Investigating the potential to retrieve cloud droplet number concentration from ship-based measurements of spectral solar radiance during EUREC^{4}A

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Summary: Ship-based cloud remote sensing observations made onboard R/V Meteor during the ElUcidating the RolE of Cloud-Circulation Coupling in ClimAte, EUREC^{4}A, campaign are presented and used to calculate cloud droplet number concentrations. The calculation is based on cloud liquid water path LWP and droplet effective radius r_{eff} retrieved from spectral measurements of transmitted solar radiance. It is shown that measurement uncertainties and retrieval assumptions impact the accuracy of the results. A case study indicates that the retrieval of LWP and r_{eff} is most affected by 3D-radiative effects in case of shallow cumulus and drizzle, which violates the adiabatic theory and plan-parallel geometry on which the radiative transfer simulations of the retrieval are based. Depending on the cloud thickness, the retrieval of r_{eff} might suffers from ambiguity.

These retrieval uncertainties and their implications on the estimated cloud droplet number concentration are investigated by a sensitivity study. The analysis showed that most of the uncertainty is introduced by r_{eff}, whereas LWP contributes significantly to the uncertainty only for thin clouds. Therefore, it is concluded that only selected cloud cases, which do not violate the retrieval assumption, such as stratiform cloud layers, are suited to apply the retrieval approach in further studies.

Zusammenfassung: Fernerkundungsmessungen von Wolken auf dem Forschungsschiff R/V Meteor während der ElUcidating the RolE of Cloud-Circulation Coupling in ClimAte, EUREC^{4}A, Kampagne werden vorgestellt und zur Berechnung der Tröpfchenanzahlkonzentration verwendet. Die Berechnung basiert auf Messungen des Flüssigwasserpfads LWP und dem effektiven Tröpfchenradius r_{eff}, welche aus spektralen Messungen der transmittierten solaren Strahldichte abgeleitet wurden. Es wird gezeigt, dass Messunsicherheiten und Annahmen bei der Ableitung der Wolkeneigenschaften die Genauigkeit der Ergebnisse beeinflussen. Eine Fallstudie zeigt, dass die Ableitung von LWP und r_{eff} am stärksten durch 3-dimensionale Strahlungseffekte von flachen Cumuli und Nieselregen beeinflusst wird. Beides wiederspricht den Idealisierungen von adiabatischen Wolken und einer planparallelen Geometrie, auf denen die Strahlungstransfersimulationen des Verfahrens beruhen. Abhängig von der Wolkendicke kann die Ableitung von r_{eff} zusätzlich durch Mehrdeutigkeiten beeinflusst sein.

In einer Sensitivitätsstudie wurde der Einfluss dieser Unsicherheiten auf die Berechnung von der Tröpfchenanzahlkonzentration untersucht. Die Analyse ergab, dass der größte Teil der Unsicherheit durch Retrievalunsicherheit von r_{eff} verursacht wird, wogegen LWP nur bei dünnen Wolken einen wesentlichen Beitrag leistet. Deshalb wird geschlussfolgert, dass nur ausgewählte Wolkenfälle, die die Annahmen der Methode nicht verletzen, wie z. B. stratiforme Wolkenschichten, geeignet sind, um die Methode in weiteren Studien anzuwenden.

1 Introduction

The cloud droplet number concentration CDNC is a key parameter, which significantly regulates the cloud radiative effects (Wolf et al., 2019). Therefore, estimates of the CDNC from observations are highly desired either from satellite or ground-based instruments (Grosvenor et al., 2018). Ground-based measurements of transmitted solar spectral radiance or irradiance have frequently been used to derive cloud properties of liquid clouds and cirrus (e.g., Brückner et al., 2014; Schäfer et al., 2013; LeBlanc et al., 2015). The spectral measurements are often converted into spectral ratios or normalized radiances to reduce the impact of calibration uncertainties. However, other studies such as, e.g. Fielding et al. (2014, 2015) or Merk et al. (2016), use absolute radiances or a combination of passive and active remote sensing to characterize clouds.

The adiabatic cloud theory provides a basis to calculate the CDNC from observed cloud properties (Brenguier et al., 2000). This approach has previously been applied for active ground-based and passive satellite remote sensing observations, using the retrieved cloud liquid water path LWP, cloud droplet effective radius r_{eff}, and adiabaticity factor. Here, we apply this approach to ship-based observations including passive spectral solar radiance measurements. The observations and uncertainties are presented with respect to their effect on the retrieval of LWP and r_{eff}. The CDNC retrieval theory is then adapted to the transmissivity observations and tested by a sensitivity study and furthermore applied to potential cloud cases.

2 Passive solar cloud remote sensing during EUREC^{4}A

2.1 Spectral solar radiance measurements

The COmpact RAdiation measurements System spectrometer system (CORAS, Brückner et al., 2014) was deployed during EUREC^{4}A in January and February 2020 on the research vessel R/V Meteor. CORAS was configured to measure spectral solar radiance in zenith viewing directions with a sampling frequency of about 6 s. Individual measurements were obtained with an integration time of 300 – 500 ms. The setup consisted of two separate radiance inlets; one of them was installed on the stabilized cloud radar platform correcting for ship motion, the second one was installed on the microwave radiometer which was operated non-stabilized.

The radiance optical inlets feature an opening angle of 2°, which results in a cross track resolution of about 35 m assuming a cloud base at 1 km altitude. The zenith radiance of the stabilized inlet is analyzed by two separated grating spectrometers, which cover the visible (380 – 1000 nm) and near-infrared (900 – 2000 nm) wavelength range. The

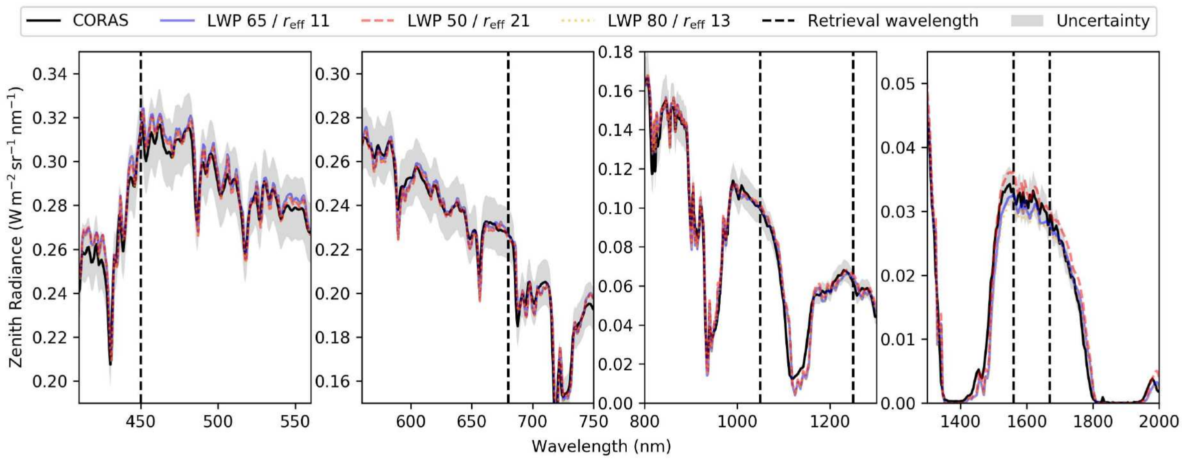

Figure 1: *Spectral zenith radiance observed by CORAS for a homogeneous stratocumulus field on 24 January 2020 around 14:20 UTC compared to radiative transfer simulations using three different combinations of LWP and* r_{eff}. *Uncertainties (gray area) and potential wavelengths used in a cloud and CDNC retrieval (vertical lines) are shown.*

non-stabilized radiance is available only for the near-infrared wavelength range, which is sufficient for most common cloud retrievals. The spectral resolution of the spectrometers was quantified by the full width at half maximum of the individual wavelength and is 2 – 3 nm in the visible and, 15 nm near-infrared range, respectively.

In order to monitor changes in the radiometric calibration of the instrument, transfer calibrations with a portable integrating sphere were regularly performed in the course of the campaign. Significant changes due to the temperature dependence of the system and the deposition of sea salt on the inlets were observed and corrected. The uncertainties of the measured radiance amount up to 8 % and are mostly dominated by the radiometric calibration as discussed in Brückner et al. (2014). For ratios of two different wavelengths the uncertainties are significantly reduced but might be prone to spectral shifts of the radiometric calibration over time, which was indicated by the transfer calibrations to introduce spectral uncertainties of about 1 %. During EUREC^{4}A the near-infrared spectrometer sometimes have been overexposed, in particular at cloud edges, where cloud side effects enhance the solar irradiance. These data were filtered carefully.

An example of the observed radiance is shown in Fig. 1. The radiance spectra obtained from the two spectrometers were merged at 990 nm. Using a LWP of 65 g m^{-2} as derived from the simultaneous microwave radiometer measurements and assuming a cloud droplet effective radius $r_{\text{eff}} = 11\ \mu$m, radiative transfer simulations with the library of radiative transfer (libRadtran, Emde et al., 2016) were performed. The simulated spectral radiances were then compared to the observations. To test the sensitivity of the spectral radiance with respect to the assumed r_{eff} and LWP, two other combinations of LWP and r_{eff} typical for trade wind cumulus were simulated ($LWP = 50$ g m^{-2} and $r_{\text{eff}} = 21\ \mu$m as well as $LWP = 80$ g m^{-2} and $r_{\text{eff}} = 13\ \mu$m, cf. Fig. 1). The simulations show almost identical radiances except for the wavelength around 1600 nm. This illustrates the ambiguity of zenith radiance with respect to their dependence on LWP, cloud particle size and also cloud droplet number concentration.

2.2 Retrieval of liquid water path and effective droplet radius

Different approaches to retrieve LWP and r_{eff} have been applied to estimate the retrieval uncertainties. All retrievals are based on spectral radiance measured by CORAS and look-up tables derived from radiative transfer simulations by libRadtran. In the simulations, clouds were constructed by the sub-adiabatic cloud model with adiabaticity factor of 0.7 and a range of LWP and r_{eff} covering the conditions experienced during EUREC^{4}A. Cloud boundaries were obtained from cloud radar observations on R/V Meteor. The retrievals use either the classic ratio method (Brückner et al., 2014) or a combination of cloud transmissity at eight wavelengths. The eight wavelengths approach includes the six wavelengths used in Brückner et al. (2014) and add measurements at 500 nm and 860 nm wavelength to improve the sensitivity to LWP. To constrain the retrieval, for both approaches, an option prescribing the LWP with observations from the microwave radiometer are tested. The ratio method was applied in two version. One using the original wavelength combination by (Brückner et al., 2014). A second version using an alternative wavelength combination, 500 nm and 860 nm, was tested. As a benchmark, LWP obtained by a passive microwave radiometer (HATPRO) are used.

Figure 2 compares LWP and r_{eff} as retrieved by all different approaches for a time period observed on 2 February 2020. The period was characterized by the presence of shallow cumulus and thin strato-cumulus with low $LWP < 100\,\text{g m}^{-2}$ and a deeper cloud field with LWP reaching up to $400\,\text{g m}^{-2}$ as indicated by the microwave radiometer. The highest LWP of around $400\,\text{g m}^{-2}$ are reached during around 12:15 – 12:30 UTC when the cloud radar still observed hydrometeors at lowest radar range gate (300 m) which likely did not evaporate before reaching the surface. Thus, the LWP from the microwave radometer might be less trustworthy if the radiometer radome got wet. It also has to be noted that the microwave radiometer was not corrected for the cloud-free offset, which typically ranges up to $30\,\text{g m}^{-2}$. However, depending on the retrieval approach, the retrieved LWP significantly differ. Both ratio retrievals overestimate the LWP of the shallow cumulus after 13:00 UTC, while they partly underestimate the LWP of the thick cloud field. The transmissivity approach shows more reasonable results, especially for shallow cumulus periods and in cloud-free conditions. The results of the retrievals, which are constrained by the LWP naturally match the LWP of the microwave radiometer best.

The retrieved r_{eff} was not filtered for spurious results, e.g., overestimations in r_{eff} close to 25 μm, which is linked to cloud edges and cloud-free conditions. In general, the shallow cumulus shows smaller r_{eff} than the deeper cloud field. Comparing the different retrieval approaches indicates that the unconstrained methods, except the one using 500 nm and 860 nm, result in higher r_{eff} than the methods that are constrained by the LWP from the microwave radiometer. These results demonstrate the sensitivity of the retrieval with respect to the choice of wavelength. This is partly linked to spectral measurement uncertainties. In addition, the vertical weighting functions of the transmissivity retrievals, and therefore, the retrieved r_{eff} depend on the location of the chosen wavelengths in the water absorption band.

This sensitivity study suggests that although nadir radiances measured by CORAS are ambiguous as a function of cloud LWP, a high information content and sensitivity is found with respect to r_{eff}, when an estimate of cloud LWP is given and used to constrain the retrieval. LWP estimates, for example provided by the microwave radiometer, allows

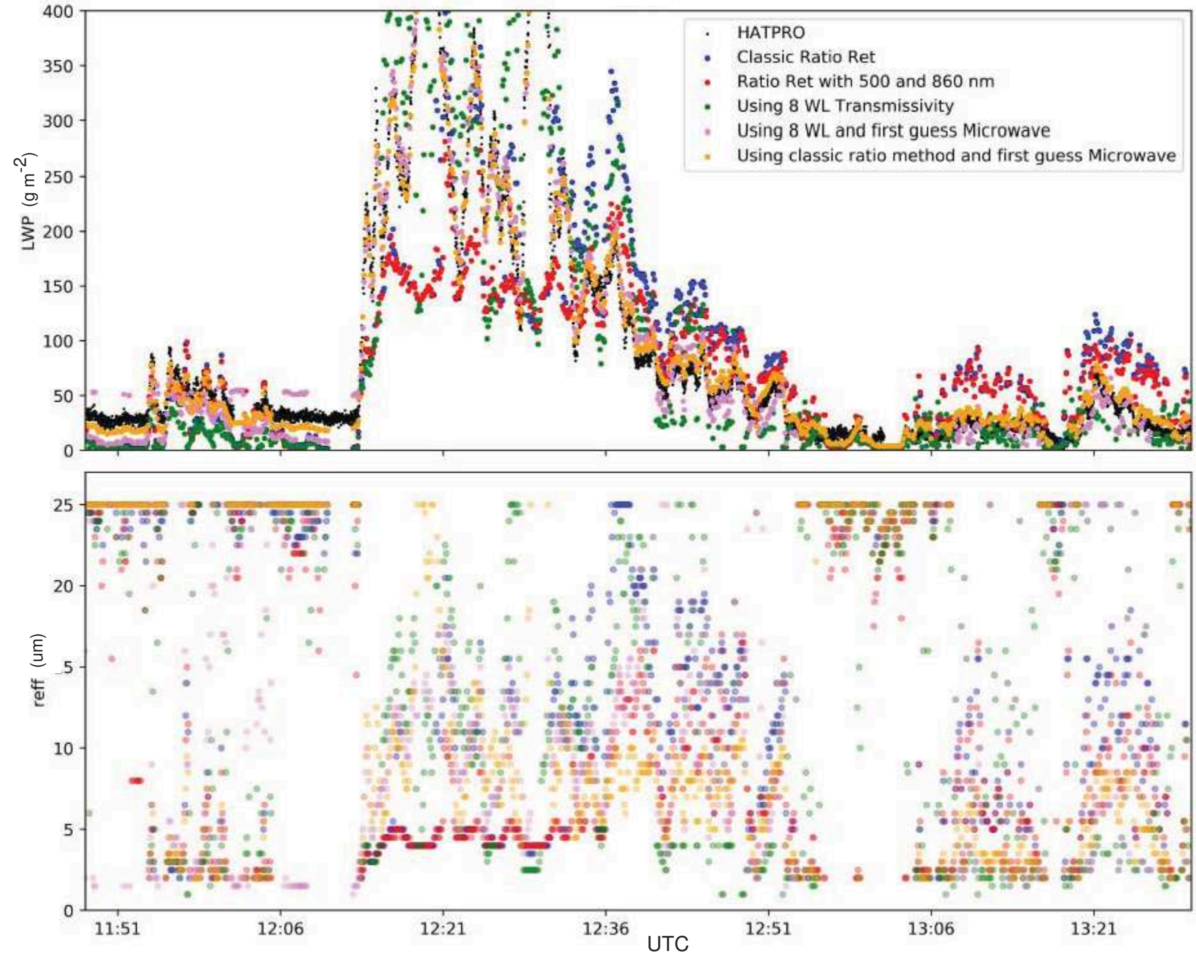

Figure 2: *Time series of LWP and r_{eff} retrieved by different methods using spectral radiance from CORAS measured onboard R/V Meteor on 2 February 2020. For LWP, the passive microwave radiometer measurements (HATPRO) are included.*

pre-separating between low and high LWP. However, the LWP threshold is not fixed and, based on thresholds, ranges between $20\,\mathrm{g\,m^{-2}}$ and $60\,\mathrm{g\,m^{-2}}$ depending on solar zenith angle and r_{eff}. Unfortunately, shallow trade wind cumulus as observed frequently during EUREC^{4}A often falls into this LWP ambiguity range.

3 CDNC Retrieval from ground based transmissivity measurements

3.1 Theory

For retrieving the cloud droplet number concentration N in stratiform clouds from satellite remote sensing, Brenguier et al. (2000) and Wood (2006) proposed a relation, which links N to the cloud liquid water path LWP and the cloud effective radius $\tilde{r}_{\mathrm{eff}}$ based on reflectivity retrievals:

$$N = \frac{3 \cdot \sqrt{2}}{4 \cdot \pi \cdot \rho_{\mathrm{w}}} \cdot \sqrt{f_{\mathrm{ad}} \cdot \Gamma_{\mathrm{ad}}} \cdot \frac{\sqrt{LWP}}{\tilde{r}_{\mathrm{eff}}^{3}} \tag{1}$$

with ρ_w the density of liquid water, f_{ad} the degree of adiabaticity, and Γ_{ad} the adiabatic rate of increase of liquid water content with respect to height.

In this relation $\tilde{r}_{eff}$ should not be mistaken with r_{eff}, the effective cloud droplet radius retrieved from transmissivity measurements as used in Section 2. Here, $\tilde{r}_{eff}$ represents the cloud droplet radius at cloud top $\tilde{r}_{eff} = r(h_{top})$, to consider the high sensitivity of satellite retrievals to cloud top layers. Vertical weighting functions of reflectivity retrievals are presented by Platnick (2000) and were shown to not penetrate deep into the clouds. Platnick (2000) also showed that the weighting functions for transmissivity measurements are more uniform and weights the profile of particle sizes equally. Therefore, retrievals based on transmissivity provide an estimate of the mean droplet radius averaged over the entire $r_{eff} = \overline{r}$.

The profile of cloud particle radius in stratiform clouds can be calculated by:

$$r(h) = B \cdot h^{1/3} \quad \text{with} \quad B = \left(\frac{3}{4 \cdot \pi \cdot \rho_w} \cdot \frac{\Gamma_{ad}}{N} \right)^{1/3} \tag{2}$$

From that, $\overline{r}$ can be derived by integration of $r(h)$:

$$r_{eff} = \overline{r} = \frac{1}{h} \cdot \int_0^{h_{top}} B \cdot h^{1/3} \mathrm{d}h, \tag{3}$$

$$r_{eff} = B \cdot \frac{3}{4} \cdot h_{top}^{1/3}, \tag{4}$$

$$r_{eff} = \frac{3}{4} \cdot \tilde{r}_{eff}. \tag{5}$$

This conversion of the reflectivity-based cloud effective radius into the transmissivity-based cloud droplet radius is used to convert Eq. 1 into a relation for calculating N from transmissivity measurements:

$$N = \left(\frac{3}{4}\right)^4 \cdot \frac{\sqrt{2}}{\pi \cdot \rho_w} \cdot \sqrt{f_{ad} \cdot \Gamma_{ad}} \cdot \frac{\sqrt{LWP}}{r_{eff}^3}. \tag{6}$$

To calculate N, measurements of LWP, r_{eff} and assumptions on the adiabaticity factor f_{ad} are required. As discussed by, e.g., Wolf et al. (2019) for reflectivity-based retrieval and Merk et al. (2016) for ground-based observations, these three parameters can be obtained by different combinations of remote sensing measurements. For example, passive spectral solar and microwave radiometer measurements, and active lidar and radar observations were combined. Here, LWP from the microwave radiometer is applied. r_{eff} is obtained from the constrained ratio retrieval of CORAS. Radar and lidar (ceilometer) cloud boundaries are used to calculate the cloud height H, which in combination with LWP is used to derive f_{ad}:

$$\Gamma_{obs} = f_{ad} \cdot \Gamma_{ad} = \frac{2 \cdot LWP}{H^2}. \tag{7}$$

This reduces Eq. 6 to:

$$N = \left(\frac{3}{4}\right)^4 \cdot \frac{2}{\pi \cdot \rho_w \cdot H} \cdot \frac{LWP}{r_{eff}^3}. \tag{8}$$

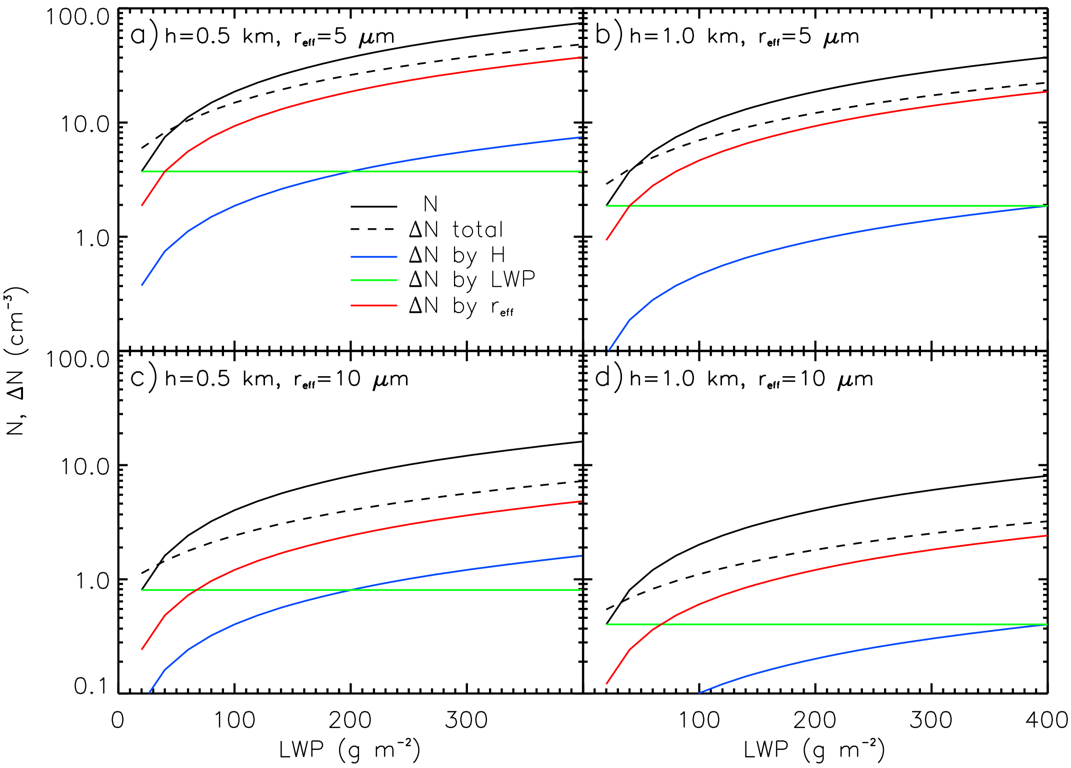

Figure 3: *Cloud droplet number concentration N and uncertainty ΔN calculated for different combinations of H, r_{eff}, and LWP.*

3.2 Sensitivity study

To analyse the potential of a CDNC retrieval and the impact of the measurement uncertainties on the retrieved cloud properties, a sensitivity study based on Eq. 8 was performed. For different combinations of H, r_{eff}, and LWP, representing the typical range of sub-tropical boundary layer clouds, the cloud droplet number concentration N was calculated. The uncertainty ΔN was estimated by Gaussian uncertainty propagation assuming that cloud altitude is measured with an uncertainty of $\Delta H = 50\,\mathrm{m}$, cloud particle size is retrieved with uncertainty of $\Delta r_{\mathrm{eff}} = 1\,\mu\mathrm{m}$, and LWP is derived with an uncertainty of $\Delta LWP = 20\,\mathrm{g\,m^{-2}}$. To estimate, which quantity contributes most to the total uncertainty ΔN, also separate uncertainties by each of the measured cloud properties were calculated.

Figure 3 shows N and ΔN for four clouds in dependence of LWP. In all cases, the total uncertainty may exceed 100 % for cloud with small LWP and low N, respectively. For higher LWP and N, absolute uncertainties increase but relatively uncertainties decrease to about 50 %. For most clouds, the strongest impact on the total uncertainty results from uncertainties of r_{eff}. Only for small LWP, the uncertainty of LWP is dominant. While LWP contributes linear to the calculation of N, the dependence on r_{eff} is cubic (compare Eq. 8) and, therefore, is more prone to uncertainties. This holds especially for clouds

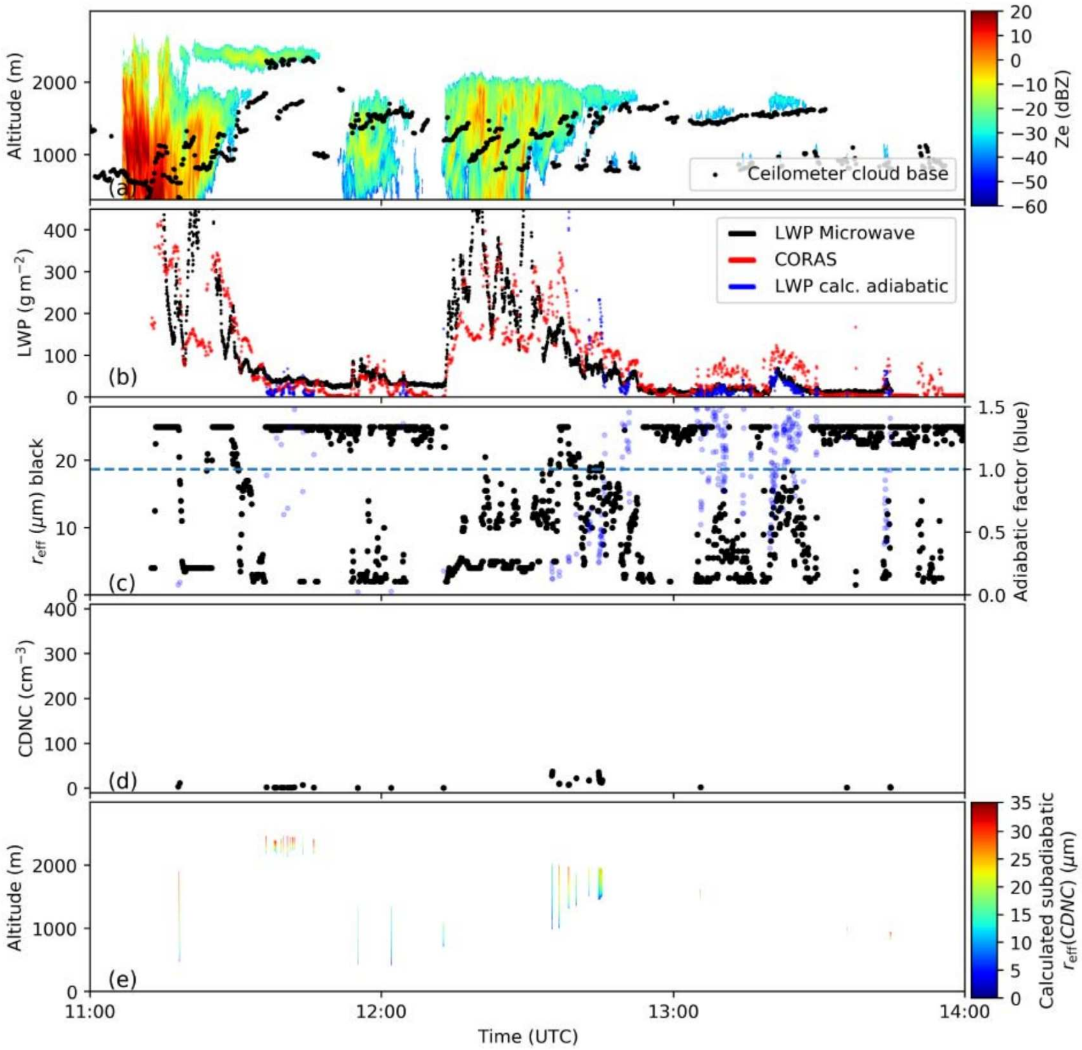

Figure 4: *Time series of cloud radar reflectivity and ceilometer cloud base altitude (a), LWP retrieved from CORAS and microwave radiometer (b), retrieved* $r_{\rm eff}$ *and* $f_{\rm ad}$ *(c), retrieved (g m-2) (d) and calculated profile of cloud droplet effective radius (e) for 2 February 2020,* 11 – 14 *UTC.*

with small droplets (i.e. small $r_{\rm eff}$) as indicated when comparing the different cases in Fig. 3.

These calculations suggest that the accuracy of the retrieval of N strongly depends on a quantitative estimation of the cloud droplet effective radius. While an accurate retrieval of LWP is important only for thin clouds, the geometrical thickness of the cloud is less critical. This indicates that the accuracy of the retrieval of cloud properties as discussed in Section 2.2 may limit the potential to retrieve N. Especially the uncertainty of $r_{\rm eff}$ in case of broken clouds, which are affected by 3D radiative effects, will not allow for deriving a reasonable N.

4 Potential cloud cases

The retrieval approach introduced above is based on several assumptions, which in reality might be violated. Horizontal homogeneous cloud layers are assumed in the plane parallel

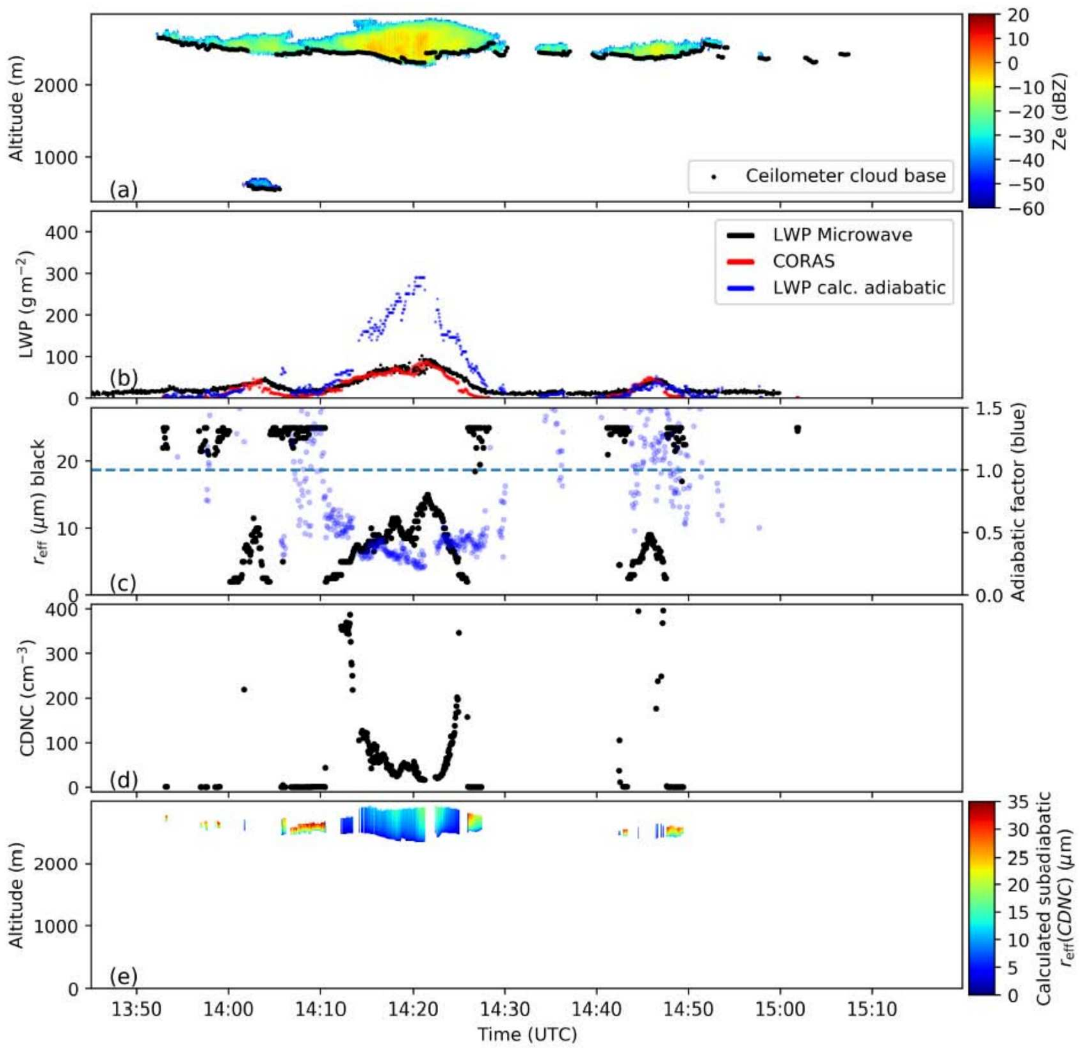

Figure 5: *Time series of radar reflectivity and ceilometer cloud base altitude (a), LWP retrieved from CORAS and microwave radiometer (b), retrieved r_{eff} and f_{ad} (c), retrieved CDNC (d) and calculated profile of cloud droplet effective radius (e) for 24 January 2020.*

radiative transfer simulations of the cloud retrieval of r_{eff}. As outlined by Fielding et al. (2014) for heterogeneous cumulus, three-dimensional (3D) radiative transfer is required to account for 3D radiative effects. Depending on the solar zenith angle and the aspect ratios of cloud height to width, the photon path length through the cloud side does change and may affect the absorption by cloud droplets imprinted in the zenith radiance. In this case, one-dimensional plane-parallel simulations inadequately represent the transmitted radiation.

The CDNC retrieval assumes the adiabatic cloud model which is not valid in the presence of drizzle. Therefore, precipitating clouds have to be filtered from the analysis. The cloud base height is often difficult do determine in case of full signal attenuation of the lidar observations, as is cloud top altitude from radar reflectivity. Due to these measurement uncertainties of the cloud boundaries, also the adiabatic factor can be biased and become non-physically. Often cases with $f_{\text{ad}} > 1$ were observed for which a calculation of N using the adiabatic theory is not possible (Merk et al., 2016).

These constrains limit the number of potential cloud cases from EUREC^{4}A. Here two cases with stratiform clouds are presented. Figure 4 shows a time series of all relevant cloud parameters measured on 2 February 2020. The three hour period is characterized by a sequence of warm convection, which partly formed precipitation. In the core of the convective cell, LWP up to 400 g m^{-2} were observed. Due to the drizzle, LWP derived from CORAS and the microwave radiometer differ significantly and the retrieved r_{eff} become non-reliable. Additionally, for this cloud, r_{eff} varies around the ambiguity range of the transmissicity retrieval, which is obvious by the absence of any solution between 7-11 μm. For smaller trade wind cumuli at low altitudes, 3D-radiative effects biased the retrieval by CORAS. Therefore, N was calculated only for short periods of the time series, e.g., the stratiform cloud layer in about 2200 m altitude observed around 11:30 UTC.

A second case, where only stratiform clouds were observed, is presented in Figure 5. For this almost ideal scenario, LWP retrieved by CORAS and the microwave radiometer agree well. The retrieved r_{eff} ranged between 4 μm in thinner cloud parts and 15 μm in the thicker parts of the cloud layer. In this section of the cloud the retrieved N become more reliable and range around 50 cm^{-3}. However, also here the retrieval of N seems to fail when the cloud layer becomes thin and LWP decreases below 30 g m^{-2}, which agrees with the sensitivity study presented in Section 3.2.

5 Summary and conclusions

Spectral solar irradiance measurements on board of R/V Meteor obtained during the EUREC^{4}A campaign are presented and used in combination with active and passive microwave remote sensing to estimate the cloud droplet number concentration. The ship-based measurements turned out to be challenging due to a non-stable radiometric calibration caused by temperature effects and deposition of sea salt, which results in a higher measurement uncertainty compared to ground-based observations (Brückner et al., 2014). The measurement uncertainty impacts the retrieval of cloud properties using transmissivity based retrieval such as presented by Brückner et al. (2014) or LeBlanc et al. (2015). Therefore, different approaches to derive LWP and r_{eff} were tested. The case study indicated that retrieval uncertainties can result from 3D-radiative effects in case of shallow cumulus and drizzle, which violates the adiabatic theory on which the radiative transfer simulations of the retrieval are based. Due to the range of LWP present during the measurements, which falls into the ambiguity range of the retrieval, the retrieval of r_{eff} may partly fail. However, synergistic approaches that combine active and passive remote sensing similar to Fielding et al. (2015) are possible.

An approach to calculate the cloud droplet number concentration N from ground-based observations using r_{eff} retrieved from the spectral irradiance measurements is presented. The approach is based on common reflectivity-based retrieval of N (e.g., Merk et al., 2016; Wolf et al., 2019) and was converted for the r_{eff} observed by transmissivity-based retrieval. A sensitivity study showed that the uncertainty of N is dominated by the uncertainty of r_{eff}, This indicates that the limitation of the retrieval of LWP and r_{eff} can significantly bias the retrieval of N. Therefore, data need to be filtered for 3D-radiative effects, the ambiguity range of the retrieval and drizzle. This was demonstrated by two cases. Cloud sections, which can potentially be used to retrieve N were identified. These sections are mostly limited to non-precipitating stratiform clouds. As discussed by e.g., Fielding et al. (2015) and Merk et al. (2016), further analysis is required to refine the retrieval approach and select the most suited approach to combine the different passive and active measurements.

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