Key Points:

- Satellite retrieval of cloud condensation nuclei in shallow marine boundary layer clouds is developed and validated
- Best validation was achieved for the cores of coupled clouds while accounting for their adiabatic fraction
- Most of the decoupled clouds had much lower CCN compared to the underlying surface

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Satellite Retrieval of Cloud Condensation Nuclei Concentrations in Marine Stratocumulus by Using Clouds as CCN Chambers

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Abstract A new methodology for the satellite retrieval of cloud condensation nuclei (CCN) in shallow marine boundary layer clouds is developed and validated in this study. The methodology is based on retrieving cloud base drop concentrations (N_d) and updrafts (W_b), which are used for calculating the supersaturation (S). The N_d is then defined as the CCN at that S . The accuracy of the satellite retrievals was validated against ship-borne measurements of CCN done in recent campaigns in the Southern Oceans (ACE-SPACE, MARCUS, & PEGASO [2015–2018]) and in the subtropics (MAGIC [2012–2013]). The satellite-retrieved N_d and S at cloud base were validated against the measured CCN at sea surface. The main findings show that (a) coupled clouds have good agreement between satellite retrievals and ship measurements of CCN; (b) the best agreement is achieved when using the brightest 10% of the clouds and accounting for their adiabatic fraction; (c) most of the decoupled clouds had much lower CCN than were present at the underlying surface. This means that most CCN in the coupled clouds originate from the surface and not from the free troposphere. This study validates the satellite retrievals of CCN and allows us to further quantify the relationships between CCN and cloud microphysical properties.

Plain Language Summary Invisibly small particles serve as sites for condensation of cloud droplets; they are called cloud condensation nuclei (CCN). In this study we present a newly developed satellite methodology to quantify these CCN particles which are the basis for the development of low-level oceanic clouds. This method was validated against near-surface measurements of particles from ships in the Southern Oceans, close to Antarctica and in the east mid-Pacific Ocean between California and Hawaii. The results show that this method works best for the brightest clouds that are coupled with the surface, that is, forming by air that originates at the sea surface. The rest of the clouds are formed with mostly lower drop concentration due to smaller CCN concentrations than at the surface. This indicates that most of the CCN particles in the coupled clouds originate from the sea surface and not from above the clouds. This validation of the satellite retrievals allows us to further quantify the relationships between particles in the atmosphere and cloud microphysical properties.

1. Background

Radiative forcing, caused by aerosol-cloud interactions (ACI), has the largest uncertainty and remains the least well-understood anthropogenic contribution to climate change. Much of this uncertainty is driven by the poor quantification of the aerosol effects in the pristine pre-industrial atmosphere, such as in the Southern Oceans (SO), which serves as the reference state for anthropogenic cloud radiative forcing (Carslaw et al., 2013).

The optical signal of aerosols has been used as a proxy for CCN. The most commonly used signal is the aerosol optical depth (AOD) (Feingold et al., 2001; Quaas et al., 2009). An improved proxy is the aerosol index, which is the product of AOD and the Ångström coefficient (Bréon et al., 2002; Gryspeerd et al., 2017; Lohmann & Lesins, 2002; Nakajima et al., 2001), because the Ångström coefficient provides a measure of the size of the particles. A recent additional improvement was achieved by using the polarimetric signal of the aerosols to obtain information on their size distribution and concentrations (Hasekamp et al., 2019). However, all these proxies still suffer from uncertainty with respect to matching between the heights of the clouds and the number concentration of the aerosols that produce the signal (Stier, 2016). In addition,

no aerosol signal is obtained in scenes with nearly full cloud cover. This undermines the possibility to relate large cloud cover to aerosols, while in some situations aerosols are responsible for the large cloud cover because they play a key role in the transitions between full and broken cloud cover of marine stratocumulus (Goren et al., 2019; Goren & Rosenfeld, 2012; Rosenfeld et al., 2006).

The accuracy of the aerosol optical signal is poorest at very small concentrations. This is important because the cloud cover of shallow marine boundary layer (MBL) clouds is strongly linked to N_d and starts to decrease substantially at $N_d < \sim 80 \text{ cm}^{-3}$ (Rosenfeld et al., 2019). At such low concentrations aerosols do not produce a useful optical signal for any of the proxies mentioned above. This occurs at $CCN(S = 0.4\%) < 130 \text{ cm}^{-3}$ (Shinozuka et al., 2015; Zamora et al., 2017). These concentrations occur at the aerosol-limited regime (Chen et al., 2016), which means that the concentration values of N_d and CCN are close to each other. Most of the ocean areas have $N_d < 130 \text{ cm}^{-3}$ (Grosvenor et al., 2018) which means that the optical aerosol signal is inadequate, even under the most sophisticated processing of its polarimetric signal. Therefore, we have to rely on other methods for quantifying CCN . In summary, there is no methodology that is based on aerosol optical signals that can approximate CCN and its activation S in a useful way, especially at low concentrations, where the accuracy matters most.

This situation motivated an alternative approach which infers the CCN and S based on their impacts on satellite retrieved N_d and updrafts near cloud base. Rosenfeld et al. (2016) showed that the CCN can be retrieved from space by using clouds as natural CCN chambers. This methodology is reviewed here with some detail for two reasons: (1) it sets a precedent for the concept, and (2) it serves to differentiate the new methodology presented in this study for marine shallow clouds from the previously published methodology for convective clouds.

The N_d is determined by W_b and by the $CCN(S)$, where S is the maximum supersaturation near cloud base and W_b is the cloud base updraft. The relationship between S , N_d , and W_b is given by

$$S = C_{(T,P)} W_b^{3/4} N_d^{-0.5} \quad (1)$$

where C is a coefficient that depends on cloud base temperature and pressure (Pinsky et al., 2012).

This method was applied to convective clouds, where N_d was calculated based on the satellite-retrieved $T-r_e$ relationship, where T and r_e are cloud temperature and droplet effective radius. The r_e increases with altitude nearly as in an adiabatic cloud (Brenguier et al., 2000). Based on that, the adiabatic N_d can be calculated at different altitudes of the cloud using the calculated adiabatic liquid water content (LWC) and r_e . The cloud base N_d is approximated by the adiabatic N_d as calculated by Equation 2 (Freud et al., 2011):

$$N_d = \frac{\alpha^3 LWC}{r_e^3} \quad (2)$$

$$\alpha = \frac{62.03 r_e}{r_v} \quad (3)$$

where r_v is the cloud drop mean volume radius assuming monodispersed cloud droplet sizes. The relation between r_e and r_v used here is $r_e = 1.08 r_v$ according to analyzed non-precipitating clouds by (Freud & Rosenfeld, 2012).

The W_b (m s^{-1}) is calculated based on the existence of linear relationships between W_b and cloud base height (H_b) (m) relationship found by Zheng & Rosenfeld (2015):

$$W_b = 0.0009 H_b \quad (4)$$

This methodology and its validation is fully described in Rosenfeld et al. (2016). Validation against ground-based CCN measurements (Figure 1) shows an accuracy of $\pm 30\%$.

The calculation of the adiabatic N_d is based on the retrieved $T-r_e$ relationship above cloud base. A useful $T-r_e$ for that purpose requires clouds with a vertical temperature span of at least 5°C . However, marine stratocumulus clouds are usually shallower than that, so retrieving CCN with the methodology of Rosenfeld et al. (2016) is not applicable to them. Therefore, we develop and present here a new methodology for

Figure 1. The relationship between satellite-retrieved CCN at cloud base, and the ground-based measurements of CCN at the same S . The validation data are collected from Oklahoma (SGP), the Amazon (MAO and ATTO), and a ship over the Northeast Pacific (MAGIC). The location is denoted by the marker shape, and S is shown by the color. Reproduced from Rosenfeld et al. (2016).

retrieving CCN in shallow marine clouds. The retrieval is still based on the same principle expressed in Equation 1. The N_d is calculated based on cloud top r_e and optical depth. The W_b is calculated based on cloud top radiative cooling rate ($CTRC$). The retrieved CCN are validated against measurements from recent cruise campaigns. The description of the methodology is provided in the next section.

2. Satellite Retrieval of CCN in Marine Stratocumulus Clouds

2.1. Retrieving Cloud Base Drop Concentrations

The retrieval of N_d and W_b for marine stratocumulus clouds is based on a different methodology than the above-mentioned one for convective clouds. The retrieval is based on the Moderate Resolution Imaging Spectroradiometer (MODIS) L2 cloud products (collection 6.0) (Platnick et al., 2017) on board the Terra and Aqua satellites. We used the r_e and the cloud optical depth (τ) products. Having r_e and τ allows us to calculate the N_d by Equation 5 (Szczodrak et al., 2001):

$$N_d = \frac{\sqrt{5}}{2\pi k} \left(\frac{C_w \cdot \tau_c}{Q_{ext} \rho_w r_e^5} \right)^{0.5} \cdot AF^{0.5} \quad (5)$$

where C_w is the condensation rate of cloud water in a rising adiabatic cloud parcel ($m^{-3} m^{-1}$), Q_{ext} is the extinction efficiency factor, which represents the ratio between the extinction and the geometric cross section of a given droplet, approximated to the value of 2, $\rho_w = 1,000 \text{ kg m}^{-3}$ is the density of

liquid water, k relates r_v to r_e ($k = \left(\frac{r_v}{r_e} \right)^3 = 0.8$), and AF is the adiabatic fraction. The AF is the ratio between the actual cloud water content with respect to the case where all condensed water in the rising cloud parcel is conserved. We initially assumed fully adiabatic clouds ($AF = 1$) (Grosvenor et al., 2018; Grosvenor & Wood, 2014; Szczodrak et al., 2001).

The retrieval of N_d is fraught with inaccuracies which are caused by deviations from the retrieval assumptions. Because the clouds are assumed to be horizontally homogeneous with flat tops, the errors are dominated by inhomogeneity of the clouds and partial pixel filling (Grosvenor et al., 2018). These errors can be minimized when selecting the cores of the clouds as defined by the brightest (largest τ) 10% of the clouds. The difference between N_d retrieved from the 10% brightest pixels and fully surrounded pixels at the same field of clouds is less than 5% (Zhu et al., 2018). The top 10% (90th percentile) of the cloud optical depth represent the cores of the clouds, where updrafts are strongest and fed most directly from the air and CCN below cloud base, thus best representing the surface CCN in well a mixed boundary layer (Figure 2).

Figure 2. Cloud selection for retrieved N_d which best represents the CCN effects. The brightest part of the cloud, the top 10%, is the convective core of the cloud, which has largest updrafts and adiabatic fraction, thus best represents the effects of subcloud CCN and complies most closely with the retrieval assumptions. Reproduced from Zhu et al. (2018).

Figure 3. The dependence of cloud base updraft on cloud base height (a) and cloud top radiative cooling rate (b). The points in the left panel are based on theoretical calculations. The curve is parameterized in Equation 7. The dashed red lines represent the observed relationships from ship-borne measurements in Zheng and Rosenfeld (2015). The left panel is reproduced from Zheng (2019). The right panel is reproduced from Zheng et al. (2016).

2.2. Retrieving Cloud Base Updrafts

The theoretical framework for the retrieval of W_b was laid out by Zheng (2019) and Zheng et al. (2020), after finding the linear relationships between cloud base height and W_b (Zheng & Rosenfeld, 2015). Marine stratocumulus clouds are propelled mainly by cloud top radiative cooling, causing an inverse convection. In a steady state, a stronger CTTC would require stronger compensating surface heat fluxes, which can be in the form of mainly latent heat over the ocean. Therefore, a stronger CTTC would require more evaporation, which then, for a given surface wind speed, requires lower relative humidity near the surface and respectively a higher lifting condensation level. It follows that a larger CTTC would result in a higher cloud base height following a nearly linear relationship. The latent heat flux is transferred upward by convective updrafts which also increase nearly linearly with CTTC (Zheng, 2019; Zheng et al., 2018). The linear relationships between W_b and CTTC were quantified as

$$W_b = -0.44 \cdot \text{CTTC} + 22.30 \pm 13 \quad (6)$$

where the W_b and CTTC have units of cm s^{-1} and W m^{-2} , respectively (Zheng et al., 2016). The maximum possible negative CTTC sets an upper limit on H_b of coupled clouds near 1,000 m.

Based on these considerations, Zheng (2019) derived a theoretical relationship between W_b and H_b , shown in Figure 3. This theoretical relationship overlaps with the observed relationship for $H_b > 400$ m, as quantified in Equation 4. Calculated W_b based on the actual H_b would overestimate the actual W_b for decoupled clouds, because the actual H_b value is larger than it would be if the clouds were coupled. Therefore, W_b is taken as the minimum of the values calculate based on CTTC and H_b .

The relationship between W_b and H_b in Figure 3a is given by

$$W_b = 0.0370888 + 1.86128H_b - 2.76877H_b^2 + 2.53356H_b^3 - 0.858022H_b^4 \quad (7)$$

where W_b is in m s^{-1} and H_b is in km.

The retrieval of H_b is based on the assumption that the clouds are convectively coupled with the surface and that they have adiabatic cores. Under such assumptions an adiabatic parcel model was used for calculating H_b as a function of cloud liquid water path, cloud top temperature, and sea surface temperature. This was done for the cloud cores, that is, highest 10 percentile of cloud optical depth (Goren et al., 2018; Rosenfeld et al., 2019). The same procedure was used also for calculating cloud geometrical thickness (CGT). The implications of deviations from these assumptions are discussed below.

Table 1
All the Research Cruises and Aircraft Campaigns Used in This Study

Campaign	Duration	Course	Platform type	Website and reference
MAGIC	10/2012–09/2013	Los Angeles, California to Honolulu, Hawaii	Ship	https://tinyurl.com/r8zxs2 (Zhou et al., 2015)
PEGASO	01/2015–02/2015	Ushuaia, Argentina to Anvers Island via South Georgia Island and South Orkney Islands	Ship	https://tinyurl.com/yygn5t6x (Simó & Dall'osto, 2015)
ACE-SPACE	12/2016–03/2017	Cape Town, South Africa, through the Indian Ocean to Hobart, Australia, via the Pacific Ocean to Punta Arenas, Chile, and through the Atlantic Ocean back to Cape Town	Ship	https://tinyurl.com/w6fpbab (Schmale et al., 2019)
MARCUS	10/2017–04/2018	Hobart, Australia to Antarctica	Ship	https://tinyurl.com/rzvw6xs (G. McFarquhar & Greg, 2016)
SOCRATES	01–02/2018	Hobart, Australia southward	Aircraft	https://tinyurl.com/r35ayww (G. M. McFarquhar et al., 2014)
CIRPAS	2005–2016	Off the coast of California	Aircraft	https://tinyurl.com/vvoq2sw (Braun et al., 2018)

The cloud base S was calculated based on the satellite retrieved W_b and N_d . The W_b can be retrieved based on either $CTRC$ (Equation 6) or H_b (Equation 7). By definition, CCN at the retrieved S are equal to the retrieved N_d .

3. Ship-Borne Validation

3.1. The Validation Campaigns

The satellite retrievals were validated against ship-borne measurements of CCN and cloud base heights. There were three cruises in the Southern Oceans and one cruise in the northeast Pacific, shown in Table 1. All the cruises measured CCN supersaturation spectra. The lifting condensation level (LCL) was calculated based on the ship-measured surface temperature and dew point. The actual cloud base height over the ships was measured by a lidar or a ceilometer. In addition, sea surface temperature was measured.

3.2. The Validation Methodology

Cloud properties over or near the ships were retrieved from regions which were manually selected as polygons containing single layer boundary layer water clouds. Only coupled clouds, that is, clouds feeding on air and aerosols originating from the surface, could be used for validation against the CCN measurements at the sea surface. A decoupled cloud is not fed from near-surface air, and therefore the ship-borne measurements do not necessarily represent the CCN in the air ingested by the cloud. Decoupling is identified when (1) the absolute difference between the ship-measured LCL and satellite-retrieved cloud base height is over 200 m. A common cause for overestimate of H_b by satellite is due to slightly elevated cloud layers that are convectively decoupled from lower cloud layers which may be coupled to the surface; (2) when the cloud base height measured by the ship's ceilometer was higher than LCL by more than 200 m; (3) when the cloud base absolute temperature difference from satellite and ship is larger than $\pm 1.8^\circ\text{C}$, which is practically the same as the first condition; (4) when the cloud base height is over 1 km, which is the maximum possible height for coupled clouds over oceans (Zheng et al., 2018); and (5) when cases are characterized by warm advection. The warm advection was inferred based on SST and wind properties from the National Atmospheric Administration (NOAA) and National Centers for Environmental Prediction (NCEP) reanalysis. When an air parcel over higher SST is pushed toward a region of colder SST , it moves above the cooler surface air and thus creates a stable state or a decoupled state, in which the measured air parcels from the ship and the satellite are not the same. A case was defined as decoupled due to warm advection, when the SST given by a back trajectory of 3 hr was warmer than at the ship location by at least 0.6°C .

The CCN counters on the ships operated with cycles of several fixed values of S , producing time series of CCN supersaturation spectra. The spectra between 2 hr prior and following a satellite overpass were used (Figure 4), assuming the same air mass over the ship for the entire time. To minimize the noise, CCN data were taken only for the second half of each new S level, guaranteeing that S had mostly stabilized.

Figure 4. Measured *CCN* supersaturation spectra. The blue line is the *S* level. The red bars are the *CCN* measured when the device temperature was not stable. The green bars are the *CCN* stable measurements that were used. The spectra were used within the 4-hr time windows, two before and after the satellite overpass. The stable measurements of each *S* level were used to produce a *CCN* spectrum of the sampled air parcel.

A *CCN* supersaturation spectrum was produced for each case using a single-error-function (erf) fit model, represented by the function:

$$N_{CCN}(S) = \frac{a_1}{2} + \frac{a_1}{2} \cdot \text{erf} \left(\frac{\ln \left(\frac{S}{S_1} \right)}{w_1} \right) \quad (8)$$

where a_1 is a measurement-dependent coefficient, S_1 is the supersaturation at which half of the maximum activated fraction (MAF) of the aerosol particles acts as *CCN*, and w_1 is the width of the erf fit (Pöhlker et al., 2018). In some cases, the satellite-retrieved *S* was below the lowest *S* in the instrument measured spectrum; thus the erf fit was not useful for the validation of those cases.

For a given cloud base N_d and W_b , *S* is smaller for higher temperature due to the increased molecular diffusion rates (Johnson, 1980). The temperature at the center of the *CCN* counter (around $25 \pm 5^\circ\text{C}$) is much higher than at cloud base, mainly over the Southern Oceans (-3°C on average). This requires a correction, where *S* for cloud base is calculated using Equation 1 with the *CCN* counter temperature instead of cloud base temperature. This correction is defined here as the ΔT correction. It is necessary because the *CCN* measurements were conducted by the Droplet Measurement Technology continuous flow *CCN* counter (Roberts & Nenes, 2005). The *S* in that instrument is determined by the flow rate and cooling rate of the air sample (Moore & Nenes, 2009), as it does at cloud

base. For a given flow rate and temperature gradient in the walls of the *CCN* chamber, the supersaturation is determined by the molecular diffusion rate, just as at cloud base (Johnson, 1980). This is not the case for a static *CCN* chamber, where molecular diffusion rates do not affect the outcome. Therefore, the relationships between *S* and N_d in the *CCN* counter and at cloud base are subject to the same dependence on temperature, and the temperature difference between them has to be accounted for. Applying the *CCN* chamber temperature to the calculation of *S* at the colder cloud base (Equation 1) would result in a smaller than actual *S* there but will match the *S* within the *CCN* chamber which produces the same N_d for the same aerosols as at cloud base.

An additional correction is made by adjusting the cloud base N_d to the difference between cloud base and surface air densities.

The surface-adjusted satellite retrieved N_d and *S* are compared to the in situ measured *CCN* supersaturation spectra according to the scheme shown in Figure 5, as was first done in Rosenfeld et al. (2014).

3.3. The Validation Results

Out of a total of 308 overpasses by the satellites, 185 had suitable clouds within an acceptable distance (<150 km) from the ship. The closest clouds to the ship within that range were analyzed. Cases with solar zenith angle larger than 65° were rejected. Only 16 cases passed all the quality and selection criteria and could be compared with the ship measurements. A large fraction of the cases was rejected because satellite-retrieved H_b was higher by more than 200 m than the ship-based measured H_b . This happens when the clouds lack vertical continuity within the scene. It implies that the cloud tops are decoupled, while the cloud bases may be coupled with the surface. In such cases cloud top properties do not reflect the aerosols effect near the surface, so they should be rejected from the comparison with surface *CCN*. The location of the scenes that passed the selection criteria is shown in Figure 6.

As shown in Figure 7b, the satellite and in situ measured *CCN* are well correlated for coupled cases, but there is a systematic underestimation (slope of 0.67) in the retrieved N_d from the satellite with respect to the in situ measured *CCN* at the cloud base *S*.

Figure 5. The matching of satellite-retrieved N_d and S with surface-measured CCN supersaturation spectra. The vertical line shows the value of the satellite-retrieved S on the abscissa. The interception of that line with the curve of the CCN spectrum shows the surface-measured CCN at the satellite-retrieved S . The satellite retrieved CCN (which is the satellite retrieved N_d) at the same S is shown by the horizontal red line. Panel (a) shows a good match, whereas panel (b) shows a large underestimation of the CCN number concentration by the satellite.

A possible explanation for this underestimation is the adiabatic fraction, AF , which represents how close the cloud is to the adiabatic assumption. This study assumed $AF = 1$ in the cloud cores, that is, in the areas with the largest 10% of optical depth.

According to Equation 5, N_d is underestimated as AF is used with an exponent of 0.5. Therefore, we define:

$$N_{d,corrected} = \frac{N_{d,original}}{AF^{0.5}} \quad (9)$$

To determine the AF from the satellite, we relate the AF to the CGT. The thicker the cloud gets the lower the AF , because there is more vertical distance for the cloud to mix with ambient air and the droplet concentration is reduced. We derived a climatology of CGT from the SOCRATES campaign, using

Figure 6. Map with the tracks of the measurement campaigns. The cyan dashed line is the MAGIC campaign between Los Angeles and Honolulu. The green dashed line is the PEGASO campaign between South America and the Antarctic Peninsula. The red dashed line is the ACE-SPACE campaign around Antarctica. The blue dashed line is the MARCUS campaign between Tasmania and Antarctica. The magenta dashed line is the SOCRATES air campaign between Tasmania and Antarctica. The black rectangle is the CIRPAS air campaign off the coast of California. The locations of the selected scenes are shown with stars.

Figure 7. Comparisons between satellite measured N_d and ship-based CCN at the same adjusted S as at cloud base. S was calculated using W_b based on H_b , including the correction for temperature differences between CCN counter and cloud base. Panel (a) shows all the cases of CGT up to 800 m marked by their filtering criteria. The black rings are cases filtered out due to $-1.8^\circ\text{C} < \Delta T_b < 1.8^\circ\text{C}$. The blue squares are the cases filtered out due to an absolute difference between satellite and ship measured $H_b > 200$ m. The cyan circles are the cases filtered out due to warm advection. The red circles are the cases filtered out due to ship-measured $H_b > 1,000$ m. The solid black circles are the remaining coupled cases not filtered by any of the criteria above. Panel (b) shows only the remaining 16 coupled unfiltered cases marked by the different validation campaigns: PEGASO (green), ACE-SPACE (red), MARCUS (blue), and MAGIC (cyan). The dashed blue lines show biases by factors of ± 1.3 . The thick dashed blue line represents the fit to the data with R being the correlation coefficient and the RMS the root mean square.

measurements from the aircraft's altitude profiles as it flew from beneath the clouds through them to their tops. The LWP was measured when N_d was at least 5 droplets cm^{-3} . The adiabatic LWP was calculated using the ambient temperature and pressure at cloud base for an adiabatic parcel rising from cloud base to the flight level. Additional data points for this climatology were obtained from the CIRPAS campaign off the coast of California (Braun et al., 2018). The fitting curve in Figure 8 shows the physically expected behavior of AF as a function of CGT .

Figure 8. Adiabatic fraction as a function of cloud geometrical thickness (CGT), as calculated from SOCRATES (blue) and CIRPAS (orange) airborne campaigns. The best fit line is in agreement with the expected behavior of decreasing AF as CGT rises, due to the larger vertical distance for the cloud to mix with ambient air and the reduced droplet concentration.

A successive application of the ΔT and AF corrections to the matching between satellite and surface-measured CCN resulted in a decreasing bias as indicated by the increasing slope from 0.56 (Figure 9a) to 0.67 (Figure 9c) and to 0.82 (Figure 9e), respectively. The correlation coefficient increased from 0.74 to 0.88 and 0.93, respectively.

The results in Figure 9e are valid only for coupled clouds with H_b retrieved from satellite and ships that are within 200 m of each other. This condition is set because the satellite retrieval of W_b relies on the satellite retrieval of H_b , which is converted to W_b according to Equation 7. However, Figure 7a shows that the vast majority of the scenes do not comply with these conditions. This can be remedied by using the dependence of W_b on $CTRC$ (Equation 6), which is affected only weakly by the coupling state (Zheng et al., 2016). The $CTRC$ based calculation of CCN behaves similarly to the H_b -based matching of CCN . A successive application of the ΔT and AF corrections resulted in a decreasing bias as indicated by increasing slope from 0.75 (Figure 9b) to 0.91 (Figure 9d) and to 1.10 (Figure 9f), respectively. The correlation coefficient increased from 0.63 to 0.79 and 0.82, respectively. Therefore, the $CTRC$ -based calculation of CCN is applicable to the satellite retrievals with some loss of accuracy compared to H_b -based calculations as indicated by the decrease in the correlation coefficient from 0.93 to 0.82, but with improved (smaller) bias (compare the slopes of 0.82 and 1.10). The advantage of the $CTRC$ -based method is that it can also be applied to decoupled clouds.

Figure 9. Comparisons between satellite measured N_d and ship-based CCN concentrations at the same S as at cloud base after application of the selection criteria shown in Figure 7 for calculations based on H_b (a) and on CTRC (b). The application of the correction of S for the temperature difference between the CCN counter and cloud base is shown for the calculations based on H_b (c) and on CTRC (d). The additional correction for the adiabatic fraction (Figure 8) is shown for the calculations based on H_b (e) and on CTRC (f).

4. Discussion and Conclusions

There is no fundamental difference in the retrieval of N_d for the coupled and decoupled clouds. There is only a minor difference in the dependence of W_b on CTRC for coupled and decoupled clouds (Zheng et al., 2016). Therefore, the validity of the retrieved CCN for the coupled clouds also validates the retrieved CCN of the decoupled clouds.

The *CCN* concentrations near sea surface under decoupled clouds are generally much larger than at cloud base (Figure 7a). This gradient of decreasing *CCN* concentrations from the surface upward indicates a dominant source of *CCN* from the surface with only a minority of cases, where more *CCN* were present in the free troposphere.

Satellite retrievals of *CTRC* and N_d at the highest 10% of cloud optical depth can be applied to retrieve cloud base *S*. By definition, N_d is then the *CCN* concentrations at that *S*. This represents surface *CCN* only when the clouds are coupled with the surface, as was shown here for 16 cases from four ship campaigns around the world. The retrieved *CCN* concentrations at the base of decoupled clouds are mostly much lower than at the surface. But these are the relevant *CCN* for the determination of aerosol cloud interactions and their impact on cloud properties.

In summary, satellite-based assessment of *ACI* in *MBL* clouds is achievable with the methodology of this study. To improve accuracy we applied (1) a better quantification of the *AF*, through exploring the relationships between *CGT* and *AF* under various conditions, and (2) a better distinction between single and multi-layer clouds and their coupling state.

Data Availability Statement

Data set including the validation cases from vessels is available in this in-text data citation reference: Efraim Avichay. (2020). Nd Validation Cases from Vessels [Data set]. Zenodo (<http://doi.org/10.5281/zenodo.3940800>). The *MODIS* data for this study are available and accessible from *NASA* website (<https://search.earthdata.nasa.gov/search>). The *SST* and wind reanalysis data are available and accessible from *NOAA* and *NCEP* websites (<https://rda.ucar.edu/>, <https://www.esrl.noaa.gov/>).

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