Investigation of the Cloud Microphysics and Albedo Susceptibility of the Southeast Pacific Stratocumulus Cloud Deck

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INVESTIGATION OF THE CLOUD MICROPHYSICS AND ALBEDO SUSCEPTIBILITY OF THE SOUTHEAST PACIFIC STRATOCUMULUS CLOUD DECK

By

David Painemal

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INVESTIGATION OF THE CLOUD MICROPHYSICS AND ALBEDO SUSCEPTIBILITY OF THE SOUTHEAST PACIFIC STRATOCUMULUS CLOUD DECK

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Marine stratocumulus cloud regimes exert a strong climatic influence through their high solar reflectivity. Human-induced changes in stratocumulus clouds, attributed to an increase of the aerosol burden (indirect effects), can be significant given the cloud decks proximity to the continents; nevertheless, the magnitude and the final climatic consequences of these changes are uncertain. This thesis investigates further the interactions between aerosols, cloud microphysics, regional circulation, and radiative response in the Southeast Pacific stratocumulus cloud deck, one of the largest and most persistent cloud regimes in the planet. Specifically, three different aspects are addressed by this thesis: The importance of the synoptic atmospheric variability in controlling cloud microphysical and radiative changes, a validation analysis of satellite retrievals of cloud microphysics from MOderate Resolution Imaging Spectroradiometer (MODIS), and the quantitative assessments of cloud aerosol interactions along with their associated radiative forcing using primarily aircraft remote sensing data.

Synoptic and satellite-derived cloud property variations for the Southeast Pacific region associated with changes in coastal satellite-derived cloud droplet number concentration \( N_d \) are analyzed through a composite technique. MAX and MIN \( N_d \) composites are defined by the top and bottom terciles of daily area-mean \( N_d \) values over the Arica Bight, the region with the largest mean oceanic \( N_d \), for the five October months
of 2001, 2005, 2006, 2007, and 2008. The MAX-$N_d$ composite is characterized by a weaker subtropical anticyclone and weaker winds than the MIN-$N_d$ composite. Additionally, the MAX-$N_d$ composite clouds over the Arica Bight are thinner than the MIN-$N_d$ composite clouds, have lower cloud tops, lower near-coastal cloud albedos, and occur below warmer and drier free tropospheres. At 85°W, the top-of-atmosphere shortwave fluxes are significantly higher (50%) for the MAX-$N_d$, with thicker, lower clouds and higher cloud fractions than for the MIN-$N_d$. The change in $N_d$ at this location is small, suggesting that the MAX-MIN $N_d$ composite differences in radiative properties primarily reflects synoptic changes.

The ability of MODIS level 2 retrievals to represent the cloud microphysics is assessed with in-situ measurements of droplet size distributions, collected during the VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx). The MODIS cloud optical thickness ($\tau$) correlates well with the in-situ values with a positive bias (1.42). In contrast, the standard 2.1 micron-derived MODIS cloud effective radius ($r_e$) is found to systematically exceed the in-situ cloud-top $r_e$, with a mean bias of 2.08 $\mu$m. Three sources of errors that could contribute to the MODIS $r_e$ positive bias are investigated further: the spread of the cloud droplet size distribution, the presence of a separate drizzle mode, and the sensor viewing angles. The sensor zenith viewing angles were found to have little impact, while the algorithm assumption about the cloud droplet spectra and presence of a precipitation mode could affect the retrievals but not by enough to fully explain the positive MODIS $r_e$ bias. The droplet spectra effects account for $r_e$ offsets smaller than 0.6 $\mu$m, 0.9 $\mu$m, and 1.6 $\mu$m for non-drizzling, light-drizzling, and heavy-drizzling clouds respectively. An explanation for the observed MODIS bias is lacking although three-dimensional radiative effects were not considered. This
investigation supports earlier studies documenting a similar bias, this time using data from newer probes. MODIS $r_c$ and $\tau$ were also combined to estimate a liquid water path (LWP) and $N_d$. A positive bias was also apparent in LWP, and attributed to $r_c$. However, when selected appropriate parameters a priori, the MODIS $N_d$ estimate was found to agree the best with the insitu aircraft observations of the four MODIS variables.

Lastly, the first aerosol indirect effect (Twomey effect) is explicitly investigated with VOCALS-REx observations, collected during three daytime research flights (Nov 9, 11, and 13), utilizing an aerosol-cloud interactions metric, and defined as $ACI=\frac{d\ln(\tau)}{d\ln(N_a)}$, with $N_a$ corresponding to the accumulation mode aerosol concentration, $\tau$ derived from a broadband pyranometer, and ACI binned by cloud LWP derived from a millimeter-wavelength radiometer. Aircraft remote sensing estimates of the ACI, during sub-cloud transects, show that the cloud aerosol-interactions are strong and close to the maximum theoretical value for thin clouds, with a decrease of ACI with LWP. Although an explanation for the dependence of ACI on LWP is lacking, we found that a decrease in ACI with LWP is associated with decreases in both surface meridional winds and $N_d$. Similar to ACI, albedo fractional changes due to $N_d$ fractional changes also tended to be smaller for higher LWPs, but with an overall radiative forcing larger than conservative global estimates obtained in global circulation models.

The findings of this thesis emphasize the strong stratocumulus albedo response to an aerosol perturbation and its dependence on the regional scale atmospheric configuration. The results presented here can be used as a benchmark for testing regional and climate models, as well as helping to improve the current parameterizations of the first aerosol indirect effect.
To my parents, Olga and Lisandro
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Chapter 1

Introduction

1.1 Challenges in our current knowledge of the aerosol indirect effects

It is well-established that atmospheric aerosols can modify the cloud physical properties. The cloud radiative changes associated with an increase in aerosols, known as aerosol indirect effects (AIE), have captured the interest of the scientific community due to their potential effects in the global climate (Lohman and Fechter, 2005). Twomey (1977) first proposed that an increase of cloud nuclei by pollution and therefore, an increase of the cloud droplet number concentration under constant liquid water content, leads to an increase in the solar radiation reflected by clouds (first aerosol indirect effect or albedo effect). This effect has been reported in many studies, encompassing satellite observation (e.g. Twomey and Platnick, 1994; Segrin et al. 2007), in-situ and land-based observation (e.g. Twohy et al. 2005; McComiskey et al. 2008), and theoretical studies (e.g. Feingold et al. 1997; Platnick and Oreopoulos, 2008). A second mechanism of cloud-aerosol interaction (second indirect effect) links an increase of aerosol concentration, and reduction of the droplet effective radius, with less precipitating clouds and increase in the cloud life-time (e.g. Albrecht, 1989), or geometrical thickness (Pincus and Baker, 1994). Others additional mechanisms have also been proposed but their contributions are debatable (Lohman and Fechter, 2005).
Despite evidence that associates an increase of aerosol concentration with changes in cloud microphysics, the exact way the AIE modify the global radiative balance is unknown, and constitutes one the largest uncertainties in our current understanding of the radiative forcing (Fig. 1.1, albedo effect only, IPCC report 2007). This difficulty in estimating the aerosol effects is manifested in the disagreement among different numerical models (Quaas et al., 2009). In contrast, although satellite retrievals provide a more accurate picture of the cloud system, with a high temporal and spatial coverage, the lack of retrieval validations make it difficult to determine if the magnitude of satellite-derived indirect effects are instrument/algorithm artifacts or the result of physical interactions.

The uncertainties in the AIE are not only limited to estimates derived from satellite retrievals and climate models. In fact, different in-situ studies also report a broad spread in the indirect effect magnitude (McComiskey and Feingold, 2008). These inconsistencies reflect measurements artifacts as well as differences in the sampling strategies; nevertheless, dependences on the cloud regime and geographical region can potentially account for some portion of the spread. This idea is supported by global studies that suggest that particular cloud regimes respond differently to changes in aerosol burden and cloud microphysics (e.g. Han et al., 1994, Oreopoulos and Platnick, 2008).

An additional aspect that hampers a more exact quantification of AIE is that the cloud physical properties are simultaneously controlled by both the availability of atmospheric aerosols that can be activated (serving as cloud condensation nuclei), and the environmental conditions that determine the general conditions favorable for cloud
development. The separation of both processes in the final cloud radiative response is not trivial, especially from in-situ observations. As a consequence, the meteorological component is generally ignored in bulk estimates of AIE (Quaas et al. 2009), where it is implicitly assumed that aerosol changes are the only factor that can modify cloud microphysics.

Figure 1.1: Global average radiative forcing (RF) in 2005 with respect to 1750 for CO₂, CH₄, NO₂, and other important agents and mechanism, together with the typical geographical extent (spatial scale) of the forcing and the assessed level of scientific understanding (LOSU). The thin black line attached to each colored bar represents the range of uncertainty value. Taken from Chapter 2, IPPC report 2007.
1.2 Indirect effects in marine stratocumulus

The subtropical cloud-capped marine boundary layer has a strong climate impact through the cloud decks’ high solar reflectivity, whereas the emitted long-wave radiation remains close to that of surface emission under clear skies (Hartmann et al., 1992). Microphysical and radiative properties in maritime warm clouds are closely linked, and highly sensitive to changes in the environmental conditions. This is very significant when considering the dramatic aerosol burden increase since the beginning of the industrialized era (Intergovernmental Panel on Climate Change, 2007). Marine low clouds, namely stratus and stratocumulus, can be especially susceptible to cloud-aerosol interactions due to their proximity to the western edge of the continents (Klein and Hartmann, 1993), close to the anthropogenic sources of aerosols. The aerosol impact in stratocumulus clouds is also suggested in satellite-derived cloud droplet number concentration retrievals (Figure 1.2) that show a remarkable increase of number of droplets near the coast.

Figure 1.2: Mean satellite-derived cloud droplet number concentration for warm clouds during Octobers 2005, 2006, 2007 and 2008. The red circle indicates the approximate location of the Southeast Pacific stratocumulus regime.
The Southeastern Pacific stratocumulus deck, under the quasi-permanent influence of the southeast Pacific anticyclone, is arguably the most persistent of the Earth’s cloud decks (Fig. 1.3). In addition to its climatic significance, the Southeast Pacific stratocumulus deck is also one region where aerosol impacts on cloud microphysics are substantial, with Bennartz (2007) and Wood et al. (2008) showing large (> 200 cm$^{-3}$) cloud droplet number concentrations retrieved from MODerate resolution Imaging Spectroradiometer (MODIS) data along the coast of Peru and Chile (17°S-32°S, Fig. 1.2). Bennartz (2007) speculated that the downwind transport of polluted air affected the microphysics of these clouds, although the processes that drive the microphysical changes near the coast are not sufficiently understood.

![Satellite-based low cloud fraction climatology in January (left) and in July (right). Climatology calculated over the period from 2003 to 2008. Red circle indicates the approximate location of the Southeast Pacific stratocumulus regime. Figure modified from Stubenrauch et al. (2010).](image)

The Southeast Pacific region is relatively void of in-situ observations, an aspect that has limited our ability to characterize the marine boundary layer and the stratocumulus cloud deck itself. This observational gap has changed gradually during the last two
decades, with the contribution of six months-long National Oceanic and Atmospheric Administration (NOAA) research cruises\(^1\) during 2001 to 2007 (Serpetzoglou et al., 2008, Zuidema et al 2009, and references therein). These rather sparse campaigns were followed by VOCALS Regional Experiment (VOCALS-Rex)\(^2\), the most comprehensive field experiment carried out over this region. VOCALS-Rex included coordinated aircraft, ship, and landsite atmospheric and oceanic observations during October-November 2008. VOCALS-REx is helping to elucidate regional mechanisms that control the evolution of the stratocumulus cloud deck and its importance in the global climate, and presents a unique opportunity to examine the aerosol indirect effects.

### 1.3 Objectives and content of the thesis

Uncertainties in the aerosol indirect effects motivate the following questions:

- Are satellite remote sensing retrievals accurate enough to investigate cloud microphysical properties in marine stratocumulus?
- What are the meteorological features associated with the variability in cloud microphysics?
- What is the combined radiative contribution of microphysics and meteorology?
- Does anthropogenic-induced \(N_d\) enhance the cloud albedo?
- When the atmospheric conditions and cloud dynamics are kept constant, how do the cloud microphysics respond to a fractional change in aerosol concentration. Does this response vary for different atmospheric scenarios?

---

\(^1\) It is fair to say that the first research cruise over the Southeast Pacific, oriented to investigate the stratocumulus cloud topped marine boundary layer, was CIMAR-5, carried out in October 1999 and funded by the Chilean National Oceanographic Committee (Garreaud et al., 2001).

\(^2\) VOCALS stands for VAMOS Ocean Atmosphere Land System. VAMOS is the acronym for Variability of the American MOnsoonS.
It is important to emphasize that the main challenge posed by the aerosol indirect effects is the accurate quantification of the cloud radiative response under different scenarios (synoptic conditions). Ultimately this knowledge can be used to improve the cloud representation of the aerosol indirect effect in regional and global models.

Here a regional approach is used to focus on answering the previous questions over the domain of the Southeast Pacific (Chile-Peru) stratocumulus regime. We rely on different observational platforms deployed during VOCALS-Rex and the 6 NOAA cruises. In addition to this, we use satellite retrievals because of their spatially extended coverage. The purpose of this synergistic approach is to constrain those results that are more likely to be consequence of instrument artifacts, rather than the consequence of real physical mechanisms.

In Chapter 2, we study the regional scale factors associated with changes in cloud microphysics. We rely on a composite analysis constructed from cases with high and low satellite-derived cloud droplet number concentration. The atmospheric conditions in the lower troposphere are analyzed, and the synoptic patterns that control changes in the boundary layer are also described. We finally use satellite retrievals of shortwave radiation to estimate the final radiative response when cloud microphysics dynamics are combined.

In Chapter 3, we carry out a validation analysis of satellite retrievals by using aircraft observations collected during VOCALS-Rex. We focus on Moderate Resolution Imaging Spectroradiometer (MODIS) instrument, and compared retrievals of optical thickness, effective radius, liquid water, and cloud droplet number concentration with in-
situ measurements. We explore possible factors that can explain the biases in the satellite observations and investigate plausible physical processes affecting the retrievals.

In Chapter 4 we investigate in detail the first aerosol indirect effect with aircraft observation of cloud microphysics. We use airborne remote-sensed liquid water and optical thickness to characterize the cloud dynamics and microphysics. The retrievals are validated against cloud probe measurements and then used to calculate cloud droplet number concentration. We calculate metrics of cloud-aerosol interactions as a function of liquid water path, and discuss the modulation of dynamical factors in the activation of the atmospheric aerosols. The albedo, calculated from a plane-parallel radiative transfer model, is used to derive the cloud albedo susceptibility to a perturbation in cloud droplet number concentration for different magnitudes of liquid water path. The implications of a change of aerosol concentration in the cloud albedo are also discussed. The final chapter corresponds to the discussion and concluding remarks.
Chapter 2
Microphysical variability, synoptic conditions, and radiative response

2.1 Motivation

Given the global radiative importance of the stratocumulus cloud regimes, a deeper understanding of the processes that affect their radiative properties, both large-scale and microphysical, is necessary for developing confidence in future climate predictions. This also includes a careful investigation of the aerosol indirect effects and their ability to modify the cloud microphysics, as well as the way the atmospheric circulation can modify the cloud dynamics.

Observational evidence indicates that changes in aerosol concentrations are generally connected to changes in the atmospheric circulation. This covariability emphasizes the importance of the meteorological control of liquid water path (LWP) in estimates of the first aerosol indirect effect. Satellite-derived examples of cloud-aerosol radiative compensations include Han et al. (2002) and Matsui et al. (2006); both show more aerosols are often associated with lower liquid water paths. This aerosol-LWP correlation is not causal; instead, it is indication that particular meteorological conditions that promote aerosol transport to the cloud layer can also decrease the cloud water content. A principle of the cloud lifetime effect, that higher LWPs are associated with longer cloud lifetimes (through precipitation suppression), has also recently been called into question (Christensen et al., 2009). Theoretical and modeling support for these
observations has come from, among others, Jiang et al. (2002) and Ackerman et al. (2004), with more discussion in Stevens and Feingold (2009).

Dynamic compensation of aerosol indirect effects emphasizes the need to control for meteorological forcings, a difficulty in both modeling and observational assessments. Atmospheric static stability is often chosen as a meteorological control variable (e.g., Matsui et al., 2006), because static stability correlates well with stratocumulus cloud fraction at seasonal scales (Klein and Hartmann, 1993). Matsui et al. (2006) observed that cloud droplet sizes tend to be the smallest under strong inversions as well as within polluted environments. Mauger and Norris (2007) further examined the static stability of parcel back-trajectories of satellite-derived aerosol and cloud properties, and found that the covariation of aerosol amount and cloud fraction with static stability could increase with meteorological history.

As described in the introduction, the Southeast Pacific stratocumulus deck is one region where aerosol impacts on cloud microphysics are potentially significant, reflected in the high cloud droplet number concentrations retrieved from MODerate resolution Imaging Spectroradiometer (MODIS) data along the Peru and Chile coast (17˚S-32˚S), also shown here in Fig. 2.1. Though difficult to quantify from satellite imagery, measurements taken during the VAMOS Ocean-Coupled-Atmosphere-Land-Study Regional Experiment (VOCALS-REx; Wood and Mechoso, 2008) provide evidence of anthropogenic emissions as far west as 85˚W (Hawkins et al., 2010). Bennartz (2007) speculated that the downwind transport of polluted air affected the microphysics of these clouds. This is plausible for the southeast Pacific where the Andes dictate along-shore winds that can transport aerosol from the more developed southern regions (the capital
city of Chile, Santiago, is located near 33˚S) to the north. Huneeus et al. (2006) hypothesized instead that the entrainment of polluted free-tropospheric air north of 25˚S could be responsible for the \( N_d \) increases. At more northern latitudes the Andes protect the deck from boundary-layer aerosol-laden continental outflow, but high-altitude copper smelters emit sulfate aerosols above the marine boundary layer.

Comprehensive regional assessments relying on satellite data, available local data, and reanalyses help provide background understanding for further regional aerosol-cloud interaction assessments. In this chapter, we focus further on understanding the meteorology and cloud macrophysical variability associated with \( N_d \) variability in the southeast Pacific stratocumulus region. An underlying premise is that the differences in cloud properties associated with the largest differences in \( N_d \) primarily reflect synoptic influences, rather than the effect of cloud-aerosol interactions. The content of this chapter (Painemal and Zuidema, 2011; published in Atmospheric Chemistry and Physics) was conceived before VOCALS-Rex, with the main objective of giving a regional context to the measurements collected during the field experiment, which form the dataset used in Chapter 3 and 4.

2.2 Data and Methods

The large-scale circulation is depicted using the NCEP/NCAR Reanalysis. The Reanalysis is capable of resolving the main circulation features away from the coastline (Garreaud et al., 2001) and has been used successfully as a boundary forcing for regional simulations (e.g., Garreaud et al., 2004). However, the Reanalysis is also known to underestimate the boundary layer depth, cloud fraction, and cloud liquid water path (e.g.,
Bretherton et al., 2004). Instead, we derived cloud properties (cloud top height, fraction, liquid water path/thickness and droplet number) primarily from MODIS data. The retrievals are aided by frequently overcast skies, relatively homogenous cloud conditions, and favorable sun-satellite viewing geometries (e.g., Kato and Marshak, 2009). Satellite-derived cloud droplet numbers (e.g., Bennartz, 2007) serve as an aerosol proxy, avoiding issues with satellite retrievals of clear-sky aerosol properties (e.g., Loeb and Schuster, 2008).

Further averaging of daily satellite data into composites also reduces the influence of random error. The analysis approach here is based on satellite and NCEP Reanalysis composites, with the composites defined through the upper and lower terciles of the satellite-retrieved cloud droplet numbers above the Arica Bight, the region with the largest mean oceanic $N_d$ (box in Figure 2.1). A composite analysis approach also allows us to keep issues with the NCEP Reanalysis representation near the coastline and Andes mountains in perspective: these issues are demonstrated in comparisons to coastal radiosonde profiles of temperature, moisture and wind, and discourage coastal Lagrangian backtrajectory calculations. The composite analysis was applied to five October months of daily data. This month corresponds to the regional climatological maximum of the stratocumulus (Sc) deck (Klein and Hartmann, 1993), and the focus on October months alone is intended to minimize influences from the seasonal progression. Our approach complements more thorough single case studies such as Huneeus et al. (2006) and provides a context for VOCALS-REx analyses. We selected four October months coincident with NOAA research cruises (2001, 2005, 2006 and 2007), while October 2008 coincides with the VOCALS Regional Experiment. This was done so that
the satellite retrievals could be compared to the ship-based measurements.

The composite analysis relies on daytime Terra level 3 data at 1°x1° spatial resolution; at this latitude, the daily-mean values for each platform are effectively regridded individual swath data. The daytime Terra overpass (10:30 LT) was previously found to be the MODIS overpass most representative of daily-mean conditions (Zuidema et al., 2009). MODIS cloud top temperatures, derived from separate day and nighttime 11 µm equivalent brightness temperatures, were used to estimate a satellite cloud top height, and rely on a depth-varying lapse rate, proportional to the inverse of the cloud top height, inferred from the open-ocean cruise radiosondes (Zuidema et al., 2009). These are shown to capture daily synoptic-scale variations reasonably well within Painemal et al. (2010) and agree well with airborne radar observations of cloud top made during VOCALS-REx (Rahn and Garreaud, 2010).
Figure 2.1: Mean $N_d$ (# cm$^{-3}$, colors) and LWP (g m$^{-2}$, contours), based on October 2001 and 2005-2008 Terra daytime MODIS level 3 data. The box indicates the area over which the daily-mean $N_d$ were averaged and the black square indicates Antofagasta.

Estimates of the all-sky shortwave fluxes (SW) at the top of the atmosphere were obtained from the Clouds and Earth’s Radiant Energy System (CERES, Wielicki et al. 1996) instrument on board the Terra platform. The CERES data allow for a net radiative assessment that is somewhat independent of the plane-parallel radiative transfer approximation. We used the radiometric measurements from the shortwave channel (0.3 - 5 µm), and spatially averaged Single Scanner Footprint data to a 1˚x1˚ spatial resolution. These data were only available for October 2001, 2005, and 2006.

The occurrence of precipitation was determined from CloudSat radar reflectivity data (the Cloud-Geometrical-Profile product; Stephens et al., 2002) from October 2006,
2007 and 2008. Surface winds were provided by the satellite scatterometer QuikSCAT at a 0.25°x0.25° resolution; we only used the evening pass (1800 LT) data. Non-satellite datasets are the NCEP/NCAR reanalysis (Kalnay et al. 1997) meteorological fields, with a horizontal resolution of 2.5°x2.5°, and daily radiosonde observations at Antofagasta (22.43°S, 70.43°W, 120 a.m.s.l.) available only at 1200 UTC (0800 LT) and interpolated to a consistent 50 hPa vertical resolution.

We combined MODIS Collection 5 cloud effective radius ($r_e$) and cloud optical thickness ($\tau$) retrievals (Platnick et al., 2003) to produce values for $N_d$ and cloud depth ($H_{sat}$), and used a temperature threshold of 273 K to select for warm, liquid-only clouds. MODIS retrievals of cloud optical depth and effective radius are routinely available, but small droplet sizes do not unambiguously indicate aerosol loading (e.g., Schuller et al., 2003). Instead, we estimated $N_d$ and cloud depth ($H_{sat}$) from a combination of $r_e$ (2.1 micron channel) and $\tau$ assuming adiabatic conditions (e.g. Bennartz, 2007, Szczodrak et al. 2001). This allows for a separation of macrophysical and microphysical cloud property measures, and in addition facilitates a comparison to the ship-based measurements of accumulation-mode aerosol $N_a$ and cloud depth $H_{ship}$.

$H_{sat}$ is estimated from the adiabatically-derived liquid water path (LWP) using

$$LWP = \frac{\Gamma_{ad} H_{sat}^2}{2} \text{ as}$$

$$H_{sat} = \sqrt{\frac{2}{\Gamma_{ad}}} \frac{5}{9} \rho_w \cdot r_e \cdot \tau$$  \hspace{1cm} (2.1)

where $\rho_w$ is the density of water and $\Gamma_{ad}$ is the adiabatic lapse rate of liquid water content with height. Sounding-derived $\Gamma_{ad}$ values reveal that the lower, warmer coastal clouds have typical values between 2.1-2.3 x10$^{-3}$ g m$^{-4}$, while clouds west of 75°W have values
between 1.8-2.0 x10^{-3} \text{ g m}^{-4}. Near-coastal liquid water content profiles in VOCALS-REx aircraft data often show diminished values near cloud top because of cloud top entrainment, which reduces the effective $\Gamma_{\text{ad}}$, while profiles from further offshore are close to adiabatic (also seen in Zuidema et al., 2005). With this in mind, we rely on a constant $\Gamma_{\text{ad}}$ value of 2.0 x10^{-3} \text{ g m}^{-4}, and approximate its uncertainty at 10%.

The satellite estimate of cloud droplet concentration is expressed as (Appendix 1):

$$N_d = \Gamma_{\text{ad}}^{1/2} \frac{10^{1/2}}{4\pi \rho w^{1/2}} \frac{\tau^{1/2}}{r_e^{5/2}}$$ (2.2)

### 2.3 First aerosol indirect effect and preliminary validation of satellite retrievals

Instantaneous swath data (level 2) from both the Terra and Aqua platforms (~1030 LT and ~1330 LT overpasses, respectively) were used for the validation comparisons to in-situ observations, complementing the validation analysis presented in Chapter 3. $H_{\text{sat}}$ and $N_d$ derived from instantaneous level 2 data were spatially averaged over an area of 25 km by 25 km, corresponding to a 7 m s^{-1} “frozen turbulence” advective speed over one hour. For overcast, opaque, mostly unbroken, warm cloud pixels, the a priori error in $H_{\text{sat}}$ (25 km^{2} scale) from a $\tau$ error of 8% (Bennartz, 2007) and a conservative $r_e$ (2.1 micron) error of ~20% is ~11% (Marshak et al. 2006). The error in $N_d$ will be dominated by the error in the MODIS-derived effective radius through the $r_e^{-5/2}$ dependence, which can account for a 50% error in $N_d$ alone. This encourages us to consider the sources for effective radius error more closely. The effective radius retrieval is most suspect in broken cloud conditions, where $r_e$ can be systematically overestimated (e.g., Marshak et al., 2006). We observed that low $N_d$ values tended to be associated with
high-LWP clouds (not shown), which are more likely to precipitate, and which corresponded poorly to the surface-based aerosol measurements for the lower values of $N_d$. The impact of precipitation and cloud inhomogeneities in $N_d$ retrievals was minimized by limiting our quantitative analysis to $N_d$ values > 50 cm$^{-3}$ for overcast pixels. In addition, averaging to a scale of 25 km$^2$ allows for compensation of the largest three-dimensional radiative transfer effects within individual pixels. If we assume an $r_e$ (2.1 µm) error of 10% based on comparisons to microwave-derived $r_e$ (Bennartz, 2007, Fig. 4), we estimate a $N_d$ error of 26% (also accounting for uncertainty in our assumed $\Gamma_{ad}$). This is less conservative than the uncertainty estimate of 50% within Bennartz (2007), but seems justified for our more restricted, regional dataset (A more comprehensive $N_d$ validation is carried in Chapter 3).

We used a more comprehensive database of in-situ observations from six NOAA cruises (October, 2001, 2005, 2006, 2007, and November, 2003 and December, 2004) in this analysis. The ship-based aerosol number concentration $N_a$ is a direct measurement, typically made from a Particle Measuring System Lasair-II instrument, which measures all particles greater than 0.1 microns, but does not control for humidity. For the 2004 cruise, data from a Differential Mobility Analyser are used (Tomlinson et al., 2007). Aerosol counts gathered from both instruments during the 2003 cruise show good agreement (Tomlinson et al. 2007). The ship-based cloud depth ($H_{\text{ship}}$) is computed as the difference between the radiosonde$^3$ inversion base height (placed at the temperature minimum) and hourly-averaged ceilometer cloud base heights. Comparisons were restricted to the one-hour time periods spanning the radiosondes.

$^3$ Radiosondes were launched at either 4 or 6 hour intervals.
$H_{\text{sat}}$ and $H_{\text{ship}}$ values correlate well with each other ($r=0.54$), increasing to 0.7 when restricting the sample to $H_{\text{ship}} < 400$ m and to overcast periods (ceilometer hourly cloud fractions $> 90\%$) (Fig. 2.2a; only overcast periods are shown). $H_{\text{sat}}$ values often exceed $H_{\text{ship}}$, with a mean bias of 59 m for $H_{\text{ship}} < 400$ m, a similar bias as the one found by Schuller et al. (2003). This result gives additional support to the idea that MODIS retrievals of LWP (cloud depth) tend to overestimate the in-situ observations. In Chapter 3, we find that this positive bias is mainly explained by the overestimate of MODIS $r_c$ of the in-situ value.

Satellite-derived $N_d$ is compared to ship-based aerosol concentrations (size $> 0.1$ micron) in Fig. 2.2b, with no constraint placed on the hourly ceilometer cloud fractions towards increasing the data sample. Most of the aerosol concentration measurements were taken near 85°W and 20°S, with a limited sampling near the coast. $N_d$ and $N_a$ show an obvious correlation ($r=0.65$, increasing to 0.8 for hourly-mean ceilometer cloud fractions $> 0.9$), with $N_d$ typically $< N_a$. The values of both $N_d$ and $N_a$ are higher when sampled closer to the coast (east of 80°W, black filled circles in Fig. 2.2b).

A quantitative assessment of the observed $N_d$-$N_a$ relationship can be expressed through an aerosol cloud index $\frac{d \ln N_d}{d \ln N_a}$ (McComiskey et al., 2009). Such observed relationships have been compared to climate model values (e.g., Quaas et al., 2009) and additional care was taken to determine the best-fit line. The mean of the $N_a$ standard deviations about the hourly-mean value was used as a mean $N_a$ uncertainty estimate (10 cm$^{-3}$) and a constant $N_d$ error of 26%. We restricted the data samples to $N_d > 50$ cm$^{-3}$ as a proxy for non-precipitating clouds and to control for gross overestimates in the effective radius retrievals caused by cloud inhomogeneities. Taking both $N_a$ and $N_d$ uncertainty
estimates into account, we find a best-fit value for $\frac{d \ln N_d}{d \ln N_a}$ of 0.56 (York et al. 2004, but using mean rather than individual datum uncertainties). For comparison, Pruppacher and Klett (1997) suggest a value of 0.7 for $\frac{d \ln N_d}{d \ln N_a}$ based on droplet activation theory, while McComiskey et al. (2009) find an empirical value of 0.48 using cloud microphysical retrievals from surface-based remote sensors of marine stratocumulus clouds at Pt. Reyes, CA. Quaas et al. (2009) find a sensitivity of MODIS-derived $N_d$ to the aerosol optical depth at Pt. Reyes that is close to the corresponding surface-based value.

Comparison to other published values of $\frac{d \ln N_d}{d \ln N_a}$ must take many factors into account. McComiskey et al. (2009) document a strong reduction with increasing scale, for example, though satellite-derived values within Quaas et al. (2009) are similar to the surface-based value. Our value is therefore fairly high given the 25 km$^2$ scale. Moreover, 42% of our variation in $N_d$ is explained by the aerosol concentration variability, compared to 15% for McComiskey et al. (2009). Some of the explanation is provided by Fig. 2.1, for which daily maps of Terra $N_d$ and LWP values were averaged over the five October months. Many of the higher $N_d$ values are associated with thinner clouds (LWP<90 g/m$^2$, contours in Fig. 2.1) that are less likely to be drizzling (e.g., Leon et al., 2008) and more likely to be adiabatic if offshore (e.g., Zuidema et al., 2005). For such clouds, values of $\frac{d \ln N_d}{d \ln N_a}$ are typically higher (McComiskey et al., 2009; Kim et al. 2008). Three-dimensional radiative transfer effects upon the $r_e$ and thereby $N_d$ retrieval, which will be more pronounced for thicker, more broken clouds with naturally higher $r_e$, may also
contribute to a higher value for $\frac{d \ln N_d}{d \ln N_a}$ by artificially lowering low values of $N_d$ even further, despite our efforts to screen for this.

This estimate of aerosol-cloud interaction does not discriminate between changes attributed to aerosol concentration and those associated with changes in the meteorological conditions that govern variations in LWP. In other words, our calculation of $\frac{d \ln N_d}{d \ln N_a}$, do not exactly explain how perturbations on aerosol concentration can modify the cloud microphysics. More detailed calculations of cloud-aerosol interaction metrics and its association with changes in LWP and atmospheric conditions are included in Chapter 4.

Figure 2.2 (a) MODIS-derived cloud depth (25 km$^2$ average) versus ship-based cloud depth (hourly averaged; ceilometer cloud fraction >0.9), and (b) MODIS-derived $N_d$ (25 km$^2$) versus ship-based accumulation-mode ($r> 0.1$ µm) aerosol concentrations (hourly-averaged; no restriction on ceilometer cloud fraction). Blacks dots indicate samples east of 80°W and crosses are samples for which the bias-corrected MODIS $H_{sat}$ is within 70 m of $H_{ship}$. The black solid line represents the best-fit line for all values with $N_d> 50$ cm$^{-3}$. Sampling domain covers 0°-30°S, 72°W-90°W. Fig 2.2a and 2.2b were constructed with 51 and 48 samples respectively.
2.4 Observational Composites

The comparisons presented in this chapter, as well as satellite-derived cloud top heights analysis within Zuidema et al. (2009), provide further support for the ability of the MODIS retrievals to represent the stratocumulus macro- and microphysics. Our composite analysis is based on the \( N_d \) variability over the apparently most-polluted region, the Arica Bight, defined here to span 71.5°W-75.5°W and 18.5°S-25.5°S (box in Fig. 2.1). Only scenes with at least 70% of their satellite pixels classified as overcast were used; this selection bias is discussed in Section 2.7. Such overcast days occurred on 77% (120 days) of the total days. As seen in Fig. 2.1, mean LWP values are small (< 70 g m\(^{-2}\)) over the region with the largest \( N_d \).

Days are composited by the spatial-mean \( \overline{N_d} \) (TERRA only) over the Arica Bight into the highest and lowest terciles (40 and 42 overcast days each), labeled hereafter as MAX (\( \overline{N_d} > 215.8 \text{ cm}^{-3} \)) and MIN (\( \overline{N_d} < 161.6 \text{ cm}^{-3} \)) respectively. MAX-MIN \( N_d \) composite differences (Fig. 2.3, colors) are a maximum by construction between 18°S-27°S with values higher than 120 cm\(^{-3}\), and a westward extension of about 8°. Near Antofagasta (23.43°S; small black square) the \( N_d \) changes reach their maximum value of 160 cm\(^{-3}\), while the maximum climatological-mean \( N_d \) values occur further north near 20°S. Even further north along the Peruvian coast (north of 15°S), the cloud droplet number concentrations are slightly higher within the MIN-\( N_d \) composite than the MAX-\( N_d \) composite (negative values in Fig. 2.3). MAX-MIN \( N_d \) composite differences in LWP reveal decreased LWPs east of 85°W (~ 20 g m\(^{-2}\)) during the MAX-\( N_d \) composite, along with increased LWPs (10-30 g m\(^{-2}\)) west of 85°W (Fig. 2.3, contours), in one early
indication of a dynamical compensation to the cloud albedo for near-coastal microphysical changes.

Figure 2.3. As Fig. 2.1 but for the MAX-MIN \(N_d\) composite differences: \(N_d\) (colors) and LWP (contours).

Time series of mean surface wind speeds (20°S-30°S, 70°W-90°W), mean cloud top heights alongshore (20°S-30°S, 70°W-80°W), and mean offshore 500 hPa geopotential heights (15°S-35°S, 70°W-100°W, approximately the location of the climatological anticyclone) are shown along with the occurrence of MAX (dark triangles) and MIN (open circles) \(N_d\) days in Fig. 2.4. These locations were chosen to exemplify synoptic differences between MAX and MIN cases: MAX (MIN) \(N_d\) days are more likely to co-occur with weaker (stronger) coastal winds (Fig. 2.4a) and shallower coastal boundary layers (Fig. 2.4b), also shown in Fig. 2.5, and enhanced offshore mid-
tropospheric geopotential heights (Fig. 2.4c). About one-half/two-thirds of the MIN/MAX \( N_d \) days, respectively, occur in groups of three or more days. Some interannual variability is evident as well, with more MIN \( N_d \) cases occurring during 2007, coinciding with a weak cool ENSO phase, and more MAX \( N_d \) cases than MIN \( N_d \) cases occurring during 2001 and 2008, when more intense observational campaigns, the Eastern Pacific Investigation of Climate (Bretherton et al., 2004) and VOCALS-REx, were conducted in the region.

Figure 2.4: Time series of: a) mean surface winds (QuikSCAT) at 20°S-30°S, 70°W-90°W, b) mean Terra-daytime MODIS-derived cloud top heights at 20°S-30°S, 70°W-80°W and c) mean 500 hPa geopotential height at 15°S-35°S, 70°W-100°W. Black triangles and open circles correspond to MAX and MIN \( N_d \) days.
2.4.1 Satellite-derived composite characteristics

The mean and MAX-MIN $N_d$ difference regional circulation patterns are shown in Fig. 2.6 panels a and b respectively. The anticyclone is strengthened offshore (85°W-95°W) during the MIN $N_d$ composite, with stronger surface winds and a strengthened coastal jet near 33°S. Surface winds are light within both composites at the northern end of the Arica Bight, where changes in $N_d$ are also modest. Mean October values along with MAX-MIN $N_d$ composite difference values are shown for the MODIS-derived cloud top height (Fig. 2.6c), cloud depth (Fig. 2.6d), cloud fraction (Fig. 2.6e), and top-of-atmosphere CERES shortwave fluxes (Fig. 2.6f). Boundary layer depths are shallower for the MAX $N_d$ composite by 200 m to 350 m over a broad domain extending beyond the Arica Bight (Fig. 2.6c). Over the Arica Bight and along 80°W, these changes are...
associated with thinner clouds for the MAX N\textsubscript{d} composite (Fig. 2.6d), by up to 50 m. West of 80°W, the pattern reverses, and clouds are slightly thicker for the MAX N\textsubscript{d} composite than for the MIN N\textsubscript{d} composite.

The mean MODIS cloud fraction is a maximum parallel to the Peruvian coast (contours, Fig. 2.6e), with the mean values in the top-of-atmosphere shortwave fluxes following a similar spatial pattern (Fig. 2.6f). Cloud fractions are diminished by about 5% over the Arica Bight for the MAX composite relative to the MIN composite, remain approximately the same off the coast of Peru, and increase significantly elsewhere – by up to 20% near 85°W, 25°S. The variations in the top-of-atmosphere shortwave fluxes (Fig. 2.6f) are broadly consistent with the cloud fraction changes. Over the Arica Bight, the MAX-MIN composite change in top-of-atmosphere SW fluxes is negative near the coast, and increases 500 km offshore. This reflects the compensation between increased brightening from a higher N\textsubscript{d}, and decreased brightening from a thinner cloud. Near the coast the cloud thinning ends up dominating radiatively, while further offshore the small increase in N\textsubscript{d} more than offsets reductions in cloud albedo from cloud thinning. The mean change over the Arica Bight region, defined by the box in Fig. 2, is 20 W m\textsuperscript{-2}, or an approximately 15% increase in the regional-mean cloud albedo. Further south, the increase in cloud fraction over the Chilean coastal jet for the MAX N\textsubscript{d} composite corresponds with an increase of approximately 50 W m\textsuperscript{-2} in the top-of-atmosphere shortwave fluxes, but over a region with small mean cloud fraction. The most dramatic changes in the fluxes are observed away from Arica Bight along 85°W, however, and are primarily associated with cloud fraction differences, with only small changes in the cloud droplet numbers.
Figure 2.6: a) and b) MAX and MIN $N_d$ composite NCEP sea level pressure (red contours), surface wind magnitude and direction (greyscale and arrows; QuikScat descending pass, 18LT). Mean (contours) and MAX-MIN composite difference (colors) in c) MODIS-derived cloud top height, d) MODIS-derived cloud depth, e) MODIS cloud fraction and f) CERES top-of-atmosphere shortwave fluxes. All MODIS data is Terra daytime level 2. Location of Antofagasta indicated by a black square, and Arica Bight domain is shown in Fig. 2.6 e and f.
2.4.2 Radiosonde

Antofagasta (23.43°S, 70.43°W) is near the location of maximum N_d variability (Fig. 2.1) and composite changes in the satellite-derived cloud properties are representative of those for the Arica Bight region (Fig. 2.3). Composites of radiosondes from Antofagasta (1200 UTC or 0800 LT) help us interpret the satellite composites (Fig. 2.7). These show a strengthening of the inversion temperature for the MAX N_d composite (Fig. 2.7a, solid black line), with the MAX N_d radiosonde composite possessing a warmer and drier free troposphere than the MIN N_d composite (solid gray line). Composite differences in boundary-layer moisture and in the zonal wind are small and not necessarily significant.

The corresponding Reanalysis profiles, shown for a location approximately 500 km offshore Antofagasta, depict the boundary layer weakly (Fig. 2.7 dashed lines). The vertical placement of the temperature and moisture inversion is approximately correct, but the underestimate of the inversion strength is pronounced, and the zonal winds not captured well. In one important feature, however, the Reanalysis agrees with the radiosondes in the sense that the MAX N_d Reanalysis composite also depicts a warmer and drier free troposphere than the MIN N_d Reanalysis composite. This is difficult to explain other than as increased free-tropospheric subsidence for the MAX N_d cases.
Figure 2.7: Antofagasta radiosonde (solid) and NCEP reanalysis (dashed) MAX-N<sub>d</sub> (black) and MIN-N<sub>d</sub> (grey) composites of a) temperature, b) mixing ratio, and c) zonal winds. Antofagasta (23.5°S, 70.5°W) radiosondes launched at 1200 UTC (0800 LT). NCEP-NCAR reanalysis data from 22.5°S, 75°W.

2.5 Regional Circulation

The spatial structure of the 850 hPa anomaly fields (geopotential height, subsidence and winds) is shown explicitly for the two composites in Fig. 2.8. The MAX N<sub>d</sub> composite is characterized by weaker subsidence offshore (negative anomalies in dp/dt, blue color), an anomalous trough (contours) west of 85°W, and northerly winds. In contrast, the MIN-N<sub>d</sub> composite presents an anomalous ridge and enhanced offshore subsidence. This pattern is consistent with the changes in the climatological anticyclone shown in Fig. 2.6b, and the subsidence changes suggested at Antofagasta in Fig. 2.7.
The important meteorological parameters governing stratocumulus cloud behavior at daily/synoptic time scales are not well-established (e.g., Klein, 1997; Zhang et al., 2009), although lower tropospheric stability suggests itself as a sensible proxy (e.g., Mauger and Norris, 2007; Matsui et al., 2006). We first evaluated the applicability of static stability as a proxy for cloud cover at daily/synoptic timescales. The static stability defined as the difference between potential temperature (θ) at 850 hPa and 1000 hPa, was found to correlate better with MODIS cloud fraction than θ_{700hPa} – θ_{1000hPa}, reflecting perhaps the proximity of the inversion base to the 850 hPa level. A spatial map of the correlation at daily time scales between θ_{850hPa} – θ_{1000hPa} and MODIS cloud fraction (Fig. 2.9) shows the highest positive correlation at 85°W and 25°S (r = 0.55), higher than that reported for the northeast Pacific at 30°N, 140°W using θ_{700hPa} – θ_{surface} (r=0.22; Klein, 1997). The correlation is insignificant over the Arica Bight, expected based on Fig. 2.7.
Figure 2.9: Linear correlation between October daytime Terra MODIS cloud fraction and daily-mean $\theta_{850\text{hPa}} - \theta_{1000\text{hPa}}$. Correlation values greater than 0.20 below 15° S pass the 99% significance level of a Student’s t test.

Figure 2.10. October-mean $\theta_{850\text{hPa}} - \theta_{1000\text{hPa}}$ (contours) and MAX-MIN $N_d$ composite differences in $\theta_{850\text{hPa}} - \theta_{1000\text{hPa}}$ (color). Arica Bight domain contoured in grey.
Fig. 2.10 shows mean values of $\theta_{850\text{hPa}} - \theta_{1000\text{hPa}}$ (contours) along with the composite differences (colors). The largest static stability difference between the two composites occurs near the southern boundary of the stratocumulus deck, at approximately 30°S, 75-77°W, slightly east of the highest composite differences in cloud fraction (Fig. 2.6e). The stability changes are large in the same general area where the correlation between cloud fraction and static stability is high (Fig. 2.9), rationalizing the choice of NCEP-defined static stability as a proxy for cloud fraction at daily timescales.

At short timescales, the static stability variability is governed by the variability in $\theta_{850\text{hPa}}$ rather than near-surface, and we can explore its relationship to the other meteorological fields through a one-point linear correlation analysis that does not rely on the $N_d$ compositing. The 850 hPa potential temperature time series at 20°S and 75°W serves as the reference point (Fig. 2.11 filled square) and is correlated with the 850 hPa geopotential height, subsidence and wind field time series at all other reanalysis grid points in Fig. 2.11. The one-point correlation analysis reveals that an increase in the reference 850 hPa temperature is associated with a local increase in anomalous northerly winds (actually a weakening of the southerly winds), as well as increases in the 850 hPa geopotential height further west between 75°-85°W. The relationship of the geopotential height anomaly field to the anomaly winds and subsidence are those of an anomalous trough to the west of South America. Fig. 2.11 suggests synoptic changes in the southeast Pacific stratocumulus deck reflect mid-latitude intrusions rather than equatorial intrusions. An example of the opposite behavior is discussed in Wyant et al. (2010) for mid-October, 2006, when enhanced southerly winds above the inversion corresponded to a decrease in cloud coverage and increase in a boundary-layer deepening at 85°W, 20°S.
At the reference point in Fig. 2.11, increases in the 850 hPa geopotential height to its west are associated with a local weakening of the southerly 850 hPa winds, reducing free tropospheric cold temperature advection, and allowing the 850 hPa temperature to rise. Drawing on Fig. 2.9, the elevated static stability corresponds to an increase in cloud cover.

Figure 2.11: One point correlation map between the potential temperature at 20°S and 75°W (square) and: subsidence (pressure velocity dp/dt, colors; positive values imply subsidence), geopotential height (contours) and wind (arrows), all at 850 hPa. Absolute values of correlation higher than 0.25 for the geopotential height and subsidence fields pass the 99% significance level of a Student’s t test. The winds are only shown if the meridional component of the wind-temperature correlation is statistically significant. Topography higher than 1500 m is indicated by the black shade.

Perhaps counterintuitively, the increased cloud coverage at 85°W during the MAX-$N_d$ composite is associated with weaker subsidence further offshore and increased
subsidence at 20°S and 72°W and near the coast. The increase in free-tropospheric subsidence near the coast is consistent with the radiosondes at Antofagasta.

While an increased subsidence-decreased cloud fraction relationship is not necessarily surprising at short time scales (e.g., Zhang et al., 2009; Mauger and Norris, 2010), it does run counter to intuition built from monthly-mean analyses (e.g., Bony and Dufresne, 2005).

Warmer temperatures during the MAX Nd composite relative to the MIN Nd composite extend fully throughout the troposphere within the NCEP Reanalysis (not shown), indicating a quasi-barotropic structure with an anomalous anticyclonic circulation. The stronger coastal subsidence during MAX Nd cases north of 25°S are reminiscent of the shallow, warm-core-low pressure cells of one-three day duration known as coastal lows (e.g., Garreaud et al., 2002; Garreaud and Rutllant, 2003). However, a coastal trough in sea level pressure is not apparent in our MAX Nd composite (Fig. 2.6a and b), the easterlies at Antofagasta remain weak in comparison to the coastal-low easterlies identified in Huneeus et al. (2006), and the region south of 25°S is marked by an increase in cloud coverage, rather than a decrease. In addition, although more of our MIN Nd episodes last only one day compared to the MAX Nd cases, the number of episodes lasting three or more days is the same for both composites (five), which is longer than a typical coastal low event. Nevertheless, our high/low Nd composites in some ways resemble the ending/leading edge of coastal lows, and the common occurrence of both suggests there must be some common associations.

In contrast to coastal lows, cutoff-lows are upper-level low pressure centers with a quasi-barotropic structure (e.g., Fuenzalida et al., 2005) and thus seem similar to our MIN
N\textsubscript{d} composite. In addition, cutoff-lows can be preceded by quasi-stationary ridging reminiscent of our MAX N\textsubscript{d} composite, with a duration exceeding that of the coastal lows. Cutoff-lows could be considered more intense manifestations of the upper-tropospheric potential vorticity perturbations identified through our composites. Although beyond the scope of the current work, the connection between cut-off lows and cloud property composites seems worthy of further investigation.

Previous studies have associated weaker surface winds at 33\textdegree S and 73\textdegree W with reduced cold surface temperature advection, in turn reducing cloud liquid water path and cloud fraction north of 20\textdegree S (Xu et al., 2004; Muñoz and Garreaud 2005; Wood et al., 2008). This contrasts with the finding that weaker alongshore winds co-occur with an increase in offshore cloud cover. The explanation may lie in our greater focus on submonthly synoptic activity and analysis of only October months, whereas the previous studies examined longer time spans that probably also captured seasonal changes. Our compositing appears to preferentially select for lifestages of mid-latitude baroclinic waves intruding upon the stratocumulus deck.

2.6 Precipitation Characteristics

Precipitation characteristics of the two composites were investigated using CloudSat reflectivity-height distributions constructed for a coastal and an offshore location. The coastal region encompasses 17\textdegree-27\textdegree S and 70\textdegree-80\textdegree W, an area slightly broader than the Arica Bight. The MAX N\textsubscript{d} reflectivity-height distribution from CloudSat is narrower, centered near -25 dBZ and 1000 m, than the MIN N\textsubscript{d} distribution (Fig. 2.12 a and b). For both composites, however, the most frequently occurring reflectivities are
around -25 dBZ with heights between 800 m and 1200 m. Few pixels have reflectivities > 0 dBZ, equivalent to a cloudbase rain rate of ~ 2 mm day⁻¹ (Comstock et al., 2004). Approximately one-sixth and one-third of the MAX/MIN N_d pixels exceed -17 dBZ (Table 2.1), equivalent to a cloudbase rainrate of 0.01 mm day⁻¹. The low radar reflectivities for both composites near the coast can be expected from the thin mean coastal cloud depths (Fig. 2.6d, contours). The slightly higher reflectivity values for the MIN N_d distribution are consistent with the deeper clouds and higher cloud tops and do indicate some drizzle, but for the MAX N_d distribution, hypotheses for cloud thinning based on depletion through precipitation seem discouraged (by the lack of observed precipitation). For the coastal clouds, little cloud thinning from precipitation is anticipated, based on previous work that clouds do not become sub-adiabatic until liquid water paths reach ~ 150 g/m³ and the frequency of occurrence of cloud radar reflectivities > 0 dBZ reaches 20% (Zuidema et al. 2005; Fig. 10). This finding will need to be modified for coastal conditions, however, to account for higher N_d and stronger cloud top entrainment.

<table>
<thead>
<tr>
<th>Domain</th>
<th>dBZ ≥ -17</th>
<th>dBZ &gt; 0</th>
</tr>
</thead>
<tbody>
<tr>
<td>MAX N_d</td>
<td>17° - 27°S and 70°- 80°W</td>
<td>973 pixels / 12.6 %</td>
</tr>
<tr>
<td>MIN N_d</td>
<td>17° - 27°S and 70°- 80°W</td>
<td>13611 pixels/ 37.4%</td>
</tr>
<tr>
<td>MAX N_d</td>
<td>20°- 30°S and 80°- 90°W</td>
<td>11596 pixels /42.9 %</td>
</tr>
<tr>
<td>MIN N_d</td>
<td>20°- 30°S and 80°- 90°W</td>
<td>26006 pixels /55.4 %</td>
</tr>
</tbody>
</table>

Table 2.1: Number and percentage of CloudSat pixels below two km of height, at two separate regions, above two different reflectivity thresholds.
Figure 2.12: CloudSat reflectivity-height distributions for 17˚S-27˚S and 70˚W-80˚W a) MAX $N_d$, b) MIN $N_d$; and 20˚S-30˚S and 80˚W-90˚W c) MAX $N_d$, d) MIN $N_d$. The colors indicate the percentage of the total pixels at each altitude; their summation over reflectivity equals total cloud fraction at that altitude. The dashed line indicates mean MODIS-derived cloud top height. Note different y-axis ranges for top and bottom panels.

The offshore CloudSat reflectivity-height distribution was constructed for the offshore region with the most pronounced change in cloud fraction between the two composites, defined by 20˚-30˚S and 80˚-90˚W (Fig. 2.12 panels c and d). A greater percentage of the offshore pixels were precipitating (reflectivities higher than -17 dBZ), consistent with Leon et al. (2008). The substantial increase in offshore cloud fraction for the MAX $N_d$ composite is associated with only a slight shift towards lower radar reflectivities (Table 2.1). In addition, the percentage of pixels $> 0$ dBZ is approximately
the same for both offshore composites. Moreover, we note the minor change in cloud top height between the offshore MAX and MIN N_d composites. This is consistent with some cancelling in the vertical and horizontal temperature advections (i.e. less subsidence but also less free-tropospheric horizontal cold temperature advection).

2.7 Plausibility of a diurnal cycle in N_d

In this section I explore the plausibility of a diurnal cycle over Arica Bight using MODIS data. It is important to clarify that the use of the diurnal term is arbitrary because 3-4 hours difference between TERRA and AQUA pass cannot resolve the diurnal features, although they could suggest diurnal variations in the cloud microphysics.

We first compute the difference in MODIS N_d between TERRA and AQUA for MAX cases (Fig. 2.13a) and MIN cases (Fig. 2.13b). For the MAX composite differences, it is clear an increase of N_d along the coast for TERRA pass, with maximum differences near 70 cm^{-3}. For the MIN composite, TERRA and AQUA N_d are indistinguishable, although the map suggests slightly higher values for TERRA (<20 cm^{-3}). Since N_d is proportional to the product of \( \tau^{0.5} \) and \( r_c^{-5/2} \), we also compared MAX TERRA/AQUA differences for these two variables to determine how \( \tau \) and \( r_c \) control the N_d inter-satellite differences (Fig. 2.14). We found that \( \tau^{0.5} \) is generally larger during TERRA pass than AQUA, with the largest differences offshore. For \( r_c^{-5/2} \) difference maps (Fig. 2.14b), the spatial pattern is more complex, with smaller values offshore for TERRA but with larger magnitudes along the coastal line. This spatial pattern in \( r_c^{-5/2} \) resembles the structure of N_d in Fig 2.13a, although the area with higher TERRA \( r_c^{-5/2} \) presents less westward extension than the N_d differences. These maps indicate that
changes in $N_d$ for the pixels right next to the coast, is modulated by the term $r_e^{-5/2}$. Thus, the afternoon increase in $r_e$ (decrease in $r_e^{-5/2}$) might not necessarily be associated with a microphysical mechanism; instead, it could be the consequence of a decrease in LWP, due to solar radiation absorption, that increases the probability of clear-sky contamination in the retrievals. This hypothesis seems to be supported by a decrease of $\tau$ during the afternoon (AQUA), even though these changes are small near the coast. Despite a possible satellite bias near the coastal line, it is encouraging that a few degrees further west, where TERRA $N_d$ is still high, $\tau^{1/2}$ differences are positive and $r_e^{-5/2}$ differences are negative (positive $r_e$ differences). These changes in $r_e$ and $\tau$ offshore are not consistent with the typical effect of broken clouds in the retrievals; moreover, the magnitude of the differences for $r_e$ and $\tau$ show that $\tau$ also modulates the $N_d$, with increases in $N_d$ for TERRA associated with increases in $\tau$, allowing one hypothesize that the afternoon decrease of $N_d$ is not completely explained by a retrieval artifact.

Figure 2.13: Mean $N_d$ differences between TERRA and AQUA: a) MAX cases, b) MIN cases.
Figure 2.14: Mean differences between TERRA and AQUA for MAX cases only: a) $\tau_{TERRA}^{0.5} - \tau_{AQUA}^{0.5}$ cases, b) $r_{e_{TERRA}}^{-5/2} - r_{e_{AQUA}}^{-5/2}$ cases.

2.8 Discussion of Chapter 2

I have applied satellite retrievals to understand how synoptic conditions can change cloud droplet number concentrations. We focused on October months only, to reduce influences from the seasonal cycle. A one-year evaluation of the seasonal cycle in cloudiness at San Felix Island (26S, 80W), located near the region with the largest cloud fraction changes (Fig. 2.6e), also found large variations in cloud cover and meso-scale cellular structure for this month, which has weaker baroclinicity than June-September but more pronounced baroclinicity than Dec.-March (Painemal et al., 2010). Comparisons between ship-based and MODIS-derived cloud depth estimates and of ship-based aerosol concentration to MODIS-derived $N_d$ values give confidence in the satellite retrievals.

Episodes with high $N_d$ over the Arica Bight are associated with a weaker anticyclone, weaker surface and free-tropospheric winds, and thinner clouds, also shown by Wood et al. (2008) and George and Wood (2010). We also find higher cloud droplet numbers are associated with more stable atmospheres, similar to Mauger and Norris
(2007) and Matsui et al. (2006). In addition, we show that higher $N_d$ values over the Arica Bight are concurrent with lower cloud top heights along with stronger inversion temperatures at Antofagasta (Fig. 2.7). Smaller differences in $N_d$ occur near 18°S, where the wind speeds are smaller and less variable, allowing aerosols to stagnate. Changes in top-of-atmosphere shortwave fluxes show a spatial gradient, with thinner clouds near the coast dominating a reduction in shortwave reflectance, while 400 km offshore, the increase in cloud droplet number accounts for a slight increase in shortwave reflectance (Fig. 2.6f).

Changes in the easterlies (from radiosondes) at Antofagasta are not significant between the MAX and MIN $N_d$ composites, and do not suggest significant changes in advection of continental aerosols to the stratocumulus deck. In addition, the stronger inversion and thinner clouds associated with larger $N_d$ along the coast would discourage entrainment of free-tropospheric aerosols. MAX $N_d$ days often occur in 2-4 days groups, during which the enhanced stability furthers multi-day aerosol transport within the boundary layer from the south, in evidence in Wood et al. (2008). This also allows high $N_d$ conditions to correlate well with enhanced stability from previous days, similar to Mauger and Norris (2007).

Huneeus et al. (2006) analyzed strong easterly events at Antofagasta (700 hPa zonal winds $> 5$ m s$^{-1}$) during austral winter, finding a connection between one episode of upper-level easterlies and the increase of $N_d$. This suggested a connection to two important copper smelters, Chuquicamata (22.3°S, 68.9°W) and Potrerillos (26.4°S, 69.5°W), that are both above the boundary layer (2700 m and 2850 m a.s.l. respectively). In this study we did not find any event with easterly winds of that magnitude; possibly
they are sporadic during austral spring when the midlatitude weather disturbances are less intense.

Approximately one-fourth of potential daily cases were excluded because they did not satisfy our criteria for overcast conditions over the Arica Bight. Given that thinner clouds are associated with the high-\(N_d\) composite, the days with low cloud cover over the Arica Bight may be more likely to resemble the high-\(N_d\) cases. Further south, around 30°S, these days are associated with increased cloud cover, a deeper boundary layer and deeper clouds, and resemble the aftermath of the coastal lows described by Garreaud et al. (2002) and Garreaud and Rutllant (2003). The leading edge of the ~2 day coastal low events encourage conditions conducive to pollution trapping in Santiago, Chile (33.3°S, 70.5°W), as well as easterly winds capable of advecting aerosol-rich air out to sea. The synoptic conditions that encourage coastal lows are similar to those of our MIN \(N_d\) composites, though the coastal low troughing in sea level pressure is not as apparent in our composite sea level pressures (Fig. 2.6 a and b). Coastal lows must undoubtedly impact the southerly manifestation of the high/low \(N_d\) composites discussed here.

Further offshore along 85°W, a large increase in cloud fraction and top-of-atmosphere shortwave fluxes is evident in the MAX \(N_d\) composite. This is synoptic, driven by mid-latitude baroclinicity rather than from the equator. As revealed in the anomalies at 850 hPa (Fig. 2.8) and the one-point correlation map (Fig. 2.11), an anomalously warm 850 hPa temperature near the coast is associated with an offshore trough-like pattern that enhances coastal subsidence but encourages anomalous free-tropospheric ascent offshore. Anomalous northerly winds along 85°W allow the above-inversion temperature to increase and increase the static stability, which is well-correlated
with cloud fraction at this location. A similar synoptic episode is discussed in Wyant et al. (2010), but with a reduction of offshore cloud fraction associated with a deeper boundary layer and stronger southerlies. It is also useful to note that the increased cloud cover is associated with decreased subsidence. Yet, as is apparent in Fig. 2.6, the increased cloud cover is associated with widespread decreases in cloud top height, because the anomalous horizontal warm temperature advection above the inversion is more than compensating for the decrease in subsidence. Numerical simulations during VOCALS-REx period (October-November 2008) indicate that the horizontal advection of the marine boundary layer height is larger than the vertical velocity at the top of the boundary layer, stressing the importance of the changes in the southerly winds over the subsidence (Rahn and Garreaud, 2010). The most significant features of the circulations associated with the two composites are highlighted in a schematic (Fig. 2.15).

An offshore albedo increase from the increase in cloud cover, along with an albedo decrease from near-coastal cloud thinning, means that a spatial gradient in albedo along 20°S would be difficult to detect in some VOCALS-REx aircraft flights (Chapter 4). Since several of the MAX/MIN cases occurred within a day of VOCALS-REx C-130 flights (see Fig. 2.16), our analysis provides context for the VOCALS-REx observations. Our Oct. 2008 MAX N_d cases ended when a mid-latitude trough moved through the region, while the Oct. 2008 MIN N_d cases loosely coincided with baroclinic troughs (Rahn and Garreaud, 2010). The LWP- N_d correlation over the Arica Bight was negative (r= -0.27), as expected, but contained significant day-to-day variability that can mask the correlation evident within a larger data sample. This explains a positive LWP-N_d correlation observed in Twin Otter VOCALS-Rex measurements at Point Alpha (75W,
20S; Zheng et al., 2010); nevertheless, satellite bias associated with thin and broken clouds near the coast, can also be responsible of a positive LWP-$N_d$ correlation.

Figure 2.15: Schematic of a) MAX $N_d$, and b) MIN $N_d$ large-scale meteorology. The 500hPa geopotential height is indicated by the dashed line, the sea-level pressure by red contours, subsidence by the shaded area, 850 hPa and surface winds by grey and black arrows respectively. The color shading over Arica Bight indicates the region with large $N_d$. 
Several avenues for further work present themselves. The implications of our data for albedo susceptibility $d(A)/d(N_d)$ will be further explored in section 4, with more accurate retrievals during VOCALS-REx. Since both satellite cloud top height and inversion temperature indicate less favorable conditions for aerosol entrainment during MAX $N_d$, we speculate that the main source of anthropogenic aerosols must be situated within the boundary layer. This hypothesis implies the contribution of several sources alongshore and perhaps includes non-point-source pollution that has been brought out to sea, more likely from the more developed southern region of Chile (see also George and Wood, 2010). The poor reanalysis representation of the Chile-Peru coastline discouraged us from using back-trajectories to study the time history of cloud parcels, however (e.g., Mauger and Norris, 2007). New reanalyses (e.g., Year of Tropical Convection; ERA-Interim; NCEP Climate Forecast System Reanalysis and Reforecast) with better resolution of the orography and of the physical processes will allow for more confident investigations of near-coastal aerosol-cloud interactions. Further investigation of the connection of our composites to the synoptic climatology of the southeast Pacific will
also help us better determine the impact of future climates upon this region, arguably the largest subtropical stratocumulus deck on the planet.
Chapter 3

Validation of MODIS cloud effective radius and optical thickness during VOCALS-REx

3.1 Background

In Chapter 2, we find a good qualitative agreement between MODIS-derived variables (Nd and LWP) and ship-based observations collected by six NOAA cruises prior to VOCAL-REx (aerosol concentration and cloud geometrical thickness). This preliminary validation shows that MODIS retrievals provide valuable physical information over the stratocumulus region, giving further support to the findings in Chapter 2. Nevertheless, the lack of microphysical observations during the NOAA cruises prevented us from a more detailed assessment of the MODIS retrievals accuracy, their limitations, and their dependence on the cloud vertical structure and the drizzle occurrence. In this chapter, we address the problem of quantitatively determining the accuracy of MODIS retrievals over the Chile-Peru stratocumulus regime, taking advantage of the comprehensive microphysical observations collected during VOCALS-REx (Chapter based on Painemal and Zuidema, 2011).

Satellite retrievals that rely on visible and near-infrared radiances are instrumental for understanding the role of the cloud system in the Earth’s radiation budget and the global climate. Cloud optical thickness (τ) and effective radius (r_e) are two commonly visible/near-infrared retrieved variables, and generally estimated with a broad spatial and
temporal coverage. \( r_e \) and \( \tau \) can be combined to form liquid water path (LWP), expressed as the proportional product of \( r_e \) and \( \tau \), if assumptions are made about the vertical structure of the liquid water content. LWP and \( r_e \) can be applied further to compute satellite-based drizzle proxies and cloud droplet number concentration (e.g. Bennartz, 2007; Wood et al., 2008). \( r_e \) and \( \tau \) are also valuable because they provide a simple way to parameterize cloud optical properties in radiative transfer models (e.g. Hu and Stamnes, 1993; Chou et al., 1998).

The retrievals algorithm is typically based on a spectral reflectance method that uses information of a visible (VIS) non-absorbing band to calculate the cloud optical thickness, and a near-infrared (NIR, slightly absorbing) band to calculate \( r_e \), providing quasi-independent measurements of each parameter (Twomey and Seton, 1980; Nakajima and King, 1990). Despite the extended use of satellite \( r_e \), \( \tau \), and their derived variables, validation of the algorithms and a better understanding of their limitations remain an important challenge. This aspect is paramount as the necessity of counting on reliable dataset for climate regional models validation demands major efforts to determine the retrievals representativeness of the cloud features.

A deeper understanding of the satellite retrievals quality can be gained when in-situ observations are available. In early works, remotely sensed \( r_e \) and \( \tau \) trended well with observations, but with an evident overestimate of \( r_e \) by about one to two microns (Nakajima et al, 1991; Nakajima and Nakajima, 1995). This bias was rationalized as the consequence of an “anomalous absorption” not reproduced by the radiative transfer models, though more recent studies did not find evidence of this model misrepresentation (e.g. Ackerman et al., 2003).
Source of errors that affect the satellite retrievals have been amply described in the literature, encompassing dissimilar processes including: three-dimensional radiative effects (e.g. Zuidema and Evans, 1998; Marshak et al., 2006; Kato and Marshak, 2009); subpixel clear sky contamination; and solar satellite viewing geometry (Varnai and Marshak, 2007; Maddux et al., 2010). Additional source of errors are associated with uncertainties in physical quantities such as: atmospheric corrections due to water vapor and thermal emission; surface albedo; and assumptions about the cloud vertical structure and the shape of the droplet size distribution (Platnick and Valero, 1994).

Retrievals errors attributed to physical uncertainties can be analyzed further with in-situ observations. Since the exact vertical structure of the cloud is unknown, the retrieval technique assumes a vertically homogeneous cloud, even though observational evidence indicates that marine boundary layer clouds exhibit a stratified (adiabatic-like) structure (e.g. Martin et al. 1994). If the cloud is stratified, the retrieved $r_e$ might be 5-10% lower than the actual $r_e$ at the cloud top (Nakajima et al., 1991). Additionally, a shape of the droplet size distribution must be assumed along with a value for the effective variance of the droplet size distribution. Different values of the effective variance can affect the final retrieval of $r_e$ in magnitudes as large as 15%, with an overestimate of $r_e$ if the prescribed effective variance used in the look-up tables is larger than the actual variance (Platnick and Valero, 1995).

The validation of satellite-derived $r_e$ with in-situ observations is challenging because in-situ cloud probes are also prone to instrument artifacts. For example, during the Atlantic Stratocumulus Experiment, satellite retrievals from Advanced Very High Resolution Radiometer and aircraft observations showed a reasonable agreement, but a
more quantitative assessment was hindered by the uncertainties of the in-situ probes (Platnick and Valero, 1995).

In-situ/satellite comparisons also need to take into account that the photon vertical penetration in the cloud varies at different NIR bands (1.6, 2.1 and 3.7 µm for MODIS). Platnick (2000) showed that the 3.7 µm wavelength retrieval captures properties closer to the cloud top, while 2.1 and 1.6 µm radiances contain more contribution from lower cloud layers. Thus, for typical stratocumulus clouds, that is, liquid water content adiabatically distributed and \( r_e \) increasing with height, the retrieved \( r_e \) at 3.7 µm would be larger than the 2.1 µm-based \( r_e \), and the latter larger than the 1.6 µm-based \( r_e \).

Nevertheless, global surveys indicate that the retrieved MODIS \( r_e \) at 2.1 and 1.6 µm are considerably larger than the 3.7 µm \( r_e \) (Nakajima et al., 2010). Sensitivity analysis with a two-layer cloud suggests that this global trait could be explained by drizzle that can modify the cloud vertical structure, and/or cloud top mixing that reduces the cloud effective radius near the cloud top (Nakajima et al, 2010).

A regional investigation over the Southeast Pacific depicts a complex pattern in terms of MODIS \( r_e \) differences associated with the three NIR wavelengths, with small difference between 3.7 and 1.6 µm \( r_e \) retrievals, and a seasonal and regional dependent bias (Seethala and Horvath, 2010). As also shown here in Fig. 3.1, the 2.1 µm retrieved \( r_e \) (\( r_{e2.1} \)) exceeds both the 1.6 µm (\( r_{e1.6} \)) and 3.7µm (\( r_{e3.7} \)) \( r_e \), with the differences decreasing near the coast. This same coastal area also has smaller \( r_e \), liquid water path, and shallower boundary layer, with reduced drizzle (Zuidema et al., 2009; Painemal and Zuidema, 2010) and typical adiabatic-like clouds. Further offshore \( r_e \) differences are more
significant (>1 µm), and might suggest the influence of drizzle in the retrievals, as proposed by Nakajima et al. (2010).

![Figure 3.1: Terra mean differences during VOCALS-REx period: a) \( r_{e2.1} - r_{e1.6} \) and b) \( r_{e2.1} - r_{e3.7} \).](image)

As mentioned previously, we investigate in this chapter the ability of Moderate Resolution Imaging Spectroradiometer (MODIS) retrievals to represent the microphysical
properties of the southeast Pacific stratocumulus deck. We compared MODIS estimates at approximately 10:30 LT (Terra satellite) and 13:30 LT (Aqua satellite) with aircraft observations collected during the Variability of the American Monsoon Systems' (VAMOS) Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx), in October-November 2008, off the coast of Chile. This quasi-permanent stratocumulus regime is ideally suited for satellite remote sensing, presenting a unique opportunity to test satellite retrievals using in-situ measurements collected by state-of-the-art airborne cloud probes.

3.2 Data and Methodology

3.2.1 General description of VOCALS-Rex

VOCALS REx was “an international field experiment designed to better understand physical and chemical processes central to climate system of the Southeast Pacific” (Wood and Mechoso, 2008). Specific goals of VOCALS encompass the study of stratocumulus cloud-aerosol interactions and their effect over the cloud microphysics and precipitation, as well as the investigation of the coupled ocean-atmosphere system. The experiment took place during October and November of 2008 and engaged over 150 scientists from 40 institutions and nations. A total of five aircraft, two research vessels, land sites and two instrument moorings operated during VOCALS-Rex, sampling the lower atmosphere and upper-ocean (Fig. 3.2). A more general goal of VOCALS is “to improve model simulations of key climate processes using the Southeast Pacific as a

---

4 This overview is mainly based on the VOCALS operation paper (Wood et al., 2011).
testbed, particularly in coupled models that are used for climate projections and ENSO forecasting” (Wood et al 2011).

![Diagram](image)

Figure 3.2: VOCALS-Rex study region showing main sampling platforms and mission types.

### 3.2.2 Aircraft observations

The main observational platform employed in this work was the NSF/NCAR aircraft C-130, equipped with a comprehensive set of probes that measured water content and droplet size distribution, at temporal resolution of 1 second, listed in table 3.1. We analyzed data from vertical profiles taken during thirteen near 8-hours daytime flights, during 10/15/2008- 11/15/2008, over an oceanic area within 70°W-80°W and 19°S-30°S, off the Chile coast, indicated in Fig. 3.3. The overall flights are representative of diverse conditions of the cloud deck: pristine and polluted days as well as days with variable LWP, drizzle occurrence, and diverse synoptic patterns (Painemal and Zuidema, 2010; Rahn and Garreaud, 2010). Climatologically, the region of study exhibits a significant
zonal gradient of cloud effective radius, with values of optical thickness that fluctuate between 10 and 15 (Fig. 3.3).

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>King hot wire probe</td>
<td>LWC</td>
</tr>
<tr>
<td>Cloud Droplet Probe (CDP)</td>
<td>Droplet size (2 ( \mu \text{m} &lt; \text{diameter} &lt; 52 \mu \text{m} )) 30 bins</td>
</tr>
<tr>
<td></td>
<td>• 1.18 ( \mu \text{m} ) bin-width for sizes &lt; 20 ( \mu \text{m} ).</td>
</tr>
<tr>
<td></td>
<td>• 2.28 ( \mu \text{m} ) bin-width for sizes &gt; 22 ( \mu \text{m} ).</td>
</tr>
<tr>
<td>Particle Measuring System’s Two-Dimensional Cloud optical array probe (2D-C).</td>
<td>Droplet size (25 ( \mu \text{m} &lt; \text{diameter} &lt; 1560 \mu \text{m} )) 64 bins: 25 ( \mu \text{m} ) bin-width</td>
</tr>
</tbody>
</table>

Table 3.1: Airborne instruments and measurements.

The C-130 probes capable of resolving droplet size distribution are: Cloud droplet Probe (CDP), the Forward Scattering Spectrometer Probe (FSSP-100), Particle Volume Monitor (PVM) 100 and Particle Measuring System’s Two-Dimensional Cloud optical array probe (2D-C). While the CDP, FSSP-100, and PVM-100 measure small diameter droplets, the 2D-C measures droplet sizes up to 1500 \( \mu \text{m} \), which makes it particularly useful for detecting drizzle. CDP, manufactured by Droplet Measurement Technologies, is a relatively new laser-based probe with two photo-detectors, in order to constrain the sample area (Lance et al., 2010). The CDP is thought to provide the best drop size data of the three particle probes (VOCALS Data quality report, available through http://data.eol.ucar.edu/master_list/?project=VOCALS). Therefore, we primarily rely on CDP probe to describe the cloud droplet spectra. We assess the performance of the CDP further by comparing it to the King LWC values, a hot-wire probe that is insensitive to how drops are sized.
Figure 3.3: Aircraft trajectories (magenta lines) and mean Terra MODIS level 2 effective radius (shaded) and optical thickness (contour) for overcast pixels during VOCALS-REx. Black dots indicate the location of the vertical profiles used in this study. The gray line corresponds to the coastal line.

3.2.3 Instruments inter-comparison

The CDP-derived LWC is calculated as follows:

\[
LWC = \frac{4\pi}{3} \rho_w \sum_{i=1}^{30} n_i \cdot r_i^3 \tag{3.1}
\]

Where \( \rho_w \) is the liquid water density, \( n_i \) the number of droplets (#/cm³) per bin i (30 bins), and \( r_i \) the center radius of the bin.

The comparison between King and CDP LWC indicates a flight dependent bias (Fig. 3.4a) with generally larger CDP LWC. This bias, attributed to the CDP probe, was removed by determining a linear regression between King and CDP LWC, and expressed as a simple linear fit: \( LWC_{CDP} = a \cdot LWC_{King} \), calculated for individual flights. The term
“a” (table 3.2) is then used to correct the CDP droplet size distribution, assuming that the bias in the CDP measurements is a droplet sizing error rather than a counting error. A calculation of a corrected LWC is equivalent to use a modified size bin ($r_{i}^{*}$) instead of $r_{i}$ in equation (3.1), that is: $r_{i}^{*} = \frac{r_{i}}{a^{1/3}}$.

<table>
<thead>
<tr>
<th>Research flights</th>
<th>a</th>
</tr>
</thead>
<tbody>
<tr>
<td>RF02</td>
<td>0.929</td>
</tr>
<tr>
<td>RF03</td>
<td>0.789</td>
</tr>
<tr>
<td>RF04</td>
<td>0.731</td>
</tr>
<tr>
<td>RF05</td>
<td>1.046</td>
</tr>
<tr>
<td>RF06</td>
<td>0.910</td>
</tr>
<tr>
<td>RF07</td>
<td>0.972</td>
</tr>
<tr>
<td>RF08</td>
<td>1.080</td>
</tr>
<tr>
<td>RF09</td>
<td>0.967</td>
</tr>
<tr>
<td>RF10</td>
<td>1.000</td>
</tr>
<tr>
<td>RF11</td>
<td>1.215</td>
</tr>
<tr>
<td>RF12</td>
<td>1.300</td>
</tr>
<tr>
<td>RF13</td>
<td>1.008</td>
</tr>
<tr>
<td>RF14</td>
<td>1.125</td>
</tr>
</tbody>
</table>

Table 3.2: Linear coefficients used to correct CDP droplet size.

Figure 3.4: a) CDP LWC versus King LWC, and b) Corrected CDP LWC versus King LWC for research flights 2 (RF02), 5 (RF05), and 12 (RF12).
The values of the microphysical parameters derived from CDP were also corrected accordingly. An example of corrected CDP LWC is presented in Fig. 3.4b.

The necessity of calculating vertically integrated quantities (LWP and $\tau$) reduces the usable dataset to the vertical profiles taken during the ascents and descents. We then calculate the $r_e$, $\tau$, and LWP by combining the droplet size distributions from CDP and 2D-C probes. Since the first bin of 2D-C probe (12.5 $\mu$m-37.5 $\mu$m of diameter) overlaps with the CDP largest bins, we did not include this first 2D-C bin into the calculations, reducing potential errors of 2D-C associated with undercounting of the smallest bins (Baumgardner and Korolev, 1997). $r_e$ is derived from eq. 3.2, where $n_i$ is the number of droplets (#/cm$^3$) per bin i (30 bins), and $r_i^*$ the corrected center radius of the bin. The mean differences between corrected and non-corrected CDP $r_e$ is small (0.17 $\mu$m), and does not affect the main finding of this chapter.

$$r_e = \frac{\sum_{i=1}^{30} r_i^* n_i}{\sum_{i=1}^{30} r_i^{*2} n_i} \quad (3.2)$$

LWP and $\tau$ were obtained by integrating liquid water content and the volume extinction coefficient ($\beta$) respectively, with $\beta$ defined as:

$$\beta = \sum_{i} \pi \cdot Q_e \cdot r_i^{*2} n_i \quad (3.3)$$

$Q_e$ is the extinction efficiency and is assumed equal to 2.

We derived $N_d$ from the CDP probe only, as the contribution of large droplets to $N_d$ is negligible. The cloud top and base of each profile were simply defined as the
highest and lowest levels with $N_d > 10 \text{ cm}^{-3}$ and CDP LWC $> 0.03 \text{ gm}^{-3}$. Typically, at one Hz sampling, the instruments are able to resolve a vertical resolution of 6 meters.

### 3.2.4 General features of the profiles

LWC, $r_e$, and $N_d$ profiles, normalized by the cloud top height ($Z_{\text{top}}$), are shown in Fig. 3.5. LWC values increase with height, with the largest values close to $0.8 \text{ g/m}^3$, and an apparently adiabatically-distributed vertical structure (Fig. 3.5a). Values of $r_e$ lie between 5 and 26 $\mu$m, though most of the profiles maxima are less than 15 $\mu$m (Fig. 3.5b), and $N_d$ profiles relatively homogeneous (Fig. 3.5c).

For inter-comparison purposes, each profile is normalized by its in-cloud maximum value ($\text{LWC}_{\text{max}}$, $r_{e\text{MAX}}$, and $N_{d\text{MAX}}$). Moreover, we normalize the height $Z_N = (Z-Z_{\text{base}})/\Delta Z$, where $Z_{\text{base}}$ is the cloud base height, and $\Delta Z$ the geometrical cloud depth. This transformation implies that $Z_N=1$ at the cloud top, and $Z_N=0$ at the cloud base. The normalized profiles for LWC, $r_e$ and $N_d$ are depicted in Fig. 3.6. The LWC profiles show the expected adiabatic-like profiles with maximum values near the cloud top ($Z_N=0.93$) and a sharp decrease above. Similarly for $r_e$, most of the profiles present a gentle increase of $r_e$ with height, but with maximum values at the cloud top ($Z_N=1$). Profiles of $N_d$ show a constant profile with height, consistent with the $r_e$ and LWC profiles.
Figure 3.5: Profiles of: a) LWC, b) $r_e$, and c) $N_d$.

Figure 3.6: Normalized profiles of: a) LWC, b) $r_e$, and c) $N_d$. Values of $Z_N=0$ indicates the cloud base whereas $Z_N=1$ the cloud top. Red lines indicate the median profiles.

3.2.5 MODIS data

MODIS level 2 retrievals (level 2, collection 5, Platnick et al. 2003), are provided at a 1 km x 1 km spatial resolution, and include $\tau$, derived from a 0.85 µm channel, and three estimates of $r_e$ obtained from 1.6 µm ($r_{e,1.6}$), 2.1 µm, and 3.7 µm ($r_{e,3.7}$) reflectance. The 2.1 µm $r_e$ corresponds to the standard MODIS $r_e$, because the water absorption at this channel is stronger than the 1.6 µm band and the thermal emission is small. Despite the
stronger water absorption at 3.7 µm, the retrievals at this wavelength are complicated by the thermal emission in the measured radiance. The cases studied here correspond exclusively to warm clouds (>0°C), confirmed by the MODIS cloud top temperature.

A difficulty of comparing aircraft profiles with MODIS observations is that the MODIS scenes are not exactly collocated in time with the observations. We therefore use MODIS pixels collocated within one hour from the aircraft vertical profile occurrence. The pixel location is then corrected by the advective distance covered by the cloud during the lapsed time between the satellite pass and the vertical profile occurrence, assuming a mean advective velocity given by the mean wind speed of each profile. The τ and r_e pixels are then averaged to a resolution of 5 km x 5km, centered at the corrected location of the collocated pixel. The 5 km averaged values are similar to the collocated 1 km resolution values, but the sample size is larger. Here we implicitly assume that the cloud deck does not undergo dramatic microphysical changes within one hour. The results presented in this chapter shows that this assumption is reasonable in most of the cases. In an attempt to reduce biases associated with broken clouds, we only compare cases with valid τ and r_e observations in at least 90 % of the pixels within the 5 km x 5 km domain (23 pixels out of 25). 20 MODIS scenes (Terra and Aqua) met the above criteria, coincident with 20 vertical profiles (black dots in Fig. 3.3).
3.3 In-situ and MODIS comparison

3.3.1 $r_e$ and $\tau$

We found relatively good agreement between satellite and in-situ measurements of $\tau$, with high correlations ($r = .83$) and a mean bias = 1.42, with larger values for MODIS $\tau$ (Fig. 3.7a).

The $r_e$ comparison requires the selection of a particular $r_e$ in the profile. As a reference, Platnick (2000) found that, for a particular stratified cloud with an optical thickness of 8, the satellite-retrieved $r_e$ would correspond to a $r_e$ associated with a cloud optical path (from the cloud-top) of 2 for the 3.7 $\mu$m reflectance, and 3.4 for the 2.1 $\mu$m reflectance. In our case, since the cloud top $r_e$ is a maximum in the profile, we adopt the use of an average $r_e$ of the 4 samples closest to the cloud top (equivalent to a geometrical cloud depth of 20-25 m). The comparison between MODIS (2.1 $\mu$m) and total in-situ cloud-top $r_e$ (Fig. 3.7b) shows a good correspondence ($r = 0.98$), though MODIS $r_e$ were systematically higher than the in-situ ones (mean bias of 2.1 $\mu$m), and a bias increasing with $r_e$.

We have highlighted in Fig. 3.7 the samples with absolute $\tau$ differences less than 2 (Fig. 3.7, gray circles). We are assuming that for these cases, the close match may reflect more optimal conditions for satellite retrievals. For this subset, the positive bias of MODIS $r_e$ is still apparent, suggesting a robustness of the MODIS $r_e$ positive offset.
Figure 3.7: Scatterplot between MODIS and in situ observations: a) cloud optical thickness b) cloud effective radius, and c) liquid water path. The bars indicate the MODIS standard deviation over the 5 km x 5km domain. Gray circles indicate those samples in which the absolute difference between MODIS and in-situ $\tau$ is less than 2.

3.3.2 1.6 µm, 2.1 µm, and 3.7 µm wavelength-dependent effective radius

We further compared the three MODIS $r_e$ retrievals, to evaluate if the retrieval differences could be physically explained by features of the in-situ vertical profiles (Fig. 3.8a). In general, $r_e$ at 2.1 µm ($r_{e2.1}$) tends to be larger than the estimates at 3.7 ($r_{e3.7}$) and 1.6 µm ($r_{e1.6}$) (Fig. 3.8a). The positive differences between $r_{e2.1}$ and $r_{e1.6}$ is consistent with the vertical structure of an adiabatically-distributed and non-precipitating cloud. In contrast, the positive differences between $r_{e2.1}$ and $r_{e1.6}$ are counterintuitive, as the in-situ vertical profiles did not show a decrease of $r_e$ near the cloud top (Fig. 3.8b). It is plausible that smaller $r_{e3.7}$ might be the consequence of incorrect modeling of the thermal emission, although some other factors cannot be discarded. Furthermore, the few cases with 2D-C (drizzle) LWP higher than 5 g/m² (crosses in Fig. 3.8a) do not show a clear tendency in MODIS $r_e$'s. At least for the type of clouds analyzed here, it is doubtful that changes in the three MODIS $r_e$ can effectively provide information about the vertical structure of $r_e$. 
As both MODIS 3.7 µm and 1.6 µm based \( r_e \) tend to be smaller than the operational MODIS \( r_e \), this might suggest that \( r_{e3.7} \) and \( r_{e1.6} \) can better agree with the observations. Nevertheless, a positive bias between the different MODIS \( r_e \) and the in-situ \( r_e \) remains regardless the \( r_e \) selected in the comparison (Fig. 3.8b).

![Figure 3.8](image.png)

**Figure 3.8:** a) In-situ \( r_e \) vs \( r_{e1.6} \) (black circles), \( r_{e3.7} \) (red circles) and \( r_{e2.1} \) (blue crosses). Crosses indicate cases with 2D-C LWP (drizzle) higher than 5 g/m². b) Scatterplot between in-situ \( r_e \) and \( r_{e1.6} \) (black circles), \( r_{e3.7} \) (red circles), and \( r_{e2.1} \) (blue crosses).

### 3.3.3 Selected cases

Three vertical profiles of LWC and \( r_e \), possessing MODIS \( r_e \) overestimates larger than 1.5 microns and varying amounts of precipitation, are shown in Fig. 3.9. These cases highlight that the MODIS overestimate occurs regardless of the vertical distribution of the drizzle amount or the cloud top entrainment.

The two profiles with little drizzle (Fig. 3.9a and b) share a similar water content profile but differing near the cloud top. While Fig. 3.9a shows relatively low LWP, with a maximum near cloud top, Fig. 3.9b exhibits relatively large LWP, with sharp decreasing water content near the cloud top, a typical signature of cloud top evaporation due to entrainment. For the \( r_e \) profiles however, the increase of \( r_e \) near the cloud top seems not to
be affected by the decrease in water content (Fig. 3.9b), a feature also seen more generally in Fig. 3.6. Although the nuances of the cloud top mixing are not investigated here, the constant increase of \( r_e \) at the cloud top, accompanied with the decrease of the water content, suggests the occurrence of inhomogeneous mixing, that is, an evaporation time scale that is slower than the mixing scale.

Analogous to Nov 02 case, the CDP LWC (red line) for the precipitating profile (Fig. 3.9c), also increases initially with height, then decreases near the cloud top, while \( r_e \) increases monotonically. The 2D-C LWC (blue line) shows precipitation close to 0.1 g/m\(^3\) equally distributed throughout the cloud, though with subtle increases near the cloud base. When adding the 2D-C droplet size distribution (Fig. 3.9f, blue line), \( r_e \) increases about 2 microns with respect to the CDP \( r_e \) (red line), with the most dramatic changes near the cloud-base.

To put the differences between MODIS and in-situ \( r_e \) in perspective, we have included the expected location of the representative \( r_e \) for 3.7, 2.1 \( \mu m \), and 1.6 \( \mu m \) wavelengths for an adiabatic cloud with \( \tau = 8 \), according to Platnick (2000), along with the respective MODIS \( r_e \) (black diamonds, crosses, and triangles respectively in Fig. 3.9d-e). As mentioned previously, the three MODIS \( r_e \) illustrates that the vertical structure is not the factor that modulates the bias. While \( r_{e3.7} \) may be less than \( r_{e2.1} \) because of the cloud top mixing, as proposed by Nakajima et al. (2010), in these examples it appears more indicative of a retrieval artifact rather than the actual vertical structure.
Figure 3.9: Vertical profiles for three selected cases: a)-c) CDP LWC (red line) and 2D-C LWC (blue line). d)-f) CDP $r_e$ (red line) and CDP+2D-C $r_e$ (blue line). Black diamonds, crosses, and triangles indicate the magnitude of MODIS 3.7 $\mu$m, 2.1 $\mu$m, and 1.6 $\mu$m $r_e$ respectively, along with the approximate theoretical location for a cloud with $\tau=8$, according to Platnick (2000). The profiles were normalized by the cloud top height. Black horizontal thick lines indicate the cloud base.
### 3.3.4 LWP and $N_d$

MODIS $r_e$ and $\tau$ are combined to calculate LWP, assuming that the liquid water content can be expressed as a linear function of the cloud depth:

$$LWP = \rho_w \frac{5}{9} r_e \cdot \tau \quad (3.4)$$

MODIS LWP based on equation 3.4 (Fig. 3.10a) is consistently larger than the in-situ one, with a mean bias = 12.3 g/m² and $\tau = 0.73$, a relatively high correlation when considering that LWP is indirectly derived from $\tau$ and $r_e$. Only one case, associated with large drizzle LWP (78.1 g/m²) and a high underestimate of MODIS $\tau$ (-3.5), has a retrieved LWP that is considerably smaller than the observed value. The overestimate of MODIS LWP is consistent with the finding in Chapter 2 that shows that the cloud depth derived from MODIS (adiabatic cloud depth) overestimates a ship-derived cloud depth. This chapter further points to the MODIS $r_e$ overestimate as the cause of the LWP overestimate.

Cloud droplet number concentration from satellites can be similarly calculated by combining $r_e$, $\tau$, under the same argument of linear LWC with respect to the cloud depth ($Z$):

$$LWC = \Gamma \cdot Z \quad (3.5), \text{ which implies:}$$

$$LWP = \Gamma \frac{\Delta Z^2}{2} \quad (3.6)$$

The parameter $\Gamma$ would correspond to the condensation rate of water vapor with height (Albrecht et al., 1990). The adiabatic $\Gamma$ is a function of temperature and pressure (weakly) only. The sub-adiabaticity of the cloud can be taken into account by multiplying the parameter $\Gamma$ by a sub-adiabatic fraction $< 1$ that reflects the dilution of the cloud due
to mixing (e.g. Bennartz, 2007). An additional assumption in the derivation of $N_d$ is that the cubic ratio between the volume mean radius and the effective radius, known as “$k$” parameter, is constant (Martin et al., 1994). If $N_d$ is assumed constant with height, $N_d$ can then be expressed as (Bennartz, 2007):

$$N_d = \Gamma_{\text{appr}}^{1/2} \frac{10^{1/2}}{4\pi \rho_w^{1/2} k} \cdot \frac{\tau^{1/2}}{r_e^{5/2}}$$  \hspace{1cm} (3.7)

$\rho_w$ is the water density and the parameter $k$ is assumed constant at 0.8 (Martin et al., 1994). $\Gamma_{\text{appr}}$ is the approximate water content lapse rate. Bennartz (2007) assumes $\Gamma_{\text{appr}}$ as the adiabatic lapse rate multiplied by 0.8 (sub-adiabatic fraction). In this study, we assume a value of 2 g/m$^3$/km for $\Gamma_{\text{appr}}$, which corresponds to a mean value for the range of variability of the adiabatic lapse rate over this region (1.8-2.2 g/m$^3$/km), but ignores the sub-adiabatic fraction. This value has also been used in Painemal and Zuidema (2010). Unlike the $N_d$ equation in Bennartz (2007), equation 3.7 is expressed in terms of $r_e$ and $\tau$, the extinction efficiency is assumed constant and equal to 2, and cloud fraction = 1. A similar equation was used in George and Wood (2010), and expressed as $N_{\text{eff}} = K \cdot \tau^{1/2} r_e^{-5/2}$, with $K$ constant at 1.125x10$^{-6}$ cm$^{-1/2}$. Their $N_{\text{eff}}$ is equivalent to calculate an effective $N_d$, defined as $N_d$ in equation 3.7 but multiplied by the $k$ parameter (that is: $N_{\text{eff}} = k \cdot N_d$), with the same constant values assumed in our study.

The calculated $N_d$ (using MODIS $\tau$ and $r_{c2.1}$) is compared to the mean profile CDP $N_d$ in Fig. 3.10b. MODIS $N_d$ agreed the best of the four MODIS variables with the aircraft observations ($r = 0.94$). This agreement seems to be independent of how well MODIS $\tau$ matches the observations, probably because the square root smoothens out the effect of $\tau$ in equation 3.7. Only the two samples with the highest $N_d$ values have a large
MODIS positive bias (>100 cm$^{-3}$). Their N$_d$ profiles reveal that these relatively polluted clouds are not well mixed (Fig. 3.10c). Another interesting feature is the morning-afternoon N$_d$ difference, with larger N$_d$ during the morning, suggested by both in-situ and satellite observations. This result gives support to the idea of a diurnal cycle, as it was also found in the MAX composites over Arica Bight in Chapter 2.

![Figure 3.10: a) Scatterplot between MODIS LWP and in-situ LWP, (b) Scatterplot between MODIS N$_d$ and in-situ N$_d$. Gray symbols as in Fig. 3.7. (c) CDP N$_d$ vertical profiles associated with the cases with the largest MODIS N$_d$ offset. Dashed lines indicate the MODIS N$_d$.](image-url)
The good agreement between MODIS and in-situ Nd is remarkable, especially if one considers the systematic overestimation in re. A plausible explanation may be found in the values used in \( \Gamma_{\text{appr}} \) and \( k \). If we assume that MODIS \( \tau \) accurately represents the real \( \tau \), then the potential error in Nd is explained by this term in eq. 3.7:

\[
\alpha = \frac{\Gamma_{\text{appr}}^{1/2}}{k \cdot r_{\text{eMODIS}}^{5/2}}
\]

(3.8)

We assign a value of 15% for the MODIS overestimate of \( r_e \) \((r_{\text{eMODIS}}=1.15 r_e)\), which corresponds to a reasonable value for the MODIS bias. Additionally, we calculated the observed lapse rate (\( \Gamma \)) as \( \Gamma = 2 \cdot LWP_{\text{CDP}+(2D-C)} / \Delta Z^2 \), analogous to equation 3.6. We found a mean \( \Gamma \) during VOCALS-REx of 1.4 g/m³/km, or equivalent to a ratio between the adiabatic \( \Gamma_{\text{appr}} \) \((2 \text{ g/m}^3/\text{km})\) and \( \Gamma \) equal to 1.43.

In terms of the parameter \( k \), its histograms (Fig. 3.11) show that the distribution of \( k \) depends on whether the selected \( k \) is at the cloud top (gray) or a profile-averaged \( k \) (thick black line). When considering the cloud-top \( k \), the distribution is narrow with a mean of 0.88. In contrast, the distribution is wider for the profile-averaged \( k \), with a mean of 0.8, consistent with Martin et al. (1994). Since the MODIS \( r_e \) is more representative of values closer to the top, a more appropriate \( k \) would also be at the cloud top, that is, \( k=0.88 \). This observed \( k \) \((k_{\text{obs}})\) would be then 10\% larger than the prescribed value used to calculate MODIS Nd in equation 7 \((k=0.9 k_{\text{obs}})\).

The three variables in \( \alpha \) can be expressed in terms of the observations \((\text{obs})\) as:

\[
\Gamma_{\text{appr}} = 1.43 \Gamma_{\text{obs}} \quad k = 0.9 k_{\text{obs}} \quad r_{\text{eMODIS}} = 1.15 r_{\text{eobs}}
\]

Expressing \( r_{\text{eMODIS}} \), \( k \), and \( \Gamma_{\text{ad}} \) in equation (3.8) in terms of the observed values, we get:
\[ \alpha = \frac{(1.43 \Gamma_{\text{obs}})^{1/2}}{0.9 k_{\text{obs}} \cdot (1.15 r_{e\text{obs}})^{5/2}} = 0.94 \frac{\Gamma_{\text{obs}}^{1/2}}{k_{\text{obs}} \cdot r_{e\text{obs}}^{5/2}} = 0.94 \alpha_{\text{obs}} \]  

(3.9)

\( \alpha_{\text{obs}} \) is analogous to \( \alpha \) but calculated from the observed values only.

This simple calculation shows that an a-priori selection of parameters based on convention allows a computation of MODIS \( N_d \) with a small bias (based on eq. 3.9, MODIS \( N_d \) is 94% of the actual \( N_d \)). In contrast, if realistic values are selected for \( \Gamma \) and \( k \), the satellite \( N_d \) would underestimate the observed value, due to the satellite \( r_e \) bias, by almost 30%. This analysis indicates that although the individual impact of the \( r_e \) bias is large, it is compensated for by systematic over/underestimates in both \( \Gamma \) and \( k \).

Figure 3.11: Histograms of \( k \) parameter at the cloud top (gray), and vertically-mean \( k \) (black contour bars).
3.4 Error Analysis

Thus far, we have investigated the association between MODIS $r_e$ and the cloud vertical structure; however, the causes of the satellite $r_e$ overestimate need to be more specifically addressed. In the following sections, we explore three different factors proposed in the literature as mechanisms that affect the $r_e$ retrievals: changes in the spread of the cloud droplet size distribution, the occurrence of a drizzle mode, and the effect of instrument viewing geometry. Errors associated with the water vapor path do not contribute to the MODIS positive bias because, as it is shown later on, the underestimated water vapor used in the MODIS algorithm, would lead to underestimates in $r_e$.

3.4.1 Precipitation and MODIS $r_e$

The necessity of prescribing a fixed log-normal droplet spectra within the retrieving algorithm, poses the question whether this assumption is appropriate for representing the optical properties of the clouds. While the droplet spectra standard deviation might be variable, a more fundamental problem arises when the algorithm monomodal droplet size distribution (cloud mode) is utilized for retrieving properties in precipitating clouds that are better represented by a bimodal droplet spectra. The investigation of the influence of the droplet size distribution shape in the retrievals requires then the identification of non-precipitating (monomodal) and precipitating (bimodal) cases. For addressing this, we complement our observations with the Wyoming 94 GHz cloud radar (David Leon, 2011; personal communication), because of its high sensitivity to large droplet sizes and larger sampling volume. We use the radar reflectivity near the cloud top and the maximum reflectivity within the cloud column, also at 1 Hz
resolution. We averaged 30 seconds reflectivity immediately before (ascent profiles) or after (descent profiles) the aircraft profile occurrence, and converted into dBZ units.

We first use the radar information to determine if the cloud probes undersample drizzle drop sizes. Fig 3.12a depicts the relationship between the radar reflectivity and the cloud probes reflectivity (derived from the sixth moment of the droplet size distribution). The scatterplot shows a relatively good agreement between radar and cloud probes cloud top reflectivity, with relatively higher values for the cloud probes reflectivity. Similarly, the agreement is also good for maximum values of the cloud column, although the higher magnitude of the cloud probes reflectivity is more apparent. Considering the substantial sampling techniques differences between the instruments, the cloud radar gives additional support to the accuracy of CDP and 2D-C probes.

The scatterplot between MODIS \( r_e \) and the radar reflectivities (Fig. 3.12b) shows the occurrence of three cloud stages: non-drizzling, light, and heavy drizzling clouds. For MODIS \( r_e < 12 \text{ \mu m} \), the radar reflectivity reaches values lower than \(-20 \text{ dBZ}\), with cloud top radar reflectivities coinciding with the maximum values of the cloud column, consistent with condensational growth dominating the cloud microphysics. In contrast, five MODIS \( r_e \) with values between 12 and 17 \text{ \mu m} have radar reflectivities between \(-18 \text{ dBZ} \) and \(-10 \text{ dBZ} \), with maximum reflectivity not occurring at the cloud top, a feature that might suggest that collision and coalescence processes have become active. This idea is supported by the 2D-C LWP that shows magnitudes larger than 2 g/m\(^2\) for the five selected cases (Fig. 3.12c, filled circles). These light drizzling clouds still present an adiabatically-distributed LWC, as it is also suggested in Fig. 3.6a and b. The case with the largest MODIS \( r_e \) is conspicuous through its high cloud reflectivity and large
difference between cloud top and maximum reflectivity; significant precipitation, is confirmed by the 2D-C LWP (78.1 g/m², Fig. 3.12c, upper right filled circle). Only one case, with MODIS $r_e$ at 22.5 µm and reflectivity lower than -18 dBZ, does not follow the positive correlation between MODIS $r_e$ and reflectivity, suggesting perhaps a LWP control in drizzle occurrence.

Figure 3.12: Radar reflectivity versus reflectivity calculated from the CDP and 2D-C probe at the cloud top (blue triangles) and the maxima of the profiles (open triangles). Only values larger than -30 dBZ are displayed. (b) MODIS $r_e$ vs maximum radar reflectivity (circles) and cloud top radar reflectivity (crosses). (c) MODIS $r_e$ vs 2D-C LWP. Filled circles indicate samples with cloud top reflectivity higher than -18 dBZ.

Our small dataset suggests that MODIS $r_e$ larger than 12 µm, associated with maximum reflectivity larger than -17 dBZ, indicates light drizzle. This reflectivity
threshold is consistent with Frisch et al. [1995] who found a reflectivity higher than –16 dBZ associated with drizzling clouds. Similarly, Wang and Geerts [2003] showed that in marine clouds the threshold varies between –19 dBZ and -16 dBZ, with Liu et al. [2008] suggesting a drizzle threshold dependent on the droplet concentration. Additionally, we find that light and heavily drizzling clouds can be distinguished by their MODIS $r_e$ value, suggesting that one $r_e$ value can be useful to classify light/heavy drizzle. A $r_e$ threshold such as 17 or 20 micron could be used to indicate a regime in which coalescence becomes highly active. This idea is supported by observations from Tropical Rainfall Measuring Mission, in which a distinctive increase in satellite $r_e$ for precipitating clouds is also apparent [Kobayashi, 2007]. Kobayashi [2007] shows that precipitating clouds have effective radii generally smaller than 30 microns, whereas in non-precipitating clouds the effective radii are smaller than 20 microns. He further suggests that a $r_e$ threshold between 15 and 20 $\mu$m (a precipitation transition zone) can indicate unstable drops that can either grow into raindrops or dissipate very quickly.

### 3.4.2 Variability of the droplet spectra of the cloud mode

Cloud effective radius differences between in-situ and MODIS $r_e$ increase with $r_e$ (Fig 3.13a), suggesting that the error in MODIS $r_e$ could be associated with changes in the shape of the droplet spectra, as it was suggested by Platnick and Valero (1995), because it directly affects the simulated radiances used in the MODIS algorithm. Changes in the droplet spectra shape are assessed through the standard deviation or spread of the droplet size distribution, assuming a log-normal distribution (Hansen and Travis, 1974). We found, in agreement with some other studies, that the distribution spread is loosely
anti-correlated with $r_e$ (Liu and Daum, 2002), with values of spread fluctuating between 0.1 and 0.5, and a mean value of 0.29 (Fig. 3.13b). To understand the effect of the distribution spread ($\sigma$) in the retrieved $r_e$, we performed retrievals of $r_e$ assuming a constant value of $\tau = 15$ with different $\sigma$. We used LibRadtran software package (Mayer and Kylling, 2005), with the discrete ordinates method (DISORT), to simulate the radiances. The radiances were calculated at the wavelength region 1.965-2.153 $\mu$m, a band slightly larger than the one used by MODIS. We assume a vertically homogeneous cloud, without atmospheric absorption nor thermal emission, and typical MODIS-Terra geometry. Additionally, the radiances associated with a particular $\sigma$ were simulated and compared with a look-up table, in order to retrieve $r_e$ ($r_{e-retr}$). The $r_e$-radiance look-up table was constructed by using the operational MODIS $\sigma = 0.35$.

We retrieved $r_e$ ($r_{e-retr}$) for $\sigma = 0.29$ (mean value) and $\sigma = 0.2$, which corresponds to a lower limit for $\sigma$. Consistent with Chang and Li (2001), the simulations indicate that when the $\sigma$ of the look-up table (0.35) is larger than the actual $\sigma$, the retrieved $r_e$ is also larger (Fig. 3.14). This bias increases with $r_e$ up to 11 $\mu$m, and reaches a relatively constant value for $r_e > 13$ $\mu$m. The maximum difference between $r_{e-retr}$ and actual $r_e$ is found for $r_e = 9$-10 $\mu$m, with magnitudes close to 0.62 $\mu$m and 0.25 $\mu$m for $\sigma = 0.2$ and 0.29, respectively.
Figure 3.13: Differences between in-situ and MODIS \( r_e \) as function of the in-situ \( r_e \). b) Scatterplot between the observed \( r_e \) and the log-normal standard deviation (\( \sigma \)). The filled circles indicate samples concomitant with MODIS observations.

Figure 3.14: Differences between the retrieved \( r_e \) (\( r_{e-retr} \)), estimated with a fixed \( \sigma \) at 0.35, and the theoretical \( r_e \) for a cloud with \( \tau = 15 \), and \( \sigma \) at 0.29 and 0.20 (gray and black lines respectively). \( r_e \) differences are depicted as function of the theoretical \( r_e \).

### 3.4.3 Effect of the bimodal distribution

We selected two cases with MODIS \( r_e \) at 13.1 \( \mu \)m and 22.6 \( \mu \)m (in-situ \( r_e = 10.9 \) and 16.7 \( \mu \)m respectively) for determining the significance of the drizzle mode in the MODIS retrievals. These cases have the highest 2D-C LWP, 11 and 78.1 g/m\(^2\), with maximum radar reflectivities of -14 dBZ and 10 dBZ respectively. We plotted the
normalized volume spectra $\Delta V/\Delta \log(r)$ as in Nakajima et al. (2010), where $\Delta V$ is the droplet volume per bin. Figure 3.15 show the droplet volume distributions of two profiles at optical cloud path from the cloud top ($\tau^*$) = 1, 3, and, 5 (Fig. 3.15, black, red, and blue lines respectively). Fig. 3.15a and b clearly show that the cloud mode always dominates, regardless the level selected, with a ratio between the largest cloud and drizzle mode approximately equal to 1:0.06 and 1:0.1 respectively. These results can be interpreted by invoking the numerical analysis of Nakajima et al. (2010). They found that for a cloud with a volume ratio of 1:0.2 the $r_e$ offset is smaller than one micron for a typical cloud mode $r_e$ of 11 micron (similar to the in-situ value $r_e$ in Fig. 3.15a), Similarly, for a cloud mode $r_e = 17$ microns (similar to the in-situ value $r_e$ in Fig. 3.15b), the satellite bias would be close to 2 microns. Since the precipitation modes here are smaller than the ones in Nakajima et al. (2010), a smaller $r_e$ bias should be expected as well. The relatively linear increase of the $r_e$ offset with the drizzle mode of the volume spectra in Nakajima et al. (2010) may suggest that, for Fig. 3.15a the drizzle mode could explain a bias of 0.1-0.3 $\mu$m while for Fig. 3.15b the bias could be 0.8-1.2 $\mu$m; values clearly smaller than the observed bias in our study (2.2 $\mu$m and 5.9 $\mu$m respectively). This analysis allows one conclude that, although the drizzle mode can affect the MODIS $r_e$, the amount of drizzle observed by the C-130 is insufficient for explaining the satellite $r_e$ bias.
**Figure 3.15:** Volume spectra for two cases during RF13. a) (MODIS $r_e$, in-situ $r_e$) = (13.1 $\mu$m, 10.9 $\mu$m), b) (MODIS $r_e$, in-situ $r_e$) = (23.6 $\mu$m, 16.7 $\mu$m). Colors indicate the cloud depth from the cloud top ($\tau^*$), $\tau^*$ = 1 (black), 3 (red), and 5 (blue).

### 3.4.4 Incidence of the solar/sensor angles

MODIS samples typically had solar zenith angle between 20° and 40°, sensor zenith angle lower than 60°, and relative azimuth angle lower than 50° (Fig. 3.16). This solar geometry should contribute to ameliorate 3-D radiative effects (Kato and Marshak, 2009). Kato and Marshak (2009) showed that for small solar zenith angle (≤ 30°), the viewing geometry error in $\tau$ due to a plane parallel bias could be less than 10%. Nevertheless, most of the sensor zenith angles (circles in Fig 15) are rather high, corresponding to samples near the edge of MODIS scan. Maddux et al (2010) found that during the period 2003-2007, the mean MODIS $r_e$ level 3 differences between near nadir and edge of the scan scenes could be up to 7 micron, but with a bias close to 1 micron over the Southeast Pacific. These differences are mainly attributed to increases of subpixel cloudiness at the edge of the scan due to a growth of a pixel size from 1 km (nadir) to 2.2 km (Brent Maddux, 2011 personal communication). A similar investigation
as in Maddux et al. (2010) but applied to five October months level 3 observations (Fig. 3.17) over the Southeast Pacific found differences between nadir and edge $r_c$ near to zero microns or slightly positive. These small differences can be explained by more overcast conditions in marine stratocumulus that make it less prone to a subpixel cloudiness effect.

Figure 3.16: Solar and sensor geometry of the MODIS scenes. Circles and triangles indicate sensor and solar zenith angles respectively in the y-axis. Note that the relative azimuth angle is repeated twice in order to fit the scatterplot format for the solar and sensor zenith angle.

Figure 3.17: $r_c$ level 3 differences between pixels with satellite zenith angle smaller than 20° and higher than 50°. The period of study corresponds to Octobers 2005 to 2008.
3.5 Summary and Discussion

Unique clouds observations during VOCALS-REx allow us assess the ability of MODIS retrievals to represent the microphysical properties of the southeast Pacific stratocumulus deck. We found a good agreement between MODIS and the in-situ \( \tau \) and \( r_e \) at the cloud-top, though a systematic overestimate of MODIS \( r_e \) was evident, with a mean bias of 2.08 \( \mu m \), and magnitudes increasing with \( r_e \).

The consistency between the bias found here and in other studies (Nakajima et al., 1990; Nakajima and Nakajima, 1995) suggests that the \( r_e \) overestimate is a general feature of the retrievals over stratocumulus regions, and not an attribute of the microphysical probes, as has previously been suggested.

The error in \( r_e \) propagates to the MODIS LWP that also shows a positive bias with respect to the observations. In spite of the errors in \( r_e \) and \( \tau \), MODIS \( N_d \) agreed well with the observations. It is believed that this agreement mainly depends on the a-priori selection of constants, namely the stratification of the cloud water content and parameter \( k \), which are able to counteract the bias in \( r_e \). Additionally, a decrease in MODIS \( N_d \) between the morning and afternoon overpasses, noted for selected cases, was also found in the in-situ \( N_d \) values. This result encourages further research of this potential diurnal (or semi-diurnal) cycle as well as the involved physical mechanisms.

A \( r_e \) positive bias is a common feature of the three MODIS \( r_e \), although the offset slightly decreases for \( r_{e1.7} \) and \( r_{e1.6} \). Additionally, we did not find evidence of vertical structure information from the three MODIS \( r_e \), such as might be expected from an adiabatically distributed cloud or one combining drizzle.
Several sources of error were investigated further: the effect of the shape of the cloud mode droplet spectra, the influence of the drizzle mode, and the impact of the viewing geometry.

Changes in the shape of the cloud droplet spectra were analyzed by using the effective variance of the cloud mode. We found in this study a mean effective variance of 0.29, a value smaller than the variance prescribed in the MODIS algorithm (0.35). Simulations made with a fixed $\tau$ show that retrievals based on the assumed MODIS effective variance would tend to overestimate $r_e$, although this offset is typically smaller than 0.6 $\mu$m when $\sigma = 0.2$.

Analysis of the volume spectra of the droplet size distributions shows that the cloud mode is significantly larger than the drizzle mode. Simulations by Nakajima et al. (2010) suggests that a positive $r_e$ bias attributed to the drizzle mode would be smaller than 0.3 micron for a case with light drizzle and 1 micron for a heavy drizzling cloud. This apparent lack of sensitivity of stratocumulus clouds to the precipitation mode was also observed in synthetic satellite data based on large eddy simulation of stratocumulus clouds (Zinner et al., 2010). A large bimodal-dependent bias seems to be more plausible in cumulus clouds, where the precipitation is well developed. This is further illustrated in the cases of bimodal distribution during the First ISCCP Regional Experiment (FIRE) used within Nakajima et al. (2010), in which cloud optical thickness varied between 20 and 40. In contrast, the stratocumulus clouds during VOCALS-REx were characterized by mean $\tau < 15$ (see regional map, Fig. 2), in agreement with the occurrence of smaller precipitation modes. Drizzle occurrence also poses the question whether the use of the parameter $k$ within the definition of satellite $N_d$ is still valid. Since the cloud mode is the
main contributor to the droplet size distribution, parameterizing $N_d$ in terms of cloud-mode-derived parameters and $k$ is adequate. This is also supported by the fact that the bias in MODIS $r_e$ is relatively insensitive to the drizzle mode, giving additional support to the use of the cloud mode $k$.

Although a more quantitative analysis of the sensor/solar viewing geometry effect is difficult with our reduced dataset, the retrievals are benefited from typical low solar zenith angles. In contrast, high sensor zenith angles (near the edge of the scan) in our samples, generally are associated with a positive (negative) bias in $r_e$ ($\tau$), have a little effect over the Southeast Pacific (Maddux personal communication, 2011). This indicates that pixels are more likely to be located over completely cloudy regions, with little effect of sub-pixel variability especially at edges of the scan, in which the pixel resolution is degraded to 2.2 km.

Given the small effect of the viewing zenith angle, the algorithm assumptions about the droplet size distribution (both cloud and drizzle mode) are the only significant sources of errors found here. The combined effect of the precipitation mode and the spread of the droplet size distribution explain a bias smaller than 0.9 micron and 1.6 micron for light and heavy drizzle cases respectively, whereas for non-drizzling clouds the explained bias is smaller than 0.6. This points to three-dimensional radiative effects as a plausible retrievals source of error.

Uncertainties in the atmospheric corrections due to errors in the water vapor used by the algorithm also need to be considered. The water absorption effect in the measured radiance is given by the vertically integrated water vapor (water vapor path) above the cloud top (Platnick et al., 2003). The water vapor path correction poses two questions:
The first one is how well the water vapor used by MODIS (from NCEP Global Data Assimilation System, GDAS) can represent the actual water vapor profile over the region of study. The second question is how well MODIS can estimate the cloud top height. Regarding the latter, MODIS cloud top heights are known to overestimate the inversion base heights for about 2000 meters (Garay et al. 2008). This MODIS offset should impact the computation of water vapor path, through a reduction of the column above cloud used for the water vapor path calculation. This effect would lead to higher corrected radiances and therefore, smaller retrieved effective radius. This underestimate in the water vapor could be counteracted only if the GDAS water vapor profile used by MODIS is unrealistically high, particularly above the cloud top. However, neglecting 2000 meters of the lower troposphere should have a high impact because the lower levels of the atmosphere contribute the most to the total atmospheric water path. In this regard, Harshvardhan et al. (2009) showed for a case over the northeast Pacific, that when the mean water vapor path above cloud from aircraft observations was 1.1 cm, the NCEP GDAS water vapor path above the MODIS-derived cloud top height was 0.2 cm. That is to say, MODIS atmospheric corrections would tend to underestimate the effect of water vapor, leading to smaller retrievals of effective radii, contrary of what is found in this study.

Additional errors in the thermal emission corrections in the 3.7 µm channel would impact the $r_{c3.7}$ retrievals. This is plausible since Zuidema et al. (2009) found that the mean MODIS cloud top temperature was 1.3 °C cooler than the actual mean temperature of the inversion base (equivalent to the cloud top). Further investigation is necessary to
understand the impact of the cloud top temperature in the thermal corrections of the 3.7 µm band.

The mean MODIS \( r_e \) bias in this study is equivalent to the 2 micron mean difference found between MODIS and Polarization and Directionality of the Earth’s Reflectance (POLDER) \( r_e \) for warm clouds over the ocean, [Bréon and Doutriaux-Boucher, 2005]. This suggests that POLDER can better agree with the observed cloud top effective radius. Since the polarization signal is better defined in homogeneous clouds, with the POLDER algorithm relatively insensitive to the droplet size distribution effective variance, part of the POLDER/MODIS difference was thought to be the consequence of an incorrect effective variance specified within the MODIS algorithm. Nevertheless, as it was shown in this paper, the effective variance error cannot account for more than 0.6 µm bias. Another mechanism that has been postulated to explain POLDER/MODIS differences is that POLDER retrieves \( r_e \) very close to the cloud top, where entrainment is more likely to reduce \( r_e \) [Bréon and Doutriaux-Boucher, 2005]. This mechanism is also discarded over the Southeast Pacific, as \( r_e \) tends to monotonically increase with height. Some other factors such as less sensitivity of the polarization method to three-dimensional radiative effects require further investigation.

The findings presented here apply to the particular clouds sampled, and caution must be taken before generalizing these results. Nevertheless, our study does lend additional support to the suggestion that MODIS overestimates \( r_e \) in marine stratocumulus clouds, regimes that present the most favorable conditions for remote sensing of \( r_e \) and \( \tau \).
Chapter 4

Aircraft based investigation of the first aerosol indirect effect

4.1 Motivation

Thus far, we have investigated synoptic aspects associated with changes in the cloud microphysics, using MODIS $N_d$ as an indicator of the aerosol influence in the cloud properties. Nevertheless, the link between aerosols and $N_d$ has not been explored yet. This is difficult from a satellite remote sensing perspective because aerosol properties (aerosol optical thickness and angstrom coefficient) are not retrieved for overcast pixels. The problem has been partially addressed in the literature by assuming that the aerosol optical thickness of a clear-sky pixel (if any) is representative of the neighboring cloudy pixel. This strategy has shown significant shortcomings such as cloud contamination or 3D radiative effects that bias the aerosol optical thickness (e.g. Varnai and Marshak, 2009). Moreover, in marine stratocumulus regimes, the neighbor-pixel approach is less applicable because of the spatial persistence of the cloud cover. A complete investigation of the first aerosol indirect effect requires then an understanding of the way aerosol modify the cloud microphysics, which in turns dictates the changes in cloud albedo for particular atmospheric environments. Although with satellite retrievals we have gained an understanding of the large-scale processes involved in the indirect effect, more quantitative estimates require the use of accurate observations with high spatial/temporal resolution in order to resolve the details of the microphysical interactions.
It becomes clear that a quantitative investigation of the first indirect effect demands the use of in-situ observations. In this chapter, we take advantage of VOCALS-REx observations collected by the C-130 aircraft to address the problem of quantifying the Twomey effect.

### 4.2 General requirements

Several metrics have been suggested to evaluate the way pollution interacts with the cloud microphysics. These metrics are based on estimates of aerosol properties (concentration, mass, or optical thickness), in combination with microphysical variables such as: cloud droplet number concentration ($N_d$), cloud effective radius ($r_e$), and optical thickness (Feingold et al., 2001; McComiskey and Feingold 2008). A simple aerosol cloud interaction (ACI) metric is expressed as partial derivatives of optical and microphysical properties with respect to changes in aerosol concentration ($N_a$) (eq. 4.1, McComiskey et al., 2009)

\[
ACI_\tau = \left. \frac{\partial \ln \tau}{\partial \ln N_a} \right|_{LWP} \\
ACI_r = \left. \frac{\partial \ln r_e}{\partial \ln N_a} \right|_{LWP} \\
ACI_N = \left. \frac{\partial \ln N_d}{\partial \ln N_a} \right|_{LWP}
\] (4.1)

Eqs. 4.1 can be understood as the microphysical response to a change in aerosol concentration. Additionally, if $N_d$, $\tau$ and $r_e$ are related through an adiabatic-like relationship (Chapter 2), then:

\[
ACI_\tau = -ACI_r = \frac{1}{3} ACI_N
\] (4.2)
McComiskey et al. (2009) indicate that ACI depends on the activation, collision-coalescence processes, aerosol size distribution, cloud adiabaticity and updraft velocities. Upper limits for the relationships in eq. 4.1 can be derived if one assumes that a fractional change in $N_a$ cannot produce a larger fractional change in $N_d$, that is: $ACI_N = 1$. Then, according to equation 4.2: $ACI_r = -ACI_r = 1/3$.

The calculation of the ACI metrics for fixed values of LWP provides the dynamical constraint necessary to isolate the aerosol effect from other dynamical processes that can also induce changes in the cloud microphysics.

It is evident that in eq. 4.1, several processes that can also potentially influence the cloud are not explicitly included. The simplified view of the first aerosol indirect effect as a function only on $N_a$ is supported by numerical analysis that shows that for relatively clean conditions, like maritime remote regions, changes in $r_c$ are mostly determined by the aerosol concentration and liquid water content (Feingold, 2003). For very polluted cases, like those found over the continents, changes in cloud microphysics also depend on the shape of the aerosol size distributions, solubility and updrafts velocity, turning the set of eq. 4.1 a less suitable estimator of the aerosol-cloud interactions.

As shown in Chapter 3, observations during VOCALS-Rex by the C-130 aircraft provided a comprehensive set of microphysical variables useful for the investigation of the first indirect effect. However, the necessity of estimating ACI metrics for different values of LWP (vertically integrated water content), reduces the dataset to less than 100 vertical profiles, a number that might not guarantee the statistically significance of the
results. The desirability of a larger dataset motivates the use of airborne remote sensing techniques to retrieve the cloud properties. These techniques primarily rely on a microwave radiometer for the estimation of LWP (Zuidema et al., 2011), and a broadband pyranometer for determining \( \tau \) (section 4.4).

In our previous exposition, we have shown that the first aerosol indirect effect is manifested in cloud microphysics changes driven by changes in aerosol concentration. The consequences of these microphysical changes are ultimately observed in perturbations of the cloud albedo. The role of \( N_d \) in modulating the albedo response can be more specifically addressed with the albedo susceptibility (S), or the change of cloud albedo (A) due to a change in \( N_d \), under constant liquid water path (LWP):

\[
S = \frac{dA}{dN_d} \bigg|_{LWP} \quad (4.3) \quad (\text{Platnick and Twomey, 1994})
\]

Or similarly expressed as relative changes in S, or relative susceptibility (\( S_R \)):

\[
S_R = \frac{dA}{dN_d} \frac{N_d}{A} \bigg|_{LWP} = \frac{d \ln(A)}{d \ln(N_d)} \bigg|_{LWP} \quad (4.4) \quad (\text{Feingold and Siebert, 2008})
\]

Which is equivalent to a fractional (relative) change of albedo due to a fractional change in \( N_d \). Since \( \tau \) and \( N_d \) are closely related, the radiative dependence of A on \( N_d \) is mainly controlled by \( \tau \), with \( N_d \) providing the link between microphysical (\( r_c \)) and optical (\( \tau \)) properties.

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\(^5\) In this chapter the term remote-sensing is applied to cloud retrievals based on airborne passive radiometers.
4.3 General description of the selected research flights

Although the C-130 operated during fourteen research flights (RF), only six of them occurred during the daytime, allowing the retrieval of a solar transmission-based cloud optical thickness. Moreover, given the uncertainty of the aircraft-based remote sensing retrievals (section 4.4), we select three flights for an initial examination, namely: RF11 (Nov. 9), RF12 (Nov. 11), and, RF13 (Nov 13). The three selected C-130 flights are representative of the typical flight missions, including westward and southward sampling, with diverse meteorological conditions. For instance, RF11 and RF12 favored the sampling of coastal regions that are more prone to anthropogenic influences (transects in Fig. 4.1a and b). In contrast, RF13 was mainly oriented to investigate pockets of open cells, and their association with drizzle, features that are better observed well offshore (Fig. 4.1c)

During RF11 and RF12, the aircraft measured infrequent drizzle and overcast conditions; features particularly desirable for investigating the first aerosol indirect effect. These days showed moderate values of $N_d$ associated with transition meteorological conditions, within a period dominated by days classified as MIN $N_d$, according to the composites defined in Chapter 2 (Fig. 2.16). It is noticeable the consistency between large satellite $N_d$ for the RF12 transect (Fig. 4.1b) and the large in-situ aerosol concentration that reaches magnitudes up to 700 cc (Fig. 4.1d middle panel). In contrast, although $N_d$ is large near the coast for RF11, the aircraft sampled more pristine areas (Fig. 4.1a), a few degrees west from the core of high $N_d$. The changes in satellite $N_d$ are consistent with the in-situ aerosol concentration time series (Fig. 4.1d, upper panel), with typical concentration lower than 300 cc.
Figure 4.1: a), b and, c): MODIS $N_d$ maps from Terra and the aircraft transect (black solid line), RF11 (Nov. 9), RF12 (Nov 11), and RF13 (Nov 13) respectively. d) Aerosol concentration time series sampled during sub-clouds legs only (heights lower than 400 m).

As expected from a more offshore flight, RF13 measured frequent precipitation and broken clouds. The presence of drizzle is also associated with very low aerosol concentrations, with magnitudes lower than 40 cc (Fig. 4.1d, lower panel). It is not clear why the aerosol measurements presented high peaks for RF13; nevertheless, they do not affect our estimates of ACI, because $\tau$ and LWP were not retrieved during the aerosol peaks occurrence.
LWP maps show a typical decrease near the coast, although LWP higher than 100 g/m² coincides with aircraft sampling intervals, particularly for RF11 (Fig. 4.2a). LWP for RF13 is generally high south of 27°S, and seems to be modulated by a synoptic pattern. In fact, it is observed during the period Nov 10-13 a conspicuous geopotential ridge at 500 hPa moving east, associated with a coastal low (Rhan and Garreaud, 2010), and concomitant with changes in the anticyclone (Fig. 4.3 red contours).
Figure 4.3: Quikscat surface winds and NCEP/NCAR reanalysis seal level pressure (red contours: a) RF11, b) RF12, and c) RF13.

The coastal low configuration explains the strengthening of the anticyclone during Nov. 11 (RF12, Fig. 4.3b), when the axis ridge is approximately 12 degrees offshore. The zonally elongated anticyclone during RF12, reaches the coast near 42˚S by Nov. 12 (not shown), connected with a migratory anticyclone (Garreaud et al., 2002). The synoptic pattern also reinforces the coastal winds between 30˚S-40˚S during Nov. 11 (RF12, Fig 4.3b, colors), and it is typically associated with a more stable and shallower boundary layer. Although the coastal lows is still present during Nov. 12 (not shown), the
displacement of the middle level ridge to the coast marks the demise of the coastal low, characterized by a weakening of the anticyclone and reduction in both subsidence and coastal winds (RF13, Fig. 4.3c). As shown in Chapter 2, the occurrence of an anomalous ridge, as in RF12, and RF13, is generally associated with increases in $N_d$. Nevertheless, RF13 is classified as a MIN $N_d$ case. This inconsistency between $N_d$ for RF13 and the MAX $N_d$ composite in Chapter 2 is difficult to reconcile, although a migratory anticyclone and a coastally confined low pressure were not observed in the composites of Chapter 2.

### 4.4 Description of the dataset

#### 4.4.1 Liquid water path

An airborne G-band (183 GHz) vapor radiometer (GVR) was mounted looking upward, and used to retrieve liquid water path. The instrument, designed by ProSensing Inc., measures radiances from double sideband channels at $\pm 1 \pm 2 \pm 7$ and $\pm 14$ GHz from the 183.31 GHz line (Pazmany et al., 2006). While the instrument is mainly sensitive to the water vapor path, the $\pm 14$ GHz band is particularly sensitive to LWP. GVR was designed for Arctic conditions, where the atmosphere is relatively dry (< 5 mm), because the 183 GHz absorption lines easily saturate for high water vapor path. This is an important issue as the typical water vapor path over the Southeast Pacific (< 20 mm) is high enough to saturate the absorbing bands. Under this circumstance, the main problem of retrieving LWP from the GVR is that it is not possible to separate the effect of water vapor from the liquid water in the GVR measurements. The algorithm to retrieve LWP requires then prescribing a value for the water vapor path. This is done by calculating the
water vapor path from the water vapor mixing ratio measured by the C-130 and assuming that the total moisture is conserved up to the radar-derived cloud top, with the liquid water removed by assuming adiabatic liquid water content from the lidar-derived cloud base (Dave Leon, 2011 personal communication). Water vapor path derived from GVR during above-clouds flights is used to specify values of the free troposphere. The retrieval algorithm, similar to the one described in Zuidema et al., (2005), is based on an iterative method that consists of finding the LWP that better simulates in a radiative transfer model the observed brightness temperature (14 GHz band) (Zuidema et al., 2011). Although a detailed validation of the GVR LWP retrieval is work in progress, comparisons against adiabatic LWP derived from the geometrical cloud depth (cloud top and base derived from the Wyoming cloud radar and lidar respectively) show a relatively good agreement, with values from GVR generally smaller than the adiabatic LWP, as it is shown in the example of Fig. 4.4 for a particular research flight and also described in Zuidema et al. (2011).

Another unique aspect of the GVR is its high sampling rate, allowing LWP retrievals at one-second temporal resolution. In contrast, the most commonly used microwave radiometer (32 GHz) retrieves LWP with a period of 20 seconds.

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6 Physical consistency between GVR and pyranometer-based $\tau$ gives further evidence of the accuracy of the retrievals (see Chapter 4.2.2.5).
4.4.2 Cloud optical thickness

An additional cloud parameter necessary to determine equations 4.1 is \( \tau \). \( \tau \) is also valuable because provides a direct way to parameterize \( N_d \) as a function of \( \tau \) and LWP, when the cloud is assumed adiabatically stratified (Appendix 1). As shown by Leontyeva and Stamnes (1994), \( \tau \) can be computed if measurements of incoming solar radiation are combined with a 1-D radiative transfer model. Since the shortwave irradiance (\( Q \)) is essentially a function of \( \tau \) and a weak function of \( r_e \), the inverse problem of estimating \( \tau \) can be solved if observations of \( Q \) are available. The necessary \( Q \) for retrieving a transmissivity-based \( \tau \) was measured by an upward-looking Eppley Precision Spectral Pyranometer (PSP) mounted in the C-130 aircraft. This \( \tau \) retrieval is conceptually different from the one derived from satellite instruments (e.g. MODIS, Chapter 2), which primarily relies on the visible radiance reflected by clouds.
### Numerical approximation

Discrete ordinates (DISORT) and two-streams.

### Spectral resolution

24 shortwave bands, 20 cm⁻¹ bandwidths in longwave.

### Clouds

Parameterization based on Hu and Stamnes (1993)

### Aerosol

Six optical models.

### Gas Absorption

H₂O, O₂, CO₂, and O₃. Trace gases include, among others, CH₄, N₂O, and CO.

| **Table 4.1: General features of Streamer.** |

#### 4.4.2.1 Radiative transfer model

The forward calculations of the shortwave irradiance were performed with Streamer model (Key and Schweiger, 1998); a versatile radiative transfer model that has been successfully applied to the study of Arctic stratus clouds (e.g. Pinto, 1997; Zuidema et al., 2005). One of the main feature of Streamer is that the optical properties, namely single scattering albedo, extinction coefficient and asymmetry factor, are parameterized following Hu and Stamnes (1993). Streamer allows simulation of solar radiation for the spectral band 0.28µm -2.91µm, which is slightly broader than the spectral band measured by the pyranometer (0.28 µm-2.8 µm). This difference should not be a significant source of error as the energy contribution of the 2.91 µm band is very small. The general characteristics of Streamer are listed in table 4.1.

#### 4.4.2.2 Testing the radiative transfer model

The radiative model was tested with in situ observation of clear-sky shortwave irradiance. We first focused on RF12, during two periods with extended clear sky conditions during sub-cloud flights, as observed by the Wyoming up-looking cloud lidar.
We used observed values of water vapor path (in the boundary layer and the free troposphere), and computed the downwelling shortwave fluxes, with aerosol optical thickness at 0.1, and summer mid-latitudes standard profiles (Fig. 4.5, red circles, standard simulation). It was found that the simulated fluxes were lower than the irradiance measured by the pyranometer. The bias was larger for the first simulated period with a mean bias of 80 W/m$^2$, whereas the differences were reduced to 20 W/m$^2$ in the second period. These changes were also associated with changes in the solar zenith angle ($\theta$), with mean $\theta$ at 14.1° and 37.1° for the first and second period respectively. In order to test the sensitivity of the simulations to the water vapor path, we also calculated the shortwave fluxes but with a 50% reduction in water vapor path (Fig. 4.5a, blue circles). Although the reduction in water vapor produces an increase in the simulated fluxes, the increase is smaller than 19 W/m$^2$, suggesting that the bias is not a consequence of a wrongly prescribed water vapor path. Similarly, we also analyzed the sensitivity of the simulated flux to aerosol optical thickness ($\tau_{aerosol}$) and the absorbing gases (but including the effect of water vapor). In order to investigate the importance of aerosols and absorbing gases in the simulated irradiance, we performed two simulations in which aerosols and absorbing gases were removed. The unrealistic removal of aerosols and gases (Fig 4.5b, red and blue triangles respectively) could not explain the bias during the first period, and a bias close to 60 W/m$^2$ remained.
Figure 4.5: Two periods (divided by the gray vertical line) during RF12 with simulated and observed (black) shortwave irradiance. a) Simulations are made with the observed water vapor path (red circles), and with water vapor path reduced in 50% (blue circles). The aerosol optical thickness is assumed constant at 0.1, and typical midlatitude summer profiles for absorbing gases are prescribed in the simulations. θ indicates the mean solar zenith angle for each leg. b) Sensitivity simulations made with the observed water vapor path but removing the aerosol optical thickness ($\tau_{aero}$) and absorbing gases (blue and red triangles respectively). The standard simulation is represented by the green solid line. Thick black line indicates pyranometer observations that occurred below 200 m, with clear-sky conditions, according to the cloud lidar.

Since the clear-sky cases were rare during VOCALS-REx, a more systematic analysis of the pyranometer-model bias was carried out by analyzing above clouds observations. We prescribed a free tropospheric water vapor path at 0.3 cm, which is a mean value for the range of variability of the free troposphere water vapor path (0.2-0.4 cm). Additionally, we used standard values of column ozone for a summer mid-latitude atmosphere, and a fixed aerosol optical thickness at 0.1.
Figure 4.6: Observed downwelling shortwave fluxes above cloud for heights between 1500 and 1800 m (black circles). Dashed red lines indicate the simulated shortwave fluxes. a) RF11, b) RF12, and c) RF13. d) Solar zenith angle.

Figure 4.6 shows the bias between observed and simulated downwelling fluxes for the three research flights. The differences tend to increase with fluxes (Fig. 4.7), indicating a dependence on the solar zenith angle, as mentioned previously and more specifically illustrated in Fig 4.6d. As anticipated in Fig 4.5, reductions in prescribed atmospheric inputs (absorbing gases, the aerosol optical thickness, and water vapor path) cannot explain the differences in the simulated irradiance.
Figure 4.7: Simulated Q versus pyranometer observations between 1600 and 1800 meters of height.

There is no sufficient evidence to conclude that the bias found here is associated with the model, because this persists regardless the prescribed magnitudes of the environmental variables. Instrument calibration is another possibility to explain the bias. Since the inconsistency between modeled and measured irradiance might bias the cloud optical thickness retrievals, we adjust the PSP irradiance \((Q_{pyr, adj})\) through a linear regression in order to fit the simulated clear-sky irradiance (eq. 4.5 and table 4.2).

\[
Q_{pyr, adj} = a \cdot Q_{pyr} + b
\]  

(4.5)

\(Q_{pyr}\) indicates the measured pyranometer irradiance. Hereafter, the calculations are made with this adjusted pyranometer irradiance, also denoted as \(Q_{pyr}\).

<table>
<thead>
<tr>
<th>Research flight</th>
<th>Coefficient a</th>
<th>Coefficient b</th>
</tr>
</thead>
<tbody>
<tr>
<td>RF11</td>
<td>0.9</td>
<td>68</td>
</tr>
<tr>
<td>RF12</td>
<td>0.91</td>
<td>41</td>
</tr>
<tr>
<td>RF13</td>
<td>0.9</td>
<td>66</td>
</tr>
</tbody>
</table>

Table 4.2: Linear regression coefficients for adjusting the pyranometer measurements based on eq. 4.5.
4.4.2.3 Retrieving algorithm

The relationship between $\tau$ and shortwave fluxes, for two different values of effective radius (5 and 15 $\mu$m) is illustrated in Fig. 4.8. This example underlines the idea that $Q$ is primarily a function of $\tau$, with a weak dependence of $r_e$ (Fig. 4.8a). A closer look at the differences between irradiances calculated with fixed $r_e$ at 15 and 5 $\mu$m (Fig. 4.8b) shows that the error is in fact smaller than 40 W/m$^2$. This difference is negligible for large $Q$ and small $\tau$; however, for large $\tau$ (associated with small $Q$), the bias could induce errors as large as 5. In order to reduce errors associated with $r_e$, we used LWP to constrain $r_e$, as it is explained in the following.

In short, the algorithm for determining $\tau$ consists of iterative calculations of irradiance during sub-cloud flights for particular values of $\tau$ and $r_e$. The optimal $\tau$ is obtained from minimizing the difference between the simulated irradiance and the observations. A better description of the algorithm is presented in Fig. 4.9 and summarized in the next steps:

1. I first prescribe the initial values for $r_e$ and $\tau$. These values are obtained from simple relationships. For the first iteration, we used a proxy for $N_d = 0.5N_a + 50$ (Jefferson Snider, 2010 personal communication), where $N_a$ is the accumulation mode of aerosols measured by an aircraft probe. GVR LWP is used along with the first guess $N_d$ to calculate $r_e$ and $\tau$ ($r_e^*$ and $\tau^*$), based on adiabatic assumptions. It is important to mention that the only purpose of selecting these initial $\tau$ and $r_e$ is to reduce the number of iterations of the method, as the final retrieved $\tau$ is independent of the initial values of $\tau$ and $r_e$. 
2. A two-streams radiative transfer is used to estimate $Q(r_e^*, \tau^*)$. If the absolute value of $|Q(r_e^*, \tau^*) - Q_{pyr}| > 3 \text{ W/m}^2$, then $\tau^*$ is increased (decreased) by 0.04 if the retrieved value $Q(r_e^*, \tau^*)$ is larger (smaller) than $Q$. If $|Q(r_e^*, \tau^*) - Q_{pyr}| < 3 \text{ w/m}^2$ the iteration ends and $r_e^*$ is upgraded $r_e^* = \frac{9}{5 \tau^*}LWP$.

3. With the new values for $\tau^*$ and $r_e^*$, another two-streams loop is made as in 2.

4. With a value of $r_e^*$ upgraded for a second time, step 2 is repeated again, but using 8-stream simulations. The result of this iteration loop is the final $r_e$ and $\tau$.

Figure 4.8: a) Differences between the calculated $Q$ for $r_e=15\mu m$ ($Q_{15\mu m}$) and $5\mu m$ ($Q_{5\mu m}$), as a function of $\tau$. b) $Q$ for $r_e=15\mu m$ (black line) and $5\mu m$ (red line), as a function of $\tau$. It was assumed a surface reflectance of 0.05 and a solar zenith angle =30°.

In the simulations, we use the boundary layer water vapor path for well-mixed conditions plus a constant value at 0.3 cm, which represent a standard value of humidity for the free troposphere. It is also prescribed $\tau_{aerosol}$ at 0.1 and typical mid-latitude summer profiles for the absorbing gases.
4.4.2.4 Algorithm error analysis

For the radiative transfer simulations, I assumed a vertically homogeneous cloud with constant values of \( r_e \) and LWC with height. Although it has been shown in Chapter 3 that stratocumulus clouds tend to present an adiabatically-distributed vertical structure, the weak dependence of \( Q \) on \( r_e \) allows one assume that the error of the homogeneous approximation should be small. In order to assess this error, I carried out a simple experiment with a 3 layer clouds (table 4.3).
<table>
<thead>
<tr>
<th>Cloud layer</th>
<th>$\tau / \tau_{\text{bottom}}$</th>
<th>$r_e$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Top</td>
<td>3</td>
<td>10</td>
</tr>
<tr>
<td>Middle</td>
<td>2</td>
<td>8</td>
</tr>
<tr>
<td>Bottom</td>
<td>1</td>
<td>6</td>
</tr>
</tbody>
</table>

Table 4.3: Three layers cloud used to analyze the stratification effect in $\tau$ retrievals. $\tau$ was normalized by the value of the bottom layer ($\tau_{\text{bottom}}$).

The cloud in table 4.3 reflects the typical conditions of stratocumulus clouds: larger $r_e$ at the cloud-top and increase of water content with height (increase of $\tau$). Based on the values in table 4.3, I created different clouds with the same $r_e$ profile as in table 4.3 but changing the total $\tau$ ($\tau_{\text{total}}$, the summation of the three layers), by multiplying the normalized $\tau$ profile in the table by different $\tau_{\text{bottom}}$. For each cloud, the synthetic pyranometer observations were obtained with Streamer (8-stream simulation), and $\tau$ was retrieved assuming a vertically homogeneous cloud. The results (Fig. 4.10) showed a remarkable agreement between homogeneous and stratified $\tau$, with a small positive bias (0.2) for the homogeneous one. This example stresses that $\tau$ is insensitive to the cloud vertical structure.

Figure 4.10: Scatterplot between $\tau$ for a stratified cloud in table 4.3 and the retrieved $\tau$ using the algorithm described here.
Errors associated with the water vapor path were also analyzed. If an error in the vapor path of ±30 % is assumed, the associated error in the retrievals is small, with typical τ bias smaller than 4 %, in agreement with Leontyeva and Stamnes (1994).

Since \( r_e \) is calculated from τ and LWP (\( r_e \sim 9/5*\text{LWP}/\tau \)), and then used in the iterative algorithm to estimate τ, errors associated with LWP can also affect the retrievals. We also analyze this error by assuming 30% error in LWP. We find that the mean error in τ is close to 5 %, with a negative (positive) bias when LWP is biased low (high).

Pyranometer uncertainties can strongly limit the accuracy of the retrievals. We show in Fig 4.11 the effect of a conservative error of 5 % in the pyranometer signal. This error would induce variations in τ of around 1 to 2.5 units of τ. This calculation also gives a general idea of the error that could arise from the mismatch between pyranometer observation and the radiative model presented in section 4.4.2.2, with underestimates between 13% and 5% for magnitudes of τ between 5 and 40 respectively.

Figure 4.11: Error in τ associated with a 5 % error in the pyranometer measurements.
If the potential errors presented here are additive, then the upper limit error in $\tau$ would be around 15%-20%. More typical error would be smaller, perhaps with magnitudes closer to 10%.

4.4.2.5 Comparison with cloud probe observations

Since $\tau$ retrievals (and GVR LWP) are not simultaneous with the in-situ $\tau$, a validation analysis is difficult. In spite of this, in-situ observations can help us to determine the physical consistency of the remote sensing retrievals. We investigate only non-precipitating cloud because we are trying to determine the aerosol effect independently from other dynamics processes. This is done by choosing samples with maximum cloud column reflectivity lower than -16 dBZ. Figure 4.12 depicts the relationship between $\tau$ and LWP for remote-sensing and in-situ estimates for the three daytime flights. In-situ and remote sensing observations show a high LWP-$\tau$ correlation with an agreement in the slope, indicating the physical consistency of the retrievals. For RF13, a correlation between LWP and $\tau$ is more difficult to observe because the frequent precipitation (offshore flight) reduced considerably the number of samples, whereas decoupling conditions prevent accurate estimates of the water vapor path, which are used as input in the GVR LWP algorithm. A similar relationship is also found between the adiabatic LWP and retrieved $\tau$ (Fig. 4.13), and a good agreement with the in-situ observations as well. Fig. 4.12 and 4.13 show further the consistency between GVR and adiabatic LWP. Unfortunately, due to radar issues for determining the cloud top, the number of adiabatic LWP observations are considerably lower than the GVR LWP
(typically 40 % less observations than the GVR LWP), a factor that can reduce the statistically significance of the results that rely on the adiabatic LWP.

As quality control, we identified and removed those sub-legs that presented a constant decoupling of the boundary layer (determined as the difference between the cloud base and the lifting condensation level). As mentioned previously, the boundary layer estimates of water vapor path are only valid for well-mixed conditions, since they are derived from humidity measured near the surface. Any significant error in the water vapor path can lead to wrong estimates of GVR LWP, so the removal of these cases will reduce potential biases in LWP. The decoupling periods were only limited to RF11 and RF13, and they are listed in table 4.4.

![Figure 4.12: τ vs LWP, for the retrieved variables (black dots) a) RF11, b) RF11, and c) RF13. The in-situ τ-LWP (red circles) corresponds to all the available observations collected by the C-130 aircraft.](image-url)
Figure 4.13: As in Figure 4.12 but for the adiabatic LWP.

<table>
<thead>
<tr>
<th>RF</th>
<th>Time [min]</th>
<th>Mean decoupling</th>
<th>Mean LWP [g/m²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>11</td>
<td>280-286</td>
<td>376 m</td>
<td>80</td>
</tr>
<tr>
<td>13</td>
<td>224-285</td>
<td>390 m</td>
<td>65</td>
</tr>
<tr>
<td>13</td>
<td>326-332</td>
<td>314 m</td>
<td>103</td>
</tr>
<tr>
<td>13</td>
<td>347-355</td>
<td>547 m</td>
<td>108</td>
</tr>
</tbody>
</table>

Table 4.4: Decoupled periods excluded from the analysis. The level of decoupling is calculated as the difference between cloud base and lifting condensation level.

It was also identified a period during RF12 in which the relationship LWP-τ departed significantly from the observations (period 11500-12100 s, Fig. 4.14, blue dots). It is not clear why τ and LWP do not follow the expected pattern, although it is thought that future reprocessing of the LWP retrievals can solve this issue. In order to avoid potential errors attributed to this period, we have exclude it from the following analysis.
Another indirect way to determine the accuracy of our retrievals is deriving a retrieved-based $N_d$, expressed as:

$$N_d = 0.0058 \cdot \frac{\Gamma^{1/2} \rho_w^2}{k} \frac{\tau^3}{LWP^{5/2}} \quad (4.6, \text{Appendix 1})$$

$\Gamma$ is the stratification of the water content with height, $\rho_w$ is the water density, and $k$ is the ratio between the cube of the effective radius and the volume radius of the droplet size distribution. Eq. (4.6) is analogous to the $N_d$ relationship extensively used in previous chapters, but expressed in terms of LWP instead of $r_e$. For $\Gamma$, we used the mean adiabatic lapse rate, multiplied by an adiabatic factor at 0.7 (Chapter 3). For $k$, we used the mean $k$ at the cloud top, (Chapter 3).

Before estimating a remote-sensing $N_d$, I used the in-situ observations to constrain the range of variability of remote-sensing $\tau$ and GVR LWP, in an attempt to eliminate retrievals more prone to be dominated by errors, which can also affect the estimates of $N_d$. Based on the linear regression of the in-situ observations, we determine an upper and lower threshold line ($LWP_{\text{high}}$ and $LWP_{\text{low}}$ respectively), and defined as:
\[ LWP_{\text{high}} = 1.5 \left[ a \cdot \tau + b \right] + 10 \]
\[ LWP_{\text{low}} = 0.5 \left[ a \cdot \tau + b \right] - 10 \]  \hfill (4.7)

The term in brackets corresponds to the in-situ linear regression. As shown in Fig. 4.14, $LWP_{\text{high}}$ and $LWP_{\text{low}}$ (solid blue line) cover the range of variability of the in-situ observations. For the particular case of RF11 (Fig. 4.15), a very reduced number of samples present GVR LWP higher than $LWP_{\text{high}}$ and lower than $LWP_{\text{low}}$.

Figure 4.15: As in figure 4.12a but including the linear best fit of the in-situ observations (blue dashed line) and the upper and lower threshold line for LWP (solid blue lines).

$Nd$ was calculated after applying the threshold technique described above. We compare the histograms of the retrieved $Nd$ with in-situ $Nd$ (from CDP probe, see Chapter 3) for three research flights (Fig. 4.16). Qualitatively, the remote-sensing (red line) and in-situ (back line) histograms show a good agreement, particularly for RF12; however, the retrieved $Nd$ presents more cases with very large and low $Nd$, magnitudes, rarely observed by the C-130 cloud probes. As explained by McComiskey et al. (2009), since equation 4.6 depends on two independent variables with their inherent errors, the parameterized $Nd$ can become unstable at times. The difference between in-situ and
remote-sensing histograms can also reflect that the in-situ and remote sensing (sub-cloud) probes did not sample exactly the same areas.

Similar histograms as in Fig. 4.16 were constructed with all the samples, without applying the thresholds described in eq. 4.7 (Fig. 4.17). It was found that the number of samples with \( N_d < 20 \text{ cm}^{-3} \) dominate RF11 and RF13, while they increase for RF12. This shows that the threshold technique essentially decreases the number of unrealistic \( N_d \), but preserves the distribution shape for values between 50-400 cm\(^{-3} \).

![Figure 4.16: Normalized histograms for retrieved \( N_d \) (red line) with threshold screening applied, and in-situ (CDP) \( N_d \) (black line).](image1)

![Figure 4.17: As in Fig. 4.16 but without using the threshold screening.](image2)
Based on the histograms analysis, we include additional constrains in the $\tau$-LWP relationship by discarding the $\tau$-LWP pairs that produce $N_d$ smaller than 50 cm$^{-3}$ and larger than 400 cm$^{-3}$.

After removing those samples more likely to be affected by instrument and algorithm artifacts, the individual contribution of each research flight to the total dataset is 16%, 77%, and 7% for RF11, RF12, and RF13 respectively.

### 4.5 Aerosol concentration

With available estimates of $\tau$ and LWP, aerosol concentration is the additional variable necessary to estimate the first aerosol indirect effect. A suitable measurement of aerosols, linked to cloud droplet activation, is the cloud condensation nuclei (CCN). Although CCN was measured during sub-cloud transects by the University of Wyoming CCN counter (Snider et al., 2006), the measurements had a relatively low temporal resolution (20 s), in contrast with the 1 Hz sampling of the other instruments used in this investigation, which prevent us from taking full advantage of the sampling capabilities of the GVR. An additional difficulty is that the CCN counter did not properly operate during RF11. Given the issues with the CCN counter we opt for using large particle size concentration (accumulation mode, diameters in the range 0.1-2.9 µm) measured by a passive cavity spectrometer probe (PCASP), sampled at 1 Hz rate during sub-cloud legs. The aerosol accumulation mode is deemed to be the most important aerosol size in cloud formation, due to the low supersaturation required for activation (Roger and Yau, 1988).
An additional implicit assumption in our analysis is that the sub-cloud aerosol measurements are representative of the concentrations found at the cloud base. This assumption is only valid under conditions of well-mixed atmospheric boundary layer. We explore the level of mixing of the boundary layer and the coupling of the cloud layer by comparing the lifting condensation level with the cloud base height derived from the airborne lidar. We found a good correspondence between these two variables (Fig. 4.18), suggesting the occurrence of well-mixed conditions during the research flights studied here. Decoupling cases were also present and more apparent during RF13 (Fig. 4.18, blue dots), associated with frequent occurrence of precipitation, as was also described in Chapter 3. The influence of decoupled conditions was limited further by eliminating periods with the largest decoupling (table 4.4).

![Figure 4.18: Lifting condensation level vs. radar-derived cloud base.](image)
Figure 4.19: Example of time series during RF12: a) GVR LWP (blue) and τ (black), b) \( N_d \) after applying the screening technique, c) accumulation mode \( N_a \).

Figure 4.19 shows a ten minutes time series of the variables used to calculate the ACI metrics: GVR LWP and \( \tau \) (Fig 4.19a), \( N_d \) (Fig. 4.19b) and \( N_a \) (Fig. 4.19c). It is also listed in table 4.5, the dataset used in this chapter. During this period, 60 km were
sampled at a mean aircraft speed of 100 m/s, explaining the apparent high variability of the time series. LWP and $\tau$ show the expected correlation, although $\tau$ retrievals are smoother, because the pyranometer integrates solar radiation from all directions. Although $N_d$ also covaries with LWP and $\tau$, $N_a$ varies at lower frequencies, and its influence in the cloud microphysics is strongly modulated by LWP, due to more active collision/coalescence and stronger vertical updrafts (Feingold et al., 2003). In general, spatial changes attributed to anthropogenic aerosols are difficult to observe over this region because aerosols, microphysics, and boundary layer properties are spatially correlated (Chapter 2). This spatial dependence stresses the importance of investigating cloud-aerosol interactions for fixed atmospheric conditions (LWP).

<table>
<thead>
<tr>
<th>Variable</th>
<th>Dependence</th>
</tr>
</thead>
<tbody>
<tr>
<td>LWP</td>
<td>GVR radiometer</td>
</tr>
<tr>
<td>$\tau$</td>
<td>PSP pyranometer</td>
</tr>
<tr>
<td>$N_a$</td>
<td>PCASP probe</td>
</tr>
<tr>
<td>$N_d$</td>
<td>LWP and $\tau$</td>
</tr>
<tr>
<td>Cloud reflectivity</td>
<td>Wyoming cloud radar</td>
</tr>
</tbody>
</table>

Table 4.5: Main variables and instruments used in Chapter 4.

4.6 Aerosol cloud interactions metrics (ACI)

4.6.1 Aircraft remote sensing

The relationship between $\tau$ and PCASP aerosol concentration ($N_a$) for LWP smaller than 50 g/m$^2$ is presented in figure 4.20. This example illustrates the idea that $N_a$
and $\tau$ (or $N_d$) can be related through a linear regression ($r = 0.58$). The calculation of ACI (eq. 4.1) is then equivalent to estimate the slope between $N_a$ and $\tau$ through a linear regression, that is:

$$\ln \tau = \alpha_{\tau} \cdot \ln(N_a) + \beta_{\tau}$$  \hspace{1cm} (4.8)

$\alpha_{\tau}$ and $\beta_{\tau}$ are the slope and the intercept respectively, and $ACI_{\tau} = \alpha_{\tau}$. Similarly, $ACI_N$ is calculated as the slope of $\ln(N_a)-\ln(N_d)$.

Figure 4.20: $\tau$ vs PCASP $N_a$ for LWP, higher than 30 and smaller than 50 g/m$^2$. $r = 0.58$.

As mentioned previously, we partition our dataset in six LWP bins, where each bin is expressed as $[\text{LWP}_i \text{LWP}_{i+1}]$, with $\text{LWP}_{i+1}$ equivalent to $\text{LWP}_{i+1} = 1.3\text{LWP}_i$ (table 4.6). LWP smaller than 30 g/m$^2$ and higher than 145 g/m$^2$ are not used because small values are more prone to retrievals errors, and also because large LWP are more likely to be associated with precipitation.

The calculated ACI for two particular LWP partitions is presented in Fig. 4.21. Two features are worth mentioning: The first aspect is the relative consistency between ACI calculated from $\tau$ and $N_d$, presenting the expected ratio (1/3). This result is not surprising since a constant LWP guarantees the consistency between $ACI_{\tau}$ and $ACI_N$ (eq.
4.6). The second point is that the slope seems to decrease when increasing the magnitude of the bin, a trait that is better illustrated in Fig. 4.22, with ACI calculated for the six LWP bins. This figure shows that ACI is high for low LWP, with the highest value for 39.5-50.7 g/m$^2$ LWP, close to the upper bounds (0.33 and 1). For bins larger than 39.5-50.7 g/m$^2$, ACI decreases monotonically until reaching ACI$=0.54$ and ACI$=0.21$ for the bin 85.7-111.4 g/m$^2$. The increase in ACI for the largest bin is connected with large aerosol concentration (Na>600 cm$^3$), occurred during RF12 at 22.4°S and 73°W. Since the number of samples for the bin 111.4-145 g/m$^2$ is small, the calculated slope is more prone to be affected by a few values that depart from the general trend. If Na larger than 350 cm$^3$ are removed, then the 11.4-145 g/m$^2$ bin would have ACI$=0.61$, but with a statistically insignificant correlation (r =0.17).

<table>
<thead>
<tr>
<th>LWP bins [g/m$^2$]</th>
<th>Number of samples</th>
<th>r(Na,τ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30-39.7</td>
<td>163</td>
<td>0.59</td>
</tr>
<tr>
<td>39.7-50.7</td>
<td>256</td>
<td>0.6</td>
</tr>
<tr>
<td>50.7-65.9</td>
<td>298</td>
<td>0.47</td>
</tr>
<tr>
<td>65.9-85.7</td>
<td>321</td>
<td>0.52</td>
</tr>
<tr>
<td>85.7-111.4</td>
<td>248</td>
<td>0.44</td>
</tr>
<tr>
<td>111.4-145</td>
<td>104</td>
<td>0.7</td>
</tr>
</tbody>
</table>

Table 4.6: Number of samples and linear correlation between Na and τ, associated with each LWP bin. Correlations are statistical significant at 99% level according to a one-tailed student’s t test.
Figure 4.21: Measurement of ACI and scatterplots for a) \( \tau \) and b) \( N_d \). The regressions (solid line) are made for the LWP bins: 30-39.5 g/m\(^2\) (red), and 85.7-111.4 g/m\(^2\) (green).

Figure 4.22: ACI as a function of LWP for ACI (black line, left axis) and ACI\(_N\) (red line, right axis). Slopes are statistically significant at the 99% level according to a one-tailed student’s t test.

4.6.2 Cloud probes

Since the inherent retrieval errors might propagate into our estimates of ACI, we use in-situ information to confirm the remote-sensing-based analysis. For this purpose, in-situ cloud measurements, derived directly from the CDP cloud probe (Chapter 3), are used to calculate the in-situ ACI. We averaged 30 seconds PCASP aerosol concentration immediately before (ascent profiles) or after (descent profiles) the aircraft profile
occurrence, and correlate them with the profiles average \( N_d \) (see Chapter 3 for a more detailed description of the aircraft sampling strategy). We only used sub-cloud measurements of the PCASP probe because the instrument is not suitable for in-cloud aerosol measurements. Before calculating the slope between \( N_a \) and \( N_d \), we discard samples with precipitating LWP (from 2D-C) higher than 5 g/m\(^2\), CDP LWP < 10 g/m\(^2\), and CDP \( N_d \) < 10 cm\(^3\). By applying this threshold screening, we are excluding cases with drizzle, as well as potentially broken clouds. When taking all the samples, without stratifying for LWP, \( ACI_N \) is 0.77, with a linear correlation at 0.82. We also attempt a coarse estimate of the dependence of ACI on LWP by dividing the dataset into variables with CDP LWP smaller than 40 g/m\(^2\) and higher than 50 g/m\(^2\). The \( N_a-N_d \) scatterplot for the small bin (LWP<40 g/m\(^2\), red) and large bin (>50 g/m\(^2\), green) exhibit high linear correlations (0.83 and 0.87 respectively) and also contrasting slopes (Fig. 4.23), with \( ACI_N = 0.85 \) for the group with the smaller LWP, and \( ACI_N = 0.53 \) for the larger bin. These results are remarkably similar to those derived from the remote sensing estimates, in which \( ACI_N \) is 0.85 for LWP < 40 g/m\(^2\) and 0.55 for LWP>50 g/m\(^2\). Although the dataset analyzed here is small (27 and 25 samples for the small and large bin respectively), the correlations are high and statistically significant, corroborating the magnitude of the remote sensing ACI, and lending support to the idea that ACI decreases with LWP.
Figure 4.23: Measurement of $\text{ACI}_N$ and scatterplots for in situ observations of microphysics. Red color indicates samples with in-situ LWP < 40 g/m$^2$ (27 samples) and green color for LWP >50 g/m$^2$ (25 samples).

### 4.6.3 Comparison with other studies

The magnitude of the in-situ and remote-sensing-based ACI found here are similar to some other aircraft based studies over stratocumulus areas. In-situ observations of microphysics during the Dynamics and Chemistry of Marine Stratocumulus-II (DYCOMS-II; Twohy, 2005) present $\text{ACI}_N = 0.83$, while observations collected by Martin et al. (1994) produce $\text{ACI}_N=0.75$ (reported in McComiskey and Feingold, 2008). In contrast, Kim et al. (2007), with dataset collected by the Department of Energy Atmospheric Radiation Measurement (ARM) Southern Great Plains site (Oklahoma), found values for the slope CCN-$r_e$ (equivalent to $N_a$-$\tau$) smaller than 0.17. Similarly, ACI measurements derived from ARM observations at Point Reyes (California, McComiskey et al., 2009) also show small ACI, with magnitudes fluctuating between 0.05 and 0.23 (compare with Fig. 4.21). The lower temporal resolution of these landsite studies (due to sampling limitations of the 31 GHz microwave radiometer used to calculate LWP) can be part of the explanation for the low ACI (McComiskey et al., 2009). Nevertheless, the
plausible average effect in ACI, should have also influenced our in-situ estimate of ACI (approximately 30 s average in \(N_a\) and \(N_d\)), suggesting that the ACI differences with respect to the ARM-based studies are not only a temporal resolution effect. Some other factors that can play a role to explain our high ACI are those associated with mechanisms that favor the aerosol activation, such as the aerosol size distribution and hygroscopicity. For instance, the ARM sites, and particularly Oklahoma site, are more likely to be affected by continental aerosols that tend to be less favorable for droplet activation, reducing ACI. The chemical characterization of the atmospheric aerosols is one of the major VOCALS-Rex goals, and its investigation is beyond the scope of this thesis.

The role of the adiabaticity has been extensively discussed in Kim et al. (2008). They showed that for adiabatic clouds ACI tends to be larger because the cloud aerosol interactions are less affected by precipitation and entrainment. The investigation of the adiabaticity is difficult in our case, as the clouds here tend to be more adiabatic than the clouds investigated by Kim et al. (2008). Additional analysis of the aircraft vertical profiles was not conclusive on the role of the adiabaticity to modulate ACI. Uncertainties in both radar-lidar-based and GVR LWP prevent us from more quantitative analysis of the adiabaticity factor.

4.6.4 Controlling factors

An interesting aspect found here is the dependence of ACI on LWP, a feature also observed by McComiskey et al. (2009). They hypothesize that collision and coalescence can be responsible for a decrease of \(N_d\), lowering the slope \(N_a-N_d\). This aspect is debatable in non-precipitating clouds because collision-coalescence is less efficient for
high $N_d$; that is to say, a collision-coalescence-induced decrease in $N_d$ could be stronger for low $N_d$ and weaker for high $N_d$, potentially increasing ACI rather than decreasing it. Another factor discussed in McComiskey et al. (2008) is the in-cloud turbulence that would favor higher super-saturation, increasing the aerosol activation and ACI. We explored the influence of the in-cloud turbulence in explaining the dependence of ACI on LWP, using in-cloud measurements of the standard deviation of the cloud-column vertical velocity ($\sigma_w$). The comparison between in-situ $N_d$ and $\sigma_w$ (Fig. 4.24a) reveals a positive correlation ($r = 0.27$), indicating that more aerosols activation is the consequence of stronger turbulence. Similarly, LWP is also correlated with $\sigma_w$, with higher linear correlation (Fig. 4.24b, $r = 0.54$). The weaker correlation between $\sigma_w$ and $N_d$ may be attributed to biases in the measurements although the effect of collision-coalescence in lowering the linear correlation cannot be discarded, as it is also suggested by the high correlation between $\sigma_w$ and LWP. In general terms, we can say that turbulence can effectively modify ACI, in agreement with Feingold et al. (2003), although the details of this relationship are modulated by collision and coalescence processes that can induce a reduction in $N_d$ and increase in LWP. It is also interesting to note that an in-situ ACI stratified by magnitudes of $\sigma_w$ do not show an expected increase of ACI with $\sigma_w$ because cases with high $\sigma_w$ are also associated with large LWP, which in turns are associated with smaller ACI.
In an attempt to elucidate the importance of meteorological factors in the interactions between cloud and aerosols, we explore the relationship between winds measured during sub-cloud transects and the remote-sensing ACI. We selected wind speed because it was the only variable that presented some degree of correlation with LWP. The link between wind speed and LWP is better illustrated in the negative correlation shown in Fig. 4.25a for RF12. For RF11 and RF13, the correlations are lower but statistically significant, and mainly explained by the meridional wind (table 4.7). The correlation is consistent with the wind-LWP configuration near the coast (Fig. 4.2 and 4.3), with the coastal jet located 5 degrees south of a coastal maximum in LWP alongshore; and high LWP at 20°S, where the surface winds decrease. The explanation for this negative correlation is not simple as many factors control the variability in cloud water content. For instance, Klein (1997) points out the temperature advection as a mechanism that promotes upward sensible and latent flux in order to maintain a high
cloud amount. As shown in Fig. 4.2 and 4.3, changes in both LWP and wind speed occur 5 degrees south from the core of the coastal jet, particularly for RF12.

![Figure 4.2: Scatterplots between: a) wind speed and GVR LWP, and b) wind speed and cloud base height/LCL differences during sub-cloud transect for RF12.]

The negative correlation between wind speed and cloud top height for RF11 and RF12 (Table 4.7) suggests the configuration of a meteorological pattern that dynamically controls the co-variability of wind speed and inversion base. Although a subsidence/anticyclone pattern seems to explain the observed correlation, we did not find satisfactory relationships between surface pressure, winds, and cloud top height.

We also investigate the association between the surface winds and the coupling of the boundary layer. We calculate the boundary layer coupling as the difference between cloud base height and LCL. The correlation between wind speed and boundary layer coupling is low for RF11 and RF12 (Fig. 4.25b) but high for RF13. The RF13 high correlation is the consequence of more westward sampling over a region that tends to exhibit more decoupling, deeper boundary layer, more frequent drizzle, and dominant
easterlies (Bretherton et al., 2010). In contrast, RF11 and RF12 favored coastal sampling that was dominated by meridional winds, shallower boundary layer, less precipitation and more coupled conditions (Bretherton et al., 2010).

<table>
<thead>
<tr>
<th></th>
<th>RF11</th>
<th>RF12</th>
<th>RF13</th>
</tr>
</thead>
<tbody>
<tr>
<td>( r ) (wind speed, GVR LWP)</td>
<td>-0.39</td>
<td>-0.64</td>
<td>-0.23</td>
</tr>
<tr>
<td>( r ) (meridional wind, GVR LWP)</td>
<td>-0.38</td>
<td>-0.63</td>
<td>-0.27</td>
</tr>
<tr>
<td>( r ) (wind speed, cloud top height)</td>
<td>-0.35</td>
<td>-0.71</td>
<td>-0.08</td>
</tr>
<tr>
<td>( r ) (wind speed, cloud top height-LCL)</td>
<td>0.23</td>
<td>0.05</td>
<td>0.61</td>
</tr>
</tbody>
</table>

Table 4.7: Linear correlation between: wind speed and LWP (first row), meridional wind component and LWP (second row), meridional wind component and cloud top height (third row), and wind speed and cloud base height/LCL differences. Gray shading indicates correlations statistically significant at the 99% level according to a one-tailed student’s t test.

The relationship between ACI, LWP, and wind speed is better depicted when taking LWP-bin-averaged magnitudes of wind speed (Fig. 4.26). Since LWP is correlated with wind speed, \( ACI_N \) also increases with wind speed. This correlation is not necessarily causal, as this relationship well can be the consequence of spatial co-variability. On the other hand, the wind-ACI correlation is not associated with a boundary layer decoupling because 90% of the samples used to compute ACI correspond to RF11 and RF12, in which we find well mixed conditions and infrequent drizzle. The lack of dependence of the atmospheric decoupling on the surface winds for RF11 and RF12 (table 4.7) also indicated that changes in ACI with LWP are not the artificial consequence of using sub-cloud \( N_a \) when the boundary layer is not well mixed. This is further supported by in-situ
observations of cloud microphysics and $N_a$ at the cloud base (Fig. 4.23) that also reproduced the dependence of $ACI_N$ on LWP.

![Figure 4.26: Scatterplot between wind speed averaged per LWP bin and remote-sensing $ACI_N$.](image)

4.7 Albedo susceptibility

Changes in $ACI_N$ are also closely connected with changes in $N_d$, indicating the modulation of more activated aerosols in $N_d$ (Fig. 4.27). The dependence of $N_d$ on LWP in Fig. 4.27 can be interpreted as the role of collision-coalescence processes that reduces $N_d$ for large LWP. These changes in $N_d$ and LWP also anticipate a control in the cloud albedo, as it is described in the following.

We first calculate the broadband shortwave albedo ($0.28\mu m$ - $2.91\mu m$) at the top of the atmosphere with Streamer (8-streams DISORT), using the retrieved $\tau$ and atmospheric profiles as described in section 4.4. The albedo is then used to investigate its association with changes in $N_d$ for different values of LWP.
Figure 4.27: $\text{ACI}_N$ (black line, left axis) and Mean $N_d$ (gray line, right axis) as a function of LWP.

Figure 4.28: Scatterplot between $N_d$ and albedo ($A$). Colors indicate the LWP intervals: 30-39.5 (blue), 39.5-50.7 (red), (green), 65.9-85.7 (black), 85.7-111.4 (magenta), and 111.4-145 (yellow). Solid lines are the adjusted linear fit as a function of the logarithm of $N_d$.

Fig. 4.28 shows the basic idea of the first indirect effect, that is, increases in $N_d$ are accompanied with an albedo enhancement. Moreover, Fig 4.28 also illustrates the strong control of LWP in the albedo ($A$), with $A$ increasing with the LWP independent of...
\( N_d \). For instance, for clouds with relatively high \( N_d \) (200 cm\(^{-3} \)) and small LWP bins (blue and red colors), their albedo is smaller in magnitude than an albedo associated with low \( N_d \) (100 cm\(^{-3} \)) and high LWP (magenta and yellow colors). Fig. 4.28 also shows that a combination of large LWP and high \( N_d \) produces the highest albedo.

The dependence of the albedo on changes in LWP can be analytically shown by using a two-streams framework, with conservative scattering (Bohren, 1980). The two-streams albedo is expressed in terms of \( \tau \) and the scattering asymmetry parameter \( (g) \) as:

\[
A = \frac{(1-g)\tau}{2+(1+g)\tau} \tag{4.9}
\]

If \( g \) is assumed constant, \( A \) is only a function of \( \tau \). Additionally, we can also express \( \tau \) as a function of \( N_d \) and LWP. If adiabatic conditions are assumed:

\[
\tau \propto N_d^{1/3} \text{LWP}^{5/6} \tag{4.10}
\]

The effect of LWP in \( A \) can be further investigated if one defines the cloud albedo susceptibility to changes in LWP \( (S_{\text{LWP}}) \). Here the focus is determining the isolated impact of LWP in \( A \), so we assume \( N_d \) constant:

\[
S_{\text{LWP}} = \frac{dA}{d\text{LWP}} \bigg|_{N_d} = \frac{\partial A}{\partial \tau} \frac{d\tau}{\text{LWP}} \bigg|_{N_d} \tag{4.11}
\]

We use the eq. (4.8) and (4.9) to obtain:

\[
S_{\text{LWP}} = \frac{5}{6} \frac{A \cdot (1-A)}{\text{LWP}} \tag{4.12}
\]

\( S_{\text{LWP}} \) complements the cloud albedo susceptibility due to changes in \( N_d \) (and fixed LWP), which expressed in terms of the two-streams approximation is \( (S_o) \):
\[
S_o = \left. \frac{dA}{dN_d} \right|_{\text{LWP}} = \left. \frac{A(1-A)}{3N_d} \right|_{\text{LWP}}
\]  
(4.13) (Platnick and Twomey, 1994)

Equations 4.10, 4.12, and 4.13 shows that changes in LWP can have and impact 2.5 stronger than those due to cloud microphysics for a same fractional change in LWP and \(N_d\). This analysis emphasizes the importance of the mean state of the cloud (controlled by large scale forcings) to investigate the first aerosol indirect effect and it should not be interpreted in the sense that the impact of \(N_d\) is negligible. In fact, Fig. 4.28 demonstrates that large changes in \(N_d\) can effectively offset or reinforce the radiative effect of LWP. Additionally, Eq. 4.13 also indicates that the absolute susceptibility decreases with \(N_d\) \(\left( S_o \propto 1/N_d \right)\), a result that can also be derived from Fig. 4.28, because \(A\) can be modeled as a logarithmic function of \(N_d\) (solid lines in Fig. 4.27).

We qualitatively assess the cloud albedo response by calculating the relative albedo susceptibility \((S_R)\); that is, the fractional changes in albedo due to fractional changes in \(N_d\). \(S_R\) is a valuable variable because it allows a computation of the albedo perturbation with respect to the background conditions in \(N_d\) and \(A\). Similar to ACI, \(S_R\) is calculated as the slope of the logarithm of \(N_d\) and \(A\), justified by the high linear correlation suggested by Fig. 4.29. In agreement with the dependence of ACI on LWP, \(S_R\) also decreases with LWP, showing that higher fractional changes in \(N_d\) (due to an increase in \(N_d\)) are also consistent with larger fractional increase in albedo (Fig. 4.30).
4.8 Discussion

4.8.1 Aerosol Cloud interactions

The negative correlation between LWP and wind speed is not consistent with the findings of Chapter 2 that showed that a decrease in meridional winds at 850 hPa (and at surface) is associated with an increase in the air temperature at 850 hPa, enhancing the
lower tropospheric stability, lowering the boundary layer height and reducing the cloud depth (or LWP). In contrast, during RF11, RF12, and RF13, an increase in wind speed is associated with a decrease of the boundary layer depth (cloud top height) and LWP. This apparent inconsistency can be explained by the fact that the correlations found here represent snapshots of spatial variability, without fully capturing the synoptic evolution described in Chapter 2. It is possible that the modulation of the winds in LWP reflect the importance of the anticyclone and subsidence in determining the conditions that dictate the evolution of LWP. It is interesting to note that the correlations are very small during RF13, a flight that sampled weak winds with a dominant zonal component (Fig 4.3c).

The puzzling negative correlation between ACI and LWP also found in Kim et al. (2008) and McComiskey et al. (2009) is not easily explained, although a close connection with the wind speed near the surface suggests a modulation of turbulence in the aerosol activation or less likely, the role of sea salt in the production of cloud droplets. Other mechanisms such as feedbacks between Nd and cloud dynamics cannot be discarded. For instance, large eddy simulations indicate that clouds with higher Nd due to pollution tend to present more evaporation at the cloud top, increasing the mixing that dries the boundary layer and reduces the LWP (e.g. Hill et al, Ackerman et al., 2004). The way these processes can affect aerosol activation is not clear as the entrainment can be reduced near the coast, as a consequence of the strong inversion and moister free troposphere (Bretherton et al., 2010).
4.8.2 Radiative forcing

This susceptibility analysis complements a satellite-based work by Oreopoulos and Platnick (2008) that emphasize the radiative importance of the cloud microphysics in a global context. In their study, they found that marine stratocumulus present the highest relative susceptibility. Nevertheless, simplifications in their treatment of the cloud microphysics used in the forward radiative calculations, namely a fixed cloud depth and liquid water content, with and a vertically homogeneous cloud, could lead to inaccurate estimates of susceptibility. In contrast, our susceptibility computations include variation of LWP, implying that $N_d$ is not only a function of $r_c$ as in Oreopoulos and Platnick (2008). Additionally, the derivatives in this chapter are estimated as linear fits, whereas Oreopoulos and Platnick derived the albedo susceptibility from the radiative transfer model after including small microphysical perturbations.

A robust result found here is that both ACI and $S_R$ decreases with LWP (Fig. 4.30), suggesting that coastal clouds are more prone to changes in albedo, due to the presence of high $N_d$ and low LWP (Chapter 2). In this chapter we have found that the high relative albedo susceptibility over the Southeast Pacific is accompanied with high sensitivity of the clouds to perturbations in aerosol concentrations. A simple computation can help to put in perspective the results presented here, by calculating the magnitude of $N_d$ and albedo response due to an arbitrary increase in $N_a$. We select a 10% increase in $N_a$\(^7\) and then calculate the effect of this increase in $N_d$, and albedo. Here we are discarding any second indirect effect, that is to say, LWP is not affected by changes in $N_a$. If we assume a typical daily mean value of shortwave radiation, representative of

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\(^7\) Oreopoulos and Platnick (2008) used a 10% increase of $N_a$. 
spring conditions over the region of study, at 450 W/m², we can estimate the radiative forcing associated with an increase of aerosol concentration (table 4.8). The magnitudes of radiative forcing calculated here are larger than those reported in Intergovernmental Panel on Climate Change (IPCC 2007, Figure 2.14 therein), which are typically smaller than – 2 W/m², even though climate models estimates of the indirect effect are made by assuming a threefold increase in aerosol mass (Menon et al., 2002). Since our estimate of radiative forcing only applies to overcast stratocumulus clouds at small zenith angle, with fixed atmospheric conditions, and derived from empirical relationships; a direct comparison with modeling estimates is difficult. However, what our results emphasize is the idea that the large aerosol activation in these clouds can induce surprisingly large changes in the planetary albedo. This simple calculation encourages further investigation on the ability of global climate models to reproduce this albedo response for different LWP scenarios.

<table>
<thead>
<tr>
<th>LWP (fixed)</th>
<th>%N_a</th>
<th>%N_d</th>
<th>%A</th>
<th>ΔQ [w/m²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>30-39.7</td>
<td>10%</td>
<td>8.4</td>
<td>2.07</td>
<td>-9.31</td>
</tr>
<tr>
<td>39.7-50.7</td>
<td>10%</td>
<td>10</td>
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Table 4.8: Changes in N_d, A, and radiative forcing associated with a 10 % increase in N_a
Chapter 5

Summary and discussion

In this thesis we have investigated the interactions between aerosol, cloud microphysics, and the role of the synoptic atmospheric conditions to modulate the cloud radiative properties over the Southeast Pacific stratocumulus regime; the most persistent low cloud regime in the planet. This work has been motivated by the uncertainties in the magnitude of the first aerosol indirect effect, that also hamper the investigation of the factors that determine the strength of this effect, and the induced change in cloud albedo due to a fractional change in aerosols.

We first investigate the meteorological factors associated with variability in MODIS $N_d$ along the Chile coast, relying on a composite technique. MAX and MIN satellite $N_d$ composites are defined by the top and bottom terciles of daily area-mean $N_d$ values over the Arica Bight, the region with the largest mean oceanic $N_d$, for the five October months of 2001, 2005, 2006, 2007 and 2008. The MAX-$N_d$ composite is characterized by a weaker subtropical anticyclone and weaker winds both at the surface and the lower free troposphere than the MIN-$N_d$ composite. The MAX-$N_d$ composite clouds over the Arica Bight are thinner than the MIN-$N_d$ composite clouds, have lower cloud tops, lower near-coastal cloud albedos, and occur below warmer and drier free tropospheres (as deduced from radiosondes and NCEP Reanalysis). CloudSat radar reflectivities indicate little near-coastal precipitation. The co-occurrence of more
boundary-layer aerosol/higher $N_d$ within a more stable atmosphere suggests a boundary layer source for the aerosol, rather than the free troposphere.

Analysis of Terra and Aqua MODIS $N_d$ differences suggest the presence of a diurnal cycle in cloud microphysics, also suggested by VOCALS-Rex observations. The inter-satellite differences in $N_d$ are more apparent over highly polluted conditions (MAX $N_d$), exhibiting differences close to 60 cm$^3$. Even though these changes may be attributed to cloud thinning and clear-sky contamination, for some cases the changes in $r_e$ and $\tau$ are not consistent with a clear-sky bias. It is uncertain the mechanisms that might drive a diurnal cycle in $N_d$, and why the signal is stronger over highly polluted environments. Further investigation is necessary to understand the effect of the environmental conditions, a region that exhibits a conspicuous coastal diurnal changes in the boundary layer depth, and strongly controlled by a diurnal subsidence wave (e.g. Garreau and Munoz, 2004; Zuidema et al., 2009).

The MAX-$N_d$ composite cloud thinning extends offshore to 80°W, with lower cloud top heights out to 95°W. At 85°W, the top-of-atmosphere shortwave fluxes are significantly higher (~50%) for the MAX-$N_d$ composite, with thicker, lower clouds and higher cloud fractions than for the MIN-$N_d$ composite. The change in $N_d$ at this location is small (though positive), suggesting that the MAX-MIN $N_d$ composite differences in radiative properties primarily reflects synoptic changes. Circulation anomalies and a one-point spatial correlation map reveal a weakening of the 850 hPa southerly winds decreases the free tropospheric cold temperature advection. The resulting increase in the static stability along 85°W is highly correlated to the increased cloud fraction, despite accompanying weaker free tropospheric subsidence.
Since both satellite cloud top height and inversion temperature indicate less favorable conditions for aerosol entrainment during MAX N_d, we speculate that the main source of anthropogenic aerosols must be situated within the boundary layer. This hypothesis implies the contribution of several sources alongshore and perhaps includes non-point-source pollution that has been brought out to sea, more likely from the more developed southern region of Chile.

In Chapter 3, we carried out a comprehensive validation analysis of satellite cloud microphysics from the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument, one of the most commonly satellite instruments utilized in cloud-aerosol research. We used cloud microphysical observations collected during VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx), over the Chile-Peru stratocumulus cloud deck, were used to assess the accuracy of the Moderate Resolution Imaging Spectroradiometer (MODIS) level 2 retrievals. The in-situ cloud effective radius (r_e), optical thickness (τ), and liquid water path (LWP) were constructed from the drop size distributions measured by the Cloud Droplet Probe (drop diameter < 52 µm) and Two-Dimensional Cloud Probe (drop diameters up to 1600 µm) during twenty vertical profiles made by the C-130 aircraft in October-November 2008 in mostly overcast conditions with little precipitation. MODIS τ correlated well with the aircraft-derived value with a positive bias (1.42). In contrast, the standard 2.1 µm-derived MODIS r_e systematically exceeded the in-situ cloud-top r_e, with a mean bias of 2.08 µm, and an error increasing with droplet size. This bias was not thought to be an attribute of the cloud probes. In addition, the r_e retrieved using the 1.6 µm and 3.7 µm reflectances
did not appear to provide further information about the cloud vertical structure. Three sources of errors that could contribute to the MODIS $r_e$ positive bias were investigated further: the cloud mode droplet size distribution breadth, the presence of a drizzle mode, and the sensor viewing angles. A slight decrease in effective radius was found for scan-edge pixels compared to nadir-view pixels, opposing the detected bias. The droplet spectra effects accounted for $r_e$ offsets smaller than 0.6 µm, 0.9 µm, and 1.6 µm for non-drizzling, light-drizzling, and heavy-drizzling clouds respectively. An additional drizzle mode could explain at most 17% of the effective radius bias in the two cases with sizable drizzle water content. Thus, differences in assumed and observed dropsize distributions cannot fully explain the MODIS bias, supporting earlier studies that find satellite visible/near-infrared $r_e$ overestimates are a general feature of retrievals over stratocumulus regions. An explanation for the observed MODIS bias is lacking although three-dimensional radiative effects were not fully considered. The MODIS $r_e$ overestimate has ramification for the secondarily-derived MODIS LWP, in that the MODIS-derived LWP also exceeds the in situ LWP values. A MODIS-derived cloud droplet number concentration ($N_d$) estimate agreed the best of the four MODIS variables with the aircraft observations, with a small offset and a high linear correlation ($r = 0.94$), although this exercise pointed to the need for a consistent definition for $N_d$ within the research community.

With a better regional picture of the changes in $N_d$ and the atmospheric circulation, our efforts were finally focus on accurately quantifying the first aerosol indirect effect, using VOCALS-Rex aircraft information. The challenge of obtaining
simultaneous high-frequency measurements of LWP and $\tau$ is approached utilizing remote-sensing techniques applied to measurements made by an airborne G band water vapor radiometer (GVR) and an airborne broadband pyranometer during sub-cloud flights. Specifically in this thesis, we have developed a technique to retrieve $\tau$ from a pyranometer. Despite the difficulty of validating the retrievals, we find that the LWP-$\tau$ pair is physically consistent, with a similar trend as the in-situ observations. In order to eliminate suspicious retrievals, we screen the data that did not reach threshold values defined from in-situ observations.

We define six different LWP bins, aimed to control for changes in the atmospheric conditions and cloud dynamics, a fundamental aspect of the first aerosol indirect effect. For each bin, we calculate the aerosol-cloud interactions metrics, using $\tau$ (ACI$\tau$) and $N_d$ (ACI$N$), as the slope between the logarithm of $N_a$ and the logarithm of $\tau$ or $N_d$. Both ACI$\tau$ and ACI$N$ are consistent and show value decreasing with LWP, with ACI close to its theoretical value when LWP is small. These magnitudes were significantly higher than some other in-situ remote sensing studies, though similar to aircraft estimates over the North Atlantic Ocean. Independent estimates of ACI based on a reduced in-situ dataset provided by the C-130 cloud probes corroborate the magnitudes of the remote sensing ACI and also confirm that ACI decreased with LWP. Additionally, we use a third independent bulk\textsuperscript{9} estimate of ACI using MODIS $N_d$ and ship-based $N_a$ (Chapter 2). This satellite-based ACI$N$ was 0.56 (for pixels with $N_d > 50$ cm$^{-3}$), which is remarkably similar to our bulk ACI estimate. Although NOAA cruises favored more coastal sampling than the C-130 flights analyzed here, it is interesting to note that three independent ACI

\textsuperscript{9} Without controlling for LWP
estimate present a remarkable agreement, giving additional support to the idea that cloud-aerosol interaction over the Southeast Pacific region are strong.

The explanation for a decrease in ACI with LWP is still uncertain. We investigate single meteorological variables measured during sub-cloud flights, in an attempt to understand the importance of the atmospheric conditions to modify LWP and ACI. It was found that wind speed near the surface was negatively correlated with LWP but positively correlated with ACI. At this stage of the analysis, any causal relationship between LWP and wind speed is only speculative and well could only reflect that most of the meteorological variables co-vary (Zuidema et al., 2009). The suggestion that the dependence of ACI on LWP can reflect a “physical artifact” due to less coupled conditions is not supported by in-situ calculations of ACI that were obtained using cloud probe measurements and \( N_d \) right at the cloud base. The dependence of LWP on the wind speed might suggest the importance of the temperature advection to increase water vapor and latent heat fluxes, as suggested by Klein (1997). On the other hand, the dependence of ACI on LWP well could imply an active role of the turbulence on the activation of the atmospheric aerosols. Additionally, Since the cloud top cooling increases with LWP, the in-cloud radiative-driven turbulence can also be higher in deeper clouds.

It is finally investigated the relationship between \( N_d \) and albedo (A) through the cloud albedo relative susceptibility (\( S_R \)) or the fractional change in A due to a fractional change in \( N_d \). The magnitudes of susceptibility found here are difficult to compare with other studies due to the scarcity of this type of analysis and because susceptibility is generally calculated without stratifying for LWP, making impossible to determine if the change in A is due to LWP or \( N_d \), especially because A is more sensitive to changes in
LWP than $N_d$. When binning for LWP, it is observed that $S_R$ decreases with LWP, indicating that coastal clouds are more prone to albedo changes due to fractional change in $N_d$. This result shows that clouds with large ACI also present higher $S_R$, suggesting that the increase in $S_R$ is the consequence of more active cloud-aerosol interactions.

When assuming typical shortwave irradiance at the top of the atmosphere, it is found that when $N_a$ is increased in 10%, the change in shortwave reflected fluxes would vary between 11 and 3 W/m$^2$, magnitudes larger than global estimates of indirect effect derived from global climate models, even though we use a rather low $N_a$ fractional change.

The findings of this thesis underline the necessity of more regional analyses, in order to have a good understanding of the global significance of the first aerosol indirect effect. Similitudes of the cloud response between different cloud regimes can be used to test and improve the indirect effects representation in global climate models.

### 5.1 Future work

A better description of the plausible diurnal cycle in cloud microphysics found in this thesis requires the use of other available datasets. Geostationary satellite such as NOAA Geostationary Environmental Satellite (GOES) can provide the necessary temporal resolution for the investigation of the diurnal cycle. For instance, Garreaud et al. (2001) found signatures of diurnal cycle in GOES $r_e$ and $\tau$ during some particular days of October 1999. While the main advantage of GOES is its high temporal resolution, some other issues need to be kept in mind in future investigations: relatively low pixel resolution, geometry viewing angles less suitable for satellite retrievals, and less accurate
instrument radiometric calibration as compared with MODIS.

Ship observations by the NOAA Research Vessel R. H. Brown during VOCALS-Rex are advantageous because they are more temporally extensive (40 days), in contrast with the six 8-hours C-130 daytime flights. The R. H. Brown observations include a pyranometer, GVR, and a 23 and 31 GHz microwave radiometer. The water vapor path derived from a 23 GHz radiometer would allow a better constraint of the GVR LWP calculations, presenting a good opportunity for a more detailed validation of LWP satellite retrievals, from both visible/near-infrared sensors (e.g. MODIS) and microwave instruments such as the Advanced Microwave Scanning Radiometer - Earth Observing System. Ship-based observations also provide a good opportunity to calculate ACI, following a similar methodology as in chapter 4. A main advantage of additional ACI from ship measurements is that the role of the boundary layer dynamics in the aerosol indirect effect can be better studied by combining wind profiler measurements, radiosondes, and radar observations. Additionally, the ship-based dataset is also well suited to understand further the influence of different synoptic configuration in the ACI metrics.

The change in albedo due to a perturbation in cloud microphysics also needs to be addressed globally. For example, Oreopoulos and Platnick (2008) calculated the albedo response to a perturbation of $N_d$ through radiative transfer simulations for fixed values of cloud effective radius and optical thickness from MODIS, providing a first global estimate of susceptibility. It would valuable to recalculate the cloud albedo response with better estimates of albedo, for instance from Clouds and the Earth’s Radiant Energy System instrument (CERES, Wielicki, 1996), which is to some extent independent of the
plane parallel approximation. If combined with MODIS $N_d$, and LWP, empirical susceptibility for different regimes of LWP can be derived from satellite observations. A more realistic treatment of $N_d$ along with more exact observations of shortwave fluxes at the top of the atmosphere, should contribute to improve the estimate of the cloud radiative forcing.
Appendix 1

Derivation of liquid water path and number of droplet for an adiabatically stratified cloud

Let $n(r)$ the cloud droplet size distribution, which represents the number of droplet per volume per droplet size. The integral of $n(r)$ for all the domain of droplet sizes should produce the total droplet number concentration per volume ($N_d$) or:

$$N_d = \int r n(r) \cdot dr \ \text{(A1.1)}$$

The third ($r_3$) and the second moment ($r_2$) of the droplet size distribution $n(r)$ are defined as:

$$r_3 = \int r^3 \cdot n(r) \cdot dr \ \text{(A1.2)}$$

$$r_2 = \int r^2 \cdot n(r) \cdot dr \ \text{(A1.2)}$$

And the effective radius ($r_e$) and optical thickness ($\tau$) are:

$$r_e = \frac{r_3}{r_2} \ \text{(A1.3)}$$

$$\tau = \int_0^Z \beta \cdot dz \ \text{(A1.4)}$$

$\beta$ is the volume extinction coefficient and is expressed as:

$$\beta = \int r n(r) \left[ Q_e(r) \cdot \pi \cdot r^2 \right] \ \text{(A1.5)}$$
Q_{e}(r) is the extinction efficiency, that reaches its asymptotic value (~2) in the visible and near-infrared for typical cloud droplet sizes. Considering this, $\tau$ can be expressed in terms of the second moment of the droplet size distribution as:

$$\tau = \int_{0}^{\infty} Q_{e} \pi r_{c} d\pi (A1.6)$$

The liquid water content (LWC) is defined as:

$$LWC = \frac{4\pi \cdot \rho_{w}}{3} \int_{r} n(r) \cdot r^{3} = \frac{4\pi \cdot \rho_{w}}{3} r_{c}^{3} \quad (A1.7)$$

Using the definition of $r_{c}$ and LWC (A1.3, A1.6, and A1.7), we can express $\tau$:

$$\tau = \frac{3}{4\rho_{w}} \int_{0}^{\infty} \frac{LWC}{r_{c}} \cdot dz \quad (A1.8)$$

Moreover, $r_{3}$ can be rewritten in terms of LWC as:

$$r_{3} = \frac{3}{4\pi \cdot \rho_{w}} LWC \quad (A1.9)$$

In order to link LWC and $r_{c}$, we first need a way to relate the third moment of the droplet spectra to $r_{c}$. Martin et al. (1994) noted that $r_{3}$ and $r_{c}$ can be related through a constant “k”, defined as:

$$k = \frac{N_{d}}{r_{c}^{3}} \quad (A1.10)$$

From (A1.9), we can express $r_{c}$ using the definition of $k$ (A1.10) as:
As shown in chapter 2, LWC increases linearly with height \( z \):

\[
LWC = \Gamma \cdot z \quad (A1.12)
\]

\( r_e \) near the cloud top is estimated from (A1.11) and (A1.12):

\[
r_e = \left( \frac{3 \cdot LWC}{4\pi \cdot k \cdot \rho_w \cdot N_d} \right)^{1/3} \quad (A1.13)
\]

\( Z \) is the cloud depth. We use \( r_e \) at the cloud top because satellite \( r_e \) is more representative of that level.

Using (A1.8), (A1.10), and (A1.12), \( \tau \) is expressed as:

\[
\tau = \frac{3}{4 \cdot \rho_w} Q \int_0^Z \frac{\Gamma \cdot z \cdot dz}{\left( 3 \cdot \frac{4\pi \cdot k \cdot \rho_w \cdot N_d \cdot \Gamma \cdot Z \right)^{1/3}}
\]

\[
\tau = \frac{3}{4 \cdot \rho_w} Q \int_0^Z (\Gamma \cdot z)^{2/3} \cdot \left( \frac{4\pi \cdot k \cdot \rho_w \cdot N_d}{3^{1/3}} \right)^{1/3} \,dz
\]

\[
\tau = C_1 \Gamma^{2/3} k^{1/3} N_d^{1/3} Z^{5/3} \quad (A1.14)
\]

with \( C_1 = \frac{9 \cdot (4\pi)^{1/3} Q_e}{20 \cdot \rho_w^{2/3} \cdot 3^{1/3}} \)

This result assumes that \( N_d \) is constant with height, which is consistent with the \( N_d \) profiles observed during VOCALS-REx.

We can note that both (A1.13) and (A1.14) have dependences on \( \Gamma \) and \( Z \). Since liquid water path (LWP) of LWC is:

\[
LWP = \int_0^Z LWC \,dz = \Gamma \cdot \frac{Z^2}{2} \quad (A1.15)
\]
We can obtain (A1.15) as a function of \( r_e \) and \( \tau \) when multiplying (A1.11) and (A1.13):

\[
r_{e} \cdot \tau = \left( \frac{3}{4\pi \cdot k \cdot \rho_w N_d} - \Gamma \cdot Z \right)^{1/3} \cdot C_1 \Gamma^{2/3} k^{1/3} N_d^{1/3} Z^{5/3} = \frac{9Q}{20\rho_w} \frac{2^{LWP}}{\Gamma \cdot Z^2}
\]

It is clear then:

\[
LWP = \frac{5}{9}\rho_w r_e \cdot \tau \quad \text{(A1.16)}
\]

This LWP is valid for a stratified cloud, independent of the value of \( \Gamma \).

Similarly, we can use further (A1.13) and (A1.14) to derive a relationship for \( N_d \), by replacing the value of \( Z \) found from (A1.13) in (A1.14)

\[
\tau = C_1 \Gamma^{2/3} k^{1/3} N_d^{1/3} \left( r_e^3 \cdot \frac{4\pi \cdot k \cdot \rho_w \cdot N_d}{3\Gamma} \right)^{5/3}
\]

\[
\tau = C_2 \cdot r_e^5 \cdot k^2 \cdot N_d^2
\]

\[
\Rightarrow N_d = \frac{C_2^{1/2} \tau^{1/2} r_e^{-5/2}}{k} \quad \text{(A1.17)}
\]

with \( C_2 = \frac{Q}{20\Gamma} \cdot \frac{4\pi^2 \rho_w}{10\Gamma} = \frac{4\pi^2}{10\Gamma} \cdot \rho_w \)

The dependence of \( N_d \) on \( \Gamma \) is now explicit in (A1.17)

\( N_d \) can also be expressed as a function of LWP and \( \tau \), using the A1.16 and A1.17

\[
N_d = \frac{C_2^{-1/2} \tau^{1/2} \left( \frac{9LWP}{5 \tau \cdot \rho_w} \right)^{5/2}}{k}
\]
\[ N_d = \frac{C_3 \tau^3 \cdot \text{LWP}^{-3/2}}{k} \] (A1.18)

with \( C_3 = \rho_w^3 \left( \frac{5}{9} \right)^{5/2} \left( \frac{10 \Gamma}{4\pi} \right)^{1/2} \)
Appendix 2

List of the vertical profiles used in Chapter 2

Time is in days (starting from the 1st of January of 2008, UTC time). The RF column shows the research flight mission names, with A and D denoting ascents and descents profiles respectively. \( r_e \) is at the cloud top, \( N_d \) is the mean value of each profile, \( \tau \) and LWP are calculated from the combined droplet spectra, whereas LWP\(_{2D-C}\) is calculated only from the 2D-C probe.

<table>
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<th>Time</th>
<th>RF</th>
<th>Lat</th>
<th>Lon</th>
<th>( r_e ) [( \mu \text{m} )]</th>
<th>( \tau )</th>
<th>( N_d ) [( \text{cm}^{-3} )]</th>
<th>LWP [( \text{g/m}^2 )]</th>
<th>LWP(_{2D-C}) [( \text{g/m}^2 )]</th>
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<td>11.49</td>
<td>5.55</td>
<td>47.60</td>
<td>43.07</td>
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<td>318.8009</td>
<td>13D</td>
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<td>1.80</td>
<td>5.84</td>
<td>23.93</td>
<td>3.60</td>
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# Appendix 3

## List of Acronyms

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
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<tbody>
<tr>
<td>2D-C</td>
<td>Particle Measuring System’s Two-Dimensional Cloud optical array probe</td>
</tr>
<tr>
<td>A</td>
<td>Albedo</td>
</tr>
<tr>
<td>ACI</td>
<td>Aerosol cloud interaction metrics</td>
</tr>
<tr>
<td>β</td>
<td>Volume extinction coefficient</td>
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<tr>
<td>Γ</td>
<td>Lapse rate of the water content</td>
</tr>
<tr>
<td>CDP</td>
<td>Cloud Droplet Probe</td>
</tr>
<tr>
<td>CTH</td>
<td>Cloud top height</td>
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<tr>
<td>f_e</td>
<td>Cloud fraction</td>
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<tr>
<td>GVR</td>
<td>G-band vapor radiometer</td>
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<tr>
<td>k</td>
<td>Ratio between the third and second moment of the droplet size distribution</td>
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<tr>
<td>LWC</td>
<td>Liquid water content</td>
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<tr>
<td>LTS</td>
<td>Lower tropospheric stability</td>
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<tr>
<td>LWP</td>
<td>Liquid water path</td>
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<tr>
<td>MAX</td>
<td>Composite of maximum Nd</td>
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<tr>
<td>MIN</td>
<td>Composite of minimum Nd</td>
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<tr>
<td>N_a</td>
<td>Aerosol concentration</td>
</tr>
<tr>
<td>N_d</td>
<td>Cloud droplet number concentration</td>
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<tr>
<td>PVM</td>
<td>Particle Monitor Probe</td>
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<tr>
<td>Q_e</td>
<td>Extinction efficiency</td>
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<td>Q_pyr</td>
<td>Pyranometer shortwave irradiance</td>
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<tr>
<td>r_e</td>
<td>Cloud effective radius</td>
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<td>r_e1.6</td>
<td>1.6 µm wavelength r_e</td>
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<tr>
<td>r_e2.1</td>
<td>2.1 µm wavelength r_e</td>
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<tr>
<td>r_e3.7</td>
<td>3.7 µm wavelength r_e</td>
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<td>σ</td>
<td>Spread of the cloud mode droplet size distribution</td>
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<td>σ_w</td>
<td>Standard deviation of the vertical velocity</td>
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<td>RF</td>
<td>Research flight</td>
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<td>S</td>
<td>Albedo susceptibility to changes in $N_d$</td>
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<tr>
<td>$S_{LWP}$</td>
<td>Albedo susceptibility to changes in LWP</td>
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<tr>
<td>$S_R$</td>
<td>Relative susceptibility to changes in $N_d$</td>
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<td>SW</td>
<td>Upward shortwave fluxes from CERES</td>
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<td>$\tau$</td>
<td>Cloud optical thickness</td>
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<td>$\tau^*$</td>
<td>Cloud optical path from the cloud top</td>
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</table>
References


Intergovernmental Panel on Climate Change, Climate Change, 2007: The physical science basis, edited by S. Solomon, D. Qin, and M. Manning, Geneva Switzerland. (Available at http://www.ipcc.ch/).


