inland. Some clouds were observed with CDNC over 1500 cm^{-3}. This long tail on the distribution is due to the effect of thick urban plumes, but it only represents a relatively small fraction of the inland data. The large majority of CDNC measurements inland were in the range $200 - 840 \ \mathrm{cm}^{-3}$ (10th and

90th percentiles), and medians were 470 cm^{-3} up to 50 km from the coast, and ~430 cm^{-3} further inland. It is possible that some of this small difference in the medians may be due to measurement of more convective sea breeze clouds near the coast, but it is relatively minor overall.

To investigate the effect of local pollution on CDNC, Fig. 4c shows the distribution of CO concentrations measured in inland clouds. The inland CO data show a mode with mean and standard deviation of 141 $\pm$17 ppbv, but the distribution is asymmetric, with a tail extending to higher values above around 160 ppbv. We therefore used this value of 160 ppbv (the 93rd percentile) as a threshold to distinguish the thickest pollution plumes. Our measurements in this polluted tail were generally in and around the major cities, which were the one of the research themes of the DACCIWA campaign. On the regional scale these are likely to be over-represented compared to the more sparsely populated regions such as Ghana and Côte d'Ivoire, but may be under-represented compared to the more polluted regions of coastal Nigeria. For comparison, the mean and standard deviation of in-cloud offshore CO concentrations were 143 $\pm$ 11 ppbv. This offshore value, and similarly the minimum inland CO concentrations of ~100 ppbv, is significantly enhanced compared to CO concentrations in the unpolluted south Atlantic, which reach lows of 60 ppbv in the absence of transported biomass burning smoke (Zuidema et al., 2018). This contrast highlights the ubiquity of transported biomass burning smoke affecting the DACCIWA region (Haslett et al., 2019).

Using the 160 ppbv CO threshold to stratify the CDNC highlights the effect of pollution plumes, which we show in Fig. 4d. Using only the inland data here removes any possible bias from differing conditions (for example dynamics) over the sea. The data in Fig. 4d show enhanced CDNC in the polluted plumes, with the entire distribution shifted around 1.5 times higher. However, these highly polluted plumes represent less than 10% of the inland data measured over the region.

There appeared to be a difference between the offshore and inland clouds that was not related to differing pollution. Inland clouds with CO < 160 ppbv (i.e. similar to levels found offshore) had a median CDNC of 430 cm^{-3}, compared to 265 cm^{-3} offshore. However, Fig. 4b shows some offshore clouds that had CDNC and CO comparable to those found inland. With the limited offshore data available we are unable to fully quantify whether there was any systematic difference, which might be expected due to different dynamical conditions.

3.4   Vertical profile of cloud microphysics

Figure 5 shows average vertical profiles of N_{250}, cloud drop number concentration, and cloud effective radius, measured at different distances north or south of the coast. The aerosol measurement here is only of particles larger than 250 nm, so it is representative of the variability of the accumulation mode, though not the total number. N_{250} does not show any variability related to smaller particles. Inland, accumulation mode aerosol concentrations were highest closer to the ground, and decreased with altitude up to around 2 – 2.5 km. There was relatively little meridional variation inland, but aerosol concentrations were enhanced by around ~40% near the ground compared to offshore concentrations. This enhancement decreased with altitude, as concentrations offshore were fairly constant up to 2.5 km, and above 2.5 km offshore concentrations were higher than those inland. Together, these aerosol profiles suggest local terrestrial sources adding to the background aerosol on a regional scale. In the boundary layer, the southwesterly winds brought offshore pollution inland, and local city emissions would be added to the pollution transported from offshore, but above the inversion at boundary layer top, the wind shifted to a regime dominated

by the African easterly jet and this linkage was broken. Above the boundary layer, distinct biomass burning plumes were encountered more often, which caused the larger variability in N_{250} in the free troposphere.

The CDNC profile inland (Fig. 5b) shows a decrease with altitude, with a similar profile to N_{250}, but the relative decrease was lower in magnitude. Inland there was high variability within all regions, but little difference between the average values of CDNC, though >100 km inland concentrations were ~10% lower than closer to the coast. Offshore concentrations at almost all levels were significantly lower than those inland, and the difference was larger than the differences in aerosol and did not correlate with differences in the aerosol vertical profiles. However, the majority of offshore bins in Fig. 5b show data from one or two individual days, so may not be representative of offshore clouds in general. If the difference is representative, it does not appear to be directly related to differences in accumulation mode aerosols.

Figure 5c shows the corresponding cloud effective radius profile. Again, there was relatively little difference inland, particularly in the boundary layer, and on the whole R_{Eff} increased with altitude. In the free troposphere there was more deviation, and R_{Eff} did not always increase with altitude. This is due to the structure of clouds over SWA; multiple cloud layers with distinct and separate bases were often present, meaning clouds at higher altitudes were not necessarily deeper than those below, and cloud observations at higher altitude were not necessarily higher above the cloud base than observations at lower altitude. Between 0.5 and 1.5 km, the clouds offshore tended to have higher R_{Eff} than those measured inland at the same altitude, which is consistent with the lower CDNC measured in this altitude range. However, throughout the rest of the lower troposphere, including near the surface, the clouds measured offshore had similar R_{Eff} to those measured inland at the same altitude, despite generally lower CDNC. This implies that these offshore clouds had lower liquid water contents than inland clouds at the same altitude, suggesting that clouds offshore may have been influenced by a different thermodynamic structure, such as different temperature or humidity profiles causing different cloud base heights.

Overall, relatively little systematic spatial variability is seen inland in aerosol or cloud parameters. There are differences in aerosol and CDNC between inland and offshore clouds, but not in R_{Eff}, meaning the differences cannot be solely due to increased aerosol activation inland. For the remainder of this analysis we will consider only clouds measured in the lowest kilometre of the atmosphere, as this covers the majority of the cloud measurements made in the morning, and to minimise the influence of the African easterly jet. Although the properties of individual clouds were variable, the average inland clouds below 1 km were fairly homogeneous regardless of the distance inland, meaning we can consider all inland clouds together and develop better statistics of the day-to-day variability and diurnal cycle of cloud properties.

3.5 Day-to-day variability

In the previous sections, we demonstrated that the ubiquitous background aerosol reaching SWA caused all clouds to be fairly polluted. Figure 6 shows the day-to-day variability in both the offshore and inland aerosol concentrations, as well as the CDNC. For the accumulation mode aerosol, the the inland concentrations of N_{250} were around 40% higher than offshore, though there was some day-to-day variability in this ratio. The average inland enhancement is in excellent agreement with Haslett et al. (2019), who also found a 40% enhancement in accumulation mode aerosol in continental background areas compared to upwind marine, using SMPS measurements averaged over the whole campaign. The day-to-day averages of the accumulation

mode aerosol number concentration from the SMPS showed good correlation with the GRIMM, with $R^2 = 0.7$, but data were only available for 9 days inland and 2 days offshore. The GRIMM data therefore show a similar trend as the SMPS, but with greater coverage. ~~concentrations offshore were 50 – 90% of those inland, and the offshore and inland concentrations were well correlated.~~ The mean and standard deviations in daily N_{250} are $170 \pm 50 \ \mathrm{cm}^{-3}$ offshore and $230 \pm 50 \ \mathrm{cm}^{-3}$ inland, meaning the average variabilities were similar. Together, this suggests that aerosol imported from offshore comprised the majority of accumulation mode aerosol inland, and also strongly influenced its day-to-day variability.

The day-to-day variability in the aerosol was not strongly represented in the CDNC measurements in Fig. 6b. There was a degree of correlation in the median CDNC and N_{250} measurements ($R^2 = 0.72$) after the 5th July, but there was no correlation in the first half of the campaign. On a cloud-to-cloud basis this correlation was dwarfed by the inherent local variability in CDNC. Comparing the inland CDNC to offshore, on four days the median values inland were 85 – 175% higher than those measured offshore, but 25% lower on one day. These differences were not correlated with differences in the offshore and inland aerosol. As was stated in Sect. 3.3, we do not have sufficient measurements to investigate or explain these differences in detail, or to assess if they are representative. On any individual day, the CDNC measurements were much more variable than the aerosol concentrations, suggesting highly localised factors such as entrainment or dynamics may have had an influence on the CDNC. Overall, the day-to-day variability in the inland CDNC and N_{250} were both ~20%, and there was a degree of correlation for some of the project, suggesting aerosol concentrations may have had some effect on daily CDNC. In the next Section, we discuss diurnal variation, to investigate these factors in more detail.

3.6 Diurnal variability of aerosols, clouds, and dynamics

The variation in inland CDNC at different times of day is shown in Fig. 7a. As some of the measurements were taken over a wide range of longitude, here we used the local solar time (LST) instead of UTC. A trend is apparent in the data, as CDNC increased inland throughout the ~~day and peaking around 15:00 LST~~ morning and remained higher through the afternoon. The inland CDNC measurements were 45 – 60% higher in the afternoon than before 08:00. The median of all CDNC measured inland in the morning was $430 \ \mathrm{cm}^{-3}$, and $540 \ \mathrm{cm}^{-3}$ in the afternoon. We draw this distinction because cloud cover peaked in the late morning, before decreasing in the afternoons as the clouds broke up. The 90th percentiles of CDNC in Fig. 7a exhibited a more dramatic increase in the afternoon, indicating a greater cloud drop concentrations in the more convective broken clouds.

CDNC is determined by the supersaturation in an updraft, and the concentration of aerosols that activate at that supersaturation. Figures 7b & c show the diurnal variations in N_{250} and in-cloud vertical velocity. The accumulation mode aerosol showed no strong diurnal variation, varying only $\pm 10\%$ during the day. In comparison, the distributions of vertical velocity showed a shift towards stronger updrafts in the afternoon. In any stable cloud layer the median vertical velocity is zero, and this was the case for all times of day shown in Fig. 7c other than the few cloud measurements made after 16:00 LST. As the median value of vertical velocity in a stable cloud is zero, the 75th percentile can be considered as a representative updraft, and is useful to investigate changes in updrafts that are present at cloud base, where the majority of aerosol activation occurs. Vertical velocity does not vary strongly with altitude in stratocumulus clouds (Wood, 2012), so although our measurements are not just from cloud base, they are still suitable to use in our simulations.

The distribution of vertical velocity was fairly stable throughout the morning, with 75th percentiles around $0.45\,\mathrm{m\,s^{-1}}$, and 90th percentiles around $1\,\mathrm{m\,s^{-1}}$. In the afternoon, the clouds became more convective, with 75th percentiles of vertical velocity of $0.5 - 1.6\,\mathrm{m\,s^{-1}}$ and 90th percentiles of $1.1 - 1.7\,\mathrm{m\,s^{-1}}$. The stronger updrafts in the afternoon suggest that dynamics played a role in the diurnal cycle of CDNC, but cannot fully explain the changes. ~~, rather than varying aerosol concentrations, were~~

5 ~~responsible for the increase in CDNC seen in the afternoons over SWA.~~ In the next Section we use parcel model simulations to quantify the effect of these factors on CDNC.

4 Parcel model simulations

The limited offshore dataset and the possibility of differing dynamics inland makes it difficult to assess the impact of local aerosol emissions on cloud properties based on measurements alone. To further investigate the relative sensitivities of CDNC

10 to aerosol properties and updraft velocities, we conducted a series of parcel model simulations using ACPIM, described in Sect. 2.1.2. We initialised the model with representative thermodynamic conditions and varied the updraft velocity and aerosol size distribution and composition based on the measurements made in the field campaign.

4.1 Aerosol size and composition

The average measured aerosol size distributions are shown in Fig. 8a – c. Here, the data coverage was more limited, and the

15 scanning time of the instrument was $90\,\mathrm{s}$, so we used a simple mean of all the data inland or offshore, averaging out any diurnal or day-to-day variation. While the Aitken mode was variable inland due to highly localised anthropogenic emissions, the accumulation mode showed minimal variability as the majority of particles in this size range were due to long-range transport (Haslett et al., 2019). This averaging approach is therefore reasonable to inform cloud activation modelling, as the accumulation mode is the source of CCN. The measured offshore average aerosol size distribution and accumulation mode composition are

20 shown in Fig. 8a & d. Two lognormal fits were used to approximate this size distribution for use in ACPIM. The measured inland average aerosol size distribution and accumulation mode composition are shown in Fig. 8c & f. The offshore size distribution was subtracted from the inland, to give an estimate of the average contribution of dispersed local emissions to regionally-averaged inland aerosol over SWA. Two lognormal fits were then used to approximate the average local emissions, and the sum of the four fitted modes makes up the measured size distribution inland. The lognormal fit parameters are listed in

25 Table 2. The utility of this approach is it allows us to scale the transported background aerosol (i.e. the offshore fits) and the local aerosol separately.

The measured size distributions in Fig. 8a & c, and the measured compositions in Fig. 8d & f, are almost identical to similar estimates presented by Haslett et al. (2019). The difference in our approach when highlighting the impact of local emissions is that we have subtracted the transported background to isolate the effect of dispersed local emissions on regional inland aerosol,

30 whereas Haslett et al. (2019) measured aerosol in the thickest urban plumes, but still on top of the transported background.

The offshore aerosol size and composition are consistent with being a mixture of aged biomass burning (Rissler et al., 2006; Capes et al., 2008; Sakamoto et al., 2014) and Atlantic marine aerosol (Zorn et al., 2008; Taylor et al., 2016). The two modes

of local aerosol were an Aitken mode containing the majority of the particle number concentration, and a smaller accumulation mode. We are unable to determine the specific source of each mode, but local emissions are largely from domestic wood burning and transport, and when combined these sources are capable of producing aerosols with this size distribution and composition (Maricq et al., 2000; Capes et al., 2008; Vakkari et al., 2014). More informative studies of aerosol size distribution in African

5   cities appear to be absent from literature. The local Aitken mode, which accounts for the majority of particle number inland, contains particles that were too small to make a significant contribution to CCN in stratocumulus clouds, and the majority of the aerosol that are large enough to activate into cloud drops were still the result of long-range transport.

4.2   Model results

We used the aerosol size distributions and composition shown in Fig. 8 and Table 2 as input for ACPIM simulations at varying
10   updraft velocities. The resultant simulated CDNCs are shown in Fig. 9a. For weak updrafts the different input aerosols makes little difference to the CDNC values, as particle activation starts from the largest sizes, where the inland and offshore size distributions were most similar. The difference between the two then increases with updraft velocity. For the representative morning updraft of $0.45 \ \mathrm{m\,s^{-1}}$, calculated as the 75th percentile of vertical velocity, the calculated enhancement in inland CDNC due to local aerosols is 44%. This enhancement is larger for stronger updrafts; for the representative afternoon updraft
15   of $0.78 \ \mathrm{m\,s^{-1}}$, it rises to 65%. In polluted clouds with relatively gentle convection, CDNC is more sensitive to changes in updraft than in aerosol (Reutter et al., 2009). However, Fig. 9a suggests the clouds over SWA are only in this regime for the lowest updrafts, and for the average updrafts the clouds are in a regime where they are sensitive to both changes in updraft and aerosol.the main sensitivity of CDNC is not so clear-cut.

A large fraction of the day-to-day variability expected in the mean inland CDNC, shown by the shadings in Fig. 9, derives
20   from day-to-day variability in the offshore aerosol brought inland. At higher updrafts, the variability from offshore aerosol causes 50% of this inland variability, but at lower updrafts this fraction increases to 100%. Using these average morning and afternoon updrafts (as defined above), the modelled CDNC is 31% higher in the afternoon than in the morning, which compares well to a 26% increase in the measured CDNC shown in Fig. 7.

Fig. 9b shows the effect of varying the local emissions on the calculated CDNCs, using these average morning and afternoon
25   updrafts. For comparison, the top axis in Fig. 9b shows the number concentration of aerosols larger than 125 nm (N_{125}) in the input size distributions, as a way of comparing the effects that scaling local emissions have on aerosol concentrations (i.e. transported background plus local emissions). The diameter of 125 nm was chosen as N_{125} is roughly comparable to the modelled values of CDNC in Fig. 9b, and is therefore an approximation of the modelled critical diameter, although the actual critical diameter will have been different in each model run, and varies as the aerosol size distributions and number
30   concentrations change.

An alternative way of looking at the impact of transported aerosols inland is to say that if local emissions were doubled, N_{125} would only be 38% higher, and CDNC would only increase by 17% in the morning and 22% in the afternoon. If local emissions were halved, N_{125} would be reduced by 19%, and CDNC would decrease by 13% in the morning and 16% in the afternoon. If local emissions were removed entirely, N_{125} would decrease by 38%, and CDNC would drop by 31% in the morning and

39% in the afternoon. Large changes in local emissions cause smaller changes in CDNC because the high aerosol background means they form only a fraction of the total aerosol. Additionally, the local emissions are smaller and less hygroscopic than the background aerosol, so they are less likely to activate. The effect of varying local aerosol emissions does not produce a linear relationship between CDNC and total aerosol concentrations. In particular, the effect of increasing total aerosol concentrations

5   has less of an impact on CDNC than decreasing aerosol. The exact comparability between the aerosol metric and CDNC will depend on the size assumed as representative of the critical diameter (125 nm in this comparison, but would vary as the aerosol changes), but the nonlinearity of the relationship between aerosol and CDNC is independent of this choice of size. This nonlinearity may be related to a reduction in supersaturation for a given updraft velocity, as CCN number concentration increases and water vapour condenses more quickly.

10   The values of CDNC produced by ACPIM are higher than the median values shown in the ambient measurements. We have acknowledged the limitations of our simplistic approach above, such as the assumptions regarding aerosol mixing state and the lack of entrainment in the model. Moore et al. (2013) summarised 36 previous studies of CCN closure, and similarly found that CCN concentrations were generally overpredicted, other than when using size-resolved measurements of aerosol composition. Additionally, a previous comparison by Simpson et al. (2014) showed that ACPIM calculated higher CDNC compared to

15   simpler parametrisations when using multiple aerosol modes. The values of modelled CDNC using the representative morning and afternoon updrafts are between the 75th and 90th percentiles of measured CDNC shown in Fig. 7a, meaning the difference between the average model and measured values is smaller than the variability in the measured data. ACPIM is an idealised simulation and we have used it here primarily to explore the sensitivity to local aerosol emissions, rather than arbitrarily try to tune factors such as aerosol mixing state, more realistic dynamics, or entrainment (of which we have limited or no

20   measurements) to make the CDNC agree perfectly.

We performed some additional simulations to explore some possible sensitivities in our assumptions. Firstly we considered the presence of a coarse sea salt aerosol mode, which we isolated from the GRIMM size distributions. The size distribution of this mode is listed in Table 2, and the composition was assumed to be NaCl. As the concentration in the sea salt mode is so low compared to the accumulation mode, this addition of a coarse sea salt mode made less than a 1% difference to the

25   derived CDNC. We also performed some illustrative runs on the inland aerosol- one possible scenario is that the local Aitken mode aerosol were from primary combustion. We assumed them to be 100% organic, and proportioned the composition of the local accumulation mode to match the total measured organic/inorganic ratio. Compared to the base case, this increased the inorganic fraction in the local accumulation mode from 31% to 41%, and the resultant CDNCs showed around a 12% increase. These simulations show that there are some differences that are attributable to assumptions in aerosol composition and mixing

30   state, but they are likely to be smaller than the differences we show related to the total amount of aerosol and updraft velocity.

It may be that entrainment ubiquitously affected clouds sufficiently to reduce the median CDNC, as the variability in CDNC on a particular day was much larger than the variability in accumulation mode aerosol. We have used a threshold for in-cloud data of $0.1\,\mathrm{g\,m^{-3}}$ and found a campaign median inland CDNC of $450\,\mathrm{cm^{-3}}$. Using a threshold of $1\,\mathrm{g\,m^{-3}}$ gives a value of $720\,\mathrm{cm^{-3}}$. This suggests that entrainment is likely to have affected CDNC, otherwise CDNC would not vary with LWC.

Convective clouds may have an adiabatic core that is relatively unaffected by entrainment (Heymsfield et al., 1978), but this is less likely to be the case in layer clouds that form the majority of our dataset.

5   Discussion

Most stratocumulus clouds are over the oceans, and most previous studies of cloud-aerosol interactions in stratocumulus clouds have focused on clouds in this setting. Allen et al. (2011) observed cloud drop number concentration in the southeast Pacific decreasing from 300 cm^{-3} near the coast to ~100 cm^{-3} 150 km downwind offshore. Their situation is simpler to visualise as the anthropogenic source regions are well defined, the ocean surface is flat (compared to the land surface), and there is no strong diurnal cycle in ocean surface temperature.the marine boundary layer. In Southern West Africa, the situation is made more complex by the combination of local sources and the variability of the dynamics, both in terms of the diurnal cycle and the difference between offshore and inland.

In very remote regions of the south Atlantic, like in the southeast Pacific, clouds are generally clean, with average cloud drop number concentration estimated at 67 cm^{-3} (or 87 cm^{-3} when corrected to STP) (Bennartz, 2007), and effective radius may be ~15 – 20 μm for clouds up to 1.5 km in height (Painemal et al., 2014). In this regime, cloud albedo is strongly susceptible to change in aerosol concentration. In DACCIWA, the limited measurements of offshore clouds were split into one day with CDNC around 300 – 700 cm^{-3}, which was ~35% higher than the corresponding inland measurements, and four days with CDNC around 100 – 200 cm^{-3}, 45 – 65% lower than inland. None of the offshore measurements were representative of clean clouds; offshore clouds were universally more polluted, with higher CDNC and lower R_{Eff}, than those in the clean regions of the south Atlantic. The ubiquitous presence aged biomass burning smoke transported from fires in western Central Africa towards SWA provides a source of CCN in greater concentrations than may be found in clean, remote regions.

One of the aims of this paper was to assess the relative impacts of local and transported aerosols on the microphysics of continental clouds over SWA. None of the offshore differences in CDNC, either between the different offshore CDNC on different days, or the different offshore/inland ratios, corresponded with differences in the measured aerosol concentrations. Therefore other factors must have been at play to cause the variability in the offshore cloud measurements, such as different dynamics, thermodynamics, or simply random biases due to our limited set of observations. Our measurements of offshore clouds were sporadic rather than systematic, and it is difficult to assess the statistical significance of the variability within these offshore clouds, or the differences observed between the clouds measured offshore and inland. Figure 2 shows that in our measurement region, the low-level cloud fraction offshore was much lower than inland, and most of the inland cloud formed over the continent, rather than forming offshore before moving inland. The complex dynamic and thermodynamic structure of the marine and continental boundary layers (Wood, 2012), and the diurnal cycle of these differences, mean that the offshore and inland clouds are not directly comparable to assess the impact of local aerosol emissions.

In contrast, the offshore aerosol measurements are much more robust. There were aerosol measurements from a larger number of days, with a much longer sampling time on each day, and aerosol concentrations are much less susceptible to changes in boundary layer thermodynamics compared to clouds. Our measurements showed an average ~40% increase in

regional accumulation mode aerosol inland compared to offshore. The accumulation mode is the most important for CCN, as particles smaller than this are too small to activate. Haslett et al. (2019) also found a 40% enhancement in the inland accumulation mode aerosol concentrations compared to offshore using SMPS data, which is in excellent agreement with our GRIMM measurements of N_{250}. In localised city plumes, highly concentrated emissions had a noticeable increase in CDNC, but on a regional scale the majority of accumulation mode aerosol were due to long-range transport.

Our model simulations allow us to isolate the effects of specific aerosol modes and dynamics. The increase in CDNC inland due to local aerosols is strongly sensitive to the updraft velocity used in the simulation, but for our representative morning and afternoon updrafts local aerosols increased inland CDNC by and 44 and 65%. As these numbers are larger than our measured increase in accumulation mode aerosol, some of the particles in the Aitken mode fits must be having an influence. Regardless, local aerosols constitute less than half of the regional CCN over SWA. Any future increases in the local anthropogenic emissions during the summer monsoon will be on top of the transported background, and this reduces the sensitivity of CDNC to local emissions, which form less of the total.

The polluted background has three main consequences for the microphysics of low-level clouds. Firstly, it means local emissions are a lower fraction of the total aerosol loading and of CCN concentrations at any given supersaturation, limiting the impact of local emissions on CDNC. Secondly, with the baseline CDNC at already-polluted values, it puts the clouds in a regime where CDNC is less sensitive to increases in aerosol concentrations than decreases. Finally, cloud albedo is not overly sensitive to changes in CDNC that do occur when the baseline is so high (Twomey 1991), so this further limits the effect of increasing local aerosol emissions on cloud albedo.

The day-to-day and diurnal variability are extra factors that complicate our analysis, but they do not detract from this overall picture. Similar results were found by Cecchini et al. (2017) in the polluted regions of southern Amazonia, affected by widespread biomass burning, in contrast to the cleaner coastal and forested areas. The modelling results in Fig. 9 show that the increase in CDNC in the afternoons is broadly that expected from the increased updraft speeds. With the limited data available, it is difficult to assess whether or not there is a systematic difference between CDNC or dynamics offshore versus inland. While Fig. 4d shows an increase in CDNC in pollution plumes, the CO distribution inland did not show any large systematic increase in pollution to the offshore mean and standard deviation of 141 ± 7 ppbv, meaning the bulk of the measurements shown in Fig. 4b were made in similar pollution conditions. It may be that our poor statistics of offshore data present a misleading distribution in Fig. 4b, or that there was a systematic and ubiquitous increase in CDNC due to some other factor such as stronger updrafts inland. Most of our offshore measurements were made over the patch of cooler water with lower cloud cover shown in Fig. 3, which may have affected the offshore measurements. Regardless, our evidence is not consistent with a ubiquitous increase in CDNC inland as a result of a large increase in aerosols due to local anthropogenic pollution.

Across the whole of SWA, anthropogenic emissions are dominated by Nigeria, which contains over half the region's population (United Nations, 2017). The Niger delta is also a large source of industrial pollution. In the boundary layer, Nigeria was downwind of our in situ measurements, so we have no measurements of Nigerian pollution. Under such conditions, Flamant et al. (2018a) and Deroubaix et al. (2018) have shown that Lagos did not influence on the air quality to the west (i.e. Benin and Togo). It is possible that local emissions have more of an effect on CDNC in Nigeria, but the baseline CDNC is still

likely to have been high enough that this will have a reduced effect on cloud albedo. Based on the satellite data there was no obvious effect on LLC fraction downwind of Lagos and the Niger delta, suggesting geographical variation in cloud formation and lifetime were still largely controlled by dynamics, orography, and the transport of cool, humid air inland by the maritime inflow.

5   Over the next few decades, the populations of coastal cities such as Accra, Lomé and Cotonou are expected to rise dramatically, as are their respective emissions. While this may mean there is more of an effect from local pollution on the cloud field, our results suggest the dominating factors for cloud radiative properties during the summer West African monsoon are likely to remain the transport of biomass burning aerosol from the southern hemisphere. Indeed, as the population of western Central Africa is also expected to rise at a similar rate to SWA (United Nations, 2017), increasing rates of agricultural burning

10 may cause this background to rise. In this study we have focused on cloud-aerosol interactions leading to the indirect effects, and have not attempted to investigate possible changes to the cloud field by the semi-direct effect or direct effect via changing dynamics, as suggested by Deetz et al. (2018b) and Kniffka et al. (2018). It is vital that future studies examining these effects take into account the transport of biomass burning emissions to SWA from the southern hemisphere.

6   Conclusions

15 This study has assessed factors affecting properties of low-level clouds over southern West Africa during the West African summer monsoon, using an approach combining satellite observations with in situ measurements from the novel DACCIWA dataset. Satellite observations of low-level cloud (LLC) cover suggested regional variation in the cloud field was determined largely by the local orography and transport of cool, humid air inland by the maritime inflow, and no obvious impact of pollution plumes was observed in the large-scale cloud field downwind of major population centres. An assessment of cloud

20 drop number concentration (CDNC) based on aircraft data acquired during the DACCIWA field campaign as well as parcel modelling showed that local emissions had a reducedlimited effect on CDNC on the regional scale, as biomass burning pollution transported from the southern hemisphere dominated regional aerosol concentrations (e.g. Haslett et al., 2019). This transported pollution also caused a high baseline CDNC inland, putting the clouds in a regime where they had a limitedreduced susceptibility to any further increases in aerosol, and also minimising the impact any enhancements in CDNC are likely to have

25 on cloud radiative effects and precipitation.

    We investigated statistics of CDNC on different days, at different times of day, and at different distances downwind of the coast (and the populated regions nearby), to assess the causes of the observed variability. Overall, the large majority of CDNC fell into the range $200 - 840 \ \mathrm{cm^{-3}}$, and relatively little meridional variation was seen inland. The day-to-day variability of CDNC showed some correlation with measured aerosol concentrations, but this was not seen across the whole of the campaign.

30 A diurnal cycle was observed, and CDNC increased by $45 - 60\%$ as clouds began to break up and become more convective in the afternoons compared to measurements made in the early morning. A systematic increase in CDNC was observed in CO plumes near the coastal cities, reaching over $1500 \ \mathrm{cm^{-3}}$ in some cases. However, these plumes represented only a small fraction of the data measured across the region, and are unlikely to be the major factor determining CDNC inland. Offshore, the

limited measurements of CDNC were split into clouds with around $100 - 200$ cm^{-3} and those with around $300 - 700$ cm^{-3}. However, relatively few cloud penetrations were made offshore, so it is difficult to assess whether any differences between offshore and inland clouds are statistically representative. In no region, and at no time of day, were clouds observed that were representative of those found in pristine environments, and our measurements were all affected by the ubiquitous biomass

5   burning pollution transported to the region from offshore.

In order to assess the impacts of local urban emissions and transported background pollution on cloud properties, we performed a sensitivity analysis with a parcel model that was initialised with representative aerosol size distribution and composition, and thermodynamic conditions based on the ambient measurements. We divided up the aerosol into different modes representing either transported pollution or local emissions, and scaled the local emissions to see the effect on CDNC. Dou-

10   bling local emissions increased the calculated CDNC by $17 - 22\%$, whereas it was reduced by $13 - 16\%$ if local emissions were halved, or by $31 - 39\%$ if they were removed altogether. The ~~high~~ aerosol background from transported smoke means local emissions only make up a fraction of total aerosol. Our results suggest that increasing local emissions over the next few decades will therefore have a ~~limited~~reduced impact on CDNC.

Compared to clean conditions, this polluted regime means local emissions have a ~~limited~~reduced effect on aerosol concen-

15   trations, enhanced aerosols have a ~~limited~~reduced effect on CDNC, and enhanced CDNC has a reduced~~limited~~ effect on cloud radiative properties. As the southern hemisphere biomass burning season and West African monsoon coincide every year, and the cloud field during DACCIWA was similar to a previous multi-year assessment by van der Linden et al. (2015), it is reasonable to assume that our observations are typical of clouds over SWA at this time of year. As the population of SWA grows in the coming decades, the transported biomass burning pollution will have a dampening effect on the impacts that growing

20   aerosol emissions have on low-level clouds via cloud-aerosol interactions. Future studies investigating other mechanisms by which aerosols may affect the cloud field, such as by changing atmospheric dynamics and thermodynamics, must take into account the transport of biomass burning smoke from the southern hemisphere to SWA.

Code and data availability. The DACCIWA field measurements are available on the DACCIWA SEDOO database (http://baobab.sedoo.fr/DACCIWA/). SEVIRI optimal cloud analysis data and associated product guides are available to download from EUMETSAT (https://www.

25   eumetsat.int/website/home/Data/Products/Atmosphere/index.html). ACPIM is available to download from The University of Manchester (https://personalpages.manchester.ac.uk/staff/paul.connolly/research/acpim01.html).

## Appendix A: Low-level cloud flag derivation

The SEVIRI optimal cloud analysis product is based on the scheme described by Watts et al. (2011). Cloud top pressure (CTP), cloud optical thickness (COT), and cloud-top effective radius, and their respective uncertainties, are reported every 15 minutes

30   with a spatial resolution of around 3 km in the DACCIWA region. Here we utilise the CTP for the upper and second cloud layers (CTP$_1$ and CTP$_2$), and the COT of the upper layer (COT$_1$).

Our scheme for determining LLC cover is shown in Fig A1. Pixels are defined as LLC if either CTP_1 or CTP_2 are statistically significantly above a threshold pressure (CTP_T). Where either pressure is within the uncertainties of this threshold, the pixel is defined as "borderline" LLC. Additionally, where the top layer of cloud is not at low level, a threshold COT (COT_T) is used to account for the fact that this thick top layer makes it difficult to detect any other cloud layers below. This extra factor may be
5   responsible for the better agreement between our scheme and ground measurements compared to others (e.g. van der Linden et al., 2015). We chose a value of CTP_T of 680 hPa (~3.5 km) because the OCA algorithm can sometimes place a cloud on the wrong side of an inversion. 680 hPa is near a minimum in relative humidity (Kalthoff et al., 2018), at a level above the inversion at boundary layer top but below any mid-level clouds.

     A comparison to LLC fraction derived from ground-based ceilometers in Kumasi and Savé is shown in Fig. A2. The ceilome-
10   ters made ground-up measurements of cloud base, compared to top-down satellite measurements of cloud top, which may cause a small uncertainty in the comparison that is difficult to quantify. Surprisingly, the best agreement was generated by ignoring the per-pixel uncertainty in the SEVIRI data (i.e. assuming all values of CTP were absolutely accurate). This comparison showed agreement in the diurnal cycle and absolute values within 10% LLC fraction, and similar levels of variability (i.e. the standard deviations). The comparison was better during the day than at night, in agreement with a similar observations by
15   van der Linden et al. (2015), though using a different LLC scheme.
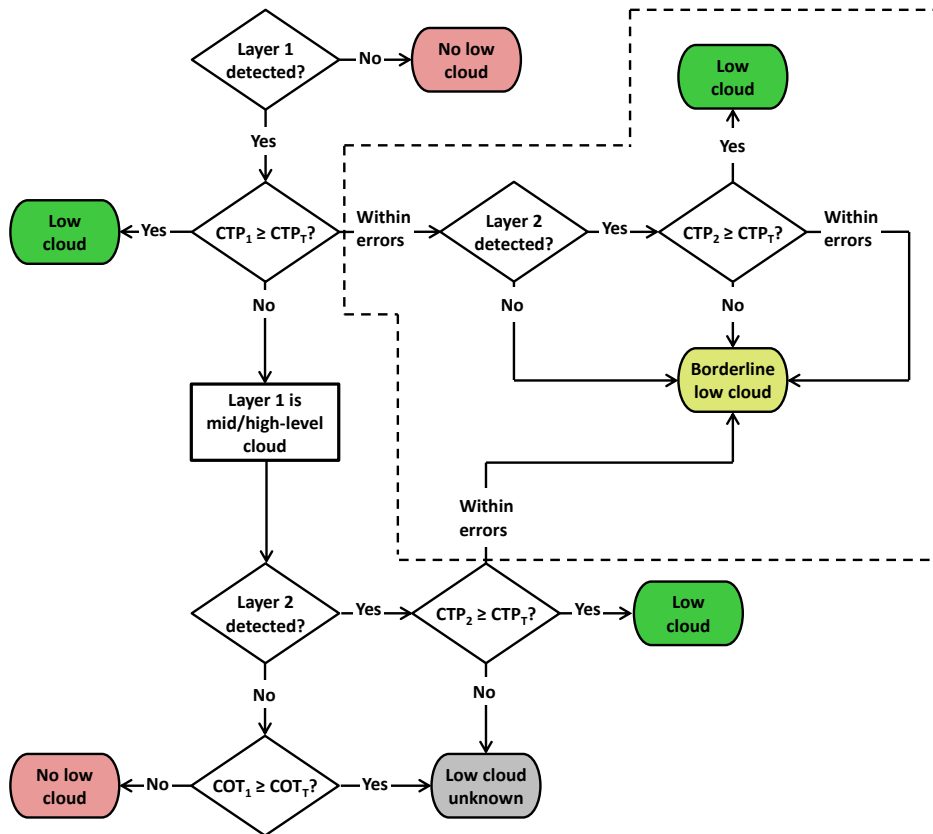
Figure A1. Schematic describing the derivation of the LLC flag from SEVIRI OCA data. We used values of $CTP_T = 680$ hPa and $\log_{10}(COT_T) = 0$. The dashed lines separate off the section of the scheme that is only necessary where the per-pixel uncertainty is considered.
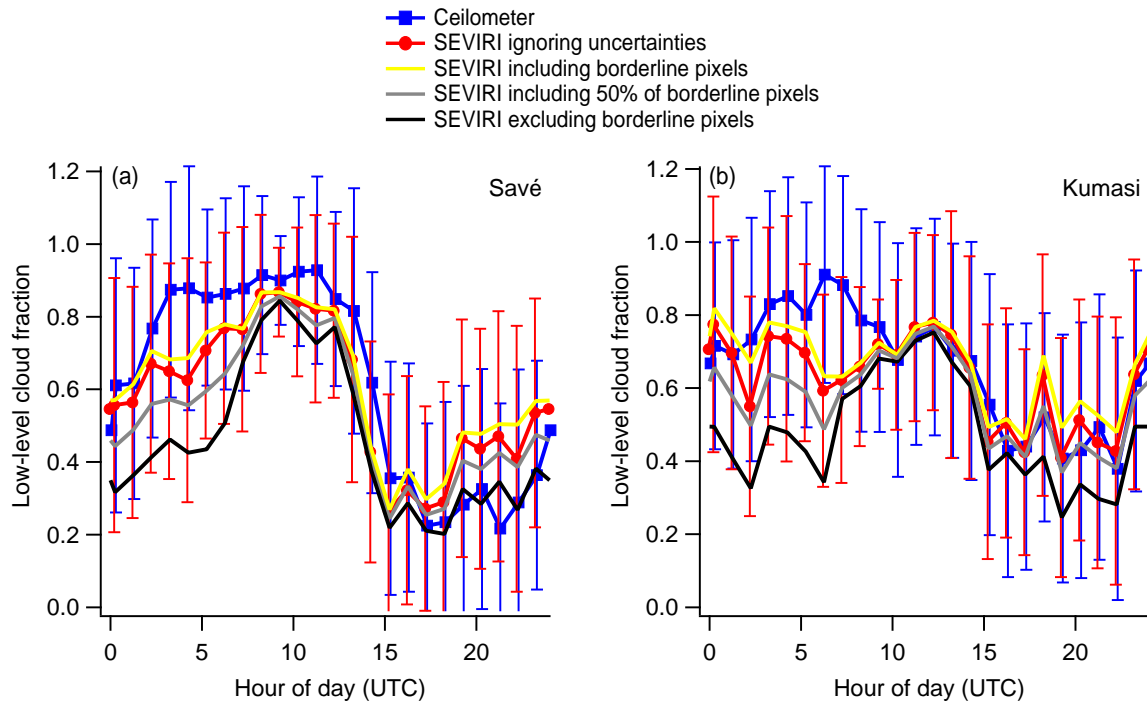
Figure A2. Comparison of LLC fraction derived from satellite and ground-based ceilometer measurements from Savé (a), and Kumasi (b). The satellite measurements were calculated from a 4×4-pixel grid centred on the ground site.

5 *Competing interests.* The authors declare no competing interests.

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Table 1. List of cities in Fig. 3.

Code	City name	Latitude (°)	Longitude (°)
AB	Aba	5.12	7.37
AC	Accra	5.55	-0.20
AJ	Abidjan	5.32	-4.03
BE	Benin	6.33	5.62
CO	Cotonou	6.37	2.43
IB	Ibadan	7.40	3.92
IL	Ilorin	8.50	4.55
JO	Jos	9.93	8.88
KA	Kano	12.00	8.52
KD	Kaduna	10.52	7.44
KU	Kumasi	6.67	-1.62
LA	Lagos	6.46	3.38
LO	Lome	6.13	1.22
OG	Ogbomosho	8.13	4.25
PH	Port Harcourt	4.82	7.03
ZA	Zaria	11.07	7.70

Table 2. Lognormal fit parameters for the fits shown in Fig. 8, using the equation $dN/dlnD_{\rm p} = \frac{N}{\sqrt{2\pi}ln(\sigma)}exp\left(-\frac{(ln(D_{\rm p})-ln(D_{\rm med}))^2}{2ln(\sigma)^2}\right)$.

	$N(\text{cm}^{-3})$	σ	D_{med} (nm)
Offshore wide Aitken mode	282	1.65	75.5
Offshore accumulation mode	570	1.50	199.1
Local Aitken mode	2829	1.66	56.5
Local accumulation mode	175	1.42	242.5
Sea salt coarse mode	1.18	1.52	1236
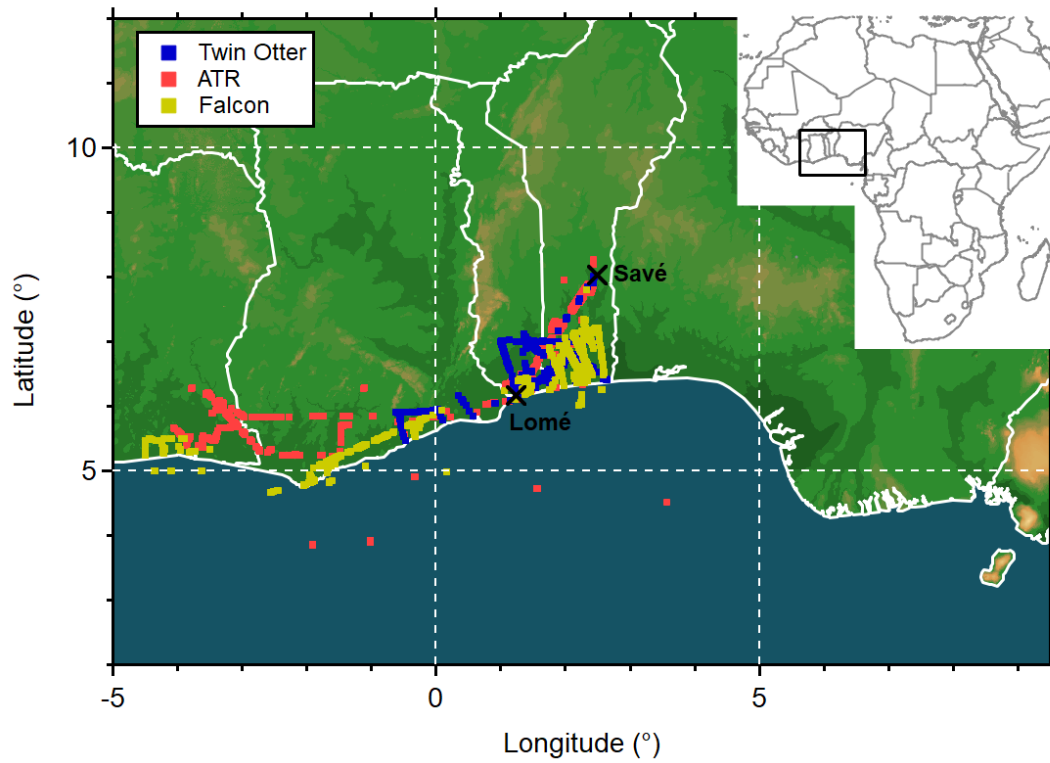
Figure 1. Map showing the parts of the flight trackers where airborne cloud measurements were made below 1 km above mean sea level. The locations of Lomé and Savé are marked as many research flights took place between these two locations.
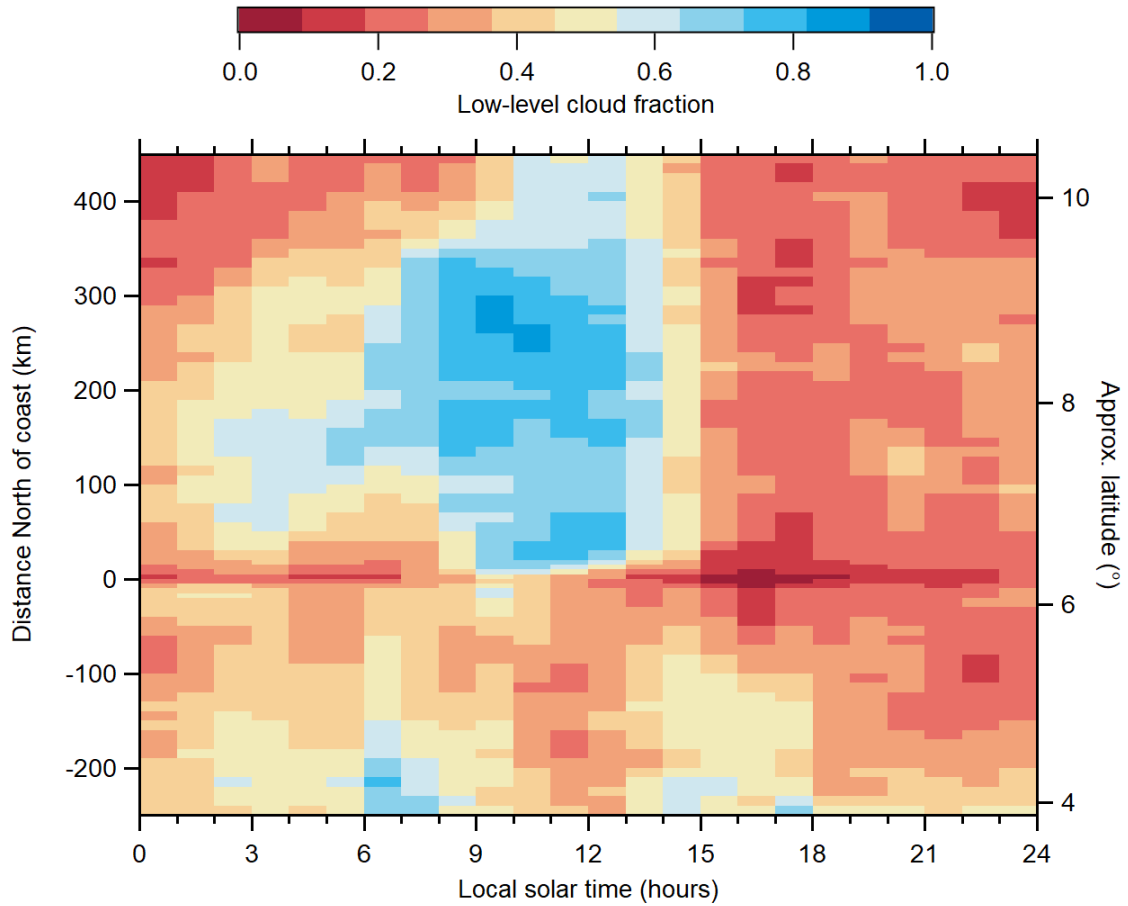
Figure 2. The diurnal cycle of mean low-level cloud fraction at different distances inland or offshore over Togo & Benin (4 – 11°N, 0.6 – 2.75°E), taken from the LLC flag derived from SEVIRI cloud data. The data are shown from the time period coinciding with the DACCIWA aircraft campaign: 29th June – 16th July 2016. The latitude scale on the right axis is approximate, as the latitude of the coastline varies by 0.6° over Togo & Benin.
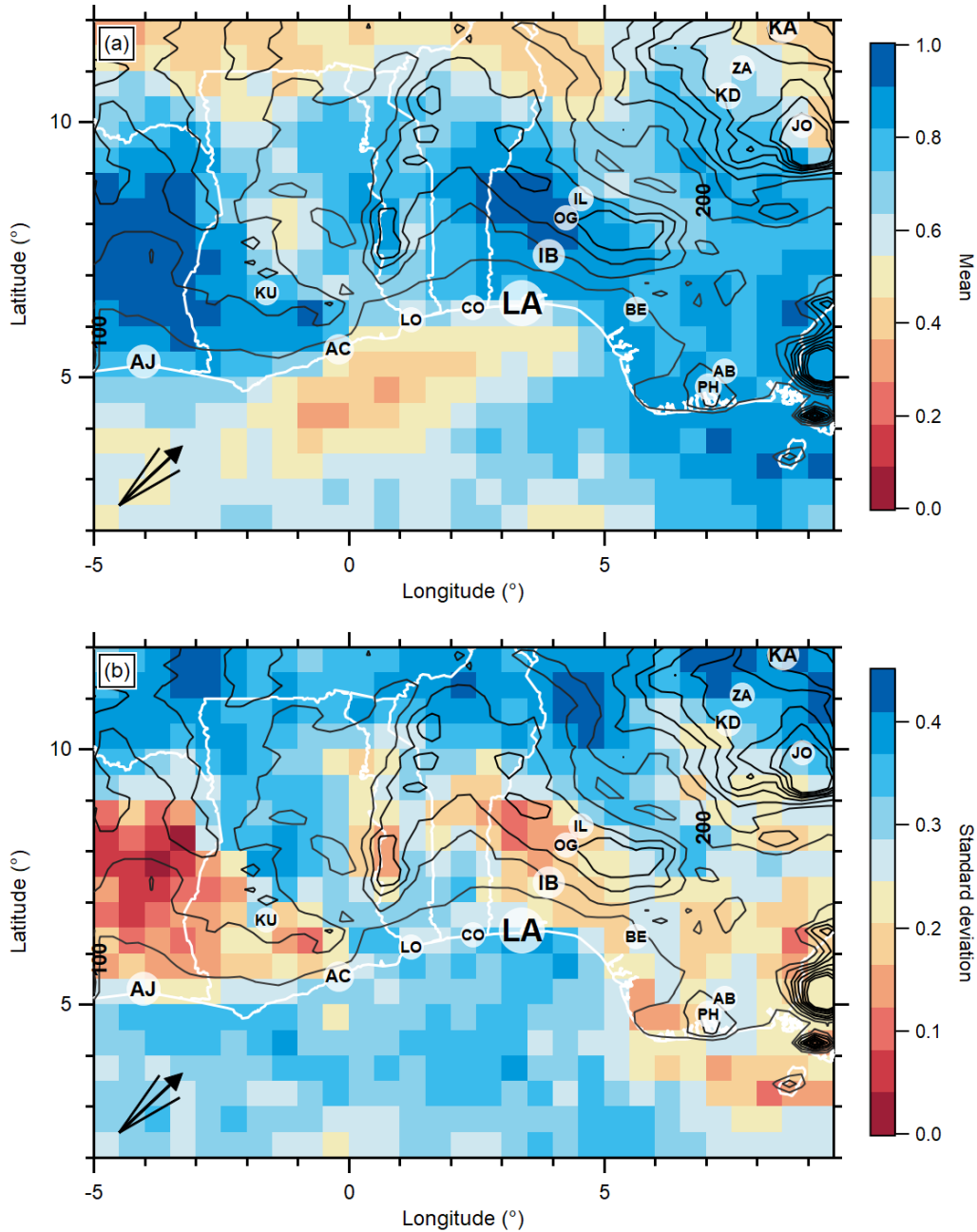
Figure 3. Mean and day-to-day standard deviation of low-level cloud fraction between 10:00 – 11:00 UTC during the DACCIWA aircraft campaign, 29th June – 16th July 2016, taken from the LLC flag derived from SEVIRI cloud data. The contours show the elevation of the land surface, and are labelled in metres above mean sea level. Text markers show the locations and abbreviated names of large cities in the region, which are listed in Table 1. The text size gives a qualitative indication of the relative population of each city. The arrow markers in the bottom left show the median and quartiles of wind direction, measured inland below 1km between 10:00 – 11:00 UTC.
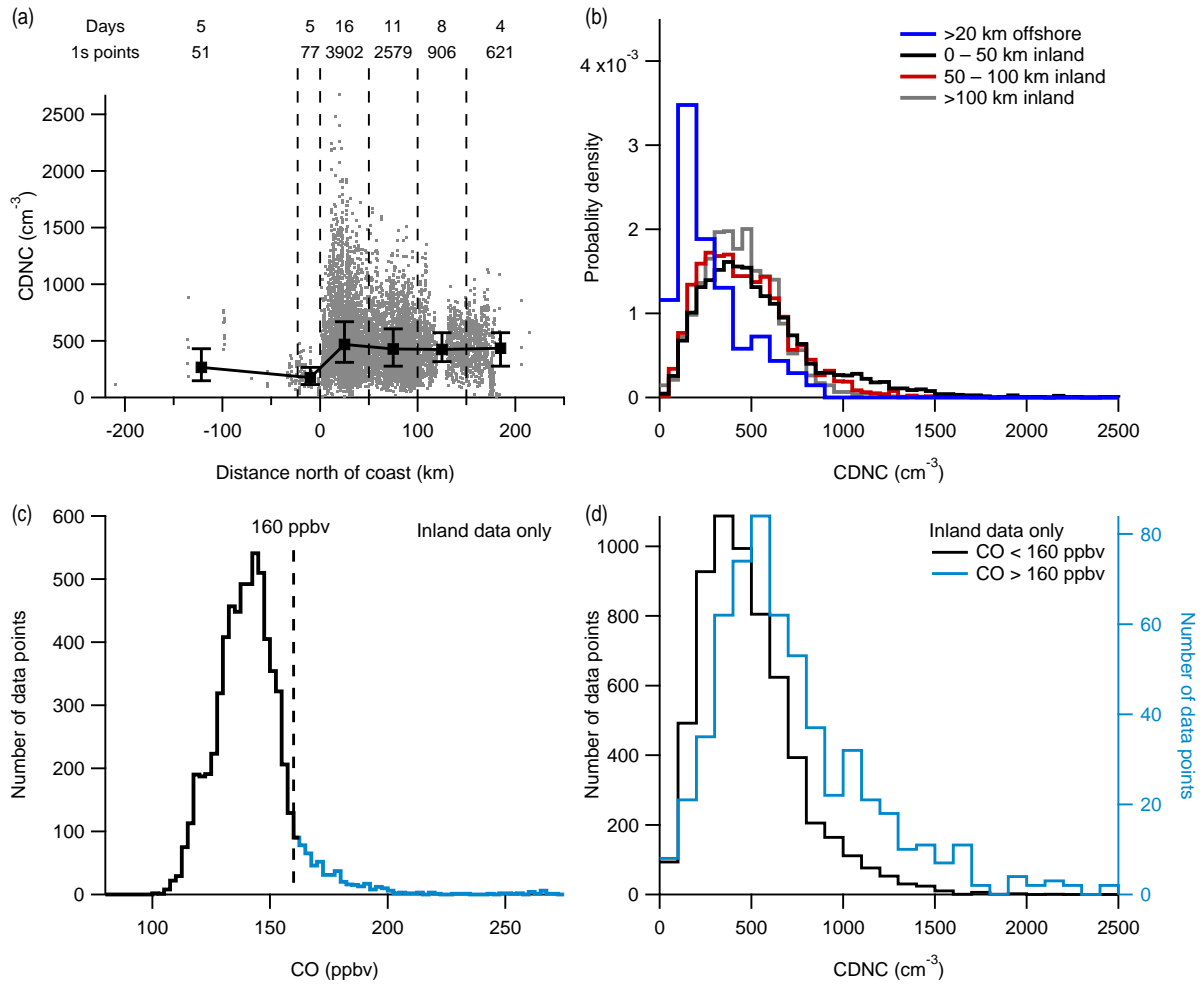
Figure 4. Measured CDNC for all data below 1 km, stratified by distance inland and pollution conditions. Panel (a) shows all data as a function of distance from the coast. The markers and error bars show the median, 25th, and 75th percentiles. The numbers at the top show the number of different days' worth of data in each bin, as well as the number of individual data points. Panel (b) shows normalised histograms of CDNC at different distances from the coast. Panel (c) shows a histogram of in-cloud pollution, and panel (d) shows normalised histograms of inland CDNC stratified by CO concentrations.
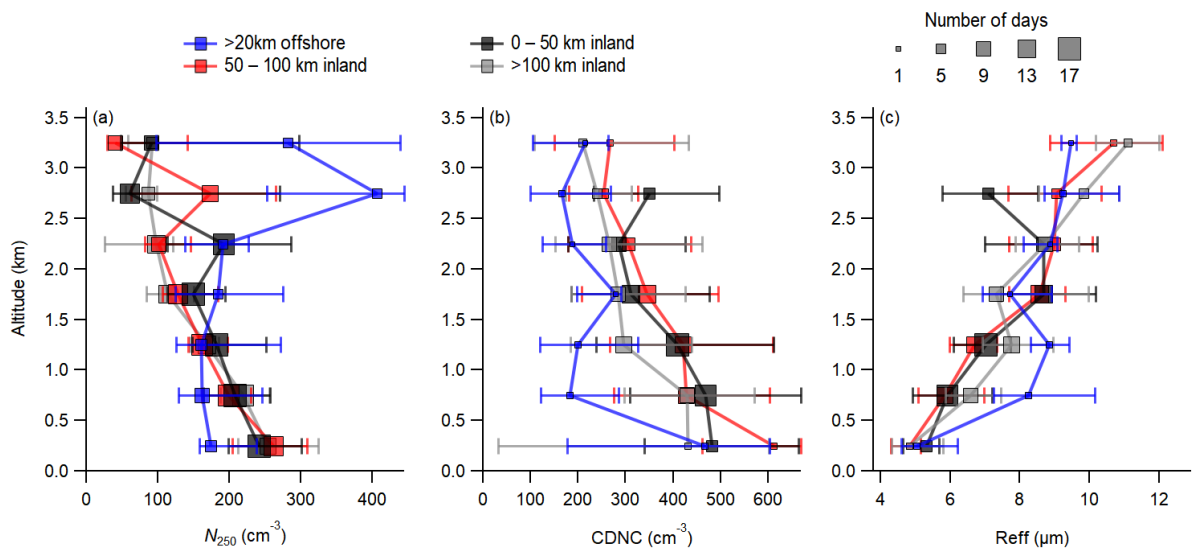
Figure 5. Average vertical profiles of aerosols larger than 250 nm (a), cloud drop number concentration (b), and effective radius. The markers are the medians, and are different sizes depending on the number of individual days the data are taken from. The error bars show the 25th and 75th percentiles, and the solid lines connect the medians. The cloud profiles show the average properties of the cloud field, which often contained multiple discreet layers.
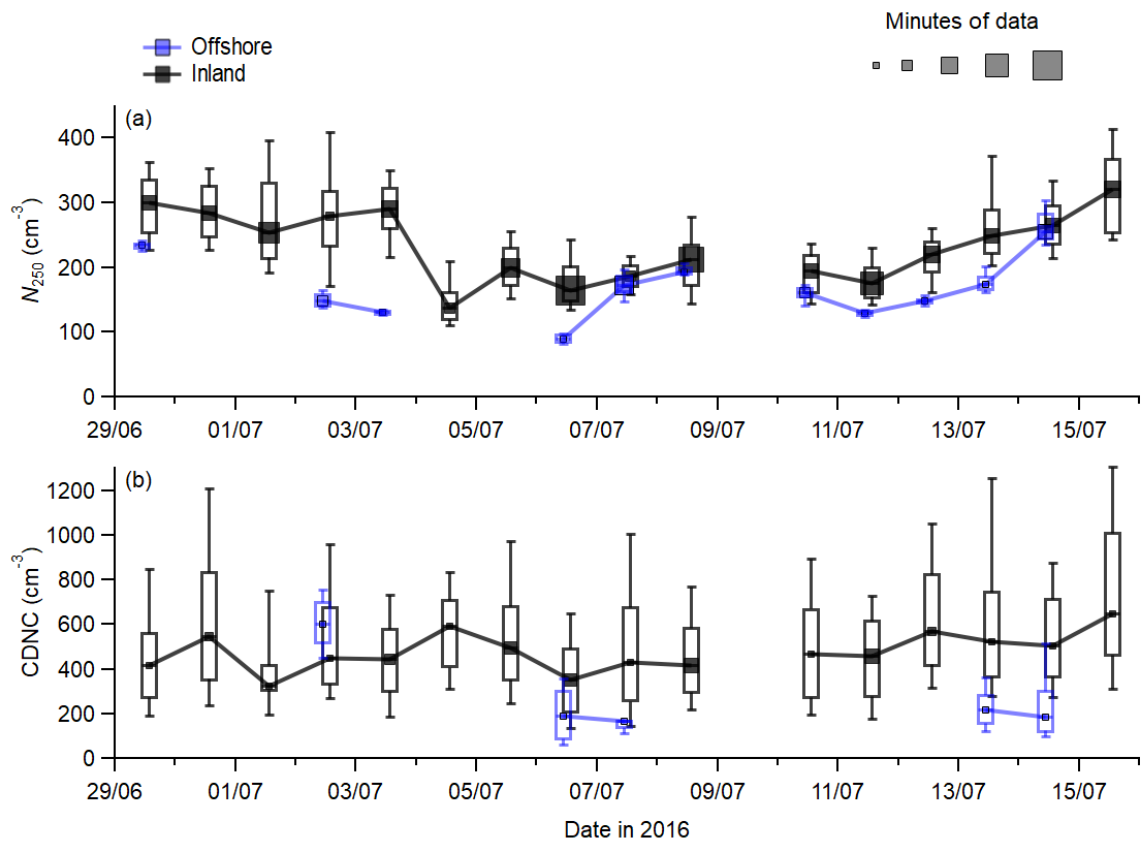
Figure 6. Day-to-day variation in N_{250} (a) and CDNC (b). The data shown are from all data measured below 1km. The markers, boxes, and bars show the medians, 25th and 75th percentiles, and the 10th and 90th percentiles respectively. Offshore data were collected over 20 km south of the coast.
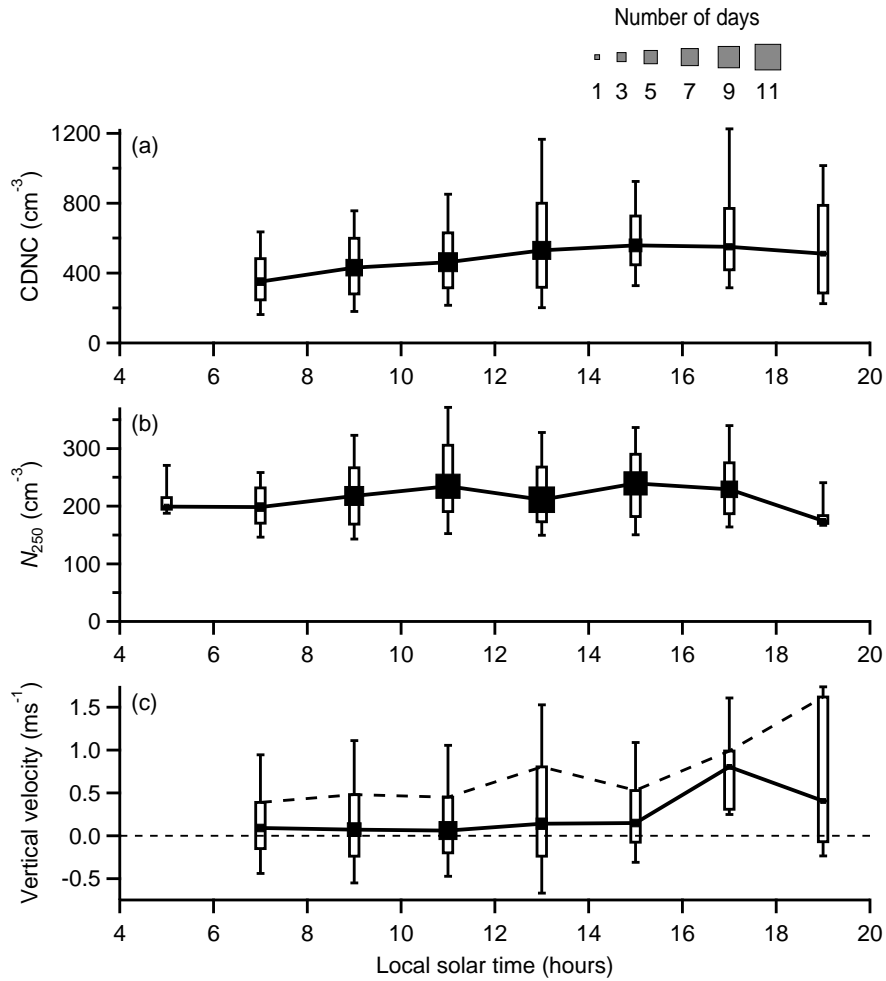
Figure 7. Inland diurnal variation in CDNC (a), N_{250} (b), and in-cloud vertical velocity (c). The data shown are from all data measured below 1km. The markers are the medians, and are different sizes depending on how many individual days the data are taken from. The boxes are the 25th and 75th percentiles, and the bars are the 10th and 90th percentiles. The solid lines connect the medians, and the dashed line in part (c) connects the 75th percentiles.
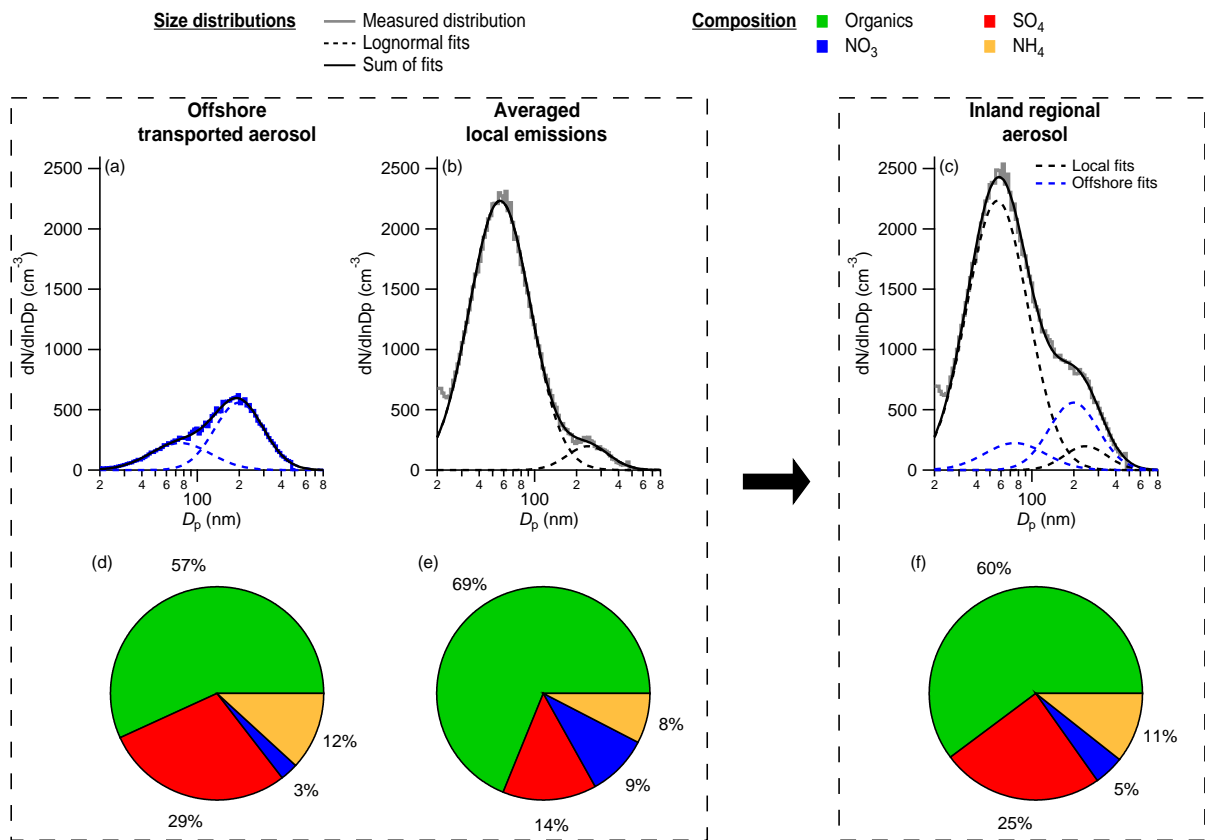
Figure 8. Aerosol size distributions (a – c) and accumulation mode composition measurements (d – f) made over SWA, that were used as input into ACPIM. The averaged local emissions were calculated by subtracting the offshore transported aerosol from the inland regional aerosol.
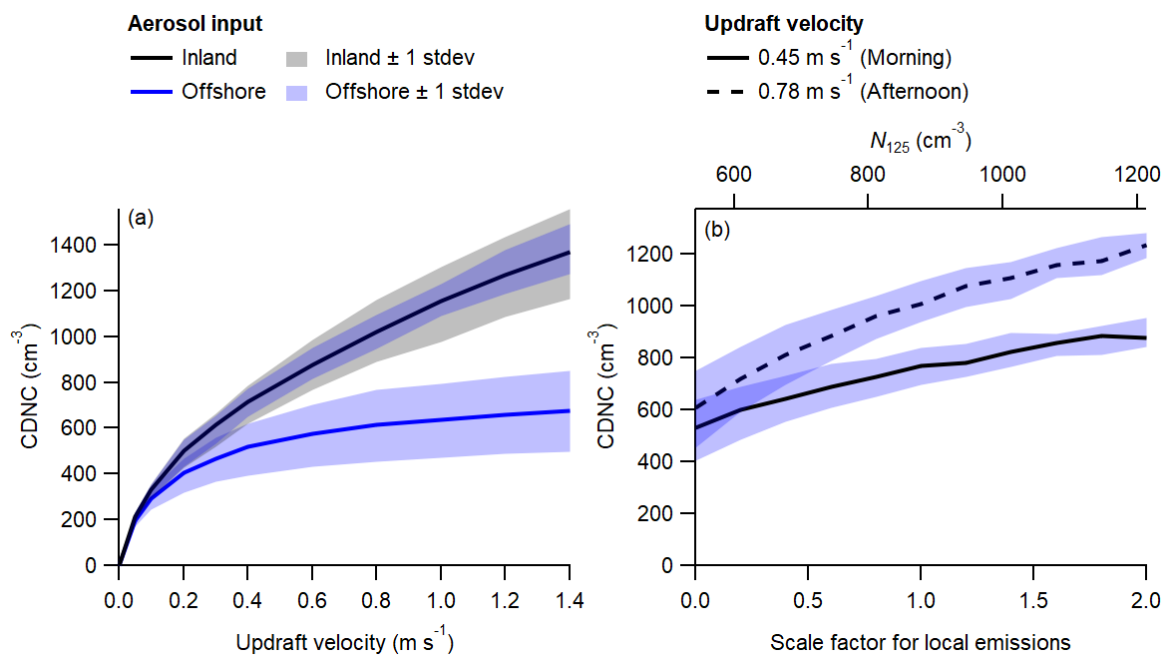
Figure 9. CDNC calculated during ACPIM parcel model runs initialised using measured aerosol size distributions and compositions shown in Fig. 8 and Table 2. The shaded regions show the expected day-to-day variability in CDNC as a result of variation in the aerosol concentrations, based on the standard deviation of measurements shown in Fig. 6a. Panel (a) shows the sensitivity of CDNC to updraft velocities, and panel (b) the sensitivity to different scale factors for local emissions in the aerosol input. The smaller and larger updraft velocities in panel (b) are representative of morning and afternoon.