Response to Referee #3

Thanks to the reviewer for the very helpful suggestions, which have allowed us to clarify and improve the manuscript. Below, we address the reviewer's comments, with the reviewer comments in black and our responses in blue. We have also revised the manuscript accordingly.

The manuscript is a rexamination of the VOCALS aerosol-cloud dataset obtained from sixteen flights of the CIRPAS Twin Otter that each profiled the below-, in-, and above- cloud environment over the southeast Pacific Ocean. Relationships between the cloud droplet number and relative dispersion to sub-cloud CCN(s=0.2

1) The authors need to do a better job of explaining what this study contributes over and above the previous papers that have been published on VOCALS. Who is the audience for the paper (i.e., who would be interested in the findings)? How does the manuscript represent a substantial contribution to scientific progress (through substantially new concepts, ideas, methods, or data) as required by the ACPD publication criteria? Right now, I would say that it does not represent a substantial contribution (which, in my opinion, makes this paper a borderline reject). Please discuss these details briefly in the abstract and more thoroughly in the last paragraph of the introduction; the current brief paper section layout discussion on Lines 62-64 is not particularly useful and could be replaced.

   Thanks to the reviewer for the thoughtful comments and suggestions. The aerosol climatic effect is one of the greatest uncertainties in climate predictions, particularly the indirect effect, i.e., aerosol-cloud interaction. Data analyses based on ground, aircraft, and satellite measurements, as well as numerical simulations, are usually conducted to investigate aerosol-cloud interactions. Aircraft measurements from VOCALS provide detailed information on the microphysical properties of aerosols and clouds and their vertical profiles; thus, these measurements are very useful in studies on the topic discussed in this manuscript. We agree that there are quite a few previous studies that have been published on VOCALS measurements. Most of these studies either described the instruments or presented the properties of aerosols and clouds, which certainly improved our understanding of the properties of aerosols, clouds and BLs over the SEP. However, few of these previous studies explored the detailed processes of aerosol-cloud interactions, especially physical processes such as dispersion effects, entrainment mixing, and the impacts of these effects on the interaction. Using the microphysical properties of both aerosols and clouds, as well as the meteorological parameters from VOCALS, we explored the detailed processes on aerosol-cloud interaction over the SEP, including (a) the controlling factors of cloud droplet formation, (b) the dispersion effect after constraining the differences in cloud dynamics, and (c) the entrainment mixing process near the stratocumulus top and its impact on clouds. To our knowledge, such kind of analysis has not yet conducted by previous studies using VOCALS measurements.

   The effect of aerosols on stratocumulus clouds is complicated by various dynamical conditions, e.g., strong wind shear within the BL, moist layers above clouds, and a strong decoupled BL. Thus, we investigate the contribution of cloud dynamics and aerosols to cloud droplet formation, which to our knowledge has not received much attention in previous studies using VOCALS measurements.

In addition to modulating the cloud droplet number, aerosols can also change the shape of the cloud droplet size distribution (i.e., dispersion effect) and thereby cloud albedo (Liu and Daum, 2002). The dispersion effect could act to either offset or enhance the well-known Twomey effect, which mainly depends on the sensitivity of the relative dispersion (ε) to the aerosol number concentration (Na). However, as shown in Table 1, the relationship between ε and Na derived from previous studies remains largely uncertain, implying that the effect of aerosols on ε is often intertwined with the effects of other factors, especially cloud dynamic conditions. Thus, it is necessary to isolate the ε response to aerosol perturbations from meteorological effects, which to our knowledge, was not considered in many previous studies. A clear comparison between 'typical well-mixed' and 'other' cases in this manuscript can aid in understanding the influence of meteorological conditions on the dispersion effect estimation. Aerosols were found to broaden the droplet spectrum, and cloud dynamic perturbations may lead to an underestimation of the aerosol dispersion effect. This result is helpful for reducing the dispersion effect uncertainty and may benefit cloud parameterizations in global climate models to more accurately assess the indirect aerosol effect.

Additionally, entrainment plays a critical role in the formation and evolution of clouds and the change in droplet spectrum, as well as aerosol indirect effects (Chen et al., 2014, 2015; Andersen and Cermak, 2015). However, it remains unclear whether the entrainment-mixing mechanism is predominantly homogeneous, inhomogeneous, or in between (Andrejczuk et al., 2009; Lehmann et al., 2009). To our knowledge, little attention has been given to the entrainment-mixing mechanism obtained during VOCALS. Using cloud observations obtained from other aircraft (G-1) during VOCALS, Yum et al. (2015) showed that both homogeneous and inhomogeneous mixing were found in their analysis and attributed it to method uncertainties. We used a completely different method to re-examine the entrainment-mixing mechanism near the stratocumulus top. As stated by Gerber et al. (2005), in marine stratocumulus clouds, entrainment occurs when the LWC begins to decrease from the bottom of the cloud. In this manuscript, entrainment and non-entrainment zones are thus defined as the regions within 20 m above and below the maximal LWC height, respectively. A comparative analysis of the difference in cloud microphysics between the two zones suggests that the entrainment-mixing mechanism is predominantly extreme inhomogeneous in the stratocumulus clouds during VOCALS. The impacts of entrainment on cloud microphysics are also investigated. Previous studies have noted that applying different assumptions to the entrainment-mixing mechanism would have a significant impact on cloud albedo (Grabowski, 2006; Chosson et al., 2007; Slawinska et al., 2008). Therefore, our results provide insights to improve the understanding of entrainment mixing in stratocumulus clouds and the assessment of aerosol indirect effects and cloud radiative forcing.

According to the reviewer's suggestions, we have added the corresponding discussions to the abstract and introduction to ensure clarity of the novelty and contribution of this study, and we removed the paper section layout discussion from the text.

2) I have a couple of concerns about the treatment of the interstial aerosol. First, the PCASP-100 misses the large fraction of sub-0.1-um aerosols that are unlikely to act as CCN and would therefore remain as intersitial aerosols. Are there data from in-cabin particle counters sampling on an aerosol inlet that could fill in this major gap? Inlet shatter may be more of an issue here, but the sub-cloud measurements would be a good place to quantify the fraction of aerosol number that is sub- and super-0.1-um diameter. It is likely that many (or most) of the interstitial aerosol number is not being captured here.

   We agree that the concentration of interstitial aerosols (> 0.1 μm) measured by PCASP-100 is less than the concentration of all un-activated aerosols, i.e., the interstitial aerosols with diameters smaller than 0.1 μm are not captured here. However, because in-cloud sampling of this part of the aerosols is problematic due to cloud droplet shatter (Hudson and Frisbie, 1991; Clarke et al., 1997; Weber et al., 1998; Kleinman et al., 2012), we do not include it into interstitial aerosols in this manuscript. As suggested by Kleinman et al. (2012), cloud droplet shatter can create a large number of spurious small particles (~ 50 nm), leading to a serious overestimation of interstitial aerosol concentration. Additionally, other previous studies also observed extremely high concentrations of these small particles in-cloud (e.g., 10^3 to more than 10^4 particles cm^{-3} smaller than 50 nm diameter) and attributed these concentrations to droplet shatter (Hudson and Frisbie, 1991; Clarke et al., 1997; Weber et al., 1998). However, the shatter contribution to the total in-cloud aerosols is minor when the diameter is greater than 0.1-0.15 μm (Kleinman et al., 2012). Therefore, treating aerosols larger than 0.1 μm as interstitial aerosols can avoid the interference of cloud droplet shatter to a large extent, which is precisely what we did in this manuscript.

   According to the reviewer's suggestions, we calculated the ratio of the sub-0.1-μm aerosol concentration to the super-0.1-μm aerosol concentration during flights on Oct. 18, where sub-0.1-μm aerosol concentration is derived from the concentration measured by CPC (size range: > 15 nm) minus that measured by PCASP-100 (size range: 0.1-2.0 μm), and the super-0.1-μm aerosol concentration is obtained from the concentration measured by PCASP-100. It is found that the average ratio for in-cloud (2.99) is significantly higher than that for sub-cloud (0.31), which further confirms the contribution of droplet shatter to the spurious increase in small aerosols in clouds. Notably, the average ratio for sub-cloud is only 0.31, indicating that some aerosols could be missed by PCASP-100, but these aerosols are not the majority. Furthermore, as the reviewer stated, most aerosols smaller than 0.1 μm are unlikely to act as CCN and thus affect cloud properties. Therefore, due to the weak connection with CCN, these aerosols that are missed by PCASP-100 may not be important when exploring the impact of aerosols on clouds. For example, the same treatment of interstitial aerosols as in this manuscript has also been employed by other studies (Kleinman et al., 2012) to examine the aerosol effect on clouds.

3) The use of effective diameters to characterize the interstial aerosol (Di) and cloud droplet sizes (De) doesn't make sense to me as this paper is largely focused on number concentrations and number size distributions. While the effective diameter is relevant for remote sensing measurements, there are no remote sensing data presented in this paper. The authors should instead use geometric mean diameters to describe these aerosol populations and better convey the aerosol and cloud diameters relevant for the number distributions.

Thanks for the comments. Our focus in this manuscript is to investigate the effect of aerosols on clouds and hence provide a reference for the assessment of aerosol indirect radiative forcing. The climatic effects of aerosols and clouds are usually estimated based on the radiatively important effective diameters rather than the geometric mean diameters. For this reason, many aircraft-based studies on aerosol-cloud interactions also used effective diameters to represent the sizes of aerosols and cloud droplets (Peng et al., 2002; Zhang et al., 2011; Vogelmann et al., 2012; Yang et al., 2019; Zhao et al., 2018, 2019), although there was no comparison with the remote sensing measurements. Additionally, effective diameters are often used to quantify the extent of dilution of the cloud caused by the entrainment-mixing process (Pontikis et al., 1993; Gerber et al., 2008, 2013; 2016). For the above reasons, we used effective diameters instead of geometric mean diameters in this manuscript.

We agree with the reviewer that the geometric mean diameter is more relevant for the number size distributions. Thus, we compared the effective diameters with the geometric mean diameters of aerosols and cloud droplets, respectively (Figure R1), and found that there is a good correlation between them. This correlation implies that using the two different diameters would not influence our conclusions.

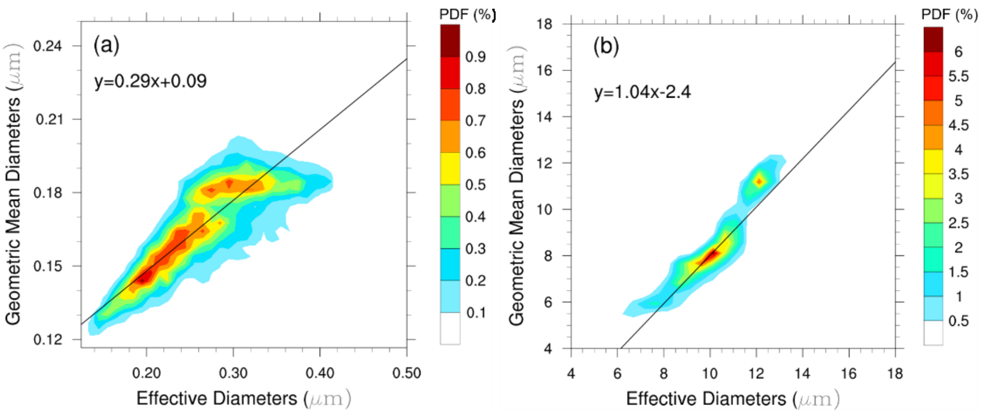

Figure R1. Correlation between geometric mean diameters and effective diameters of (a) aerosols and (b) cloud droplets, respectively. Colors represent the frequency (units: %).

4) In a number of instances associations between sub-cloud and in-cloud variables are misinterpreted to suggest causal relationships that are inconsistent with our understanding of cloud physics. For exampele, on Lines 13-15, it is stated "Our analysis suggest (sic) that the increase in liquid water content (LWC) is mainly contributed by cloud droplet number concentration (Nd) instead of effective radius of cloud droplets in the polluted case, in which more droplets form with smaller size, while the opposite is true in the clean case." On Lines 142-144, it is stated: "This may imply that the increase of LWC induced by sub-CCN is mainly caused by increasing Nd instead of Re. Fig. 3d indicates a positive correlation between cloud depth and sub-CCN..." These statements are either misleading or just not correct. LWC is known to be driven by changes in environmental conditions (i.e., profiles of temperature and total water content as well as entrainment mixing); microphysics are not a primary driver. Similarly, changes in these environmental conditions will also change the cloud base altitude (and hence cloud depth if the cloud top is driven by a constant

inversion height). There is a causal link between sub-CCN and droplet number, while the in cloud supersaturation (again driven by environmental conditions) can also affect Nd. What the analysis shown in Figure 3 does suggest is that there is a correlation between higher sub-CCN loadings and wetter (or colder) environmental conditions, which should be discussed. The old axiom that correlation does not imply causation certainly holds here. These conclusions (on the lines cited above and elsewhere in the manuscript) need to be either revised or removed from the manuscript.

Thanks for the comments. We agree with the reviewer that both environmental conditions and microphysics (i.e., aerosol effects) can affect cloud properties, and usually, the former is the primary driver. On this point, we distinguished between the flights of the typical mixed BL and others to ensure relatively similar meteorological conditions (similar inversion heights, and the jump of potential temperature and total water mixing ratio across the inversion). In addition, the in-cloud dynamics (i.e., vertical velocity) for the 16 non-drizzling flights were also compared (Figure 9 and Table 2). The result indicated that the in-cloud dynamic differences between the typical well-mixed boundary flights is very small, which confirms the assumption of similar meteorological conditions. Therefore, the interference of environmental conditions on the relationships between the cloud and sub-CCN shown in Figure 3 would be minimal.

LWC is a function of both the number (Nd) and size (Re) of cloud droplets, i.e., both Nd and Re contribute to the change in LWC. The objective of the analysis on the relationships between LWC and Nd/Re (Lines 13-15; Figure 4) is to understand which contributed more to the growth of LWC under polluted and clean conditions, i.e., cloud formation under different aerosol loadings. Our analysis shows that the low aerosol concentrations in the clean case inhibit the increase in Nd with LWC, which promotes the rapid increase in Re with LWC. In contrast, there are enough particles that may potentially be activated into cloud droplets under polluted conditions; thus, Nd increases rapidly with LWC, while the increase in Re is suppressed. We agree with the reviewer that the sentences on lines 13-15 and lines 142-144 might be misunderstood, and thus, these sentences have been removed from revised manuscript as suggested.

Minor Comments:

1) On Line 76, it is stated that the PCASP-100 measures the aerosol dry diameter. How was this accomplished? Was some sort of unique inlet heater or dryer used to dry the aerosol? While there will be some ram heating effects that will lower the relative humidity in the PCASP-100 optics region, I don't think that this would be enough to say that the aerosol size is dry.

   Thanks for this comment. The relative humidity of the air sampled by PCASP can be reduced to 40 % by using deicing heaters, which increases the temperature by approximately 10 to 20 °C (Strapp et al., 1992; Hallar et al., 2006; Snider and Petters, 2008). Cabin heat also contributed to the drying of aerosols measured inside the aircraft. We agree with the reviewer that this may be not enough to say the diameter is dry, although the relative humidity is already very low. Thus, we have revised “dry diameter” to “diameter” on line 104.

2) Too many significant figures reported on Line 158. Reporting aerosol concentration as integer

values would be appropriate.

Thanks for the suggestion. In the revised manuscript, the number concentrations of aerosols and cloud droplets were reported as integer values throughout the paper.

3) The manuscript would benefit from some additional proofreading to improve grammatical or typo errors.

We have carefully proofread the manuscript and asked a native English speaker to read and edit the language of the manuscript.

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Exploring aerosol cloud interaction using VOCALS-REx aircraft measurements

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Abstract. In situ aircraft measurements obtained during the VAMOS Ocean-Cloud-Atmosphere-Land Study-Regional Experiment (VOCALS-REx) field campaign are analysed ~~employed~~ to study the aerosol-cloud interactions ~~between aerosol and~~ in the stratocumulus clouds over the southeast Pacific Ocean, with a focus on three understudied topics~~factors~~ (separation of aerosol effects from dynamic effects, dispersion effects, and turbulent entrainment mixing processes)~~as well as entrainment process near the top of stratocumulus and its possible impacts on aerosol-cloud interaction~~. ~~Our analysis suggest that the increase of liquid water content (LWC) is mainly contributed by cloud droplet number concentration (N_d) instead of effective radius of cloud droplets in the polluted case , in which more droplets form with smaller size, while the opposite is true in the clean case. By looking into the influences of dynamical conditions and aerosol microphysical properties on the cloud droplet formation, it is confirmed that cloud droplets are more easily to form under the conditions with large vertical velocity and aerosol size.~~ Our analysis suggests that ~~An~~ an increase in aerosol concentration tends to simultaneously increase both cloud droplet number concentration (N_d) and relative dispersion (ε), while an increase in vertical velocity (w) often increases N_d but decreases ε. After constraining the differences of cloud dynamics, the positive correlation between ε and N_d becomes stronger, implying that perturbations of w could weaken the aerosol influence ~~of aerosol~~ on ε, and hence ~~may~~ result in an underestimation of ~~aerosol~~ dispersion effect. A comparative analysis of t~~T~~he difference of cloud microphysical properties between entrainment and non-entrainment zones ~~confirms~~suggests that the entrainment-mixing mechanism is predominantly extreme inhomogeneous in the stratocumulus that capped by a sharp inversion, whereby ~~, namely the entrainment reduces N_d and LWC by 28.9 % and 24.8 % on average, respectively, while~~ the variation of liquid water content (25 %) is similar to that of N_d (29 %) and the droplet size ~~of droplets is~~ remains ~~relatively unaffected~~approximately constant. In entrainment zone, ~~smaller aerosols and~~ drier air entrained from the top induces less cloud droplets with respect to total in-cloud particles (0.56 ± 0.22) than the case in non-entrainment zone (0.73 ± 0.13) by ~~inhibiting aerosol activation and~~ promoting cloud droplet~~s~~ evaporation. This study is helpful in reducing uncertainties in dispersion effects and entrainment mixing for stratocumulus, and the results of this study may benefit cloud parameterizations in global climate models to more accurately assess aerosol indirect effects.

1 Introduction

Stratocumulus clouds play~~s~~ a key role in the radiative energy budget of the Earth by reflecting incoming shortwave radiation and thus cool~~s~~ the planet surface~~ of the planet~~ and offset~~s~~ the warming by greenhouse gases (Hartmann et al., 1992). Stratocumulus clouds are susceptible to aerosols, i.e., aerosol indirect effect (Twomey, 1974; Albrecht, 1989), which currently remains large uncertainties (Lohmann and Feichter, 2005; Chen and Penner, 2005; Carslaw et al., 2013; McCoy et al., 2017).

Globally, ~~The~~ marine stratocumulus clouds overlaying the southeast Pacific Ocean (SEP) ~~is~~ are the largest and most persistent clouds ~~in the world~~ (Klein and Hartmann, 1993; Bretherton et al., 2004). Sources of anthropogenic aerosols from the Chilean and Peruvian coasts, in contrast with the relatively clean air masses from the Pacific Ocean, make the SEP an ideal region to explore the interaction between~~of~~ aerosols and stratocumulus cloud topped boundary layers. The cloud properties from satellite retrievals exhibit a gradient off the Northern Chile shore~~ of Northern Chile~~. For example, the cloud droplet number concentration decreased from 160 to 40 cm^{-3} (George and Wood, 2010), and the cloud droplet effective radius increased from 8 to 14 µm from the coast to approximately~~about~~ 1000 km offshore (Wood et al., 2006). This gradient is plausibly attributable~~attributed~~ to anthropogenic aerosols near the coast. Huneeus et al. (2006) found that during easterly wind events, sulfate increased by one order of magnitude over SEP, which ~~results~~ resulted in a 1.6 to 2~~-~~fold increase in cloud droplet number concentration. Based on observations from satellites and cruises, Wood et al. (2008) suggested that open cellular convection within an overcast stratocumulus is associated with reduced aerosol concentration~~,~~ and ~~an~~ air masses not passing through the Chilean coast, which further confirms the impact of aerosols on stratocumulus over the SEP. However, it is difficult to establish the generality of previous studies based on satellite remote sensing due to the absence of in situ observations that provide vertical profiles of cloud and aerosol and detailed in-cloud processes.

The VAMOS (Variability of the American Monsoons) Ocean-Cloud-Atmosphere-Land Study-Regional Experiment (VOCALS-REx), which includes multiple aircraft missions, ship and land-based measurements, took place in the region extending from the near-coast~~al~~ of northern Chile and southern Peru to the remote ocean in the SE~~-~~P~~acific~~ during October–November 2008 (Wood et al., 2011). The data collected during this campaign were examined to ~~Studies based on this field campaign provided more information about~~ investigate the properties of aerosols, clouds, and marine boundary layer over the SEP. For instance, Bretherton et al. (2010) found ~~the multi-platform observations during VOCALS revealed~~ that the boundary layer was shallow and fairly well mixed near shore but deeper and decoupled offshore~~ (Bretherton et al., 2010)~~. Twohy et al. (2013) found that the clouds near the shore exhibited higher aerosol concentrations, ~~near shore were associated with more greater droplet concentrations,~~ ~~but~~ smaller ~~cloud~~ droplet sizes and~~, less~~ smaller liquid water path (*LWP*), and suggested ~~thus attributed to~~ a combination ~~ed effect~~ of anthropogenic aerosols and ~~the~~ physically thinner clouds near the shore. ~~Nevertheless~~However, Zheng et al. (2010) found an increase in *LWP* with ~~the~~ cloud condensation nuclei (CCN) concentrations ~~was found during~~under the similar meteorological conditions~~ (Zheng et al., 2010)~~. Additionally, chemical components and sources of aerosols during VOCALS-REx campaign have been discussed in several studies (Chand et al., 2010; Hawkins et al., 2010; Allen et al., 2011; Twohy et al., 2013; Lee et al., 2014).

Although these studies have improved our understanding of some aspects related to aerosol, cloud and boundary layer properties over SEP, several important factors remain understudied or unexplored. ~~the mechanisms of the detailed processes on interaction between aerosol and stratocumulus cloud is still unclear.~~ First, the aerosol effect on clouds is often intertwined with the effects of other factors, especially meteorological conditions (Fan et al., 2009; Koren et al., 2010). Currently, the impact of aerosols on the shape of cloud droplet size spectrum (i.e., dispersion effect) is reported to remain large uncertainty. The observed correlations between relative dispersion (ε) and N_d can be positive, negative, or not evident (Table 1), which could be largely attributable to the coincidentally changing cloud dynamics. Thus, it is necessary to isolate the response of ε to aerosol perturbations from meteorological effects, which to our knowledge, has not received adequate attention in many previous studies. Second, applying different assumptions to the entrainment mixing mechanism can have a significant impact on cloud albedo (Grabowski, 2006; Chosson et al., 2007; Slawinska et al., 2008). Additionally, more recent studies suggested that entrainment mixing may be a possible physical interpretation for the observed anti-Twomey effect (Ma et al., 2018; Jia et al., 2019). However, it remains unclear whether the entrainment-mixing mechanism is predominantly homogeneous, inhomogeneous, or in between (Andrejczuk et al., 2009; Lehmann et al., 2009). By using cloud observations obtained from G-1 aircraft during VOCALS−REx, Yum et al. (2015) found both homogeneous and inhomogeneous mixing in their analysis and attributed it to the uncertainty in the methods they used. Uncertainty in the entrainment mixing mechanism could lead to the inaccurate assessment of aerosol indirect effects. Thus, more attention should be paid to this topic.

Based on the useful information on the microphysical properties of aerosols and clouds provided by previous studies, in this study, we conduct additional explorations regarding aerosol-cloud interactions over the SEP by employing in situ aircraft data collected by CIRPAS Twin Otter aircraft during VOCALS-REx, which include the following: (a) investigating the controlling factors of cloud droplet formation (e.g., cloud dynamics and aerosols), (b) evaluating the dispersion effect under relatively constant cloud dynamical conditions, and (c) re-examining the entrainment-mixing mechanism by using a different approach from that of Yum et al. (2015).

~~By employing in situ aircraft data collected by CIRPAS Twin Otter aircraft during VOCALS-REx, we investigate the following issues in this study: (a) the relationships between aerosol and cloud properties; (b) cloud droplet formation and its influencing factors; (c) dispersion effect (i.e., the influence of aerosol on the shape of cloud droplet size spectrum), and (d) entrainment process near the top of stratocumulus and its impact on cloud. This paper is organized as the follows: The instruments and measurement data are described in Sect. 2, and the main results are discussed in Sect. 3. A summary and discussion is given in Sect. 4.~~

2 Data and method

2.1 Aircraft Data

The Twin Otter operated by the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) was aimed to observe aerosol and~~,~~ cloud microphysics, and turbulence near Point Alpha (20° S, 72° W) off the coast of Northern Chile from 16 October to 13 November 2008. A total of 19 flights were carried out, each of which conducting about 3 hours of sampling at Point Alpha and including several soundings and horizontal legs near the ocean surface, below the cloud, near the cloud base, within the cloud, near the cloud top, and above the cloud (Fig. 1). Since all flight tracks are similar, only one track (Oct. 18) is shown in Fig. 1. As cloud and aerosol probe measurements failed during the flight on 5 November and drizzle processes occurred on the flights on 1 November and 2 November, only the observations from other 16 non-drizzling flights are included in this paper.

Both t~~T~~he aerosols below and above clouds and the interstitial aerosols in-cloud~~ data was~~ were obtained by Passive Cavity Aerosol Spectrometer Probe (PCASP-100), which counted and sized particles from 0.1–2.0 μm ~~dry ~~diameter with 20 bins (Zheng et al., 2011; Cai et al., 2013; Twohy et al., 2013). The CCN number concentration was observed by the CCN Spectrometer at a supersaturation of 0.2 % and 0.5% respectively. The cloud data include cloud droplet number concentration (N_d, size range: 2.07–40.2 μm with 20 bins) from the Cloud, Aerosol and Precipitation probe (CAS), effective radius of cloud droplets (R_e), and liquid water content (LWC) from the PVM-100 probe (Gerber et al.,1994). All data sets used in this study are at a frequency of 1 Hz. ~~The calibrations of the onboard instruments were carried out so as to provide standard meteorological variables, aerosol, and cloud observations.~~ Zheng et al. (2011) pointed out that uncertainties of aerosols and clouds measured by these probes are within 15 %. More detailed information about the ~~observation ~~instruments and measurements on board the CIRPAS Twin Otter aircraft during VOCALS-REx can be found in Zheng et al. (2010) and Wood et al. (2011).

2.2 Data processing

In this study, the data collected near the land~~,~~ during both take-off and landing~~,~~ are removed to ensure that only the measurements close to Point Alpha (20° S, 72° W) are analysed. Only t~~T~~he ~~occurrence of ~~clouds ~~is defined by the following criterion, i.e., ~~with $LWC > 0.05$ g m^{-3} and $N_d > 15$ cm^{-3} are selected for analysis. We averaged the CCN number concentrations during the legs within 200 m above the cloud top to obtain the average above-cloud CCN, and within 200 m below the cloud base to obtain the mean sub-cloud CCN. During the study period, the CCN Spectrometer constantly measured CCN at a supersaturation of 0.2 % except on the first four flights at a supersaturation of 0.5 %. In order to have a consistent comparison between all flights, we adopted the method by Zheng et al. (2011) to adjust the CCN concentration from supersaturation of 0.5 % to 0.2 % on the first four flights. ~~Since t~~The effective ~~diameter ~~radius of aerosol particle ~~is not measured directly, so we ~~is calculated from ~~it according to ~~the PCASP-measured ~~ments of ~~aerosol size distributions based on following equation:

$$\cancel{D}Ra = \sum n_i \cancel{d}r_i^3 / \sum n_i \cancel{d}r_i^2 \qquad (1)$$

where n_i is the aerosol number concentration in the ith bin of PCASP, and ~~d_i~~ r_i represents the arithmetic mean ~~diameter~~ radius of ith bin.

To investigate the impact of the entrainment mixing processes on cloud properties and aerosol-cloud interactions, we defined entrainment zone and non-entrainment zone, respectively. Gerber et al. (2005) showed that, in the marine stratocumulus, entrainment occurs when LWC begins to decrease from the bottom of the cloud. In this manuscript, entrainment and non-entrainment zone are thus defined as the regions within 20 m above and below the height of maximal LWC, respectively. Given that the two zones are both thin layers, there is little difference in the dynamical and thermos-dynamical conditions. It is therefore assumed that the difference of cloud microphysical characteristics between the two zones is only caused by entrainment.

3 Results

3.1 Vertical profiles of aerosol, cloud and meteorological variables

Figure 2 shows ~~The~~ the vertical profiles of temperature (a), relative humidity (b), liquid water content (c), cloud droplet effective radius (d), cloud droplet number concentration (e), aerosol effective radius (f), and the ratio of CCN to condensation nuclei (g) ~~aerosol, cloud and meteorological variables~~ during the 16 flights. Note that the vertical altitude is normalized ~~-are scaled~~ by the inversion height (z_i) ~~(Fig. 2), which is~~ defined as the height of the maximum~~where~~ ~~the~~ vertical gradient of liquid water potential temperature (θ_L) ~~is the largest~~ (Zheng et al., 2011~~, Betts, 1973~~). ~~θ_L is conservative for water phase changes, but same as potential temperature when no liquid water exist (Betts, 1973).~~ The~~is~~ normalization ~~could~~ minimizes ~~exclude~~ the effect of the variation of z_i between flights, ~~and~~ allowing for ~~-hence~~ better exploration of ~~for exploring~~ the average boundary layer (BL) structure during VOCALS-REx.

As shown in Fig. 2a, temperature (T) decreased~~s~~ sharply with ~~the~~ height within the BL, which is close to the dry adiabatic lapse rate. A strong inversion occurred~~s~~ at the top of the BL, with ~~the~~ an average temperature ~~change~~ increase of approximately~~about~~ 10°C. Due to the decrease of~~reduced~~ T with height and the nearly constant water vapor mixing ratio within the strong mixing BL, the relative humidity (RH) increased~~s~~ rapidly with ~~the~~ increasing height (Fig. 2b). T and RH reached the minimum and maximum, respectively, when z/z_i was~~is~~ close to 0.9. Near the top of the BL ($0.9 < z/z_i < 1.0$), the entrainment of the dry and warm air from the free atmosphere aloft resulted~~s~~ in a slight increase in T and a slight decrease in RH. ~~When~~ As z/z_i varied from ~~>~~ 1 to 1.1, T increased~~s~~ from 11 to approximately~~about~~ 18°C, and RH rapidly decreased to approximately~~about~~ 16 % ~~rapidly~~ (Fig. 2a, b). The vertical profiles of T and RH are overall consistent with the observations of other marine stratocumulus clouds (Martinet et al., 1994; Keil and Haywood, 2003). ~~Corresponding to vertical variation of RH, the N_d gradually increases with the height, reaches the maximum when RH is maximum (z/z_i = 0.9), and then decreases when 0.9 < z/z_i~~

~~< 1.0, indicating that more cloud droplets are nucleated in high supersaturation. The profile of *LWC* and R_e is similar to that of N_d (Fig. 2d, e).~~ For the cloud properties, an average of all profiles that normalized by z_i only may be insufficient to indicate the vertical variation of clouds due to different cloud base heights of each profile. Thus, the average profiles are not shown in Fig. 2c, d, and e, and the vertical variation of cloud properties can easily be seen from the single profile. Figure 2c shows that the *LWC* first increased with height from the cloud base, reached the maximum at z/zi = 0.9 and then decreased with further increasing height when 0.9 < z/zi < 1.0. The profile of R_e is similar to that of *LWC* (Fig. 2d). The profile of N_d remains relatively constant, with a slight increase and decrease near the base and top, respectively (Fig. S1), which is consistent with the results from other VOCALS-REx observations (Painemal and Zuidema, 2011). It is interesting to note ~~Fig. 2f reveals~~ that the effective radius of aerosol particles (R~~D~~$_a$) below cloud is larger than that above cloud, which is probably attributable~~attributed~~ to the differences in aerosol sources and aerosol properties (e.g., chemical composition) ~~t chemical composition and sources of aerosols~~ (Fig. 2f). The profile of *CCN/CN* is similar to that of ~~D_a~~ R_a (Fig. 2g), suggesting that aerosols with large sizes are more likely to become CCN (Dusek et al., 2006; Zhang et al., 2011). Larger ~~D_a~~ R_a and *CCN/CN* values are also found in polluted case than clean cases.

3.2 Relationships between aerosol and cloud properties

~~Aerosol indirect effect is one of the largest uncertainties in current climate assessments.~~ The relationships between aerosol and cloud properties are essential to understanding and evaluating aerosol-cloud interactions. Most studies based on satellite data have employed aerosol optical depth or aerosol index as a proxy for ~~agents of~~ CCN number concentration to investigate ~~the~~ aerosol-cloud interactions (Koren et al., 2005, 2010; Su et al., 2010; Tang et al., 2014; Ma et al., 2014, 2018; Wang et al., 2014, 2015; Saponaro et al., 2017). However, not all aerosols ~~on~~ in the vertical column are actually involved in cloud formation~~,~~; thus, this assumption is ~~relatively rough~~ questionable, especially when the cloud layer is decoupled from the aerosol layer. ~~.~~ For example, a few studies have shown ~~Several studies revealed~~ that aerosols have little effect on cloud properties when aerosol and cloud layers are clearly separated (Costantino and Bréon, 2010, 2013; Liu et al., 2017). ~~In this study,~~ To further investigate this issue, the ~~impact of~~ CCN number concentration both below cloud (*sub-CCN*) and above cloud (*abv-CCN*) ~~near cloud layer~~are examined for their impacts ~~, e.g. below and above cloud respectively,~~ on the cloud properties. ~~is assessed.~~

Figure 3 shows t~~T~~he relationships between ~~sub-cloud CCN number concentration (~~*sub-CCN*~~)~~ and cloud properties during all 16 non-drizzling flights ~~during all flights are shown in Fig. 3~~. The red dots ~~signify~~ denote the ten flights with typical ~~well~~ well-mixed boundary layer (BL)~~and non-drizzling cases~~. These flights ~~, which have relatively~~ also shared similar meteorological conditions, such as ~~similar~~ inversion heights~~,~~ and the jump of potential temperature and total water mixing ratio across the inversion (Zheng et al., 2010), and thus can be used to isolate the response of cloud properties to aerosol perturbations. The blue dots represent the other cases~~,~~ in which the conditions except typical ~~well~~ well-mixed ~~boundary~~

~~layer~~BL~~ and non-drizzling~~, such as strong wind shear within the BL, moist layers above clouds, strong decoupled BL and so on, are involved (Table 2). ~~In~~ For the cases ~~of~~ with typical well~~-~~mixed ~~boundary~~ BL~~with non-drizzling~~, both *LWC* (Fig. 3a) and N_d (Fig. 3b) exhibited ~~the~~ positive ~~relationships~~ correlations with *sub-CCN*, with correlation coefficients of 0.60 and 0.79, respectively. It is worth highlighting that the similar increases of N_d and *LWC* led to ~~, while~~ R_e ~~has~~ having no evident correlation with *sub-CCN* (Fig. 3c), as expected from the conventional first aerosol indirect effect whereby a constant LWC is assumed.~~. This may imply that the increase of *LWC* induced by *sub-CCN* is mainly caused by increasing N_d instead of R_e. Fig. 3d indicates a positive correlation between cloud depth and *sub-CCN*, with correlation coefficient of 0.71. As cloud top height is mainly determined by the temperature inversion condition, there is no obvious correlation between cloud top height and sub-CCN, with correlation coefficient of only −0.13 (Fig. 3e). However, the correlation coefficient between cloud base height and *sub-CCN* is −0.69 (Fig. 3f), suggesting that CCN thickening cloud is mainly induced by lowering cloud base. It is noted that the above conclusions are only valid in the typical mixed boundary layer.~~ For the ~~In~~ other cases (~~i.e.~~ blue dots), the *sub-CCN* impacts ~~of aerosols~~ on the cloud properties were ~~is~~ not evident due to the large differences in the meteorological conditions and the ~~boundary layer~~BL structure.

Compared to sub-cloud CCN, the influence of above-cloud CCN on cloud properties is very weak, even for the cases with typical well-mixed BL. The absolute values of the correlation coefficient between the ~~above-cloud CCN number concentration (~~*abv-CCN*~~)~~ and cloud properties are all less than 0.4 (not shown~~figure omitted~~), and~~,~~ none of them ~~which~~ passed the significance test (α = 0.05). In this study, the above-cloud aerosol number concentration is very low (129~~.8~~ ± 60~~.1~~ cm^{-3}), and the inversion ~~capped~~ capping the cloud top is extremely strong, which weakens the aerosol mixing ~~of the aerosol~~ with cloud layer and hence the aerosol effects ~~of aerosol~~ on cloud properties. Some previous studies based on aircraft observations for stratocumulus clouds also found that N_d exhibits a significantly positive correlation with *sub-CCN*, but no correlation with *abv-CCN* (Martin et al., 1994; Hudson et al., 2010; Hegg et al., 2012).

Figure 4 contrasts the relationships of N_d (a) and R_e (b) as ~~a~~ functions of *LWC* between ~~In order to investigate cloud formation in different aerosol loadings,~~ the most polluted (Oct. 19) and ~~the~~ cleanest (Nov. 09) cases with aerosol concentrations of 647~~.78~~ ± 60~~.47~~ cm^{-3} and 268~~.97~~ ± 35~~.67~~ cm^{-3}, respectively. Also shown are the corresponding power-law fits~~, are selected in this study~~. ~~Vertical profiles for the two cases are highlighted in Fig. 2, showing that N_d and *LWC* in polluted case are larger than those in clean one, but R_e remains the same.~~ Although N_d and R_e both increased with first increasing *LWC* and then levelled off, there were significant detailed differences between the polluted and clean cases. The polluted case exhibits a steeper increase of N_d with increasing *LWC* than the clean case when *LWC* is small, whereas the opposite was true for R_e. The low aerosol concentrations under the clean case inhibit the increase of N_d with *LWC* (Fig. 4a), which hence promotes the rapid increase of R_e with *LWC* (Fig. 4b). In contrast~~On the contrary~~, there are enough particles ~~which~~ that may potentially activated into cloud droplets under the polluted case~~,~~; thus, N_d increases rapidly with *LWC*. As ~~the~~ a certain amount of water is shared by large amount particles, the increase of R_e is limited. ~~It is suggested that the increase of *LWC* is mainly controlledcontributed by N_d instead of R_e when aerosol concentrations is high, in which large number of cloud droplets are formed with smaller size, but the opposite is true when aerosol concentrations is low.~~ The result is consistent with the study in Beijing by Zhang et al. (2011), but the difference ~~of~~ in cloud formations between the clean and polluted conditions is less evident, which is ~~probably~~ likely attributable~~attributed~~ to the much smaller difference in ~~lower~~ aerosol concentration ~~difference between clean and polluted cases~~ in this study (approximately~~about~~ 400 cm^{-3}) than that in Zhang et al. (2011) (approximately~~about~~ 7000 cm^{-3}).

3.3 Cloud droplet formation and its controlling factors

Sub-cloud CCN is considered ~~as~~ to be a good proxy for ~~the~~ aerosols entering a cloud. However, during the actual flight, it is difficult to simultaneously collect enough samples of sub-cloud CCN and cloud droplets ~~simultaneously~~, which may result in ~~uncertainty in~~ statistical analysis uncertainty. This limitation can be remedied ~~overcome~~ by ~~employ~~using the total particle concentration, which equals the aerosol concentration outside the clouds and the sum of the droplet concentration and interstitial aerosol~~s~~ concentration inside the clouds. Interstitial aerosols are particles observed inside clouds that either have never activated into cloud droplets or have been deactivated ~~but then return~~ into aerosols after ~~evaporation of~~ cloud droplet evaporation. Kleinman et al. (2012) pointed out that the number concentration of interstitial aerosols (N_i) can be obtained either directly from the observation of in-cloud aerosols~~,~~ or indirectly from a number balance between sub-cloud and in-cloud particles. In this study, the interstitial aerosol properties are derived from direct measurements in the cloud. By employing aircraft observations over both land and ocean, Gultepe et al. (1996) found that the difference ~~of~~ in the number concentration between the total in-cloud particles ($N_d + N_i$) measured directly and sub-cloud aerosols is very small. Thus, ~~It is thus assumed that~~ the total in-cloud particles ~~can~~ are assumed to characterize the overall level of in-cloud aerosol concentration before activation. Figure 5 shows an example ~~The flight~~ of the ~~on~~ Oct. 18 flight ~~is singled out as a case study~~ to support this assumption ~~(Fig. 5)~~. It is shown that the number concentrations of sub-cloud aerosols and total in-cloud particles are very close, with ~~the~~ values of 583~~.7~~ ± 55~~.4~~ cm^{-3} and 567~~.4~~ ± 59~~.1~~ cm^{-3}, respectively. Similar results are also found in the other flights. The average ratio of $N_d + N_i$ to the sub-cloud aerosol concentration during all flights is 0.94, which is ~~much~~ smaller than the value (1.29) found by Kleinman et al. (2012) based on G-1 aircraft measurements during VOCALS-REx. Therefore, the ~~observation of~~ interstitial aerosol~~s~~ observations in this study ~~is~~ are unlikely to be significantly interfered with by factors such as cloud droplet shatter and cloud droplet evaporation due to instrument heating, as discussed by Kleinman et al. (2012), which has the potential to create spurious~~more~~ extra aerosols in-cloud.

The relations between N_d and $N_d + N_i$ during the 16 non-drizzling flights are shown in Fig. 6, ~~in which~~where the color~~s~~ represents the in-cloud vertical velocit~~y~~~~ies~~. All flights exhibited p~~P~~ositive correlations between N_d and $N_d + N_i$ ~~are found in all flights~~, representing the aerosol-cloud interaction (IPCC, 2001, 2007, 2013; Hegg et al., 2012). In addition, the effect of dynamical conditions on cloud droplet formation is evident. As ~~presented~~ shown in Fig. 6, the data are close to the 1:1 line when the vertical velocity is relatively large, namely, the ~~in-cloud~~ aerosols ~~are~~ were almost entirely activated into cloud droplets. However, the data deviate from the 1:1 line when the vertical velocity is small or negative. For example, for all flights, the average ratio of N_d to $N_d + N_i$ with vertical velocity greater than 1 m s^{-1} is 0.84 ± 0.12, which is much larger than that with vertical velocity less than –1 m s^{-1} (0.64 ± 0.14). The regime-dependent behaviour ~~This~~ is likely ~~possibly attributable~~~~attributed~~ due to the high supersaturation caused by the adiabatic uplift when ~~under conditions with large~~ the vertical velocity is large (Reutter et al., 2009; Chen et al., 2016). ~~High supersaturation not only induces more aerosols to reach critical supersaturation and then activate into cloud droplets, but also inhibits cloud droplet evaporation.~~

In addition to the dynamical conditions, aerosol microphysical properties~~,~~ such as size distribution and chemical components can~~,~~ also significantly affect activation process ~~significantly~~ (Nenes et al., 2002; Lance et al., 2004; Ervens et al., 2005;Dusek et al., 2006; McFiggans et al., 2006; Zhang et al.,2011; Almeida et al., 2014; Leck and Svensson, 2015). Since part of aerosol~~s~~ population ~~in the cloud have~~has activated~~to cloud droplets~~, it is difficult to obtain ~~the~~ information ~~of~~ about aerosol size before activation. According to the Köhler theory, larger aerosols have smaller ~~the~~ critical supersaturations~~ of aerosol with large size is relatively low~~, and thus, they activate preferentially, suggesting that~~,~~ ~~i.e.~~ the effective ~~diameter~~ radius of interstitial aerosols (~~D_i~~R_i) is smaller than that of the ~~initial~~ aerosols before activation. Li et al. (2011) compared the difference ~~of~~ in size distribution between interstitial aerosols and aerosols that have been activated to cloud droplets, and found that the peak diameter of the former (0.45 µm) was much smaller than that of the latter (0.8 µm). It can be thus inferred that the size of aerosols activated to cloud droplets, and thus the size of initial aerosols would be larger with ~~the~~ an increase ~~of~~ in ~~D_i~~R_i, though the quantitative relationship depends on in-cloud dynamics. Therefore, it is assumed that~~,~~ when compared with the data measured at different sampling locations during flight, the size of the interstitial aerosols can still represent the size of ~~initial~~the aerosols before activation to some extent. As indicated in Fig. 7, the larger ~~D_i~~ R_i is, the closer the data are~~is~~ to the 1:1 line, i.e., the higher proportion of cloud droplets in total in-cloud particles ($N_d/(N_d + N_i)$) is. The averaged $N_d/(N_d + N_i)$ for all flights is 0.76 ± 0.13 when ~~D_i~~ R_i is larger than ~~1.0~~0.5 µm, but only 0.64 ± 0.23 when ~~D_i~~ R_i is less than 0.25 µm. It is because ~~that~~ those aerosols with large sizes are more likely to be activated into cloud droplets. Additionally, as larger aerosol particles form into larger cloud droplets (Twohy et al., 1989, 2013) that are relatively difficult to evaporate, large particles can also inhibit cloud droplet evaporation to a certain extent.

3.4 Dispersion effect

In addition to modulating the cloud droplet number concentration, aerosols also affect the shape of cloud droplet size spectrum (referred to as the "dispersion effect") and thereby affect the cloud albedo (Liu and Daum, 2002). When the dispersion effect is taken into account, the estimated aerosol indirect forcing could be either reduced (Liu and Daum, 2002; Peng and Lohmann, 2003; Kumar et al., 2016; Pandithurai et al., 2012) or enhanced (Ma et al., 2010), i.e., the dispersion effect could act to either offset or enhance the well-known Twomey effect, which mainly depends on the sensitivity of the relative dispersion (ε, the ratio of the standard deviation to the mean radius of the cloud droplet size distribution) ~~on~~ to the aerosol number concentration (N_a). However, the dependence of ~~relationship between~~ ε ~~and~~ on N_a ~~still~~ is much less studied and remains even more ~~large~~ uncertain~~uncertainty~~ than that of $N_{\cancel{c}d}$. Table 1 ~~s~~summarize~~how~~s ~~that~~ the ~~the~~ observed correlations between ε and N_d (or N_a), being ~~can be~~ positive, negative, or no obvious correlations ~~not evident~~. The d~~D~~ifferent relationships are indicative of the fact that the effect of aerosol on ε is often intertwined with the effects of other factors, especially cloud dynamical conditions (Pawlowska et al., 2006; Lu et al., 2012). In this section, the relationship between ε and N_d based on ~~the~~ in-flight and ~~the~~ flight-averaged data are discussed ~~respectively in order~~ to distinguish the influences of aerosol and cloud dynamics on ε.

Within an individual flight, the aerosol number concentration and chemical components can be assumed to be similar, providing an opportunity to focus on the effect of cloud dynamics to the extent possible. Here, we employ the vertical velocity (w, m s^{-1}) as a proxy for cloud dynamical conditions. As shown in Fig. 8, the correlations between ε and N_d based on in-flight data ~~is~~ are significantly negative during all 16 non-drizzling flights, which is mainly modulated by w, i.e., a larger w corresponds to a smaller ε but a larger N_d. High supersaturation leads to more cloud droplets to activate and grow to the same size (i.e., narrowing the droplet spectrum) when w is relatively large, but a portion of the cloud droplets may evaporate into smaller sizes and even deactivate into interstitial aerosols when w is small or even negative, resulting in ~~the~~ a decrease of N_d and ~~the~~ broadening of the droplet spectrum.

It is interesting to see from Table 1 that the correlations between ε and N_d based on in-flight data are generally negative, while the correlations~~one~~ based on the flight-averaged data could be either positive, negative, or even uncorrelated. The latter uncertain relationships ~~of the later~~ may result from variations of the strength of cloud dynamics between flights, which would disrupt or even cancel the real influence of aerosols on relative dispersion (Liu et al., 2006; Peng et al., 2007; Lu et al., 2012). However, many previous studies did not consider~~take~~ the difference ~~of~~ in cloud dynamics ~~in~~ between flights ~~into account~~ when correlating ε and N_d, which could result in some degree of overestimation or underestimation of dispersion effect. In this study, the data of~~in~~ all flights were sampled over the same location, i.e., Point Alpha, which can reduce the difference ~~of~~ in dynamical conditions caused by variations of horizontal sampling locations. In addition, we also distinguish between the flights of typical mixed ~~boundary layer~~BL and ~~the~~ others to ensure relatively similar meteorological conditions (see section 3.2). Fig. 9 further shows the probability distribution function of w with mean values and standard deviations for 16 non-drizzling flights. The related statistics are given ~~shown~~in Table 2. ~~It can be found that, e~~Except for other cases (~~gray shadow~~crosses; especially Oct. 24, Oct. 29, Nov. 8, and Nov. 13), the difference ~~of~~ in the in-cloud dynamics between typical ~~well~~ well-mixed ~~boundary~~ BL flights is very small, which confirms the assumption of similar meteorological conditions. As indicated in Fig. 10a, ε and N_d were~~are~~ positively correlated (correlation coefficient of 0.29 and ~~the~~ slope of 1.9 × 10^{-4}) in the case of the typical ~~well~~ well-mixed BL~~boundary~~, indicating that increased aerosols concurrently increase~~d~~s ε and N_d ~~at the same time~~. However, the correlation coefficient and slope ~~reduce~~ decrease to 0.11 and 7.7×10^{-5}, respectively, in the all cases (i.e., *w* is not ~~to~~ constrained ~~*w*~~), implying that the influence of aerosols on the ε-N_d relationship tends to be weaker after ~~intertwined~~ intertwining with the effects of cloud dynamics. Although the perturbations of cloud dynamics have been eliminated as ~~far~~ much as possible, N_d is still likely determined by both aerosol~~s~~ number concentrations and updraft velocity together. Therefore, a similar statistical analysis is~~are~~ also conducted for sub-cloud CCN. Similar positive correlations ~~The relationship~~ between ε and sub-cloud CCN were found~~, is similar to that between ε and N_d~~, with much improved ~~but, as expected, the~~ correlation coefficients (slopes). The correlation coefficients (slopes) were 0.67 (3.1×10^{-4}) and 0.31 (2.1×10^{-4}) for ~~in~~ the cases with ~~of~~ typical ~~well~~ well-mixed BL~~boundary~~ and all cases ~~increase to 0.67 (3.1×10^{-4}) and 0.31 (2.1×10^{-4})~~, respectively (Fig. 10b).

3.5 Entrainment in stratocumulus

Entrainment is a key process that occurs in the clouds~~, which~~ and plays an important role in the formation and evolution of clouds and the change of droplet spectrum, as well as the aerosol indirect effect (Chen et al., 2014, 2015; Andersen and Cermak, 2015). The nature of entrainment is related to the cloud type. Entrainment in cumulus is primarily lateral with strong dilution of the cloud, which induces *LWC* to decrease rapidly to approximately~~about~~ 20% of its adiabatic value (Warner, 1955). Entrainment in stratocumulus is mainly determined by the strength of the gradients in buoyancy and horizontal winds (Wang and Albrecht 1994; Gerber et al. 2005; de Roode and Wang 2007; Wood, 2012), and proceeds from the top and mostly affects ~~mostly~~ a thin layer (Gerber et al., 2005), whose dilution effect is much weaker than that in cumulus (Warner, 1955, 1969a, 1969b; Blyth et al., 1988; Gerber et al., 2008; Burnet and Brenguier, 2007; Haman et al., 2007). Aircraft observations of marine stratocumulus clouds showed that the vertical profile of *LWC* is essentially the same as the adiabatic profile, i.e., the cloud is almost adiabatic (Keil and Haywood, 2003). Furthermore, it remains unclear whether the subsequent entrainment-mixing mechanism is predominantly homogeneous, inhomogeneous, or in between (Andrejczuk et al., 2009; Lehmann et al., 2009). Some previous studies have shown that stratocumulus is generally dominated by the inhomogeneous mechanism (Pawlowska et al., 2000; Burnet and Brenguier, 2007; Haman et al., 2007; Lu et al., 2011; Yum et al., 2015). By employing a different vertical description in characterizing the region near cloud top (Malinowski et al., 2013), Gerber et al. (2016) noted that both extreme inhomogeneous mixing and homogenous mixing play a role in unbroken stratocumulus, but the reduction in cloud droplet effective radius appears to be secondary in comparison to the dilution process that preserves the relative shape of the droplet spectrum.

~~In order t~~To explore ~~the~~ entrainment in stratocumulus during VOCALS-REx, we first~~ly~~ compare~~d~~ the differences ~~of~~ in cloud microphysics between the entrainment and non-entrainment zones near the cloud top. Here, the entrainment and non-entrainment zones are defined as the regions within 20 m above and below the height of the maximal *LWC*, respectively. As anticipated, the adiabatic fraction (*AF*, the ratio of the measured *LWC* to its adiabatic value) in the entrainment zone (AF_{ent}) is generally lower than that in the non-entrainment zone ($AF_{non\text{-}ent}$), with ~~the~~ mean values ~~of~~ for all flights of 0.64 and 0.77, respectively (Table 2), which further confirms the rationality ~~in~~ of dividing the two zones. Compared with the non-entrainment zone, the peak ~~diameters~~radius of cloud droplets in the entrainment zone has little changes (Fig. 11), and the effective ~~diameters~~ radius of cloud droplets (~~D_e~~R_e) increases ~~only~~ by only ~~1.8~~2 % (Table 2). However, N_d and LWC decrease significantly on average, by ~~28.9~~29 % and ~~24.8~~25 %, respectively ~~on average~~ (Table 2), especially during the flights on Oct. 18, Nov. 04, Nov. 09 and Nov. 13, where N_d decreases by 60~~.1~~ %, 56~~.3~~ %, 56~~.1~~ % and 59~~.2~~ %, and LWC decreases by 56~~5.7~~ %, 62~~.1~~ %, 56~~5.8~~ % and 5~~8.7~~9 %, respectively (Table 2). It is suggested that dry and warm air entrained from cloud top dilutes N_d and LWC by ~~a~~ similar amounts, while the size of droplets is relatively unaffected, which is thought as extreme inhomogeneous entrainment-mixing process. Additionally, both P_{LWC} and P_{Nd} are negatively correlated with $AF_{ent}/AF_{non-ent}$, with correlation coefficients of -0.60 and -0.47, respectively, indicating the dependence of the LWC and N_d changes on the adiabatic fraction changes (Fig. S2), where P_{LWC} and P_{Nd} are the reduction percentages in LWC and N_d within the entrainment zone relative to the non-entrainment zone. ~~Although iIt is still unclear whether the entrainment-mixing mechanism is predominantly homogeneous, inhomogeneous, or in between (Andrejczuk et al., 2009; Lehmann et al., 2009),~~ . ~~sSome previous studies showed that stratocumulus is, in general, dominated by the inhomogeneous (Pawlowska et al., 2000; Burnet and Brenguier, 2007; Haman et al., 2007; Lu et al., 2011; Yum et al., 2015).~~

The flight on Oct. 18 with strong entrainment is chosen to investigate the difference between the entrainment and non-entrainment zones. As shown in Fig. 12b, dry and warm air entrained from the top reduced the RH in the entrainment zone by 9 % on average and hence acted to accelerate cloud droplet evaporation. Consequently, $N_d/(N_d + N_i)$ in the entrainment zone (0.56 ± 0.22) is much lower than that in non-entrainment zone (0.73 ± 0.13) (Fig. 12c). Additionally, the relative dispersion in the entrainment zone is generally larger than that in the non-entrainment zone (Fig. 12d), implying that drier air entrained from the top could broaden the cloud droplet spectrum by promoting cloud droplet evaporation. Some previous observations also showed that ε with a low AF tends to be larger than that with high AF, and attributed it to the effect of entrainment mixing (Pawlowska et al., 2006; Lu et al., 2009). It is noted that the occurrence frequency of R_i in the entrainment zone is significantly higher than that in the non-entrainment zone when $R_i < 0.38$ µm, but the opposite is true when $R_i > 0.5$ µm (Fig. 12a). This result suggests that in addition to dry and warm air, small particles are also entrained into clouds from the top (Fig. 2f) and large particles are detrained out of the clouds simultaneously. However, the inversion capping a typical stratocumulus is usually too strong to excite significant updrafts near cloud top (Stevens, 2002; Wood, 2012; Malinowski et al., 2013). Ghate et al. (2010) found that vertical velocities near the top of stratocumulus tend towards zero overall with only approximately 4% of updrafts being stronger than 0.5 m s^{-1}. Therefore, although smaller aerosols are entrained into the entrainment zone, these aerosols seem unlikely to influence droplet formation. The effect of entrainment mixing on stratocumulus is mainly governed by the entrained dry air rather than small aerosols.

~~As shown in previous studies, nucleation of cloud droplet mainly occurs near cloud base, and sub-cloud aerosols are the major source of cloud droplets (Pinsky and Khain, 2002; Ghan et al., 2011). However, de Rooy et al., (2013) pointed out that entrainment mixing at the cloud edge and cloud-top contribute significantly to the amount of entrained air and hence aerosols. Therefore, activation of aerosols is not restricted to the cloud base, where the central updraft enters the cloud (primary activation). Slawinska et al. (2012) found that a significant part (40%) of aerosols is activated above cloud base (secondary activation), which is dominated by entrained aerosols. By using large eddy simulations (LES), Hoffmann et al. (2015) suggested that, in a shallow cumulus, sub-cloud aerosols and laterally entrained aerosols contribute to all activated aerosols inside the cloud by fractions of 70% and 30%, respectively. Although entrainment in stratocumulus, discussed in this manuscript, is weaker than that in cumulus, entrained aerosols is still a possible source of cloud droplets. In this study, the flight on Oct. 18 with strong entrainment is chosen to investigate the difference of cloud droplet formation between entrainment and non-entrainment zone. As presented in Fig. 12a, the probability in entrainment zone is significantly higher than that in non-entrainment zone when $D_i < 0.75$ μm, but the opposite is true when $D_i > 1.1$ μm. This result indicates that small particles are entrained into cloud from the top (Fig. 2f) and large particles are detrained out cloud at the same time. The decrease of D_i by 0.18 μm may inhibit aerosol activation into cloud droplet. Furthermore, dry and warm air entrained from the top reduces the relative humidity by 8.8 % on average (Fig. 12b), and accelerates the cloud droplets evaporation. As a result, $N_d / (N_d + N_i)$ in entrainment zone (0.56 ± 0.22) is much lower than that in non-entrainment zone (0.73 ± 0.13) (Fig. 12c). It is also noted that the relative dispersion in entrainment zone is overall larger than that in non-entrainment zone (Fig. 12d), implying that smaller aerosol particles and drier air entrained from the top could broaden cloud droplet spectrum by influencing nucleation and evaporation of cloud droplets. Some previous observations also showed that ε with low AF tends to be larger than that with high AF, and attributed it to the effect of entrainment mixing (Pawlowska et al., 2006; Lu et al., 2009). According to the discussion in Sect. 3.3, although the impact of above-cloud aerosol on whole cloud is much weaker than sub-cloud aerosols, the entrainment of above-cloud aerosols may affect the cloud droplets nucleation, and hence change cloud properties near the cloud top to some extent.~~

4 Summary

By using in situ aircraft data collected by CIRPAS Twin Otter aircraft at Point Alpha during VOCALS-REx from 16 October to 13 November 2008, aerosol-cloud interactions are investigated with a focus on understudied factors including separation of aerosol effects from dynamic effects, dispersion effects, and turbulent entrainment mixing processes. ~~we investigated the interaction between aerosol and marine stratocumulus over the southeast Pacific Ocean, especially the dispersion effect. We also explored the entrainment process near the top of stratocumulus and its impacts on cloud properties and aerosol-cloud interaction.~~

Vertical profiles of aerosol, cloud and meteorological variables indicated~~presented~~ that the BL ~~is~~ was ~~well-mixed and~~ capped by a sharp inversion during 16 non-drizzling flights. Cloud properties~~variables~~, such as LWC~~,~~ and N_d~~, and cloud depth~~, are ~~all~~ positively correlated with sub-cloud CCN number concentration, with~~having the~~ correlation coefficients of 0.60~~,~~ and 0.79~~ and 0.71~~, respectively. No evident correlation was found between cloud properties ~~with~~ and above-cloud CCN number concentrations. This is mainly due to the low aerosol number concentrations above-cloud (129~~.8~~ ± 60~~.1~~ cm^{-3}) and the extremely strong inversion ~~capped~~ capping the cloud top, which inhibits the mixing of the above-cloud aerosols with the cloud layer. Therefore, the influence of the above-cloud CCN on cloud properties is~~ very~~ weaker ~~compared to~~than the sub-cloud CCN. ~~Additionally, the comparison of cloud formation under different aerosol number concentrations conditions suggested that the increase of~~ ~~LWC~~ ~~is probably contributed by~~ ~~N_d~~ ~~instead of~~ ~~R_e~~ ~~in the polluted case due to abundant CCN, in which more but smaller cloud droplets form, while the opposite is true in the clean case.~~

The results showed that both dynamical conditions and aerosol microphysical properties have significant effects on cloud droplet formation. In the case of large vertical velocity and aerosol size, the ~~proportion of~~ cloud droplet proportion of total in-cloud particles is relatively high (e.g., 0.84 ± 0.12 and 0.76 ± 0.13, respectively), i.e., cloud droplets ~~are easier to~~ form more easily. Although aerosol chemical components ~~of aerosol~~ are also critical to cloud droplet formation (Nenes et al., 2002; Lance et al., 2004; Ervens et al., 2005; McFiggans et al., 2006; Wang et al., 2008; Almeida et al., 2014), this topic was not discussed in this study due to the unavailability~~le~~ of measurements.

The correlations between ε and N_d based on the in-flight data, ~~used to~~ representing the w-induced correlation, ~~is~~ are significantly negative, while the correlations derived from flight-averaged data (i.e., aerosol-induced correlation) ~~is~~ are positive. This finding implies that an increase in aerosol concentration tends to concurrently increase ε and N_d~~ at the same time~~, while an increase in w often increases N_d but decreases ε, which ~~is in agreement~~ agrees with the theoretical analysis (Liu et al., 2006). After constraining the differences ~~of~~ in cloud dynamics between flights, positive ε- N_d correlations ~~between~~ ~~ε~~ ~~and~~ ~~N_d~~ become stronger, indicating that perturbations of w could weaken the influence of aerosols on ε, and hence may result in an underestimation of aerosol dispersion effect. Thus, ~~it~~ this finding highlights the necessity~~requires more attention~~ ~~to~~ of ~~isolate~~ isolating the~~ response of~~ relative dispersion response to aerosol perturbations from dynamic~~al~~ effects when investigating aerosol dispersion effect and estimating aerosol indirect forcing.

~~The~~ Overall, the entrainment in stratocumulus is ~~overall~~ quite weak~~,~~ and close to being adiabatic in some cases. In this study, the difference in~~of~~ cloud microphysics between the entrainment and non-entrainment zones indicated that the entrainment in stratocumulus is mostly dominated by extreme inhomogeneous entrainment-mixing mechanism. On average, the entrainment reduced N_d and LWC by 29~~8.9~~ % and 25~~4.8~~ %, respectively, while had little effect on ~~D_e~~ R_e (only increases by 1.8 %). During the flights on Oct. 18, Nov. 04, Nov. 09 and Nov. 13, the entrainment was~~is~~ relatively strong and ~~dilutes~~ diluted N_d and LWC by about 50 %. In the entrainment zone, the~~ smaller aerosols and~~ drier air entrained from the top resulted in ~~the~~ a smaller $N_d/(N_d + N_i)$ (0.56 ± 0.22) than that in the non-entrainment zone (0.73 ± 0.13). This implies that entrainment may significantly influence cloud droplet formation and therefore influence the~~hence~~ cloud properties near the top by ~~both inhibiting aerosol activation and~~ promoting cloud droplet~~s~~ evaporation. Furthermore, we ~~also~~ found that the relative dispersion in the entrainment zone is larger than that in the non-entrainment zone. In addition to the dry and warm air, aerosols with smaller sizes are also entrained into the entrainment zone, but these aerosols seem unlikely to influence cloud droplet formation due to the negligible droplet nucleation near the stratocumulus top. That is, the effect of entrainment mixing on stratocumulus is mainly determined by the entrained dry air instead of the aerosols with properties that are different from those near the cloud base. These results seem at odds with some studies on cumulus clouds. Slawinska et al. (2012) found that in a shallow cumulus, a significant part (40 %) of aerosols is activated above cloud base (secondary activation), which is dominated by entrained aerosols. Using large-eddy simulations (LES), Hoffmann et al. (2015) suggested that sub-cloud aerosols and laterally entrained aerosols contribute to all activated aerosols inside the cloud by fractions of 70 % and 30 %, respectively. Evidently, the topics of how and to what extent entrained aerosols with properties that are different from sub-cloud aerosols can affect the formation and evolution of clouds merits further exploration. ~~As stated above, although entrainment in stratocumulus is much weaker than that in other cloud types, e.g., cumulus (Warner, 1955, 1969a, 1969b; Blyth et al., 1988; Gerber et al., 2008; Burnet and Brenguier, 2007; Haman et al., 2007), entrainment in stratocumulus still impact cloud droplet formation near cloud top significantly by entraining ambient dry air as well aerosols with physical and chemical properties different from that in cloud. Therefore, entrainment is important to take into account in studying aerosol-cloud interaction, even in stratocumulus with relatively weak entrainment. However, a quantitative contribution of entrained dry air and aerosols to cloud droplet formation, is difficult to determine only using pure aircraft measurements.~~

Data availability. The aircraft measurements data during VOCALS-REx was obtained from the public ftp at http://data.eol.ucar.edu/master_list/?project=VOCALS.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1. Correlations between ε and N_d (N_a) from observation studies.

Observations	Observation type	Location	Data for correlation analysis	Correlation
Liu and Daum, 2002	Aircraft	Ocean & coast	Flight-averaged	Positive
Peng and Lohmann, 2003	Aircraft	Coast	Flight-averaged	Positive
Pawlowska et al., 2006	Aircraft	Ocean	In-flight Flight-averaged	Negative Positive
Zhao et al., 2006	Aircraft	Land, ocean, and coast	In-flight	ε converges to a small range of values with increasing N_d
Lu et al., 2007	Aircraft	Ocean	In-flight Flight-averaged	Negative None for N_d; Positive for N_a
Lu et al., 2012	Aircraft	Land	In-flight Flight-averaged	Negative Negative
Hudson et al., 2012	Aircraft	Ocean	Flight-averaged	Negative
Ma et al., 2012	Aircraft	Land	Flight-averaged	Negative
Pandithurai et al., 2012	Aircraft	Land	Flight-averaged	Positive
Kumar et al., 2016	ground-based	Land	—	Positive

Table 2. Flight information and parameters that represent the properties of entrainment during all 16 non-drizzling flights.

Flight number	RF01	RF02	RF03	RF04	RF05	RF06	RF07	RF08	RF09
Date	10.16	10.18	10.19	10.21	10.22	10.24	10.26	10.27	10.29
BL type	**Typical**	**Typical**	**Typical**	**Typical**	**Typical**	**Other** Wind shear	**Typical**	**Typical**	**Other** Decoupled
w ave[a]	0.09	0.08	0.11	0.08	0.08	-0.06	0.06	0.08	-0.13
w std[b]	0.42	0.55	0.58	0.51	0.51	0.30	0.56	0.41	0.61
w skew[c]	-0.38	-0.16	-0.27	-0.21	-0.27	0.00	-0.23	0.08	-0.27
P_{LWC}[d~~a~~]	~~25.8~~26	~~55.7~~56	33~~.4~~	~~24.8~~25	~~24.6~~25	29~~.3~~	-~~2.7~~3	11~~.2~~	3~~.1~~
P_{Nd}[d~~b~~]	32~~.1~~	60~~.1~~	30~~.1~~	~~38.6~~39	28~~.2~~	34~~.4~~	~~4.9~~5	~~19.6~~20	6~~.4~~
$P_{R\cancel{D}e}$[f~~e~~]	-2~~1.9~~	-~~5.7~~6	~~0.9~~1	-~~6.7~~7	-~~1.9~~2	0~~-0.1~~	-4~~.1~~	-2~~.4~~	-~~1.8~~2
AF_{ent}[g~~d~~]	0.77	0.52	0.58	0.85	0.49	0.52	0.51	0.76	0.81
$AF_{non\text{-}ent}$[h~~e~~]	0.95	0.84	0.82	0.77	0.74	0.78	0.73	0.82	0.80

Flight number	RF10	RF11	RF12	RF13	RF14	RF15	RF16	Total
Date	10.30	11.04	11.08	11.09	11.10	11.12	11.13	
BL type	**Other** Wind shear	**Other** Wind shear, Decoupled	**Other** Decoupled	**Typical**	**Typical**	**Typical**	**Other** Wind Shear	
w ave	0.02	0.07	0.02	0.08	0.09	0.09	-0.02	
w std	0.45	0.41	0.42	0.47	0.49	0.51	0.41	
w skew	-0.13	-0.48	-0.03	-0.48	-0.26	-0.27	-0.42	
P_{LWC}	10~~.5~~	62~~.1~~	3~~2.5~~	~~55.8~~56	~~2.9~~3	-~~1.8~~2	~~58.7~~59	25~~4.8~~
P_{Nd}	~~7.6~~8	56~~.3~~	24~~.0~~	56~~.1~~	-2~~1.6~~	8~~7.5~~	59~~.2~~	2~~8.~~9
$P_{R\cancel{D}e}$	0~~.2~~	4~~.4~~	-8~~.4~~	-2~~.1~~	3~~.4~~	-3~~2.5~~	-1~~.2~~	-~~1.8~~2
AF_{ent}	0.73	0.66	0.84	0.28	0.70	0.67	0.56	0.64
$AF_{non\text{-}ent}$	0.82	0.97	0.77	0.50	0.79	0.60	0.64	0.77

[a, b, c] *w* ave, *w* std, and *w* skew are the average, standard deviation, and skewness of in-cloud vertical velocities, respectively..

[d~~a~~, e~~b~~, f~~e~~] P_{LWC}, P_{Nd}, and $P_{R\cancel{D}e}$ are the percentages of reduction in *LWC*, N_d and $R\cancel{D}_e$ within entrainment zone relative to non-entrainment zone.(unit: %)

[g~~d~~, h~~e~~] AF_{ent} and $AF_{non\text{-}en}$ are adiabatic fraction in entrainment zone and non-entrainment zone, respectively. Here, adiabatic fraction is defined as the ratio of the measured to its adiabatic *LWC* that is calculated using pressure and temperature near cloud base.

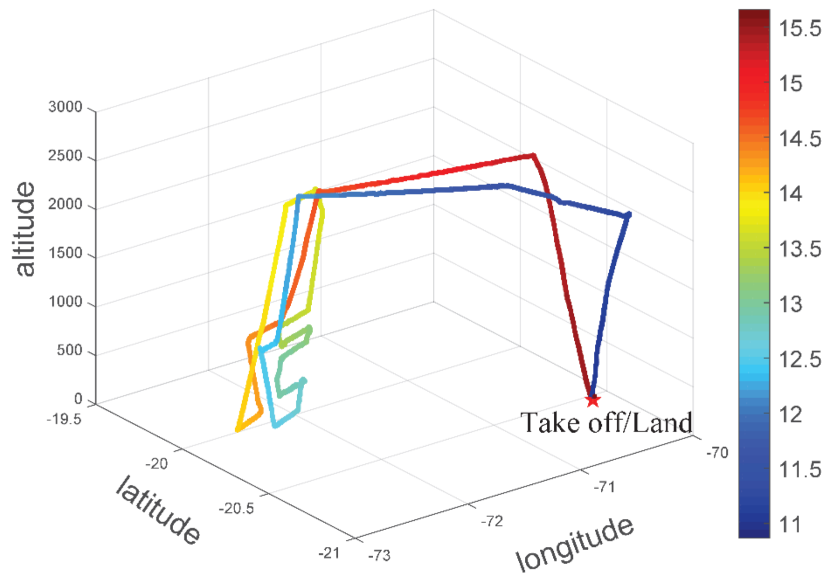


Fig. 1. The flight track in Oct. 18, and the colors represent flight time in hour (UTC).

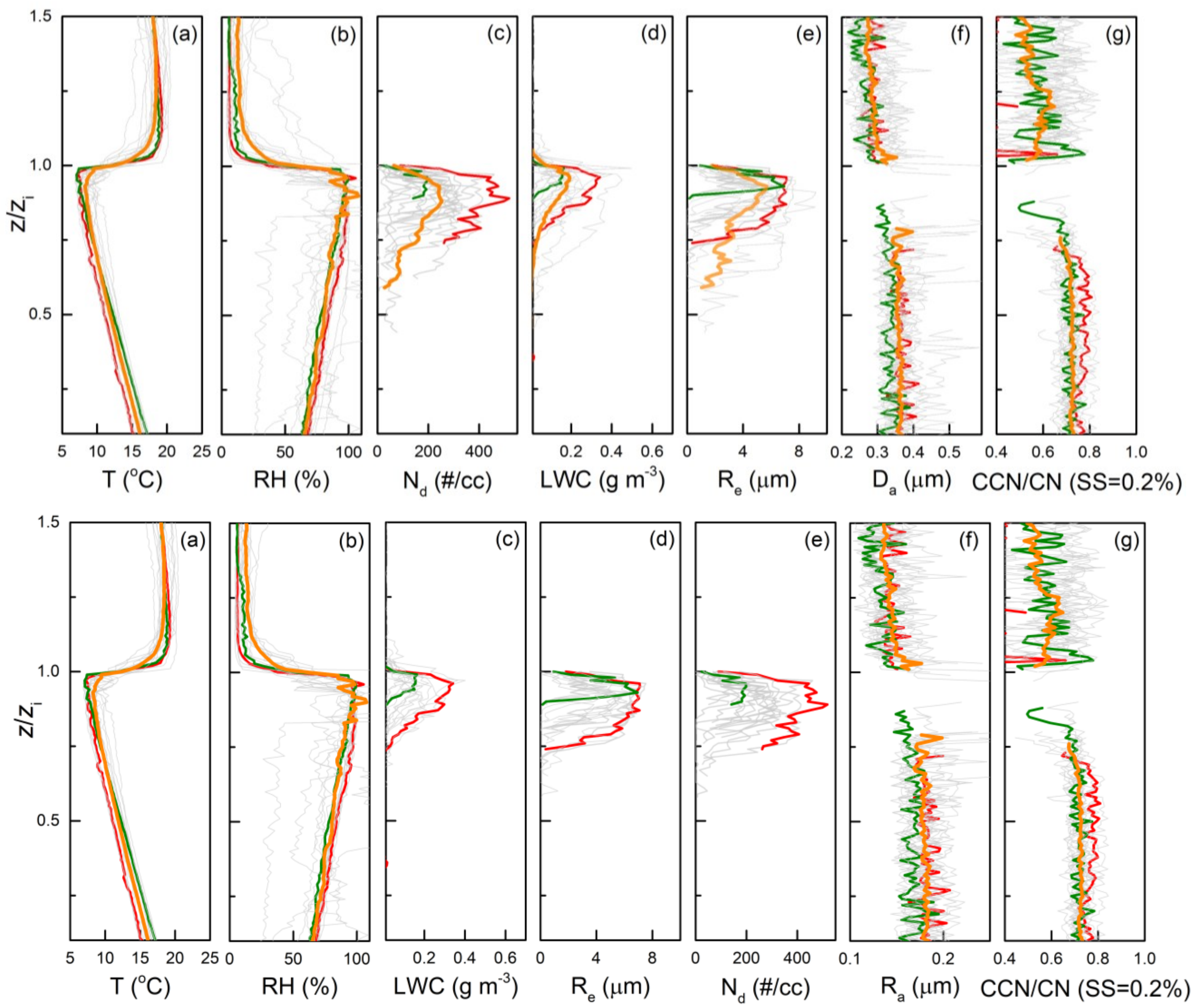
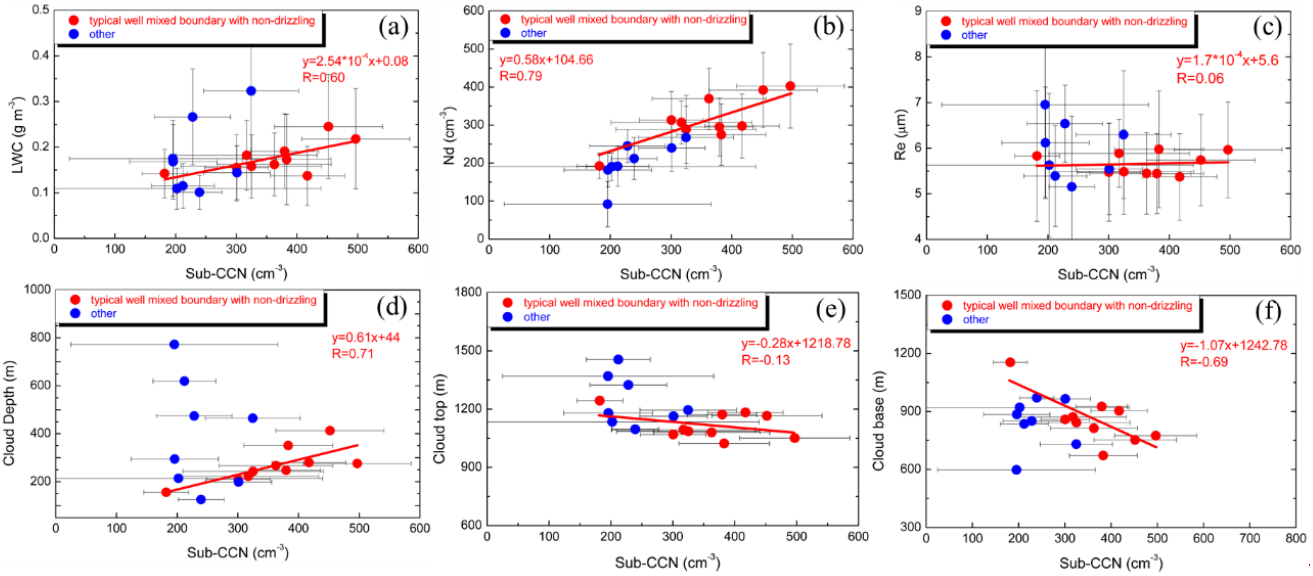


Fig. 2. Vertical profiles scaled by the inversion height. (a) temperature (K); (b) relative humidity (%); (c) liquid water content (g m $^{-3}$)~~cloud droplet number concentration (cm $^{-3}$)~~; (d) cloud droplet effective radius (μm)~~liquid water content (g m $^{-3}$)~~; (e) cloud droplet

number concentration (cm $^{-3}$)~~effective radius of cloud droplets (μm)~~; (f) ~~effective diameter of~~ aerosols effective radius (μm), and (g) the number concentration ratio of CCN to aerosols for all 16 non-drizzling flights. The gray lines show all individual flights, and the orange lines indicate the average profiles. The red and green lines represent the polluted (Oct. 18) and clean (Nov. 9) cases, respectively.



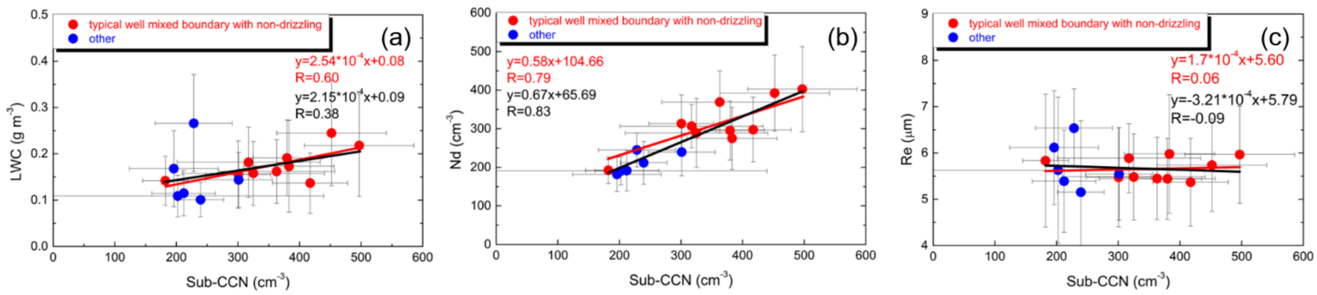


Fig. 3. (a) *LWC* (g cm $^{-3}$); (b) N_d (cm $^{-3}$); (c) R_e (μm)~~; (d) cloud depth (m); (e) cloud top height (m); (f) cloud base height (m)~~ as a function of sub-cloud CCN concentrations (SS=0.2%) for all 16 non-drizzling flights. The error bars through these symbols indicate the standard deviation. Red symbols are the cases with typical well-~~-~~mixed ~~boundary~~BL ~~-with non-drizzling~~ discussed in Zheng et al. (2011), and blue symbols for others. Red (black) texts are the correlation coefficient for typical well-mixed cases (all cases).

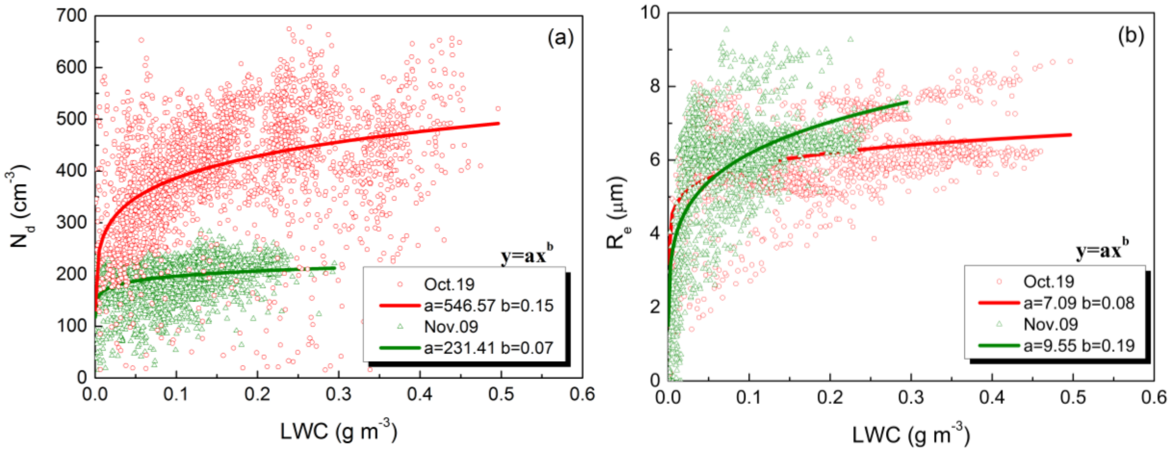


Fig. 4. Correlations between (a) N_d (cm^{-3}), (b) R_e (µm) and LWC (g m^{-3}) for clean (green) and polluted (red) cases, respectively.

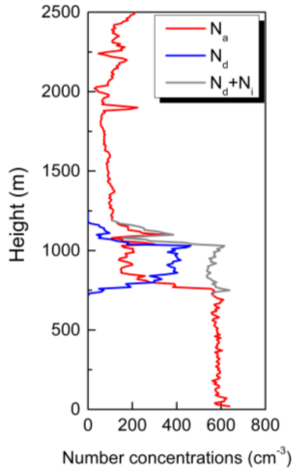
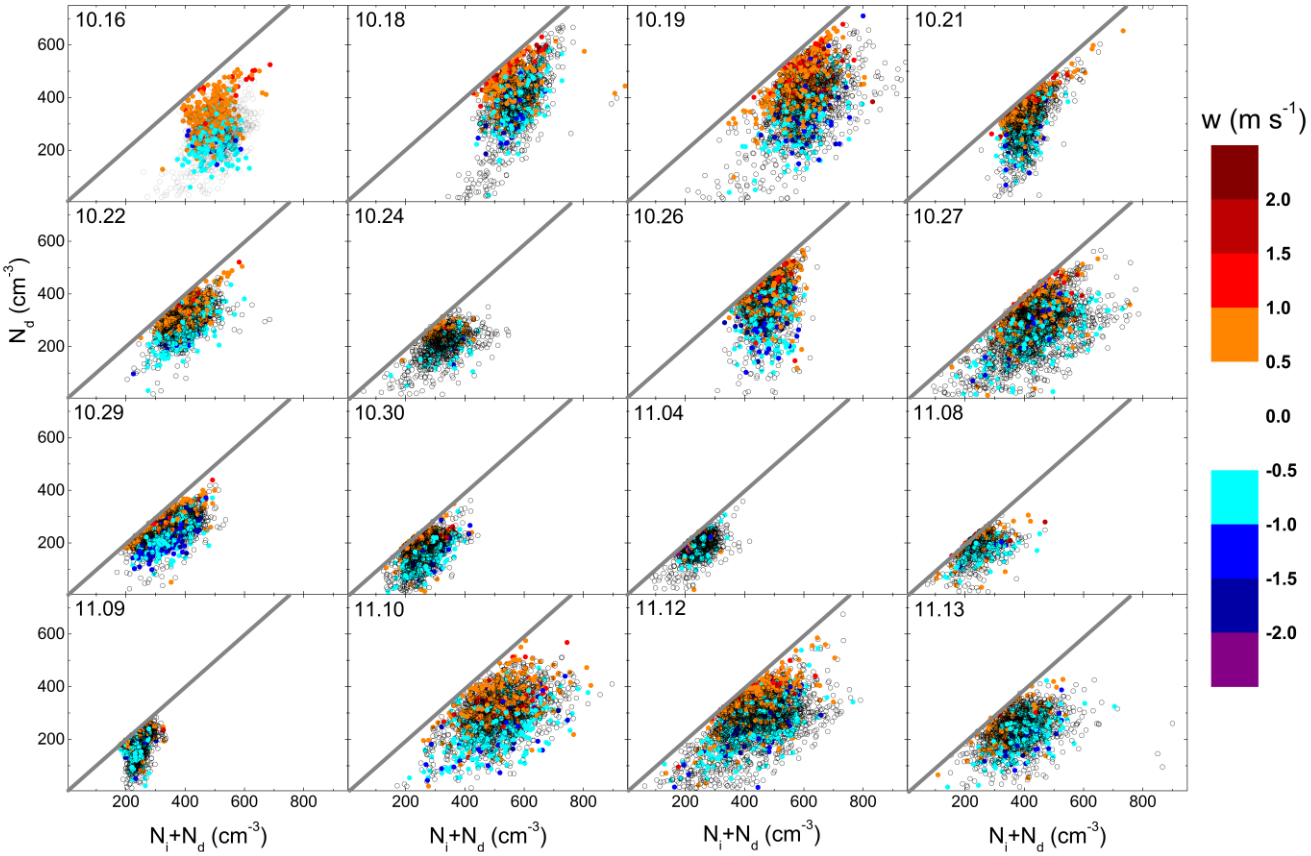


Fig. 5. Vertical profiles of number concentrations of aerosols (N_a), cloud droplets (N_d) and total in-cloud particles (N_d + N_i) during the flight on Oct. 18.



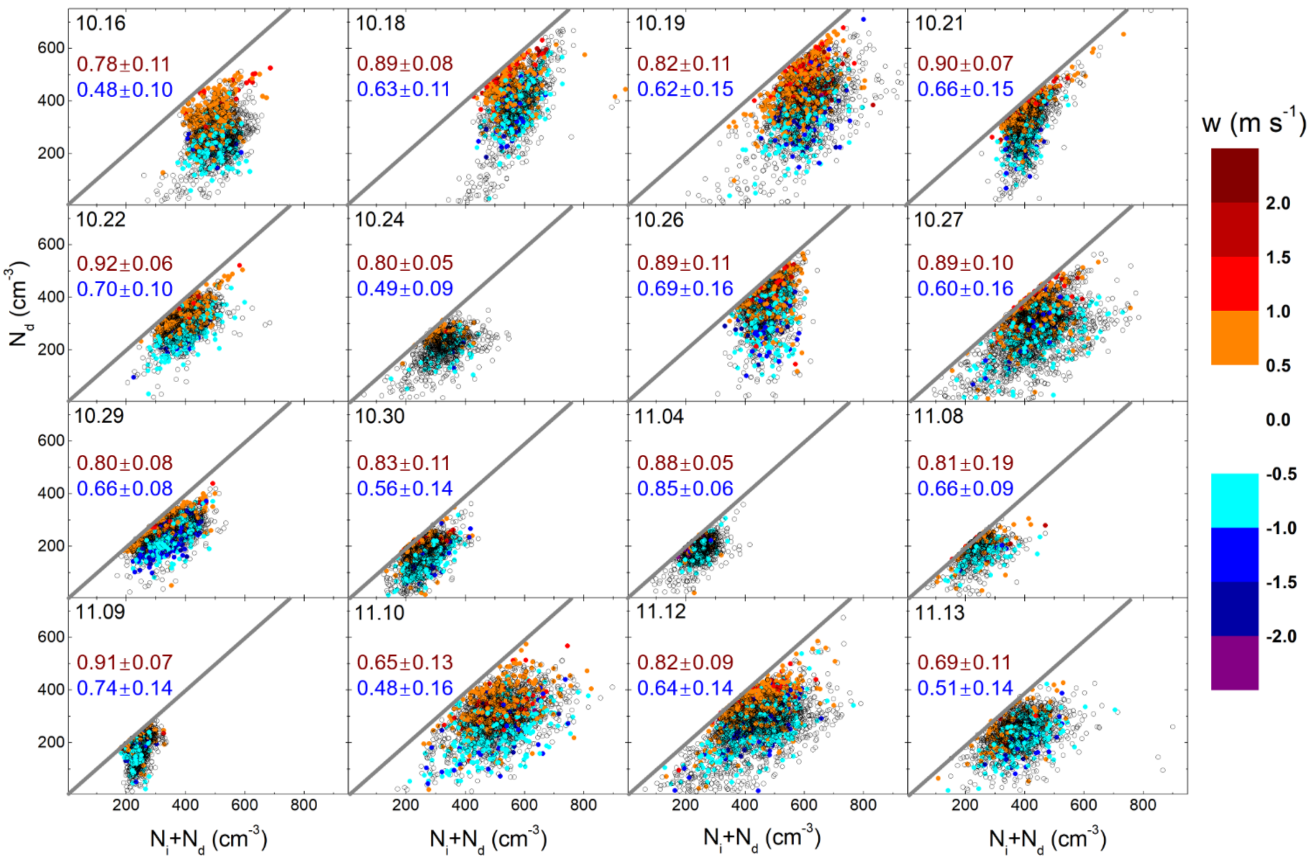


Fig. 6. Relationships between N_d and $N_i + N_d$ during all 16 non-drizzling flights. The color~~s~~ represents in-cloud vertical velocities (m s^{-1}), and gray line is 1:1 line. The mean and standard deviation of $N_d/(N_d+N_i)$ for vertical velocity greater than 1 m s^{-1} (red) and less

than -1 m s^{-1} (blue) are shown.

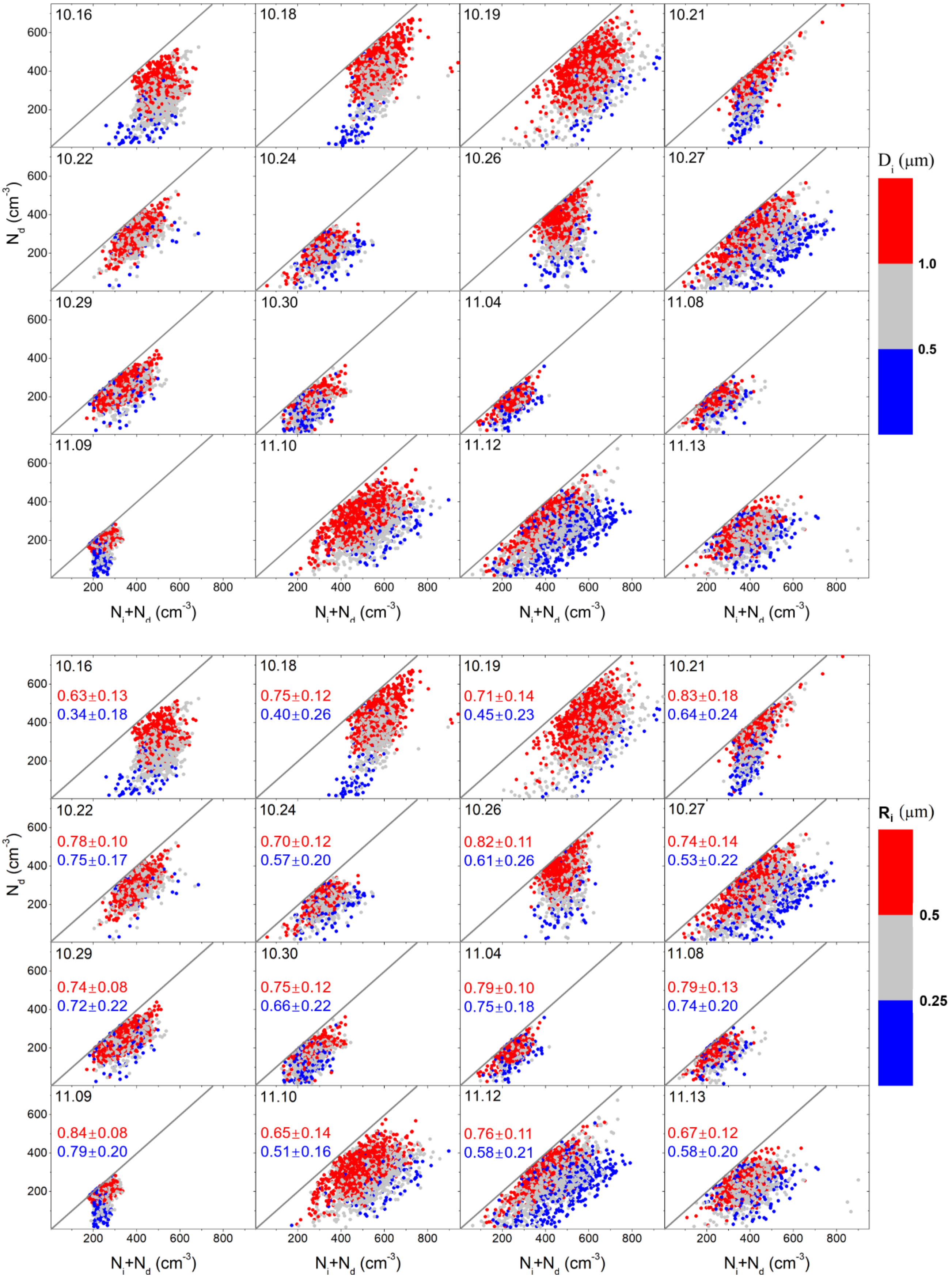


Fig. 7. Same as Fig. 6, but the color~~s~~ represents the effective ~~diameter~~ radius of interstitial aerosol (~~D_i~~R_i) (μm). The mean and standard deviation of $N_d/(N_d+N_i)$ for R_i greater than 0.5 μm (red) and less than 0.25 μm (blue) are shown.

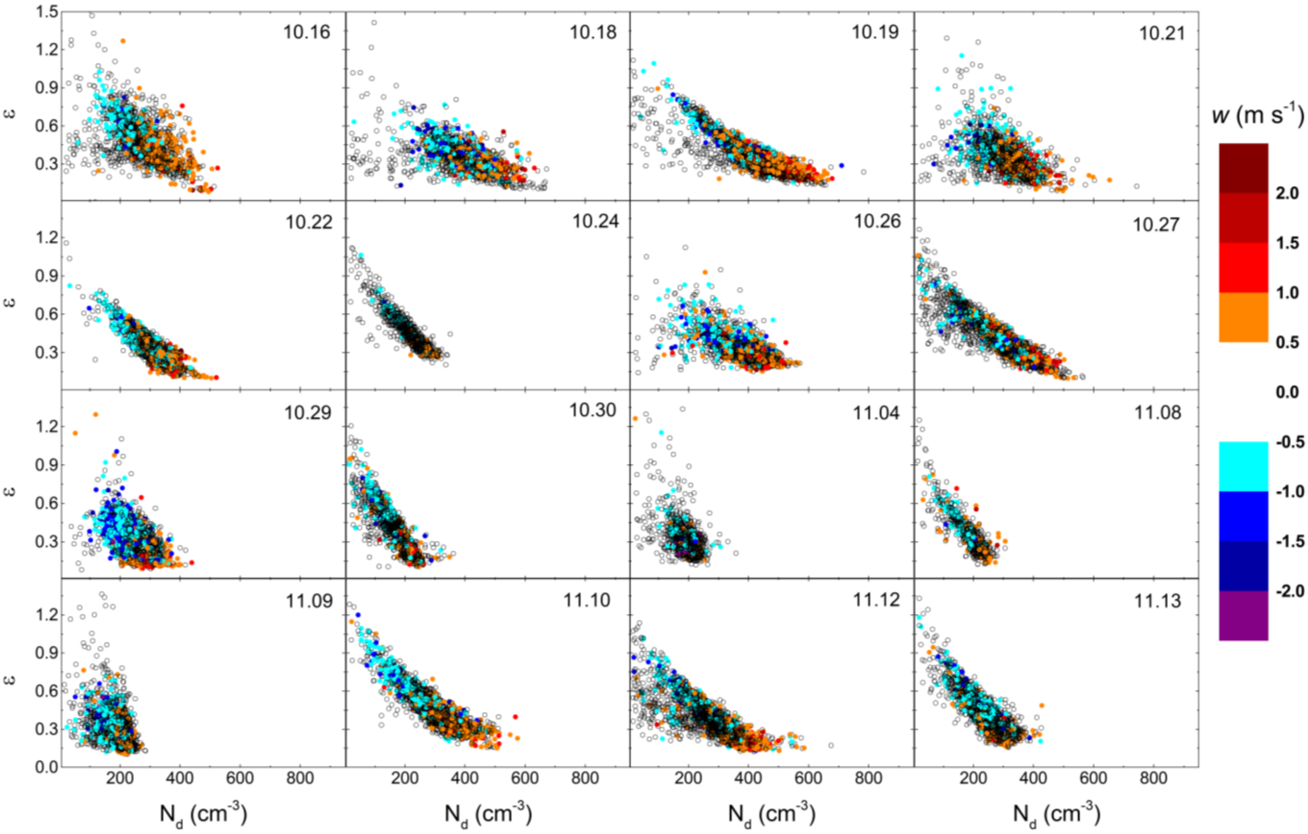
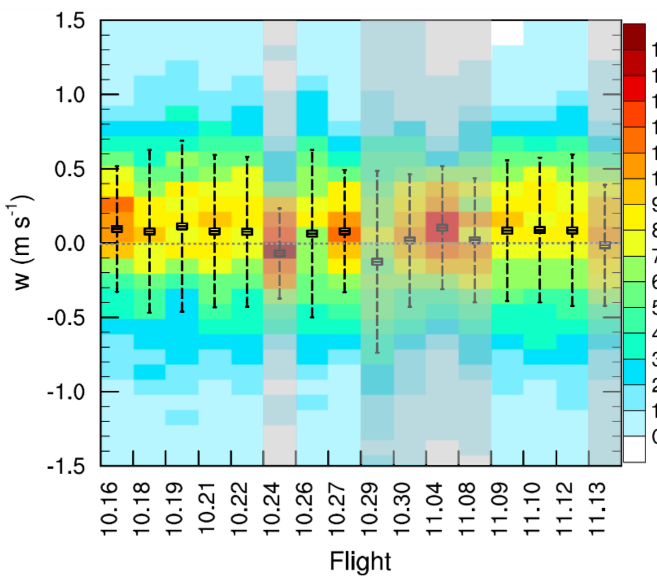


Fig. 8. Relationships between relative dispersion (ε) and N_d during all 16 non-drizzling flights, in which the colors ~~representing~~ represents in-cloud vertical velocities (m s^{-1}).



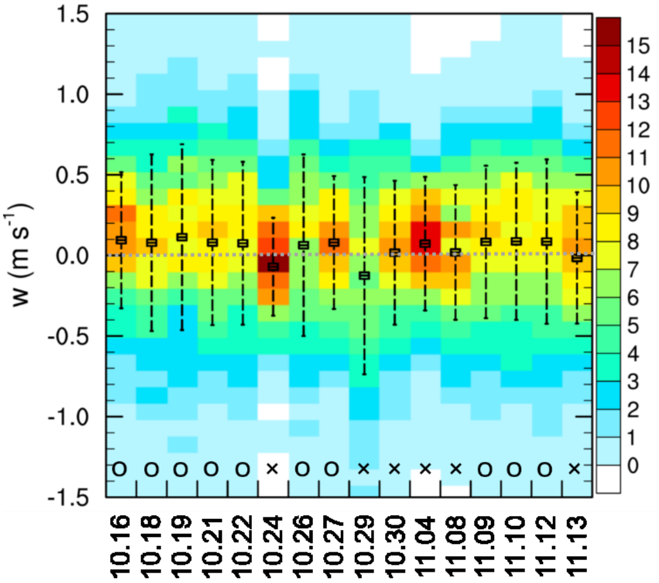


Fig. 9. Probability distribution function (units: %) of vertical velocity (*w*) for 16 non-drizzling flights. Black symbols are mean values of *w*, and error bars through these symbols indicate the standard deviation. ~~Gray shadow represents the flights other than typical well mixed boundary with non-drizzling.~~Circles are the cases with typical well-mixed BL, and crosses represents the other cases.

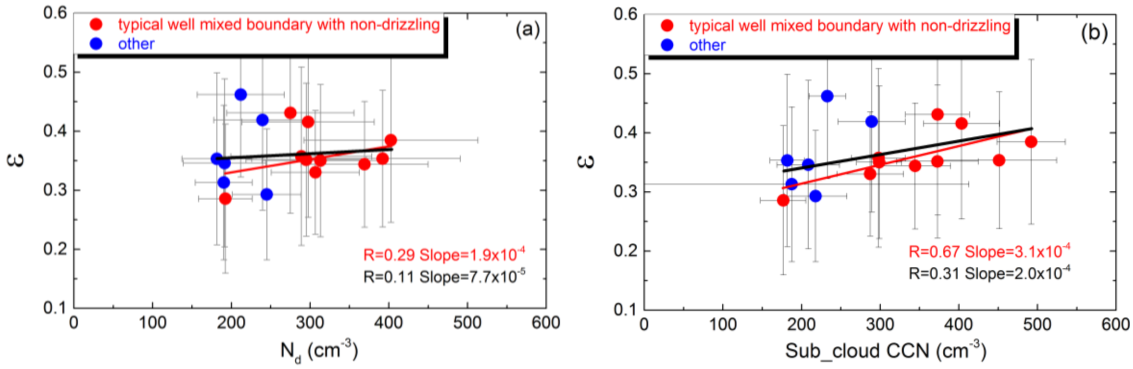


Fig. 10. Relative dispersion (*ε*) as a function of (a) N_d and (b) sub-cloud CCN concentrations (*SS*=0.2%) for all flights. The error bars through these symbols indicate the standard deviation. Red symbols are the cases with typical ~~well~~ well-mixed ~~boundary~~BL ~~with non-drizzling~~, and blue symbols for others. Red (black) texts are the correlation coefficient and slope for typical well~~-~~mixed cases (all cases).

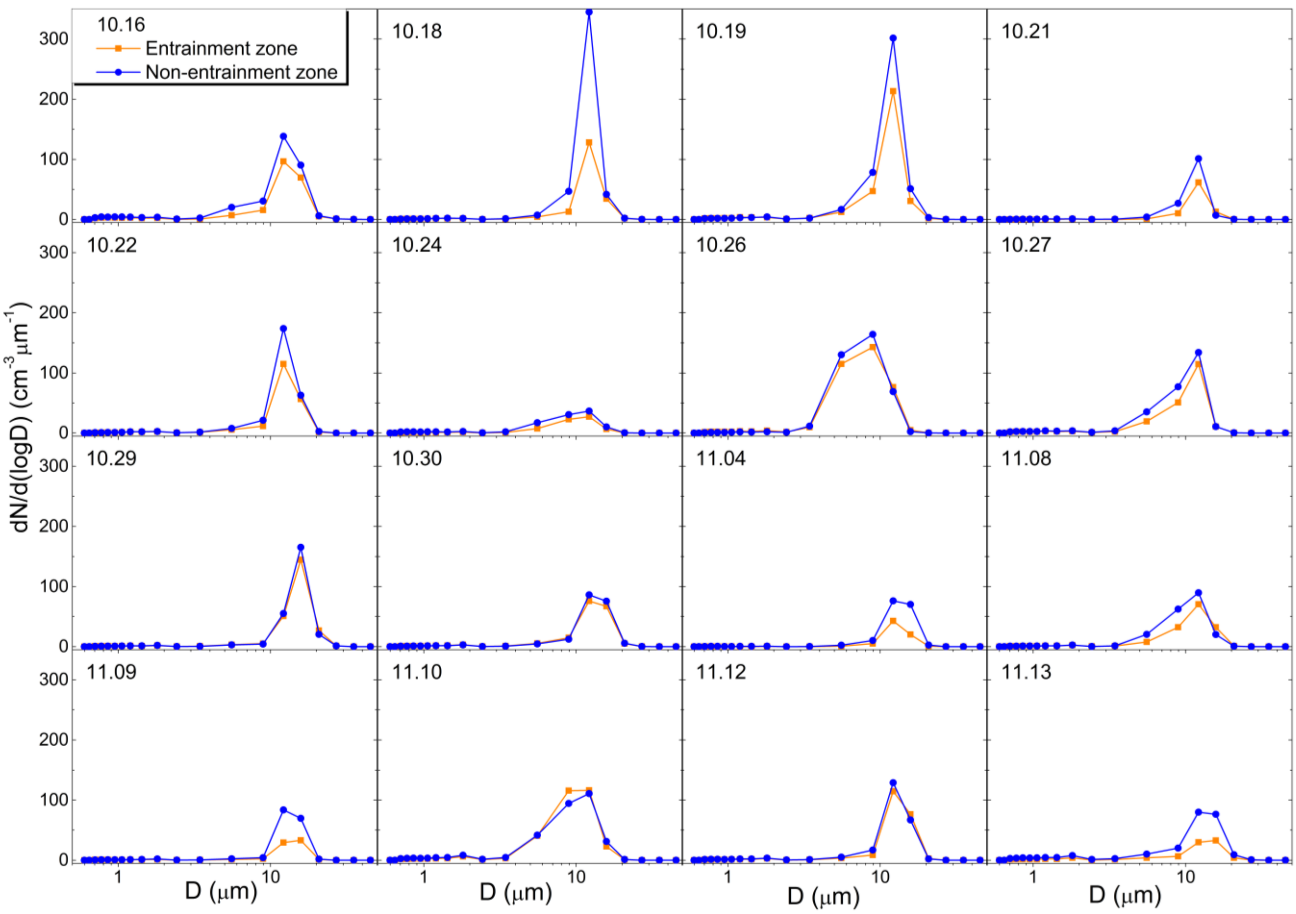
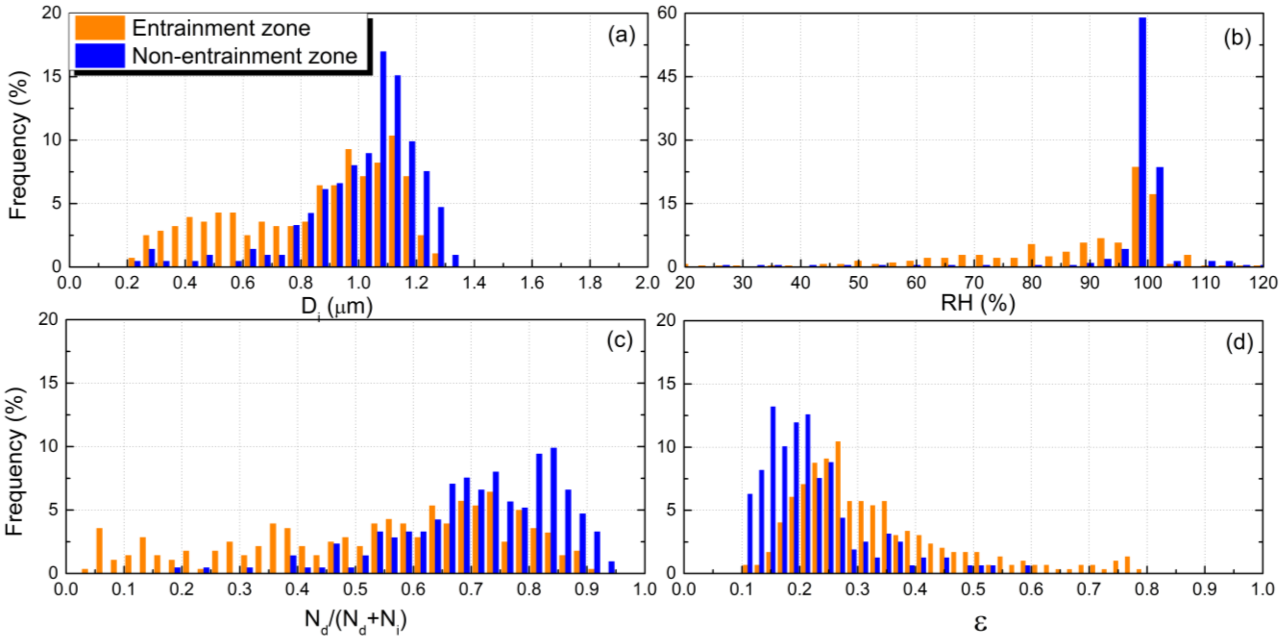


Fig. 11. Number size distributions of cloud droplets in the entrainment (yellow) and non-entrainment zones (blue) during all 16 non-drizzling flights.



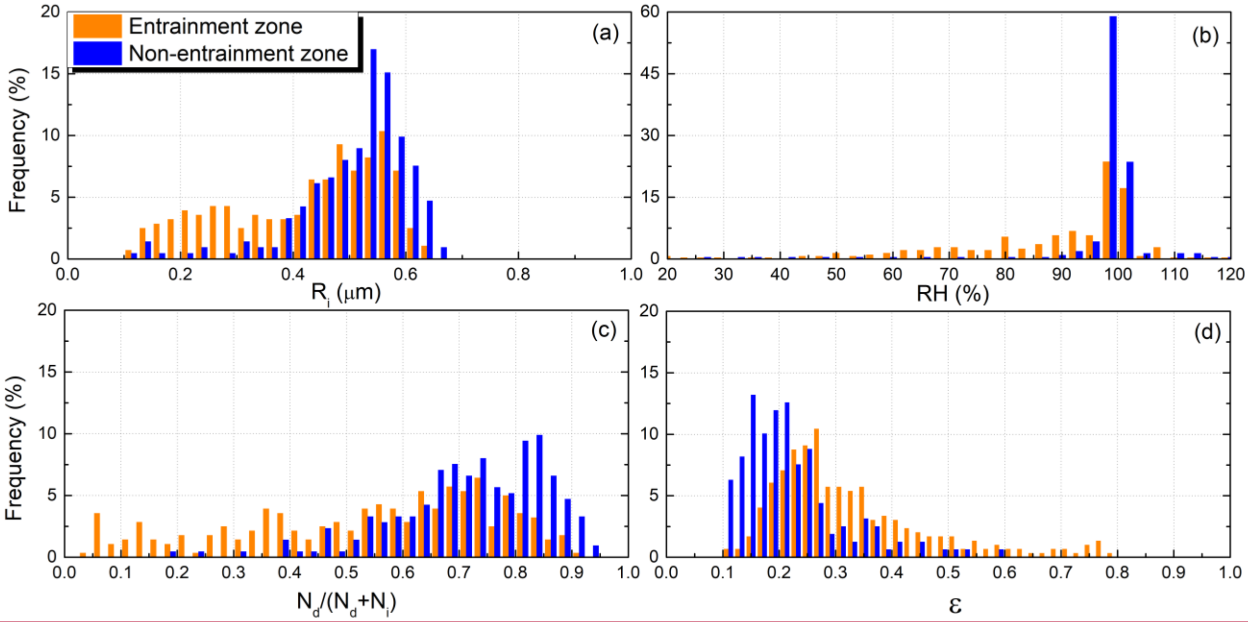


Fig. 12. Probability density functions of (a) ~~D_i~~ R_i (μm), (b) RH (%), (c) $N_d/(N_d + N_i)$, and (d) ε in the entrainment (yellow) and non-entrainment zones (blue) during the flight on Oct. 18.

Supplement of

Exploring aerosol cloud interaction using VOCALS-REx aircraft measurements

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Figure List

Figure S1. Normalized profiles of N_d. Values of Z_N=0 indicates the cloud base whereas Z_N=1 the cloud top. Orange line indicates the average profiles.

Figure S2. (a) P_{LWC} and (b) P_{Nd} as a function of $AF_{ent}/AF_{non\text{-}ent}$ for all 16 non-drizzling flights.

Figure S1

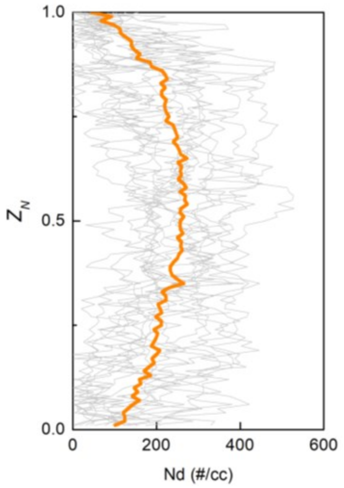

Figure S2