

We thank both anonymous reviewers for their helpful comments. Below, we have answered all their remarks point-by-point, with the reviewers comments in black, [our replies in blue](#), *quotes from the manuscript in grey italic with changed text in red italic*.

Anonymous Referee #1

The authors have addressed my comments sufficiently so that I find it suitable for publication, with one exception.

The introduction has been greatly improved; however, there is a reference that should be added that is relevant to this study. In the sentence "However, few studies have attempted to combine analysis of regional numerical models with measurements (Ten Hoeve et al., 2011)" in line 59 should also include a reference to Fan et al. Science (2018). This paper also uses a regional model combined with measurements to look at cloud-aerosol interactions.

[A reference to Fan et al. \(2018\) has been added.](#)

That paper also argues that secondary activation can occur in deep convection from aerosols entrained at cloud base. Some of those aerosols (small) may not be activated until higher altitudes where the supersaturation is different than at cloud base. Polonik included some text in the revision on secondary activation (which I encouraged them to do) that implied that it occurs via lateral mixing, but that is not the only means of secondary activation.

[We have added more text to highlight the potential further effects on cloud microphysics do to the activation of ultrafine particles.](#)

*"[...] Secondary, in-cloud activation of aerosol particles to cloud droplets is only considered to the extent that entrainment and in-cloud supersaturation is represented on the grid-scale. **Other sources of secondary activation such as ultrafine particles (Fan et al., 2018) are not considered.** Cloud chemistry and limited heterogeneous processes are included as [...]"*

Anonymous Referee #2

This paper documents a modeling study of CCN in the Amazon with WRF-Chem, evaluated with ACRIDICON-CHUVA observations. The authors find biomass burning aerosols influence the Amazon clouds, but also suggest a saturation of the effect in very polluted conditions. The authors have gone some way towards addressing the most important of my previous comments. The review responses were reasonably comprehensive and well organized. Both the introduction and conclusion of the paper are improved. However, the paper text still needs some important changes before it is suitable for publication.

Major comments

Abstract: The authors change “underestimation” of CDNC to overestimation, but Figure 2 hasn’t changed, and still shows that the model underestimates CDNC. I am confused as to why this was changed and why the abstract says the slope is two when it is 0.334.

We thank the reviewer for carefully re-reading the manuscript. This was an erroneous edit. The model does indeed underestimate CDNC, just as the reviewer remarked. We have corrected this and ask the reviewer to refer to the updated abstract in our reply to your next comment.

The last sentence of the abstract still needs changing, in line with the modified conclusions (but see below).

The reviewer is referred to our answers to his/her comments below. The updated abstract now reads:

*“[...] on cloud microphysical **and optical** properties (droplet number concentration and effective radius).*

*We found agreement between modeled and observed median cloud droplet number concentrations (CDNC) for low values of CDNC, i.e., low levels of pollution. In general, a linear relationship between modelled and observed CDNC with a slope of **0.3** was found, which **implies a systematic underestimation** of modeled CDNC as compared to measurements. Variability in cloud condensation nuclei (CCN) number concentrations **was also underestimated** and cloud droplet effective radii (r_{eff}) **were overestimated** by the model. Modeled effective radius profiles began to saturate around 500 CCN per cm^3 at cloud base, indicating an upper limit for the model sensitivity well below CCN concentrations reached during the burning season in the Amazon Basin. **Additional** CCN emitted from local fires did not cause a notable change in modelled cloud **droplet effective radii**. **Finally, we also** evaluate a parameterization of CDNC at cloud base using more readily available cloud microphysical properties, **showing that we are able to derive CDNC at cloud base from cloud-side remote sensing observations.**”*

I did not find the promised supplement.

The reviewer is correct, we mistakenly referenced a supplement that we ultimately deemed unnecessary. The evaluation plot mentioned was actually directly included in the responses to reviewers, and is reproduced here below:

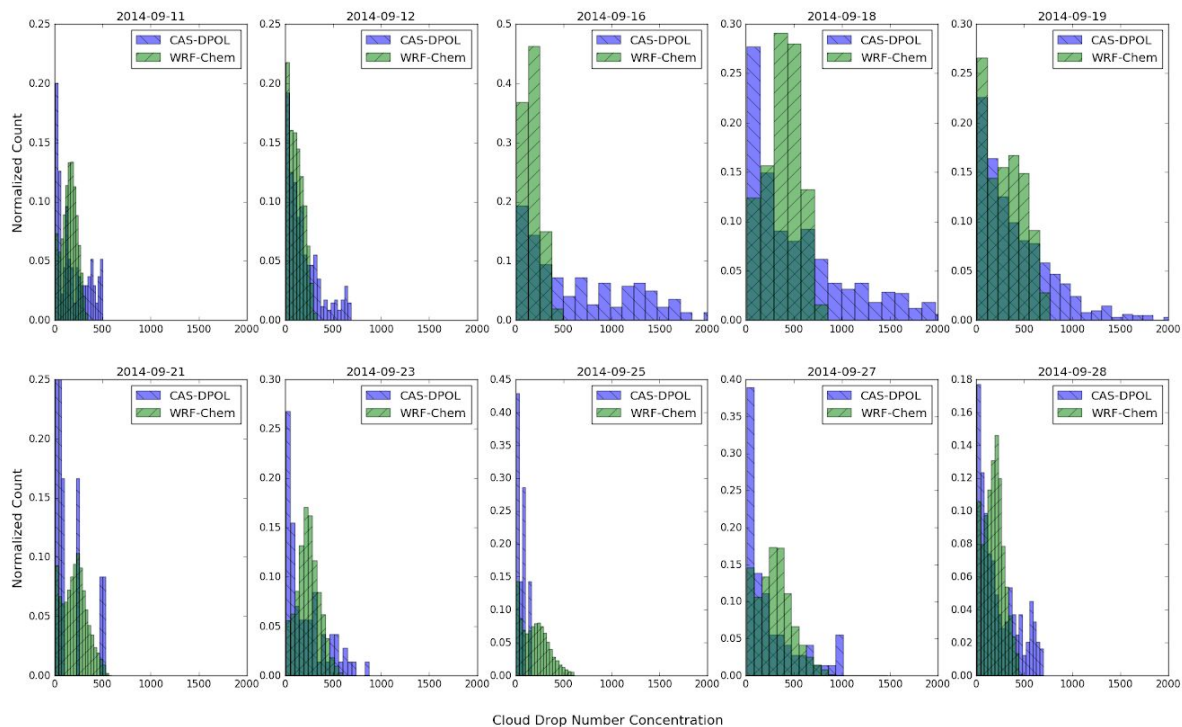


Figure: Normalized PDFs of in situ CAS-DPOL and modeled WRF-Chem track cloud drop number concentration (CDNC) data from the entire inner domain. A direct comparison of measurements and model output is not feasible, because the modeled clouds do not occur at the same place and time as those in reality. The modeled and measured CDNC agree reasonably well, but do not reach extreme values above about 1000 per cm^3 .

In my previous review, I said:

“While the effective radius is indeed the critical quantity that determines cloud albedo and the Twomey effect, it is cloud droplet number that determines the ‘microphysical effects’ of aerosols (on warm rain formation, droplet freezing rates, and droplet evaporation), and simulated CDNC apparently does not saturate (line 277). This apparent saturation of effective radius in the model is not sufficient grounds to say the model is in disagreement with observed aerosol-cloud microphysical interactions above 500/cc, as is stated in the conclusion.”

I don’t feel this comment has been adequately addressed. The authors claim to have separated Twomey effects from microphysical effects, but they only do this in the discussion, not in the abstract or the conclusions. The authors still say “the additional CCN emitted from local fires did not cause a notable change in modelled cloud microphysical properties” in their abstract, but the additional CCN clearly leads to increased CDNC – which is an obvious and observed change in microphysical properties.

The reviewer is correct, the addition of biomass burning resulted in negligible changes in cloud droplet effective radii, not cloud microphysical properties as originally stated. This has been corrected, please refer to our reply above for a reproduction of the updated abstract.

Again in the conclusions, the authors say “Our model results are in disagreement with observations of microphysical effects at much higher aerosol loading from previous campaigns”, and this statement is not at all justified. The simulations clearly do show microphysical effects, but they may not be the same effects as the microphysical effects observed.

We agree with the reviewer that this statement is too general. It has been amended, and the reviewer is referred to our updated conclusion reproduced below these comments for reference.

In fact, in polluted conditions, if CDNC increases when aerosol concentrations increase, while r_{eff} does not increase, that means LWC must increase, because of the relation $r_{\text{eff}} \sim (LWC/N)^{0.333}$. Increasing LWC with increasing CDNC is an aerosol-cloud microphysical interaction, in fact one quite commonly observed in models (e.g. McCoy et al, ACP 2018), and not a saturation of anything.

Because it probably is not the same aerosol-cloud interaction as seen in observations, the structure of the paper may not need changing. However, I really do think the authors should make a much more fundamental change to their conclusions than they did in response to my previous review. The saturation of the Twomey effect in polluted conditions due to the saturation of effective radius is obvious because of the $r_{\text{eff}} \sim (LWC/N)^{0.33}$ relation, and adds nothing new to our understanding of atmospheric science.

We agree that corresponding changes of CDNC, LWC, and r_{eff} are microphysical interactions. Still, we find that the modelled r_{eff} profile does not change anymore above 500 per cm^3 , which is frequently even below the observed regional background, and this has clear implications (no further Twomey effect due to biomass burning emissions) for the radiative impact of clouds. We agree that this saturation is not a new finding, but consider it important to show that this occurs in the model (already) at CCN roughly above 500 per cm^3 . It is actually vital to be able to determine the point (in terms of LWC and CDNC) at which this happens and whether model and observations agree, as this determines the ability of the model to accurately represent the radiative impact of biomass burning events through the cloud-albedo feedback. We have therefore adapted the text accordingly (see below).

On the other hand, other findings in the paper, for example, the testing of the Freud parameterization, the finding that CDNC is underestimated, are legitimate new findings that are worth publishing. The conclusion should be rewritten to emphasise these instead, and the abstract changed to match.

We have changed the wording on this topic to state our findings without making them sound surprising. We have also softened the language about the implications for other modeling studies. However, we clearly demonstrate that there is a systematic overestimation of effective radii in the model compared to in situ measurements, which would translate directly into an underestimation of cloud reflectivity. As suggested, more emphasis has been placed on the new application of the parameterization. The updated conclusions now read:

*“Aerosol-cloud interactions have been the focus of field campaigns and measurement development due to the large associated model uncertainty. Here we used novel observations taken on board the HALO aircraft during the ACRIDICON-CHUVA field campaign to evaluate cloud representation in a numerical model to **help reduce** this uncertainty. We demonstrated that we can reproduce realistic cloud properties (i.e., cloud droplet effective radius profiles) with a regional online-coupled chemistry-transport model at convection-permitting scales for the Amazon region during the biomass burning season.*

*As expected **from** theory, the number of CCN at cloud base has a major influence on cloud droplet size and the shape of the **vertical profile of cloud droplet effective radius**. Increasing CCN leads to decreasing cloud droplet sizes, and we **demonstrated that the model and the observations exhibit quantitatively similar behavior**. We also observed a saturation effect at high aerosol concentrations **in the model** (number concentration of CCN larger than 500 cm^{-3} at STP), above which we find no further change in modelled effective droplet size or the shape of the droplet size profile. **Observations** from previous campaigns (Reid et al., 1999; Andreae et al., 2004) and from the ACRIDICON-CHUVA campaign (Braga et al., 2017b) **have demonstrated substantial Twomey effects at much higher aerosol loadings. Additionally, the relation between modelled and observed CDNC is linear and has a slope of 0.3, indicating a considerable underestimation of cloud droplet number concentrations by the model.***

*Although we only tested one microphysics scheme, we demonstrated that a modern, complex parameterization does not imply accurate representation of **all** cloud microphysical properties and suggest that calculations of the radiative forcing of these phenomena **may be biased under polluted conditions like those found during the Amazon biomass burning season.***

Evaluation of the parameterization of Freud et al. (2011) proved to be successful in deriving N_a from cloud-side remote sensing data collected by the specMACS instrument. We note a high sensitivity of the method at low N_a and its dependence on an average mixing factor. We were able to gain these insights by applying a previously developed parameterization in a new context. Our study demonstrates that, despite some inherent challenges, existing techniques can be applied for model-measurement comparisons to improve our understanding of model biases.”

Minor comments

The Reid et al, 1999 paper is highly relevant to this study and should not be brought up for the first time in the conclusion. The authors should discuss the main findings of the paper in the introduction, and put their results more fully in the context of Reid’s work in the discussion.

We agree that Reid et al. (1999) and their findings should be mentioned earlier, and have therefore added a paragraph to the introduction:

“[...] cloud droplet effective radius (r_{eff}) vertical profiles, since r_{eff} profiles represent the micro-physical development of a cloud and can be derived from in situ and remote sensing observations.

Reid et al. (1999) similarly investigated the effects of biomass burning in Brazil. In their simulations, they found no further changes in r_{eff} from additional biomass burning aerosol when regional background accumulation-mode aerosol concentration reached $3000\text{-}4000\text{ cm}^{-3}$. r_{eff} was then merely a function of the liquid water content. They also showed that r_{eff} for clouds affected by biomass burning smoke are considerable smaller than those of clouds in more pristine environments like a marine boundary layer. Though r_{eff} profiles describe the vertical evolution[...]

L37 “nuclei”->“nuclei”

Corrected.

L371 “measured by”->“measured by”

Corrected.

The challenge of simulating the sensitivity of the Amazonian clouds microstructure to cloud condensation nuclei number concentrations

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Abstract. The realistic representation of ~~cloud-aerosol~~aerosol-cloud interactions is of primary importance for accurate climate model projections. The investigation of these interactions in strongly contrasting clean and polluted atmospheric conditions in the Amazon region has been one of the motivations for several field ~~observations~~campaigns, including the airborne Aerosol, Cloud, Precipitation, and Radiation Interactions and Dynamics of CONvective cloud systems - Cloud Processes of the Main
5 Precipitation Systems in Brazil: A Contribution to Cloud Resolving Modeling and to the GPM (Global Precipitation Measurement) (ACRIDICON-CHUVA) campaign based in Manaus, Brazil in September 2014. In this work we combine in situ and remotely sensed aerosol, cloud, and atmospheric radiation data collected during ACRIDICON-CHUVA with regional, online-coupled chemistry-transport simulations to evaluate the model's ability to represent the indirect effects of biomass burning aerosol on cloud microphysical and optical properties (droplet number concentration and effective radius).

10 We found agreement between modeled and observed median cloud droplet number concentrations (CDNC) for low values of CDNC, i.e., low levels of pollution. In general, a linear relationship between ~~modeled~~modelled and observed CDNC with a slope of ~~two~~0.3 was found, which ~~means a systematic overestimation~~implies a systematic underestimation of modeled CDNC as compared to measurements. Variability in cloud condensation nuclei (CCN) number concentrations was also underestimated and cloud droplet effective radii (r_{eff}) ~~was also underestimated~~were overestimated by the model.

15 Modeled effective radius profiles began to saturate around 500 CCN per cm^3 at cloud base, indicating an upper limit for the model sensitivity well below CCN concentrations reached during the burning season in the Amazon Basin. ~~Regional background aerosol concentrations were sufficiently high such that the additional~~Additional CCN emitted from local fires did not cause a notable change in modelled cloud ~~microphysical properties.~~

~~In addition, we droplet effective radii. Finally, we also~~ evaluate a parameterization of CDNC at cloud base using more readily
20 available cloud microphysical properties. ~~Our study casts doubt on the validity of regional scale modeling studies of the cloud
albedo effect in convective, polluted situations where the number concentration of CCN is greater than 500, showing that we
are able to derive CDNC at cloud base from cloud-side remote sensing observations.~~

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1 Introduction

25 Aerosol particles influence the formation of cloud droplets, and thereby the microphysical and macrophysical properties of
clouds. Cloud droplet sizes and number concentrations determine the effect of clouds on atmospheric radiation and, therefore,
also on weather and climate. Increased aerosol concentrations increase the cloud albedo (Twomey, 1991) and possibly the life-
time (Albrecht, 1989) of clouds by decreasing droplet size if the total liquid water mass is assumed constant. Cloud alterations
by aerosol (i.e. indirect effects) can therefore lead to enhanced reflection of solar radiation under high aerosol loading, and
30 therefore cause a net cooling of the sub-cloud layer. However, the magnitude of these effects is not well constrained, which
causes major uncertainties in current climate projections (IPCC, 2014).

Representing aerosol-cloud interactions in numerical models that form the basis of these projections is challenging because
two of the most dynamic and complex atmospheric systems (aerosol and clouds) must be adequately represented individually
before considering an accurate representation of their interactions (Ghan et al., 2016). Correctly modeling cloud condensation
35 ~~nuclei~~ nuclei (CCNs) number concentration requires accurate representation of aerosol chemistry and size, which depend on
parameterizations of emissions, relevant chemical reactions, microphysical interactions like coagulation, and removal processes
like dry deposition (Zaveri et al., 2008). In sufficiently complex parameterizations the calculated CCNs will then influence the
formation of droplets under saturated conditions and conversely, the droplets may remove the aerosol from the atmosphere.

Cloud microphysical parameterizations with varying levels of complexity have been incorporated into numerical models of
40 the atmosphere (e.g., Khain and Sednev, 1996; Seifert and Beheng, 2006; Morrison et al., 2005; Grützun et al., 2008; Thompson
and Eidhammer, 2014), which provides opportunities to better understand the underlying physical processes. It is difficult,
however, to disentangle benefits in forecast-relevant quantities (e.g., 500 hPa pressure field deviation, storm track accuracy, or
accumulated precipitation) from an actual improvement in the modelled cloud macro- and microphysical characteristics and its
impact on the atmospheric radiation budget. Testing such parameterizations on a mechanistic level requires direct comparisons
45 of model output to a variety of data sources (Seinfeld et al., 2016) as well as situations in which a noticeable aerosol signal can
be expected. Events like volcanic eruptions (Malavelle et al., 2017; McCoy and Hartmann, 2015), desert dust outbreaks (Levin
et al., 2005; Sassen et al., 2003), or wildfires (Rosenfeld, 1999; Brioude et al., 2009) provide strong signals that facilitate such
process-level analysis of aerosol-cloud interactions.

We focus on the Amazon, which has been a historically popular location for aerosol-cloud investigations, largely because both very high and very low aerosol concentrations can exist in the region and because convective clouds are somewhat predictable. There have been multiple efforts to quantify Amazonian aerosol-cloud interactions from remote sensing (Kaufman and Nakajima, 1993; Kaufman and Fraser, 1997; Lin et al., 2006; Wall et al., 2014), in situ measurements (Andreae et al., 2004; Martin et al., 2017; Andreae et al., 2018), combinations of measurement types (Rosenfeld et al., 2012; Gonçalves et al., 2015), and models (Feingold et al., 2005; Zhang et al., 2008; Martins et al., 2009). However, few studies have attempted to combine analysis of regional numerical models with ~~measurements~~ (Ten Hoeve et al., 2011) measurements (Ten Hoeve et al., 2011; Fan et al., 2018). The specific comparison of modeled and measured microphysical quantities have previously not been done. Aerosol-cloud parameterizations and computational power have recently improved to allow for such a study, but the direct comparison of modeled and measured cloud parameters remains challenging.

We use simulations and novel measurements from a recent field campaign in the Amazon to explore aerosol-cloud-radiation effects of biomass burning from a microphysical perspective. We first evaluate whether numerical simulations on convection-permitting scales can accurately represent observed cloud microphysical properties. For this purpose we focus on cloud droplet number concentration (CDNC) and cloud droplet effective radius (r_{eff}) vertical profiles, since r_{eff} profiles represent the microphysical development of a cloud and can be derived from in situ and remote sensing observations.

Reid et al. (1999) similarly investigated the effects of biomass burning in Brazil. In their simulations, they found no further changes in r_{eff} from additional biomass burning aerosol when regional background accumulation-mode aerosol concentration reached 3000-4000 cm^{-3} . r_{eff} was then merely a function of the liquid water content. They also showed that r_{eff} for clouds affected by biomass burning smoke are considerable smaller than those of clouds in more pristine environments like a marine boundary layer.

Though r_{eff} profiles describe the vertical evolution of cloud microphysical properties, it is actually the number of activated cloud condensation nuclei at cloud base, N_a , that provides the link between cloud development and aerosol availability (Khain et al., 2005). Parameterizations have been developed to determine N_a based on observations of r_{eff} since N_a is a somewhat elusive quantity to observe using remote sensing (Rosenfeld et al., 2012). Therefore we then also evaluate the applicability of the parameterization from Freud et al. (2011) using in situ, remote-sensing and model-derived r_{eff} profiles along with modeled and measured N_a .

Though many measurements and modeling studies have focused on the Amazon, they have not attempted to directly compare regional model output and measured cloud microphysical parameters. This comparison is a step towards bridging the gap between the observations used to improve physical understanding and the numerical models used to predict future climate.

2 Methods

2.1 Field Campaign

The Aerosol, Cloud, Precipitation, and Radiation Interactions and Dynamics of CONvective cloud systems - Cloud Processes of the Main Precipitation Systems in Brazil: A Contribution to Cloud Resolving Modeling and to the GPM (Global Precip-

Table 1. Dates of flights conducted during the ACRIDICON-CHUVA campaign, with basic information about each flight compiled from Wendisch et al. (2016) and the campaign blog (<https://acridicon-chuva.weebly.com/>; last accessed: July 10, 2018). CCN levels during each research flight are binned into low (“+”), medium (“++”) and high (“+++”).

Date	Flight #	CCN level	Description
2014-09-11	AC09	+	Clean conditions for cloud profiling
2014-09-12	AC10	+	Satellite coordination and several in situ clouds sampled in relatively clean conditions
2014-09-16	AC11	++	Tracer experiment near Manaus, with some fires in the vicinity
2014-09-18	AC12	+++	Polluted conditions but relatively few large clouds sampled
2014-09-19	AC13	+++	Polluted conditions, sampling of complete cloud profiles
2014-09-21	AC14	++	Satellite coordination, GoAmazon GI aircraft coordination, medium pollution
2014-09-23	AC15	++	Surface albedo measurement early, cloud sampled later, medium pollution
2014-09-25	AC16	++	Tracer experiment near Manaus, fires in the vicinity
2014-09-27	AC17	+++	Sample clouds over different land surfaces, compare to GPM satellite, polluted conditions
2014-09-28	AC18	+	Medium sized cumulus samples and full cloud profiles in clean conditions

itation Measurement) (ACRIDICON-CHUVA) field campaign (Wendisch et al., 2016), was conducted over the Amazon in September 2014 during the dry season, when biomass burning from regional agricultural practices creates strong perturbations of cloud condensation nuclei (CCN) number concentration (Pöhlker et al., 2018). Researchers collected data on aerosol size and composition, CCN concentration, cloud phase and droplet size, and trace gas concentrations, and other atmospheric quantities. Both remote sensing and in situ data were collected aboard the High Altitude and Long Range Research Aircraft (HALO), operated by the German Aerospace Center (DLR). HALO flew underneath and within clouds to reconstruct vertical profiles. Typically, HALO research flights began with a ferry from Manaus to a region of interest, followed by sampling in that region, and ending with the trip back to Manaus (Figure 1, Table 1). The regions of interest were areas with forecasted presence of convective clouds above specific surface conditions, such as intact forest or polluted agricultural burning areas. Many of the HALO flights were conducted in regions where medium or high aerosol number concentrations from biomass burning were suspected to influence cloud microphysical and radiative properties.

2.2 Model

We attempted to reproduce the measurements conducted during the HALO flights using numerical simulations with the Weather Research and Forecasting model with Chemistry (WRF-Chem, Grell et al., 2005) at convection-permitting scales. The model simulated atmospheric motion with online calculations of trace gases and aerosol chemical and physical properties in a nested domain setup. One degree resolution, six-hourly updated meteorological boundary conditions were taken from analyses of the National Center For Environmental Prediction Global Forecast System (NCEP GFS), and chemical boundary conditions

were provided by forecasts of the global chemistry model MOZART (<https://www.acom.ucar.edu/wrf-chem/mozart.shtml>, last
100 accessed February 6th, 2018).

The simulations feature a size-resolved description of the full lifecycle of ambient aerosol, including biomass burning emis-
sions, secondary particle formation through trace gas oxidation, and dry and wet deposition. Specifically, we used the Model
for OZone And Related chemical Tracers (MOZART) gas-phase chemistry (Emmons et al., 2010; Knote et al., 2014) and the
Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosol module (Zaveri et al., 2008), with a volatility
105 basis set parameterization for organic aerosol evolution (Knote et al., 2015). Anthropogenic emissions data were taken from
the Emissions Database for Global Atmospheric-Research from the task force for Hemispheric Transport of Air Pollution
(EDGAR-HTAP, Janssens-Maenhout et al., 2012). Biogenic emissions are calculated online using the Model of Emissions of
Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006). The Fire Inventory from NCAR (FINN) module was used
for the fire emissions data (Wiedinmyer et al., 2011).

110 Radiative properties of the aerosol population are considered based on size distribution and component-resolved optical
properties (Barnard et al., 2010). The modeled aerosol description is linked to the double-moment microphysics scheme of
Morrison and Gettelman (2008), and no convection parameterization was applied in the nested domain. The Morrison and
Gettelman (2008) scheme has five hydrometeor classes (cloud droplets, rain, cloud ice, snow, and graupel), with each size
distribution parameterized by a Gamma function. The cloud droplet effective radius is calculated through integration over the
115 droplet size distribution:

$$r_{eff} = \frac{\int_0^\infty r^3 N(r) dr}{\int_0^\infty r^2 N(r) dr} \quad (1)$$

with r cloud droplet radius, and $N(r)$ droplet number concentration at radius r .

Effects of aerosol particles on atmospheric radiation (direct effect) are considered as presented in Fast et al. (2006). The
number of CCN available for cloud formation as well as their physiochemical properties (size distribution and hygroscopicity)
120 are provided to the cloud microphysics scheme based on the online-calculated aerosol properties. Activation of aerosol par-
ticles as cloud droplets is calculated based on the aerosol size distribution and chemical composition using κ -Koehler theory
(Abdul-Razzak and Ghan, 2000, 2002), with relevant aspects of the implementation in the version of WRF-Chem used here
presented in Gustafson Jr et al. (2007) and Chapman et al. (2009). The life cycle of activated aerosol particles is modelled
explicitly; i.e., they are removed from the interstitial aerosol population and their evolution is modelled in accordance with that
125 of the cloud droplets in which they are incorporated, including processes like washout from precipitation or re-evaporation.
Secondary, in-cloud activation of aerosol particles to cloud droplets is only considered to the extent that entrainment and
in-cloud supersaturation is represented on the grid-scale. Other sources of secondary activation such as ultrafine particles
(Fan et al., 2018) are not considered. Cloud chemistry and limited heterogeneous processes are included as presented in Knote
et al. (2015). Chemistry and aerosol processes are included in an operator-splitting fashion, in which individual processes up-
130 date model fields sequentially. For each WRF-Chem time step, advection is calculated first, followed by droplet activation and
then chemistry and aerosol processes.

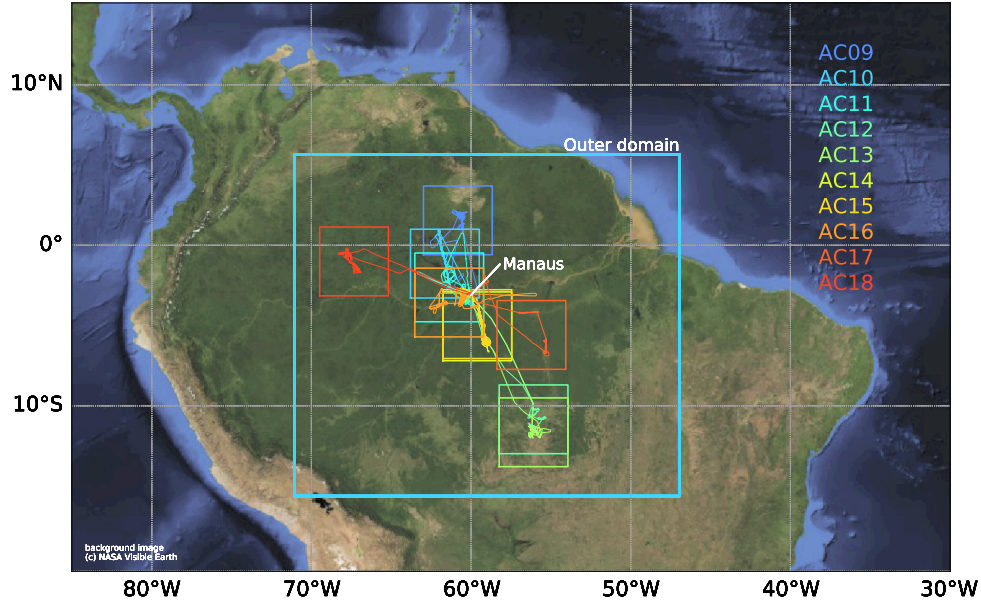


Figure 1. A map showing the campaign area, with all ACRIDICON-CHUVA research flights considered in this study as color-coded lines, the continuously-run outer simulation domain (blue box) as well as the individual nested domains used for analysis of each research flight, identified by the flight labels (Table 1). The outer domain resolution is 15 km and the inner domain resolution is 3 km.

The above-described WRF-Chem simulations were conducted over the Amazon region for the ACRIDICON-CHUVA mission period between 8 - 30 September 2014. A continuous simulation with 15 km horizontal resolution, covering an area of approximately $3000 \times 2700 \text{ km}^2$ (200×180 grid points), and 36 vertical levels up to 50 hPa, was conducted for the full campaign period (see Figure 1 for domain overview). To keep the large-scale meteorology in line with reality, WRF-Chem was restarted every 24 hours (at 0 hours UTC) from GFS analyses. Concentrations of trace gases and aerosol quantities were carried over, however, to allow for multi-day pollution build-up and aging. Each 24 hour period was simulated with a 6 hour meteorological spin-up with nudging and a chemical restart file from the previous day. Meteorology was then allowed to evolve freely within the WRF-Chem domain (i.e. no nudging was applied) to enable the model to develop the implemented aerosol-cloud-interactions. Three additional days before the study period were simulated to spin-up trace gas chemistry and aerosol.

Convection-permitting, 3 km horizontal resolution domains (180×180 grid points, approx. $540 \times 540 \text{ km}^2$) were then “nested” into this simulation during days with HALO flights. Two-way interactions were allowed between the parent and the nested domains. The location of these “nests” varied and were chosen so that they covered the area of interest sampled by HALO in each flight (Figure 1, see also Section 3.1). On each flight day, the nested domain was started (by interpolating the current state of the outer domain) at 09:00 UTC and run until 21:00 UTC, hence covering the full time frame of each HALO research flight. All model results presented in this study are from the nested, convection-permitting domains.

2.3 Measurements

2.3.1 Cloud in situ measurements

150 The cloud combination probe (CCP) combines the cloud imaging probe (CIP) and the cloud droplet probe (CDP) to measure the cloud particle size distribution by detecting their forward-scattered laser light (Lance et al., 2010). During the ACRIDICON-CHUVA campaign, the CCP measured at 1 Hz frequency from underneath the right wing of the HALO aircraft (Wendisch et al., 2016). A correction for the high flight velocities was applied to improve data quality (Weigel et al., 2016). The CCP measures particles with diameters between 2 - 960 μm , but here we only used the 14 bins for particle diameters from 3 - 50 μm
155 (from the CDP) to calculate the cloud particle effective radius. Except for the details of the selection of appropriate data points, the data used here is the same as described in Braga et al. (2017a). To filter the data we calculated liquid water content from binned effective diameter measurements and only included those with at least 1 g kg^{-1} liquid water content. This threshold is consistent with the one used to define “cloudy” points in model output.

Like the CCP-CDP, the Cloud and Aerosol Spectrometer with Depolarization (CAS-DPOL) measures cloud particle size
160 distributions at 1 Hz frequency (Baumgardner et al., 2011; Voigt et al., 2017). The CAS-DPOL measures the intensity of forward-scattered light between 4 - 12 degrees in 30 size bins from particles with diameter 0.5 - 50 μm . The polarized backward-scattered light is used to analyze the sphericity and thermodynamic phase of the measured particles (Baumgardner et al., 2014; Järvinen et al., 2016), but this capability was not used for our analysis. Our calculation of the cloud particle effective radius (Schumann et al., 2017) was again limited to particles between 3 - 50 μm , which corresponds to 10 Mie-
165 ambiguity corrected size bins, to account for consistency with the CDP. Further details on CAS-DPOL data evaluation are given in Kleine et al. (2018).

Profiles of r_{eff} were derived using data from both the CAS-DPOL and the CDP. Braga et al. (2017a) demonstrated that the CDP and CAS-DPOL instruments are comparable within their expected measurement uncertainties. Flamant et al. (2018) and Taylor et al. (2019) also found good agreement between CAS-DPOL and CDP measurements in shallow clouds. Here,
170 we combine measurements from both instruments into one in situ dataset to construct effective radii profiles. Therefore, the concentration of activated cloud condensation nuclei N_a , is derived using all in situ r_{eff} measurements with their respective adiabatic liquid water content (see further description in Section 2.3.4). Treating in situ measurements from the two instruments as independent is justifiable in part because they are located on opposite wings of the aircraft.

2.3.2 CCN in situ measurements

175 The number concentration of CCN was measured with a continuous-flow streamwise thermal gradient CCN counter (CCNC, model CCN-200, DMT, Longmont, CO, USA) (Roberts and Nenes, 2005; Rose et al., 2008). Activated CCN that grow to a diameter of at least 1 μm at a set water vapor supersaturation between 0.1 - 5% are counted by the instrument at 1Hz. Two sample inlets were used during the ACRIDICON-CHUVA campaign, but here we only use data from the HALO aerosol sub-micron inlet (HASI), which collected data at a constant supersaturation of 0.55 %. The uncertainty of the CCN measurements

180 is dominated by the counting statistics and ranges between 10% for high CCNs and 20% for low CCNs (Krüger et al., 2014).
The supersaturation uncertainty is also about 10% (Braga et al., 2017a).

2.3.3 Cloud remote sensing measurements

The spectral imager of the Munich Aerosol and Cloud Scanner (specMACS) was installed on the HALO aircraft during ACRIDICON-CHUVA. specMACS is a hyperspectral line camera that measures at visible and near-infrared wavelengths
185 (Ewald et al., 2016). Marshak et al. (2006) and Martins et al. (2011) suggested using the solar radiation reflected by illuminated cloud sides to derive the vertical profile of effective radius and cloud phase, but the ACRIDICON-CHUVA campaign was the first time that passive cloud side remote sensing was applied systematically for a large number of cases. Zinner et al. (2008) and Ewald et al. (2018) developed a cloud side retrieval and demonstrated the application using ACRIDICON-CHUVA data. Jäkel et al. (2017) derived phase information from cloud-side reflectivity measurements during ACRIDICON-CHUVA.
190 specMACS was mounted on HALO at a sideward viewing port to observe clouds passed by the aircraft. Cloud vertical profiles were then retrieved using the method by Ewald et al. (2018) along the flight route akin to a push-broom satellite instrument. Results for three cases are compared to in situ and WRF-Chem model data.

specMACS cases shown in this paper are first example cases and mainly presented to showcase the capability of airborne remote sensing to provide effective radius profiles and cloud droplet number concentration (CDNC). They are not representative
195 for whole flights or flight regions as the used in situ or modelled data, but show specific example local situations along a few minutes of flight time. In this respect they complement the large scale picture provided by modelled data averaged over $540 \times 540 \text{ km}^2$ or the in situ data collected over several hours flight time. specMACS cloud scenes were selected based on favorable data collection conditions. This includes minimal turning of the aircraft, favorable sunlight conditions, and high cloud coverage.

200 2.3.4 Derivation of N_a from in situ, remote sensing, and model cloud data

The central quantity to determine the influence of aerosol on cloud development and lifetime is the number of activated cloud condensation nuclei at cloud base, N_a (e.g. Khain et al., 2005; Freud et al., 2011). During ACRIDICON-CHUVA, HALO directly sampled N_a during their cloud profile flights, providing a valuable comparison to remotely sensed and modeled data. As the collection of in situ data is expensive and spatial coverage is limited, Rosenfeld et al. (2012) suggested to infer N_a at cloud
205 base using other more readily available observations like satellite retrievals. Freud et al. (2011) proposed a parametrization that derives N_a from the vertical profile of droplet radii. To do this, cloud base temperature and pressure are first used to calculate an adiabatic liquid water content (LWC_a) under the assumption that all water vapor above the saturation vapor pressure is condensed during the moist adiabatic ascent of a parcel. Then, LWC_a can be combined with an empirical relation between

r_{eff} and the volumetric radius, r_v (i.e., $r_v = 1.08 \cdot r_{\text{eff}}$ as in Freud et al. (2011)), and the density of water ρ_w to derive a fixed
210 N_a :

$$N_a = \frac{1}{\rho_w} \cdot \frac{3}{4\pi} \cdot \frac{LWC_a}{r_v^3} \cdot 0.7 \quad (2)$$

The ratio of LWC_a and r_v^3 is found as the slope of a linear regression through all available point pairs of LWC_a and r_v^3 in the droplet size profile, forced through the origin. An additional mixing factor of 0.7 accounts for the imperfection of the adiabatic assumption (Freud et al., 2011; Braga et al., 2017a). Freud et al. (2011) empirically derived this factor using in situ effective
215 radius and LWC data from multiple previous field campaigns, including one in the Amazon. Although there was geographic diversity in the data used for the derivation, only one estimation was made which may introduce an unknown error in our studies. This could be especially relevant for remotely sensed data that measure cloud sides rather than a cloud cross-section. Nonetheless, we apply the same derivation and same mixing factor to all three available r_{eff} datasets: remotely sensed, in situ, and model output. Applying this method to multiple data sources provides insights into the validity of this concept. The
220 resulting N_a can also be used for direct comparison of the different input r_{eff} profiles.

3 Representation of cloud microphysics in the model

3.1 Deriving comparable quantities for model-measurement evaluation

Comparing the three different sources of information on cloud microphysical properties (model, remote-sensing, and in situ observations) is not straightforward. Colocating in situ and remote-sensing observations required observing a cloud using
225 the side-facing specMACS, and then flying into this cloud to obtain respective in situ measurements. During ACRIDICON-CHUVA, cloud clusters had been identified for each research flight, which were then passed several times to allow for remote-sensing observations before probing these clusters in situ. This precludes direct comparison of individual clouds without diligent data selection, but allows for a statistical comparison of in situ data collected near the cluster and the corresponding remote-sensing observations. Simulations will not reproduce an individual (observed) cloud, but they will create a comparable,
230 realistic regional environment with comparable clouds. Hence, the nested domains were chosen such that they are center on the cloud cluster chosen as target for an ACRIDICON-CHUVA research flight. Assuming a homogeneous environment within the model domain, a statistical comparison of all modelled clouds in the model domain with observations taken of the cloud cluster within the domain is reasonable. Therefore, we used all clouds within the respective nested model domain to derive model statistics. Observation statistics are based on all data collected within the spatial domain of the model nest. As mentioned
235 above, statistics pertaining to in-cloud variables are restricted to data points with a liquid water content of more than 1 g kg^{-1} in both model and observations.

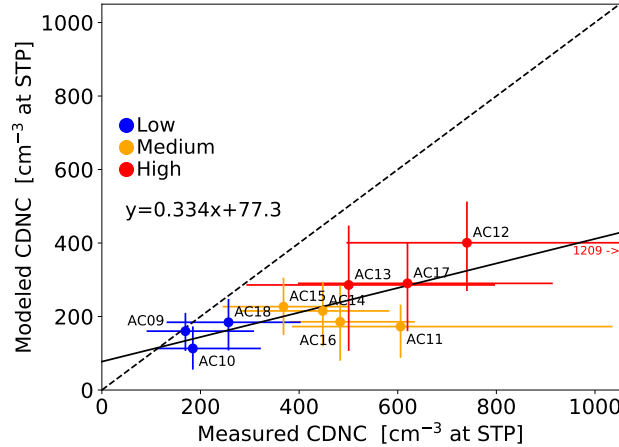


Figure 2. Median cloud droplet number concentration from the WRF domain and in situ measurements. The colors correspond to the CCN level labels in Table 1. Error bars depict the interquartile range (25 - 75% of all values). The equation describes the (solid black) regression line. The dashed black line is a 1-to-1 line for reference. STP refers to standard temperature (273.15K) and pressure (1000hPa).

3.2 Cloud droplet number concentrations

Figure 2 shows median in situ measurements of CDNC during flights and the median CDNC values from the entire nested model domain corresponding to the flight. Modeled and measured CDNC match for lower values of 200 cm⁻³ (AC09), but
 240 diverge for higher values. There is a linear relationship between WRF-Chem results and observations, albeit below the one-to-one line, leading to a factor of two of underestimation of CDNC for the most polluted case investigated (AC12 with about 750 cm⁻³ observed).

3.3 Variability in modeled r_{eff} profiles

All WRF-Chem modeled r_{eff} data from the ten nested domains was combined and binned by cloud-base CCN concentration
 245 (Figure 3). Cloud-base CCN is defined as the modeled CCN concentration at 0.5 % supersaturation directly below the lowest cloudy pixel in a model column.

The binning of r_{eff} profiles shows that the modeled profiles correspond to theoretical expectations; clouds with more available CCNs have a r_{eff} profile that is shifted towards smaller values relative to those with fewer available CCNs. The response to CCN concentration saturates in the model around 500-600 cm⁻³, indicating that biomass burning effects will be nonlinear
 250 and strongest in relatively clean conditions. We did not find such a saturation effect for CDNC (Figure 2). Between 2 - 4 km above sea level, where the most model clouds occur, the slope of the profile also scales with available CCNs. The radius grows quickly with height to a maximum r_{eff} under low CCN (clean) conditions, whereas under high CCN (polluted) conditions the radius does not reach a maximum until much higher in the atmosphere. The profiles reach a maximum and then remain roughly

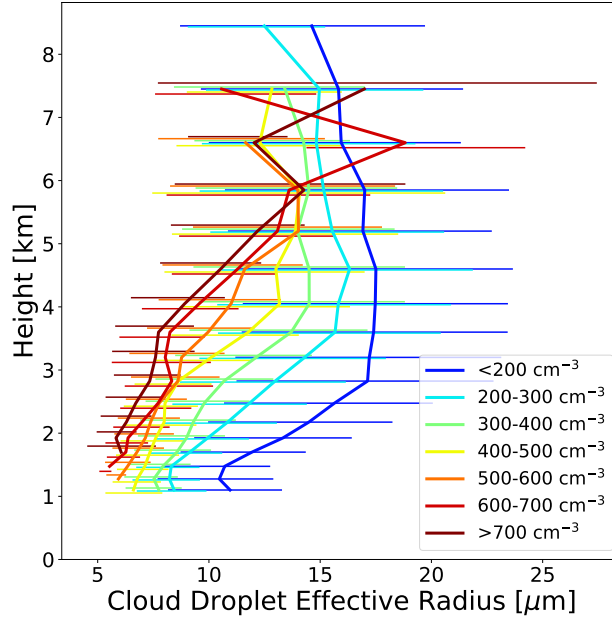


Figure 3. WRF-Chem simulated median cloud drop effective radius vertical profiles from all nested domain output during the study period, binned by below-cloud CCN concentration [cm^{-3} at STP]. Error bars represent the 20th to 80th percentile for each level and are offset vertically for readability.

constant at higher elevations. Under clean conditions, the maximum r_{eff} is larger and is reached at lower elevations. Profiles
 255 for the cleanest conditions also exhibit the largest maximum median r_{eff} of about $17 \mu\text{m}$.

3.4 Comparison of modeled and observed r_{eff} profiles

WRF-Chem modelled r_{eff} profiles were compared to remote-sensed and in situ measured profiles. In Figure 4 we show snapshots of the spatial variability of modeled CCN concentrations at cloud base for three different days. This figure demonstrates the influence of the fires on the regional CCN concentrations and highlights the CCN variability at large and small scales. Three
 260 dimensional CCN fields were simulated, but below-cloud concentrations (i.e. CCN concentration below the lowest cloudy point in a column) are most relevant for cloud droplet size. Figure 5 a-c then shows r_{eff} profiles derived from specMACS from two-minute cloud scenes on these three days, below-cloud-CCN binned WRF r_{eff} profiles from three hours near the specMACS data collection time, and all in situ r_{eff} profile measurements within the nested model domain. Figure 5 d-f shows the known modeled and in situ CDNCs. No CDNC are available for the specMACS observations since those data are remotely sensed.

265 Note that this is an approximate comparison, as no exact colocation can be expected between in situ and remote-sensed clouds, and we cannot compare individual modelled clouds directly to observed ones. Visual inspection of the slope and magnitude of median r_{eff} profiles measured by specMACS suggests that they match reasonably well to those from WRF-

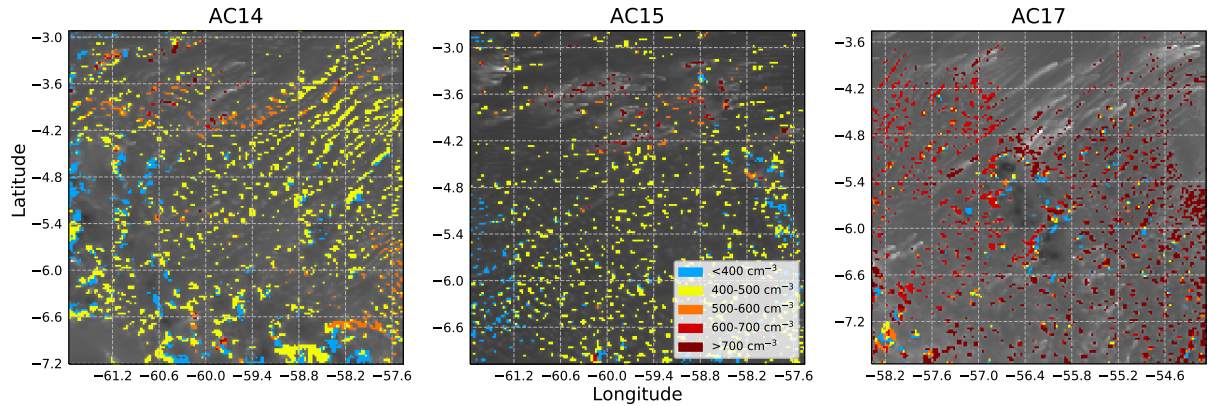


Figure 4. Spatial variability in modeled concentration of CCN at cloud base on three days (at 18Z) for the entire nested domain. Modelled aerosol optical depth (AOD) is shown as grey shading in the background, with brighter colors indicating higher AOD values. CCN concentrations are only shown where clouds were present.

Chem, though in situ r_{eff} tend to be smaller than both the modeled or the ones retrieved by specMACS for all three cases investigated here.

270 The relatively small differences between r_{eff} profiles at larger CDNC are expected because the theoretical relationship between r_{eff} and CDNC is $r_{\text{eff}} \sim (\frac{LWC}{CDNC})^{1/3}$ (Morrison and Gettelman, 2008). A linear relationship between LWC and CDNC therefore results in saturation of r_{eff} . However, at what CDNC this saturation occurs is not equally well described.

3.5 Number of activated cloud condensation nuclei at cloud base

As a more quantitative comparison of the different profiles, the number of activated CCNs at cloud base (N_a) were derived
 275 for each profile based on the methodology proposed in Freud et al. (2011). Braga et al. (2017a) already showed a comparison against in-situ measurements, which we use as a starting point here for an evaluation against remote sensing and regional model results. For the same three days as in Figure 5, Figure 6 a-c shows the regressions between adiabatic liquid water content (LWC_a) and mean volume radius (r_v) that result (using Eq. 2) in the calculated $N_{a,\text{calc}}$ values shown in Figure 6 d-f. LWC_a for the modeled profiles was calculated in model clouds at the same points as used for the r_{eff} values. For specMACS,
 280 a nested domain averaged LWC_a profile was used since the below-cloud CCN is unknown for those measurements. The same profile was used for the in situ LWC_a to allow for direct comparisons. Only the increasing portion of the WRF-Chem profiles were used for the fits in Figure 6 a-c; points above the first decrease that occurs above 4 km are excluded. The known CDNCs (Figure 5) and calculated N_a (Figure 6) matched well given that CDNC is being viewed as equivalent to N_a , although N_a is an upper limit for CDNC since CDNC can be influenced by processes like collision and coalescence. A direct comparison of
 285 the true and derived CDNC are shown in Figure 7. This comparison demonstrates the effectiveness of the Freud et al. (2011)

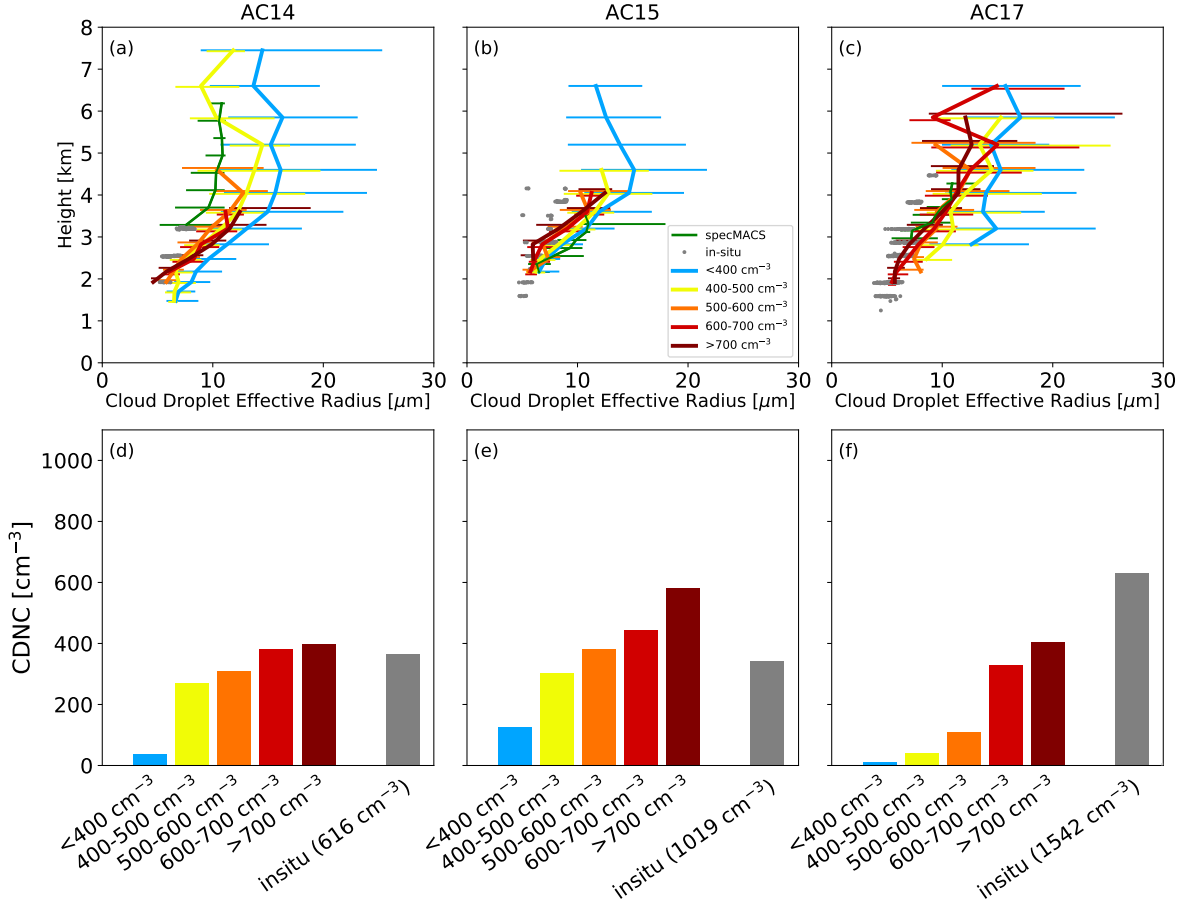


Figure 5. r_{eff} profiles and associated cloud droplet number concentration (CDNC) on three days during the field campaign. (a-c) show a comparison of median WRF-Chem, specMACS, and in-situ r_{eff} profiles. (d-f) show the “true” below-cloud CCN-binned CDNC from WRF-Chem simulations and CDNC from in situ cloud profiling. Average in situ CCN concentrations (below 2 km) are presented in the bar label for the in situ derived N_a . See Section 3.1 for details regarding the definition of “average”.

method for model data. The relationship is linear, but there is a systematic positive bias of derived CDNC. The factor of 0.7 as taken from the literature may be an underestimation for the modeled clouds. Sensitivity of the derivation to cloud base height may explain why using modeled LWC_a resulted in high derived CDNC for two of the in situ derivations. Another contributor could be the high low-level CCN concentrations that were not reached in the model and in part by the use of an average model LWC_a rather than a “true” LWC_a . Even though $N_{a,\text{WRF}}$ and $N_{a,\text{calc}}$ do not match exactly, general trends are captured. The N_a derived from the specMACS r_{eff} profiles ($N_{a,\text{spec}}$) fall within the range of modeled CDNCs (Figure 6 d-f). Compared to modeled CDNCs, specMACS-derived $N_{a,\text{spec}}$ are relatively high, low, and central for AC14, AC15, and AC17, respectively.

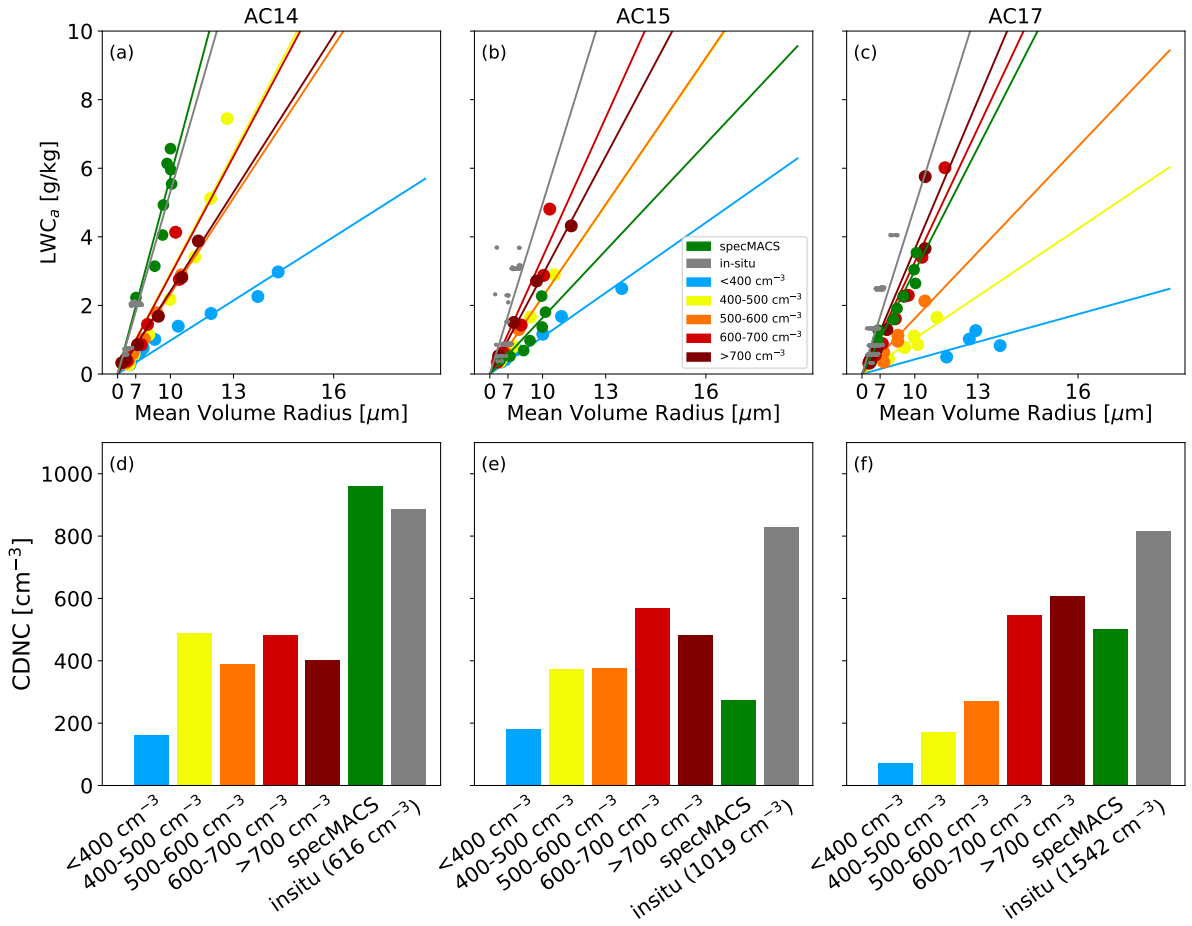


Figure 6. Derived cloud droplet number concentration (CDNC) on three days during the field campaign. (a-c) show the regressions between mean volume radius and adiabatic liquid water content (LWC_a) used to derive the CDNC as shown in (d-f). Average in situ CCN concentrations below 2 km are shown below the in situ derived N_a . (d-f) were derived from the slopes in (a-c), whereas Figure 5 d-f were more directly determined.

With the available data it is not possible to know the aerosol or below-cloud properties for the clouds sampled by specMACS. We suggest, however, that we can use the model results to deduce that the specMACS observed relatively polluted clouds during AC14 (Figure 6 a,d), relatively clean clouds during AC15 (Figure 6 b,e), and medium polluted clouds during AC17 (Figure 6 c/f). The N_a derived from the in situ profiles is higher than the others. While the calculated N_a depends on the theoretical adiabatic liquid water content (LWC_a), the measured LWC might in fact be lower. This finding should be explored further but is out of scope of this work.

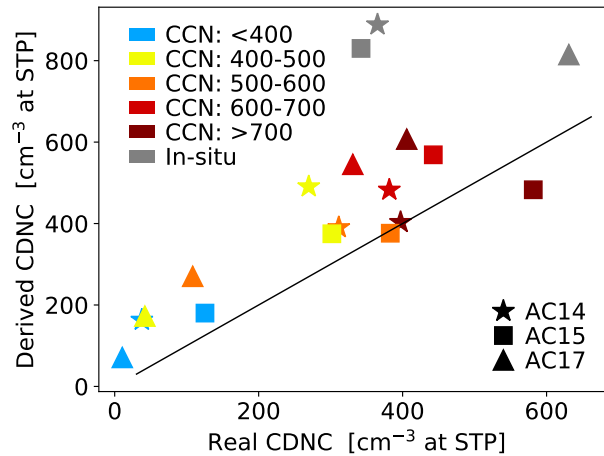


Figure 7. Comparison of real (i.e. CDP and CAS measured) CDNC with CDNC as derived using the Freud et al. (2011) method. Real CDNC for model data are average modeled CDNC in the model domain. Symbols indicate date, colors indicate model bin or in situ data. The one to one line is for reference. These are the same data as Figure 5 d-f and Figure 6 d-f.

3.6 Discussion

Modeled r_{eff} tended to be larger than in situ measurements of r_{eff} . Subsequently, directly modeled and model-derived CDNC concentrations were lower than in situ measurements and derivations. Partly, these differences can be accounted for by the low modeled CCN concentrations (Figure 2). However, the 20th to 80th percentile range of modeled profiles with high below-cloud CCNs do overlap with the in situ data. The modeled r_{eff} profiles began to saturate around 500 cm⁻³ at STP below-cloud CCN, with only small differences at higher concentrations (Figure 3), meaning that the modeled cloud albedo or Twomey effect saturates at approximately that concentration. A sensitivity study in which we artificially doubled the amount of biomass burning emissions showed the same saturation in modelled r_{eff} , further corroborating our findings. The concentration of around 500 cm⁻³ at STP below-cloud CCN is well below the CCN concentrations characteristic of the dry season in the southern half of the Amazon Basin, which are typically in the range of 1000 to 7000 cm⁻³ (Andreae et al., 2004; Andreae, 2009; Andreae et al., 2018). No such saturation was observed in the evaluation of modelled CDNC.

Increased model spatial resolution could potentially provide better agreement for these high-pollution situations, but a variety of hurdles (input data resolution of emissions and static data like land use, vegetation cover and topography, model formulation of turbulence, statistical methods for output analysis) need to be overcome before reliable simulations at higher resolution are feasible. The horizontal grid resolution of 3 km is at the fine end of what regional modeling systems were designed for, reaching for 'terra incognita' (Wyngaard, 2004) in terms of resolution. Sensitivity simulations in which we simply increased the horizontal and/or vertical resolution by a factor of two did not lead to improved agreement with observations.

More complex parameterizations of cloud microphysics, such as spectral bin microphysics (e.g. Grützun et al., 2008; Khain and Sednev, 1996), have been developed and used before in case studies. Such more complex parameterisations might improve

the representation of the cloud droplet size spectra and hence also modelled r_{eff} . Such parameterisations are, however, still computationally too expensive to be used on a regular basis or in the context of a climate study.

320 Estimating the radiative forcing due to biomass burning is of central importance to evaluate its impact on the climate system. Calculating the top of atmosphere radiative forcing leads to an campaign average daytime cooling of -0.9 W m^{-2} (not shown), which is comparable to previous estimates (e.g. Archer-Nicholls et al., 2016) and shows that our model behaves similar to existing studies. However, given the demonstrated lack of skill of the modeling system in representing the very strong CCN perturbations due to biomass burning, we refrained from further exploring their climate impacts.

325 We deem our modeling study is representative for other regional scale chemistry-transport modeling studies of aerosol-cloud interactions of convective clouds in situations strongly affected by biomass burning (e.g., Martins et al., 2009; Wu et al., 2011; Archer-Nicholls et al., 2016). WRF-Chem is a widely used modeling system and similar to other regional modeling systems. Our setup contains state of the art representations of clouds, aerosols, and aerosol-cloud interactions because we used a two-moment cloud microphysics scheme with a sectional aerosol module, and the cloud activation scheme of Abdul-Razzak and
330 Ghan (2000).

Comparisons between entire model domains and in situ measurements are inherently difficult since the exact measured clouds will never be realistically simulated due to the randomness of modeled clouds and the difference in scales. There are a variety of challenges involved with this comparison. However, especially at high CCN, the model overestimates r_{eff} and, therefore, underestimates N_a . The specMACS data experience similar comparison difficulties since each set only spans a cloud
335 scene ($\sim 50 \text{ km}$) over a short time ($\sim 2 \text{ minutes}$). However, the retrieved r_{eff} profiles still fall within the in situ measurements and the model output. Profile values derived from specMACS measurements also tend to be smaller than the data from in situ sampling, which is expected based on previous tests (Ewald et al., 2016).

We have demonstrated that the method by Freud et al. (2011) to derive cloud base CDNC from r_{eff} observations can successfully be applied in conjunction with simulated clouds to derive N_a from remotely sensed hyperspectral data of the specMACS
340 instrument. The method is limited by its high sensitivity at low N_a due to the mathematical nature of the slope (i.e. steep slopes in Figure 6 a-c) and we are unable to verify its accuracy with the available data. It also uses an average mixing factor that may vary for the cloud scenes measured ~~my~~ by specMACS. However, using Figure 7 as a guide to the accuracy of the method, the uncertainties appear to be smaller than those from satellite retrievals, which are about 78 % at the pixel level (Grosvenor et al., 2018). We therefore propose that model results can be used to differentiate specMACS observations into clean and polluted
345 conditions, which will need to be verified in future studies.

4 Conclusions

Aerosol-cloud interactions have been the focus of field campaigns and measurement development due to the large associated model uncertainty. Here we used novel observations taken on board the HALO aircraft during the ACRIDICON-CHUVA field campaign to evaluate cloud representation in a numerical model to ~~aid in reducing~~ help reduce this uncertainty. We
350 demonstrated that we can reproduce realistic cloud properties (i.e., cloud droplet effective radius profiles) with a regional

online-coupled chemistry-transport model at convection-permitting scales for the Amazon region during the biomass burning season. ~~As expected by-~~

As expected from theory, the number of CCN at cloud base has a major influence on cloud droplet size and the shape of the ~~effective-radius-vertical-profile~~ vertical profile of cloud droplet effective radius. Increasing CCN leads to decreasing cloud droplet sizes, and we ~~could show that both model and~~ demonstrated that the model and the observations exhibit quantitatively similar behavior. We also observed a saturation effect at high aerosol concentrations in the model (number concentration of CCN larger than 500 cm^{-3} at STP) ~~in the model~~, above which we find no further change in modelled effective droplet size or the shape of the droplet size profile. ~~Our model results are in disagreement with observations of microphysical effects at much higher aerosol loading~~ Observations from previous campaigns (Reid et al., 1999; Andreae et al., 2004) and from the ACRIDICON-CHUVA campaign (Braga et al., 2017b) ~~-This finding casts doubt on the validity of using a setup like ours for regional-scale modeling studies of the cloud albedo effect (Twomey, 1991) of convective clouds for biomass burning situations at high CCN concentrations~~ have demonstrated substantial Twomey effects at much higher aerosol loadings. Additionally, the relation between modelled and observed CDNC is linear and has a slope of 0.3, indicating a considerable underestimation of cloud droplet number concentrations by the model. Although we only tested one microphysics scheme, we demonstrated that a modern, complex parameterization does not imply accurate representation of all cloud microphysical properties and suggest that calculations of the radiative forcing of these phenomena ~~would therefore be unreliable. We conclude that there is a need for further~~ may be biased under polluted conditions like those found during the Amazon biomass burning season.

Evaluation of the parameterization of Freud et al. (2011) proved to be successful in deriving N_a from cloud-side remote sensing data collected by the specMACS instrument. We note a high sensitivity of the method at low N_a and its dependence on an average mixing factor. We were able to gain these insights by applying a previously developed parameterization in a new context. Our study demonstrates that, despite some inherent challenges, existing techniques can be applied for model-measurement comparisons to ~~better understand~~ improve our understanding of model biases.

Code and data availability. Model data, the source code used in the evaluation, as well as all observational data, are available from the authors upon request.

Author contributions. PP ran the simulations and conducted the analysis under the supervision of CK and TZ. PP, TZ and CK wrote the manuscript, with input from BM, MA, DR, RW and MW. MA, CP, MP, UP, DR, RW and MW contributed through fruitful discussions. FE, TKo, TJ, TKI, CM, SM, CP, MP, CV and RW provided measurements essential for this manuscript.

Competing interests. The authors declare no competing interests.

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