Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-420-AC1, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol liquid water content in the moist southern West African monsoon layer and its radiative impact" *by* Konrad Deetz et al.

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Received and published: 4 August 2018

Answer to Referee #1 Konrad Deetz 25 July 2018

Dear Referee (Atmospheric Chemistry and Physics),

thank you for your report from 3 July 2018. We have accounted for the comments and suggestions in the revised manuscript version. Please find our replies (marked with #) to the individual comments in the following.

Sincerely, Konrad Deetz on behalf of all coauthors

Referee comments: (0) Hygroscopic growth could alter the optical properties of

aerosol. This manuscript reported the follow up simulation study based on Deetz et al. (2018) setup within the COSMO-ART modeling framework for a summer monsoon event in Southern West Africa and estimated the aerosol liquid water content (ALWC) and its impact on radiative transfer. The process was separated into three characteristic phases during commonly Atlantic Inflow event over this region to detailize the ALWC-radiation interactions. It was shown that the accumulation mode particles are the dominant contributor to aerosol liquid water and aerosol growth led to the increase of aerosol optical depth from 0.2 to 0.7. The increased aerosol optical depth can lead to around 20 W/m2 decrease in shortwave radiation. Bootstrapping technique was used to derive the linear relationship between ALWC and radiation and found a stronger correlation for in-cloud conditions. This modeling study highlight the importance of including the relationship of RH dependency of aerosol optical depth in atmospheric model, which can significantly impact the local radiation balance, especially over moist tropical environment. The whole manuscript is well structured and the modeling discussion is adequate. I recommend publishing this work as a valuable component of the DACCIWA special issue in ACP after the authors address the following comments.

(1) Page 1, Line 23: ALWC = aerosol liquid water content?

We have changed the manuscript accordingly.

(2) Page 4, Line 26-27: the "coarse modes of marine origin" should be (7-9) and the following "coarse modes of mineral origin" should be (10-12)?

We have changed the manuscript accordingly.

(3) Page 5, Line 5: ISORROPIA II does not include fresh soot for calculation. Did the model assume aged soot is internally mixed with sulfate in the calculation of optical properties and radiative transfer?

Yes, fresh soot is not included in ISORROPIA II. In COSMO-ART it is therefore handeled separately (as denoted on p. 4 l. 3). And yes, as soon as the soot is treated as

aged, it is an internal mixture within ISORROPIA II and with respect to the calculation of optical properties and radiative transfer.

(4) Page 5, Line 30-32. It is better to mark down the approximate area of "Ivory Coast" (7.5 W – 3W, 4N-10N, should be a subset of 2.5km modeling domain) in Figure 1(b) since nearly all the Figures follow on (e.g. Figure 2, : : :) are focus on this area.

We agree on that and have changed the manuscript (figure and figure caption) accordingly.

(5) Page 6, Line 21: Where is the geographic location of radiosounding site at "Lamto", please provide the locations in Figure 1. Also, look at the Figure B1-B3 in the Appendix, why there is no sounding comparison for location at "Lamto" for July 2-3. The radiosounding for RH vertical profiles at the two sites are not synchronized and with different time interval? Also, the Figure B2, may be due to the compress the the aspect ratio, the grey shading regions at certain place are not consistent with the description of uniformly 4% uncertainty assigned for radiosondes.

We added Lamto as a magenta dot in Figure 1b. For Lamto, no sounding data is available for 2-3 July. Indeed, the soundings of Lamto and Abidjan are not launched at the same times and with different time intervals. We double-checked the shaded area enveloping the uncertainty of +-4 % relative humidity. This is correct. The shading just appears inhomogeneous when the black line is rather horizontal.

(6) Page 7, line 22-23: ALWC was influenced by aerosol types and RH. Are the aerosol type and RH all the same in North China plain and southern West African, so they are comparable? The authors refer this study with China campaigns (e.g. HaChi, PRIDE-PRD) heavily in the introduction section and the following discussion, maybe in some place in the introduction section, the author need to point out the similarity of this DACCIWA campaign with China campaigns such as aerosol loading, RH conditions, atmospheric oxidation capacity, cloud coverage.

This is an interesting question. We try to elaborate this by relating to the study of Bian et al. (2014) as you have proposed. When focusing on the study of Bian et al. (2014), the observations are related to the time period July-August 2009 and focusing on the chinese provinces Shandong, Hebei, Pekin and Tianjin. The climate in this area is in between humid subtropical and humid continental Summers are hot and rainy with temperatures around 24-28 °C in July with the precipitation maximum in summer via influences from the monsoon. A qualitative analysis of Terra Modis satellite images (of course only one overfly per day) revealed that in the 62 d period of July-August 2009 Shandon was fully covered by clouds on 55 d and partly covered by clouds on 7 days. Therefore the weather conditions during the DACCIWA campaign and HaChi campaign are very similar. Both studies focus on the NH summer. Both areas are located in the NH summer monsoon area with high temperatures and are very frequent covered by clouds. The measurement site for the study of Bian et al. (2014) is Wuqing. For this location, Liu et al. (2011) [Figure 3] shows measurements of temperature and relative humidity for July-August 2009. Temperature variations are between 20 °C and 32 °C. Relative humidity variations are between 40 % (mostly 60%) and 95 %. The latter is similar to what is modeled for southern West Africa (Fig. 3 in our manuscript) and to what was observed in southern West Africa at Save supersite (Kalthoff et al., 2018, Fig. 3).

Wuqing is about 90 km away from the Gulf of Bohai. So also HaChi focuses on the area near the coast. Wuqing is surrounded by large cities (Peking (80 km away, 21.5 million inhabitants, megacity), Langfang (30 km away, 4.4 million inhabitants), Tianjin (40 km away, 15.5 million inhabitants, megacity), Tangshan (100 km away, 7.6 million inhabitants)). Also southern West Africa has several large cities especially near the coast. However, the populations are generally smaller but on the same order of magnitude (Lagos: 13.7 million inhabitants, Abidjan: 5 million inhabitants). Based on MODIS observations, Bian et al. (2014) show that the averaged AOD values are generally above 0.6 in the research area and 0.7 above Wuqing. For the DACCIWA region we found averaged MODIS AOD values of 0.4-0.7, slightly smaller to what was observed in the HaChi region. However, the validity over land is limited because southern West Africa is virtually always covered by clouds, restricting the observations to a few days.

Based on these findings we came to the conclusion that the general meteorological and aerosol conditions are similar for HaChi and DACCIWA and therefore allow a qualitative comparison e.g. of the ALWC values between both sites.

We added the following passage in the conclusions to account for your remark: "HaChi and DACCIWA both focus on the northern hemispheric monsoon season, capture coastal areas that are frequently covered by clouds, have similar temperature and relative humidity conditions (Liu et al., 2011; Kalthoff et al., 2018) as well as similar aerosol loadings (Bian et al. (2014); Deetz et al. (2018a), allowing for a qualitative comparison of modeled ALWC with measurements during HaChi."

(7) Page 8, first paragraph: any explanation why OC dominate the aerosol mass composition? was it a biomass burning event? Also, for Figure D1, is the July 6-7 aerosol component vertical profiles similar to the July 2-3 shown here?

The aerosol mass composition is subject to current research in the DACCIWA research community. Therefore the main outcomes with respect to this question are not yet available/published. However, also the DACCIWA observations (e.g. aircraft measurements) show this dominance of organic carbon (e.g. Flamant et al., 2018). Biomass burning is an important source of OC and likely is responsible for the dominance of OC over Ivory Coast. Based on the experience we obtained with COSMO-ART during our two month (June-July 2016) of forecasting the atmospheric composition (with coarser grid mesh size), we observed that the biomass burning plumes over the Gulf of Guinea (coming from central Africa) frequently swash into the DACCIWA domain. To account for your remark, we repeated the composition analysis for 6-7 July 2016. The corresponding plot (Review-figure-1) is given as appendix to our review answer. For the non-OA, the situation is comparable with 2-3 July but OA is about twice as high compared to 2-3 July with a more distinct vertical gradient, indicating a stronger

influence of biomass burning.

(8) Page 8, Line 15. In contrast, AIT particles are lacking in size and COARSE particles are lacking in number.

We have changed the manuscript accordingly.

(9) Page 8, Line 30-31. Can you also provide the boxplots for median aerosol number concentrations for Aitken, accumulation and coarse mode in Figure E1?

We have changed Figure E1 and the manuscript accordingly. Now the panels (a) and (c) show the median aerosol number concentrations for Aitken, accumulation and coarse mode in addition to the aerosol diameters (b,d). The revised figure is added as appendix (Review-figure-4).

(10) Page 9, Line 8: the total water column is the full integration of model layer (e.g. 30km in Table S1) or below 1500m AGL that this study focused?

Yes, in this case the full integration of model layer is considered and not just the lowest 1500 m. This is done on purpose because the total cloud water column is a widely used measure for the quantification of clouds and with this figure we want to provide some guide values to allow for comparison between the water contribution from clouds and the water contribution from aerosol. Furthermore, Figure 7 is the basis for Figure 8 and in Figure 8 we also analyze the contribution of the in-cloud AOD to the total AOD. Since the total AOD is related to the total vertical column, it is necessary to focus on the total vertical column in the model to ensure consistency.

(11) Page 9, Line 13-15: where is the location of the model realized NLLS and convective clouds in the fouced Ivory Coast region? In Figure 3 and Figure 4, the authors showed the double peak of ALWC during phase 2 period, one near coast and the another one in hilly terrain to the north. Are the peaks for ALWC at different locations also strictly correlated with the model simulated clouds?1

We attached Review-figure-2 to emphasize the location of clouds over Ivory Coast

and the total DACCIWA domain in general. The figure shows an overview of the low-level cloud temporal evolution between 2 July 21 UTC and 3 July 10 UTC (a-f). Blue shading denotes low-level clouds via the existence of cloud water in the lowest 1.3 km AGL. Brown shading indicates the topography above 250 m ASL. The arrows show the wind speed (m s-1, scale is given below) and direction at 250 m AGL. For 21 UTC and 23 UTC the Atlantic Inflow front is shown in red. From a-c a clear separation between the cloud band directly behind the Atlantic Inflow front and at the coast is visible. This figure is published in Deetz (2018a). The ALWC is primarilly correlated with the relative humidity, therefore cloudy areas (with a presence of sufficient amounts of aerosol, which is fulfilled over the entire DACCIWA domain) are areas with the highest amounts of ALWC. Review-figure-3 shows the ALWC at 500 m AGL (where we can find the NLLS) over land (for the entire DACCIWA domain) on 3 July 6 UTC. (a) Total ALWC (mg m-3, shading) and RH of 95% (black contour) and (b) pie chart of the ALWC contribution from the single aerosol modes (%) to the total ALWC in (a). For the entire DACCIWA domain highest ALWC values can be found in areas with highest relative humidities (location of the NLLS). This figure is also published in Deetz (2018a).

(12) Page 10: Line 9-10. “... sharpen condition substantially decrease selected area”, can you provide the percentage instead of the subjective description on simulated clouds grids versus non-cloud grids in the Ivory Coast area? From page 9, line 18-19, I may know only 3%-9% of total grids realized the clouds in July 2-3. So between the two sensitivity runs, the “ALWC” and “no-ALWC” case, how many percentage were excluded from further radiation analysis due to the model simulated the displacement of clouds?

We calculated the ratio a/b with (a) the number of gridboxes which are related to clouds in both realizations by restricting to gridboxes with a total cloud water difference below 0.1 g m-2 (masking cloud displacement) (b) and the number of gridboxes which are related to clouds in both realizations without any restrictions (by ignoring cloud displacement). This ratio is between 0.04 and 0.18 in the 25 hour period with a median

of 0.076. So on average only 7.6 % of the cloud grid points (clouds in both realizations) can be used for the radiation analysis. We adapted the corresponding sentence in the manuscript as follows: "Consider that the sharpened condition substantially decreases the selected area (on average only 7.6 % of the cloudy area can be considered) and therefore makes the results less representative for the cloudy area."

(13) Page 10, Line 28-29: where is the fixed SST value from COSMO-ART coming from?

The fixed SST is coming from the driving model ICON. For ICON, the SST fields are derived daily at 0 UTC based on observations. A detailed description of the handling of the SST in COSMO can be found in the "COSMO Documentation Part III - Data assimilation" (http://www.cosmo-model.org/content/model/documentation/core/cosmoAssim.pdf) at page 89f.

(14) Page 11, Line 24. The AOD is higher -> the difference of AOD is higher

We have changed the manuscript accordingly.

(15) Page 11, Line 33-34. In what percentage are the outliers for ALWC-radiation linear fitting (e.g. “less data, large spread, extra low ALWC ...”)?

The following tables summarize the percentages of ALWC data that are not included in the linear fitting (red curves in Fig. 15 (2-3 July) and Fig. H1 (6-7 July)).

2-3 July 2016 (Fig. 15): see Table presented in Review-figure-5

6-7 July 2016 (Fig. H1): see Table presented in Review-figure-6

This analysis shows very similar results when comparing 2-3 July and 6-7 July underlining some robustness in these characteristics. The upper outliers are generally noncritical and negligible (never greater than 0.5%). Lower outliers are only relevant when focusing on longwave radiation and in-cloud areas because there a nonlinear behavior is obvious for which we have no explanation. In this case about one-fifth of the data is not considered. We added/adapted the following passage in the manuscript to meet your concerns: "The fitting omits bins with large ALWC (less data and large spread). A detailed analysis revealed that not more than 0.5 % of the data are omitted. Figure 15c and Figure 15e show a nonlinear behavior for low ALWC. Therefore also these parts are omitted in the linear fitting. This affects 3.5-23.3 % of the data."

(16) Page 12, Line 3-7. What the total size n for the linear fitting based on the grouping of ALWC versus radiation difference with the increment of 0.01 g m-2. If there existed similar linear reasonaliship either derived from observation or model from other regions, it is worthing mentioning here and discussing the possible reason for the difference considering during the DACCIWA campaign the aerosol components are dominated by OC (Figure D1) and the water uptake are most significant for coarse mode (Figure 6).

To the first part of your question: In the style of our tables of remark (15) we show again the two tables that now include the total number n of gridboxes that are used for the linear fitting. n_max is the maximum number for day and night spanned by the dimensions lon x lat x hours.

2-3 July 2016 (Fig. 15): see Table presented in Review-figure-7

6-7 July 2016 (Fig. H1): see Table presented in Review-figure-8

To the second part of your question: As far as we know, our study is the first approach assessing the linear relationship between ALWC and the radiation difference. We don't have opportunities for a comparison with observations or model results from other regions. (A prerequisite for a comparison of our results with model results from other regions is the availability of a model run that excludes the ALWC effect in the radiative transfer calculations that can be compared with a reference run. From our knowledge, this is not available from other research groups.) Zieger et al. (2017) made an approach with a global model to underline how the hygroscopicity of sea salt affects the AOD (and with that the radiative transfer which is not shown in that work). However, we

have serious doubts that a global model is able to appropriately consider the aerosol growth due to water uptake and their impacts on radiation. Nevertheless we added a reference of this work in our introductory section as follows: "Ziegler et al. (2017) assess the effect of hygroscopicity of sea salt on AOD with a global model approach. They modeled latitudinal averaged reductions in the AOD of up to 14 % when reducing the hygroscopicity of sea salt from 1.5 to 1.1." It is not unusual that OC is the dominating aerosol component, especially when regions are affected by locally emitted or long-range transported biomass burning plumes. E.g. Brito et al. (2014) characterize the ground-based aerosol during the South American Biomass Burning Analysis (SAMBBA) field experiment and found that OC is the dominating aerosol in the submicron size range. With respect to the significant water uptake of the coarse mode it has to be considered that in our radiation analysis the coarse mode only consists of sea salt. Generally, the coarse mode in COSMO-ART consists of sea salt, mineral dust and coarse mode anthropogenic particles. But the latter two are not related to ALWC in COSMO-ART. It is not a new finding that sea salt is extremely hygroscopic. Sea salt aerosol particles take up significant amounts of water at RH < 75%, due to the presence of the highly hygroscopic salts of Ca2+ and Mg2+ (Ziegler et al., 2017). Therefore we have expected most significant water uptake with respect to the coarse mode.

(17) Page 14. Line 1-5. The authors mentioned before the RH underestimation may suggest the model derived ALWC value from this case study is the lower bound (Page 6, line 28- 29), how it compared with the double counting of aerosol activate in the model, which tends to overestimate the AWLC, and the uncertainty for the corresponding radiation change calculation?

The comparison of the modeled RH with soundings at Abidjan and Lamto (Figure B1-B3) indicate that COSMO-ART tends to underestimate the RH, although there is no systematic bias. This is a source of uncertainty for the calculation of ALWC and the radiative transfer. However, it has to be considered that the increase in water uptake

is most sensitive to RH in the narrow range of RH >95 % and less sensitive for RH below 95 %. Therefore, potential deviations should not be overrated. The conception in COSMO-ART, not to remove the activated aerosol from the aerosol population, is done by reason. Model tests in the past that remove the aerosol after activation leads to a very fast (unrealistic) cleaning of the atmosphere. But conception of not removing the activated aerosol from the aerosol population does not lead to an overestimation of the ALWC. Instead it is the consideration of two different aspects: (a) Aerosol that take up water, (b) A cloud droplet or ice crystal that has an aerosol particle (CCN/IN) inside. The activated aerosol particle is a cloud droplet (or ice crystal) and the radiative interaction is only related to its quality being a cloud droplet (the negligible small aerosol particle and its ALWC is not considered when we talk about the interaction between cloud droplet and radiation). On the other hand we have the aerosol in the aerosol population that can take up water when it is hygroscopic. In this case there is an interaction between the aerosol particle (combination of aerosol and ALWC) and the radiation. Therefore we expect that we do not per se overestimate the ALWC with our model concept. But of course, we see uncertainties in the corresponding radiative transfer calculations. With our existing model system and the model realizations we have conducted for this study it is not possible to quantify these uncertainties or to set them in relation to the uncertainty that comes from deviations in the RH.

(18) Page 18, Figure 4. The caption. “Same ass for Fig. 3”??

We have changed the manuscript accordingly.

(19) Page 34, in the row of “vertical levels”, sometime in the main content the notation is “AGL” but here it is “ASL”. make it consistent.

We have changed the manuscript accordingly.

Additional References: Flamant et al. (2018): THE DYNAMICS–AEROSOL–CHEMISTRY–CLOUD INTERACTIONS IN WEST AFRICA FIELD CAMPAIGN Overview and Research Highlights, BAMS, pp. 83-104,

https://journals.ametsoc.org/doi/pdf/10.1175/BAMS-D-16-0256.1

Zieger et al. (2017): Revising the hygroscopicity of inorganic sea salt particles, Nature communications, Vol. 8, Article number: 15883, https://www.nature.com/articles/ncomms15883

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-420, 2018.

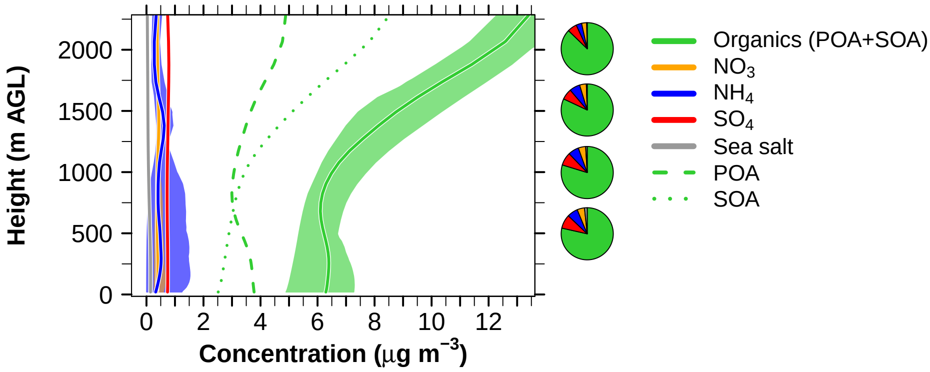

Fig. 1. Vertical profiles (m AGL) of aerosol concentrations (ug m-3) for the median over Ivory Coast (7.5° W–3° W, 4–10° N) with respect to the time period 6 July 15 UTC and 7 July 15 UTC.

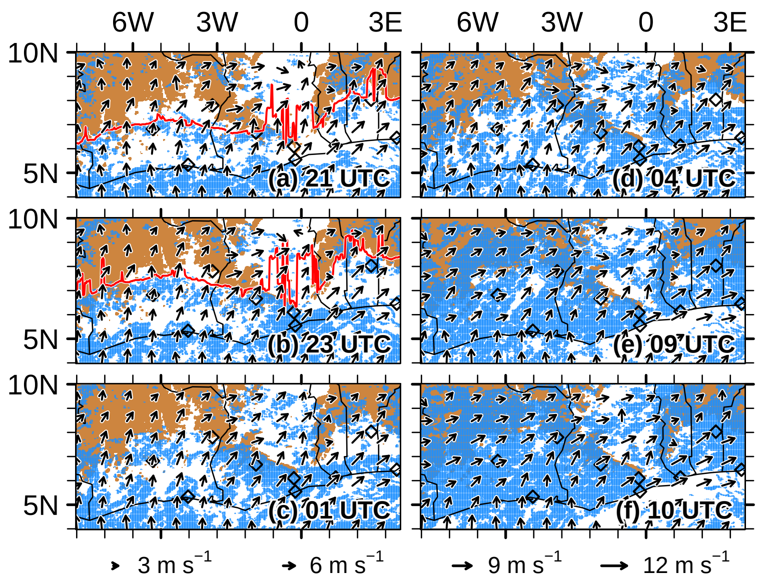

Fig. 2. Overview of the low-level cloud temporal evolution between 2 July 21 UTC and 3 July 10 UTC (a-f). Blue shading denotes low-level clouds via nonzero cloudwater below 1.3 km AGL.

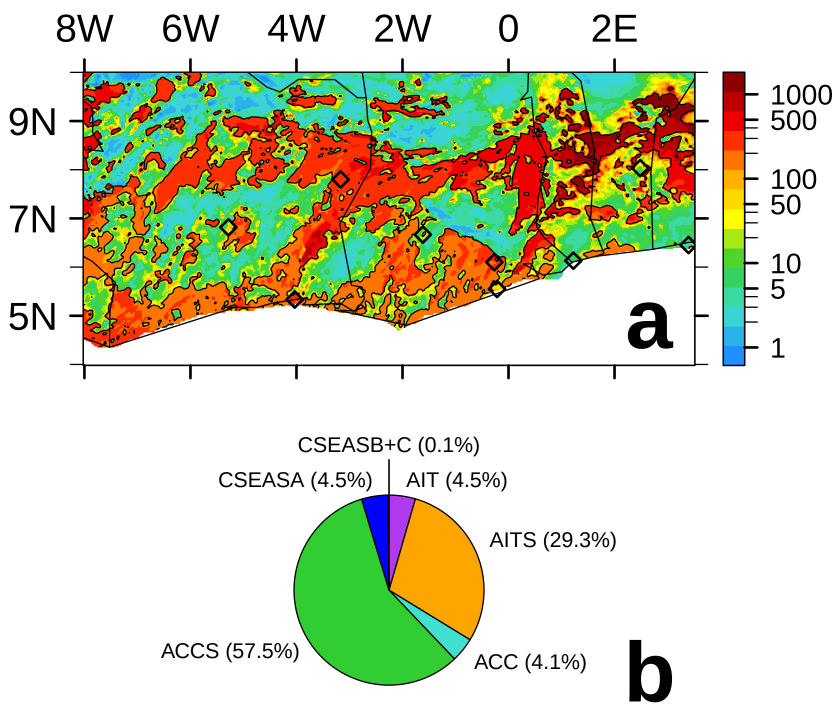

Fig. 3. ALWC at 500 m AGL over land on 3 July 6 UTC. (a) Total ALWC (mg m-3, shading) and RH of 95% (black contour) and (b) pie chart of the ALWC.

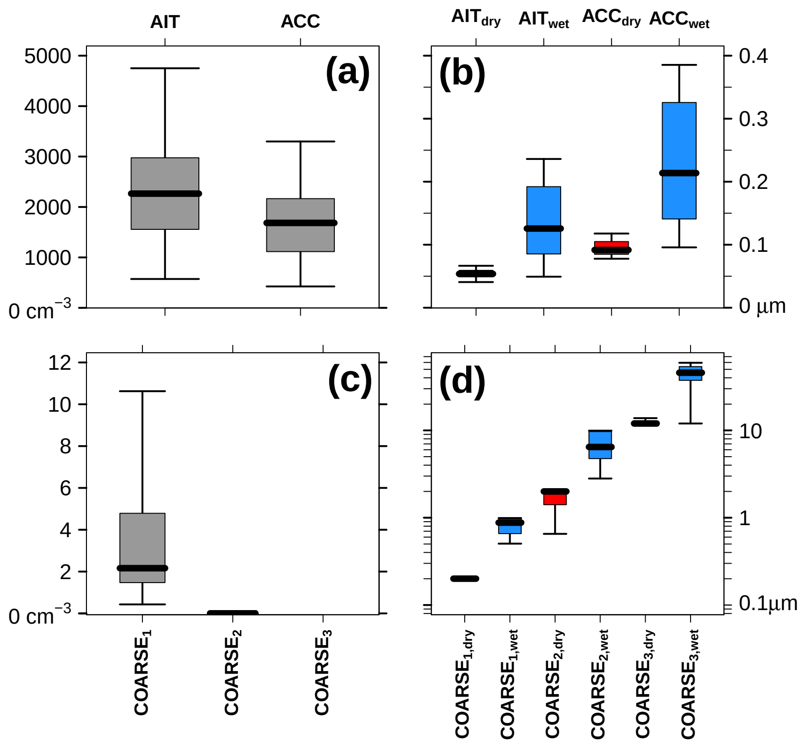

Fig. 4. Boxplots of (a) aerosol number density (cm-3) and (b) dry (red) and wet (blue) aerosol diameters (um) for AIT and ACC and boxplots of (c) aerosol number density (cm-3) and (d) dry and wet diameter.

2-3 July 2016 (Fig. 15):

	In-loud ALWC (ICA)		Off-cloud ALWC (OCA)	
	lower outlier	upper outlier	lower outlier	upper outlier
SW day	-	0.4%	-	0.2%
LW day	23.3%	0.4%	-	0.05%
LW night	3.5%	0.5%	-	0.02%

Fig. 5. Table summarizing the percentages of ALWC data that are not included in the linear fitting (red curves in Fig. 15 (2-3 July)).

6-7 July 2016 (Fig. H1):

	In-loud ALWC (ICA)		Off-cloud ALWC (OCA)	
	lower outlier	upper outlier	lower outlier	upper outlier
SW day	-	0.1%	-	0.2%
LW day	16.7%	0.1%	-	0.05%
LW night	3.5%	0.2%	-	0.06%

Fig. 6. Table summarizing the percentages of ALWC data that are not included in the linear fitting (red curves in Fig. H1 (6-7 July)).

2-3 July 2016 (Fig. 15):
n_max_day = 472680
n_max_night= 436320

	In-loud ALWC (ICA)	Off-cloud ALWC (OCA)
	n	n
SW day	16560	55120
LW day	12687	55230
LW night	9310	114402

Fig. 7. Table showing the total number n of gridboxes that are used for the linear fitting for 2-3 July 2016 (Fig. 15).

6-7 July 2016 (Fig. H1):
n_max_day = 472680
n_max_night= 436320

	In-loud ALWC (ICA)	Off-cloud ALWC (OCA)
	n	n
SW day	17086	48518
LW day	14236	48583
LW night	8903	109723

Fig. 8. Table showing the total number n of gridboxes that are used for the linear fitting for 6-7 July 2016 (Fig. H1).