Response to Referee #1

Comments are in black and responses are in blue.

1 Overview Comments

This paper uses data from a field campaign in the south eastern Pacific to investigate the aerosol dispersion effect and entrainment in stratocumulus clouds. The cases have been described in other work previously and so the new aspect here is to analyse those data in a new way to look at different properties.

The paper is well structured, and the limited information in the data and methods section is mitigated by previous published work. Some reference to entrainment in stratocumulus clouds specifically should be added.

The changes made from the original document have improved the manuscript, and it is much closer to publication. Where I still have comments or questions they are within the body of this report. The manuscript would still benefit from being more specific in places for clarity - some occasions identified in technical corrections.

We thank the reviewer for taking the time to assess the manuscript and for providing helpful comments and suggestions to improve the manuscript. We have revised the manuscript carefully according to the reviewer's comments. At the same time, we are grateful for the important references provided by the reviewer. These and other references related to entrainment in stratocumulus have been cited in the revised manuscript. Please see the following detailed point-by-point responses.

2 Specific Comments

2.1 Section 2

I would like to see more information on interstitial aerosol observations. The size looks very large.

In this study, the size distribution of interstitial aerosol is obtained directly from the observation of in-cloud aerosols by Passive Cavity Aerosol Spectrometer Probe (PCASP-100), which counted and sized particles from 0.1–2.0 μm dry diameter with 20 bins. The description has been added in section 2.1 accordingly (line 75 in the revised manuscript). For an explanation of the large size, please see the detailed responses to section 2.4 (Results Section 3.3).

2.2 Results Section 3.1

It is interesting and somewhat unusual that the number concentrations increase with height above cloud base, rather than remaining relatively constant. I suggest noting this comparing to some of the VOCALS cloud observations perhaps.

Thanks for reminder. We agree that, in most cases, N_d profiles should be close to relatively constant, but this is not always the case (Keil et al., 2003). We realize that the normalization by cloud-top height only may be insufficient to indicate the vertical variation of clouds when all profiles are averaged, because each profile has a different cloud base. Thus, the average profiles are removed from the Fig. 2c, 2d, 2e, and the vertical variation of cloud properties can be seen easily from the single profile. As depicted in Fig. 2c, the green N_d profile remains relatively constant, and the red one shows a slight increase with height. Furthermore, to get the average profile of all flights reasonably, we normalize the height $Z_N=(Z-Z_{base})/\Delta Z$, where Z_{base} and ΔZ are the cloud base height and the geometrical cloud depth, respectively (Fig. R1). This transformation implies that $Z_N=1$ at the cloud top, and $Z_N=0$ at the cloud base. As shown in Fig. R1, the average profile of Nd remains relatively constant with a slight increase and decrease near base and top respectively, which is consistent with results in other VOCALS-REx observations (Painemal and Zuidema, 2011). We have modified the main text related to N_d profile accordingly (lines 121-129 in the revised manuscript).

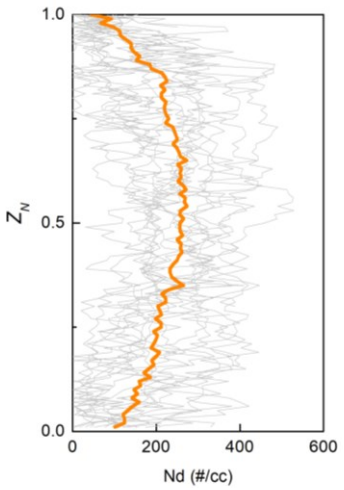

Fig. R1. Normalized profiles of N_d. Values of $Z_N=0$ indicates the cloud base whereas $Z_N=1$ the cloud top. Orange line indicates the average profiles.

2.3 Results section 3.2

Section 3.2, paragraph one. In the south eastern Pacific most of the aerosol optical depth will be within the marine boundary layer and so the assumption from the satellite studies is probably good here, as the aerosol and cloud layer are not well separated. Is there anything specific about the satellite studies that results in a large bias in this region? Otherwise it is not that relevant.

As reported in previous studies (Allen et al., 2011; Shank et al., 2012), biomass burning serves as a potential source of aerosol to the free troposphere above cloud over the South East Pacific (SEP) region. Under the influence of biomass burning plume which carry elevated organic combustion aerosol, the aerosol concentration above cloud becomes comparable to that below cloud (Allen et al., 2011). By using satellite data, Costantino et al.(2010) pointed out that aerosols from biomass burning are often separated from the underlying stratocumulus cloud layers, and thus have little effect on cloud properties. Therefore, in this case, AOD as a proxy of CCN number concentration to investigate the aerosol-cloud interactions could induce biases. It is necessary to investigate the impact of CCN number concentration near cloud layer on cloud properties.

Line 145 onwards: What altitude is the level of decoupling in these clouds? Is it below the level where sub-CCN measurements are made? In the case of Nd and LWC, and cloud base even the "other" cases look well correlated apart from 2 - possibly the ones with precipitation? The decoupling will only have an impact if it is above the level where you make the sub-CCN measurements. Do you have measurements of the decoupling altitude?

The decoupling is characterized by a vertically non-uniform distribution of total water mixing ratio from the surface to the capping inversion, or a cumulus cloud underlying stratocumulus (Zheng et al., 2018). Based on this, we derived the decoupling altitude (Table R1). The two outliers are 1st Nov (drizzling case) and 29th Oct (decoupling case). As shown in Table R1, the decoupling height for 29th Oct is indeed above the level where the sub-CCN measurements were made.

Furthermore, we have removed the drizzling cases from Fig. 3 in revised manuscript, and reanalyzed the relationships between sub-CCN and cloud properties for all flight and well mixing flights, respectively. It is found that the correlation coefficients between sub-CCN and LWC (Fig. 3a) and cloud base height (Fig. 3f) for all non-drizzling flights are 0.38 and -0.52, respectively, which are significantly lower than those for well mixing cases (0.60 and -0.69), confirming that the aerosol effect could be confounded by various dynamics. However, the change in correlation coefficient between sub-CCN and Nd is very small (0.83 vs. 0.79). One possible explanation is that, the impact of aerosol on Nd is relatively linear and direct, while LWP is a function of both Re and Nd, which depends not only on the number of condensation nuclei, but also on the subsequent growth process of cloud droplets, and thus is more sensitive to dynamics. Similarly, the relative dispersion is also strongly dependent on dynamics (Fig. 8). Therefore, even if Nd does not show a clear difference between the well mixing and other cases, it is still necessary to distinguish meteorological categories. In this study, all ‘other’ cases that could confuse the aerosol effect are eliminated, such as decoupling and wind shear, which affect the feeding of water vapor and energy from the surface.

Table R1. The heights of decoupling and cloud base for three decoupling cases.

Date	**10.29**	11.04	11.08
Decoupling Height (m)	**810.3**	631.7	844.2
Cloud Base Height(m)	**850.4**	920.5	1238.3

2.4 Results Section 3.3

October 18th Case study: do all results here apply to this case? Is it possible to get aerosol particle size distribution for the sub-CCN layer, and the interstitial aerosol? It is a surprise that the unactivated aerosols are larger than 1 micron in size (for example in Figure 7. Is this because they are in a saturated environment? For example, during the VOCALS measurements (for example Twohy ACP2013, Impacts of aerosol particles on the microphysical and radiative properties of stratocumulus clouds over the southeast Pacific Ocean) observed much smaller interstitial aerosols of 150 nm, and below cloud 135 nm.

In section 3.3, only Fig. 5 applies to 18th Oct case, and the rest of the results are for the average of all cases. This has been specified in the revised manuscript. For better understanding, the average Nd/(Nd+Ni) applied to different conditions for each individual flight have been also added to Fig. 6 and Fig. 7 in the revised manuscript.

The size distributions for the sub-cloud aerosol and the in-cloud (interstitial) aerosol are shown in Fig. R2. By directly comparing, it seems that the size of in-cloud aerosol in this study is larger than that in Twohy et al.(2013). However, it should be noted that the size shown in Fig. 7 is the individual sampling at specific locations in the cloud (instantaneous sampling), while the size of 150 nm in Twohy et al.(2013) is an average of a flight, where some large values might be smoothed. Another possible explanation is that, we use the effective diameter (Zhang et. al, 2011) to represent the aerosol size distribution rather than geometric mean diameter utilized in Twohy et al.(2013). For comparison purposes, the averaged geometric mean diameters of sub-cloud (blue, 184 nm) and in-cloud (red, 181 nm) aerosols during 18th Oct is also calculated (Fig. R2), which is much closer to the size in Twohy et al.(2013), but with a slight overestimation (~ 40 nm). This might be attributed to the difference in the measurement range of the instruments, i.e., 0.055–1.0 μm for Ultra High Sensitivity Aerosol Spectrometer (UHSAS) in Twohy et al.(2013), but 0.1–2.0 μm for Passive Cavity Aerosol Spectrometer Probe (PCASP-100) in our study. The latter is unable to observe the Aitken mode that is less than 0.1 μm, thus its geometric mean diameters is larger. In summary, comparing the aerosol in this study with that in Twohy et al.(2013) under the same conditions, the two are very close.

We agree that the aerosol size might be overestimated in a saturated environment (Fig. R2). Thus, in order to eliminate the influence of strong supersaturation on aerosol size, we exclude the samples with RH larger than 97%, and reanalyze the dependence of Nd/(Nd+Ni) on Di (Fig. R3). It is found that, without strong supersaturation, Nd/(Nd+Ni) still tend to increases with Di, so it seems saturated environment might not influence our conclusion significantly.

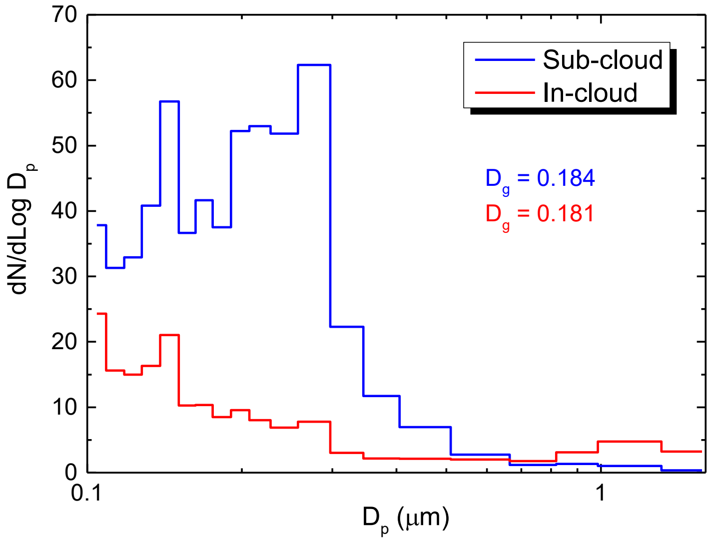

Fig. R2. The size distributions for the sub-cloud aerosol (blue) and the in-cloud aerosol (red) during the flight on 18th Oct.

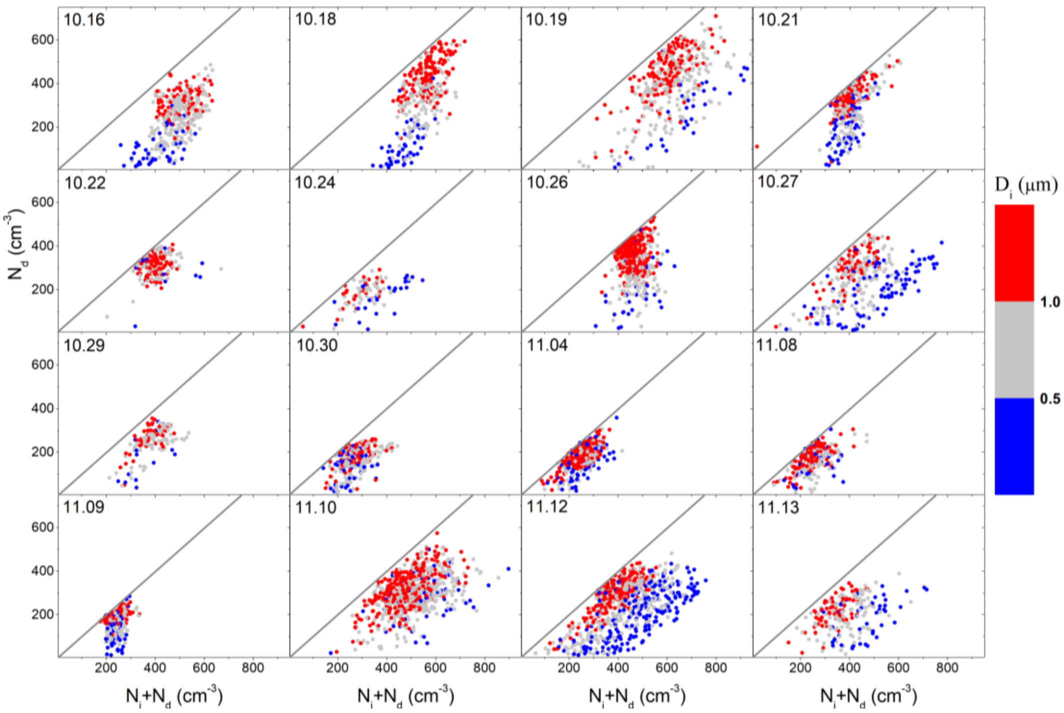

Fig. R3. Relationships between Nd and Ni + Nd during all 16 non-drizzling flights when RH is larger than 97%. The colors represent the effective diameter of interstitial aerosol (D_i) (μm), and gray line is 1:1 line.

It looks as though the vertical velocity effect is limited for low total aerosol concentrations which seems interesting. Is this worth noting? Is the effect limited by low aerosol number?

To check if this limitation exists, we compared the difference of Nd/(Nd+Ni) between large and small vertical velocity for each flight (Fig. 6). It is found that there is no significant difference between low and high aerosol concentrations cases. Thus, this might be caused by visual effects, because in the case of low aerosol concentrations, most of the data concentrate and hence overlap each other in Figures.

Line 208. Is the average here for the whole flights worth of data for October 2018? Again - is it possible to show aerosol size distributions?

The Nd/(Nd+Ni) here is the average of all flights. The aerosol size distributions is shown in Fig. R2. For a more detailed discussion of aerosol size, please see the response to the previous section.

Why do some flights show a reduced effect, e.g. 22nd Oct, 29th, 30th, 4th Nov, 8th. Are the data able to explain?

The Nd/(Nd+Ni) for each individual flight are calculated and shown in Fig. 7. It is demonstrated that these flights do not show a reduced effect.

2.5 Results Section 3.4

I still do not think there are strong difference in the vertical velocity PDFs between the well mixed and other cases. The grey shading does not help in figure 9, it might be easier to see if the shading is removed, and those cases are identified with a symbol above the axis. The standard deviations do not look different within the other category compared to well mixed, and if the skewness is not different, then what is? If anything I might expect the skewness to be the parameter that varied, when in a decoupled boundary layer, dominated by turbulence from cooling at cloud top, rather than the ocean surface thermals.

Thanks for suggestions. We have removed the shading from Fig. 9. The well-mixing and other cases are marked as circles and crosses, respectively. The means, standard deviations, and skewnesses of vertical velocities for all flights have been added in Table 2. Indeed, there is no significant difference in standard deviations between well mixing and other cases, but the means of other cases are overall smaller than that of well mixing cases. However, 4th Nov is an exception with a mean value close to well mixing cases, but its skewness is relatively large. That is, there are some differences in the vertical velocity between the well mixed and other cases (Table 2), implying the importance of distinguishing the well mixing cases from other cases.

A see that the correlation reduces when the other cases are included, and so the dynamics are important (in Figure 9), but again - it looks like there are two strong outliers - which are these? Do they have to most skewed w PDFs or most different standard deviation of w? Or else precipitation, or wind shear?

The two strong outliers in Fig. 10 are 24th Oct and 13th Nov, which are characterized by a strong wind shear (Fig. R4). For these two cases, the average in-cloud *w* are smaller (-0.06 and -0.02) and the relative dispersions are larger (0.46 and 0.41), showing the dependence of relative dispersion on *w* (as indicated in Fig. 8), which further highlights the importance of minimizing the influences of meteorological conditions by excluding the other cases.

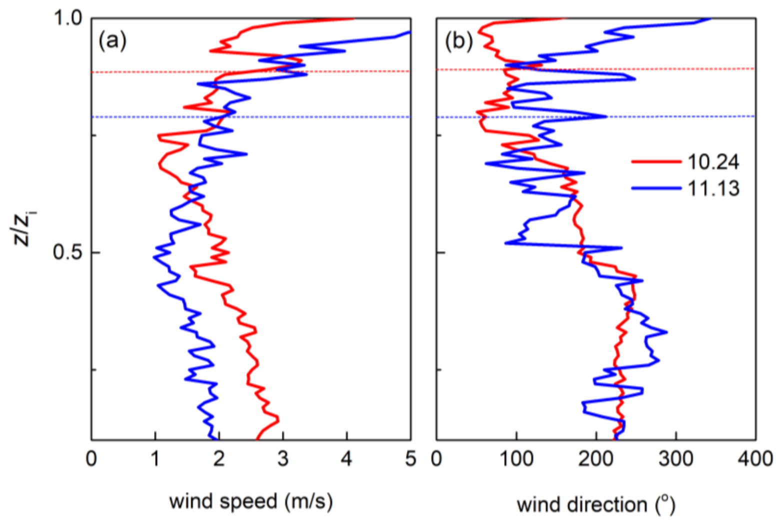

Fig. R4. Vertical profiles of (a) horizontal wind speed and (b) wind direction during flights on 24th Oct (red) and 13th Nov. Dashed lines indicate the height of the cloud base.

2.6 Results Section 3.5

This section is interesting and appears to show some evidence for inhomogeneous mixing. It is difficult to isolate this, and I wonder if there is enough precision in the observations to look at 20 m deep layers. However the size distributions in Figure 11 show some reasonably convincing evidence. Does the degree of change in the size distribution correlate with the AFdent fraction in Table 2? For a quick look it appears to - is there a way to quantify this?

As shown in Fig. R5, the vertical speed of the CIRPAS Twin Otter aircraft ranges from -5 to 5 m s^{-1}, most of which are concentrated between -1 and 1 m s^{-1}. Therefore, it is sufficient to observe the 20 m deep layers, especially during the horizontal legs near the cloud top where we distinguish the entrainment and the non-entrainment zone.

Thanks for suggestions. In order to check the relationship between the degree of change in the size distribution and adiabatic fraction, we correlated $AF_{ent}/AF_{non\text{-}ent}$ with P_{LWC} and P_{Nd}, respectively, where $AF_{ent}/AF_{non\text{-}ent}$ indicates the change of AF in the entrainment zone relative to that in the non-entrainment zone (Fig. R6). It is shown that both P_{LWC} and P_{Nd} are negatively correlated with $AF_{ent}/AF_{non\text{-}ent}$, with correlation coefficients of -0.60 and -0.47, respectively, implying the dependence of the changes in the size distribution on the changes in adiabatic fraction. The result has been added in section 3.5 accordingly (line 283-286 in the revised manuscript).

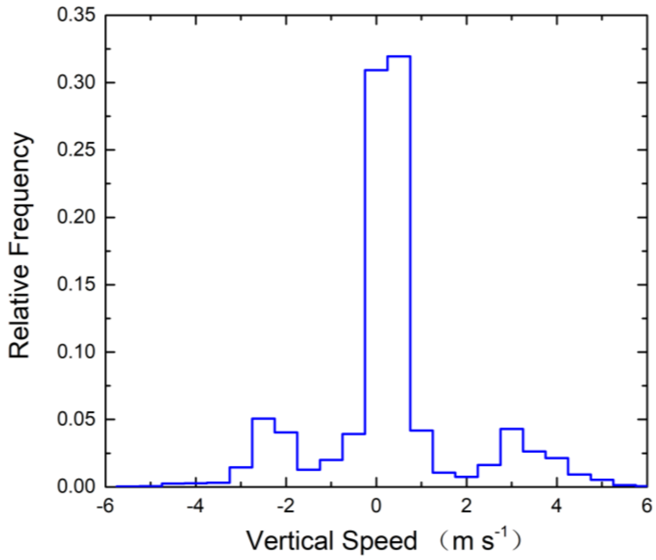

Fig. R5. Probability density functions of the vertical speed of the CIRPAS Twin Otter aircraft during the flight on 18th Oct.

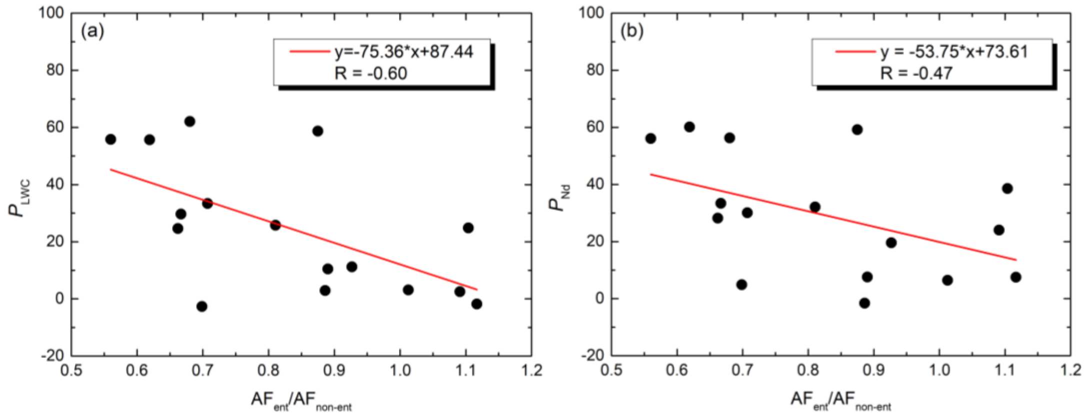

Fig. R6. (a) P_{LWC} and (b) P_{Nd} as a function of $AF_{ent}/AF_{non-ent}$ for all 16 non-drizzling flights.

There are a number of references to entrainment in cumulus clouds, but these are not relevant here. The clouds are not still developing vertically at the inversion level, whereas in cumulus, at cloud top, the clouds are still growing. Lateral entrainment is important in cumulus, but not here.

Some reference include Malinkowski ACP2012 Physics of Stratocumulus Top (POST): turbulent mixing across capping inversion, Wood Monthly Weather Review 2012 Stratocumulus Clouds, and Stevens QJ2002, Entrainment in stratocumulus-topped mixed layers.

Thanks very much for valuable suggestions. We agree that vertical velocities at the top of stratocumulus are much weaker than that of cumulus, and hence there might not be much cloud nucleation here. We have modified the text in section 3.5 accordingly, and those references have been also included to support the conclusion.

Line 285 - you suggest that entrainment of above cloud aerosol could be important, but elsewhere state it isn't, and showed this with the previous Figure 4.

We agree that entrainment of above cloud aerosol might be not important due to the negligible cloud nucleation here. Also, we have modified the text in section 3.5 accordingly.

Line 287 - probability of what?

It is the probability of Di.

Line 288 onwards - drier air would also case reduction in size.

In Figure. 2f, it is clearly shown that the effective diameter of aerosol particles above cloud is smaller than that below cloud. To minimize the effect of saturated environment on aerosol size, we excluded the data with relative humidity greater than 97%, and found that the aerosol size in the entrainment zone is still smaller than that in the non-entrainment zone. This result implies that small particles are indeed entrained into cloud from the top.

Line 312 - is this the increase in LWC from increased sub-CCN?

This part of the analysis is intended to illustrate the cloud formation in different aerosol loadings, i.e., for the polluted condition, the increase of LWC is mainly contributed by Nd instead of Re, in which large number of cloud droplets are formed with smaller size, and the reverse is true for clean the condition. Of course, this can also be used to support the conclusion that LWC increases with sub-CCN due to more cloud droplet.

line 325 - do dynamical considerations mask the dispersion effect or is the effect lower once vertical velocity is considered?

In general, the different dynamics mask the aerosol effect on relative dispersion. As indicated in Fig 10, if do not constrain the differences of cloud dynamics, the positive slope of aerosol concentration versus relative dispersion tends to be weaker, i.e., an underestimation of dispersion effect.

Line 334, 335 - the stratocumulus entrainment references may assist here. At cloud top vertical velocities will tend towards zero, and entrainment will dry the cloud and evaporate particles. There will not be much cloud nucleation here.

Thanks for suggestions. As reviewer stated, 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 overall tend towards zero with only about 4% of updrafts 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 by inhibiting activation due to the negligible cloud nucleation here. The effect of entrainment mixing on stratocumulus is mainly governed by the entrained dry air rather than small aerosols. These discussions have been included in section 3.5. The text has been revised accordingly.

3 Technical corrections

There are numerous errors of tense and grammar that should be corrected. Line 122, attributable Line 130, aerosols in, not on. Line 153, replace figure omitted with not shown line 163, As the certain... suggest re-writing for clarity line 164, replace contributed with controlled line 186, remove more, replace with spurious? As those extra aerosol area an artefact. line 196, Since part of.. suggest: Since part of the aerosol population has activated, or similar. line 200, and thus THEY activate line 209 Those aerosol, not that line 210 for INTO larger cloud droplets(Twohy

There are others to consider as well.

All revised. Thanks.

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Exploring aerosol cloud interaction using VOCALS-REx aircraft measurements

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Abstract. In situ aircraft measurements during the VAMOS Ocean-Cloud-Atmosphere-Land Study-Regional Experiment (VOCALS-REx) field campaign are employed to study the interaction between aerosol and stratocumulus over the southeast Pacific Ocean, 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 controlled~~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. An increase in aerosol concentration tends to increase both 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, positive correlation between ε and N_d become stronger, implying that perturbations of w could weaken the influence of aerosol on ε, and hence may result in an underestimation of aerosol dispersion effect. The 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, namely the entrainment reduces N_d and *LWC* by 28.9 % and 24.8 % on average, respectively, while the size of droplets is relatively unaffected. In entrainment zone, ~~smaller aerosols and~~ drier air entrained from the top induce less cloud droplet 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 droplets evaporation.

1 Introduction

Stratocumulus plays a key role in the radiative energy budget of the Earth by reflecting incoming shortwave radiation and thus cools the surface of the planet and offsets 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).

The marine stratocumulus overlaying the southeast Pacific Ocean (SEP) is the largest and most persistent clouds in the world (Klein and Hartmann, 1993; Bretherton et al., 2004). Sources of anthropogenic aerosol from the Chilean and Peruvian coasts, in contrast with relatively clean air masses from the Pacific Ocean, make the SEP an ideal region to explore the interaction of aerosol and stratocumulus cloud topped boundary layers. The cloud properties from satellite retrievals exhibit a gradient off the shore of Northern Chile. For example, cloud droplet number concentration decreased from 160 to 40 cm^{-3} (George and Wood, 2010) and cloud droplet effective radius increased from 8 to 14 μm from the coast to about 1000 km offshore (Wood et al., 2006). This gradient is plausibly attributable~~attributed~~ to anthropogenic aerosol near the coast. Huneeus et al. (2006) found that during easterly wind events, sulfate increased one order of magnitude over SEP, which results in 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 overcast stratocumulus is associated with reduced aerosol concentration, and an air mass not passing through the Chilean coast, which further confirms the impact of aerosol on stratocumulus over 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-coastal of northern Chile and southern Peru to the remote ocean in the SE Pacific during October–November 2008 (Wood et al., 2011). Studies based on this field campaign provided more information about the properties of aerosol, cloud and marine boundary layer over SEP. For instance, 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 higher aerosol concentrations near shore were associated with more but smaller cloud droplets, less liquid water path (*LWP*), and thus attributed to a combined effect of anthropogenic aerosol and the physically thinner clouds near shore. Nevertheless, an increase in *LWP* with the cloud condensation nuclei (CCN) concentrations was found during 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 improved our understanding of aerosol, cloud and boundary layer properties over SEP, the mechanisms of the detailed processes on interaction between aerosol and stratocumulus cloud is still unclear.

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, 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 aerosol below and above clouds and the interstitial aerosol 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 cloud measured by these probes are within 15 %. More detailed information about the observation instruments 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 only the measurements close to Point Alpha (20° S, 72° W) are analysed. The occurrence of clouds is defined by the following criterion, i.e., $LWC > 0.05$ g m^{-3} and $N_d > 15$ cm^{-3}. 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 the effective diameter of aerosol particle is not measured directly, so we calculated it according to the measurements of aerosol size distribution based on following equation:

$$\mathrm{Da} = \sum n_i d_i^3 / \sum n_i d_i^2 \quad (1)$$

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

To investigate the impact of the entrainment process on cloud properties and aerosol-cloud interaction, 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

The vertical profiles of aerosol, cloud and meteorological variables during 16 flights are scaled by the inversion height (z_i) (Fig. 2), which is defined as the height where the vertical gradient of liquid water potential temperature (θ_L) is the largest (Zheng et al, 2011). θ_L is conservative for water phase changes, but same as potential temperature when no liquid water exist (Betts, 1973). This normalization could exclude the variation of z_i between flights, and hence better for exploring the average BL structure during VOCALS-REx.

As shown in Fig. 2a, temperature (T) decreases sharply with the height within the BL, which is close to dry adiabatic lapse rate. A strong inversion occurs at the top of the BL, with the average temperature change about 10℃. Due to reduced T and nearly constant water vapor mixing ratio within strong mixing BL, relative humidity (RH) increases rapidly with the height (Fig. 2b). T and RH reach the minimum and maximum, respectively, when z/z_i 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 results in a slight increase in T and a slight decrease in RH. When $z/z_i > 1$, T increases to about 18 ℃ and RH decreased to 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 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 the different cloud base height of each profile. Thus, the average profiles are not shown in Fig. 2c, d, e, and the vertical variation of cloud properties can be seen easily from the single profile. Corresponding to vertical variation of *RH*, the *LWC* gradually increases with the height, reaches the maximum when *RH* is maximum (z/zi = 0.9), and then decreases when 0.9 < z/zi < 1.0. The profile of R_e is similar to that of *LWC* (Fig. 2e). For N_d, the green profile remains relatively constant, and the red one shows a slight increase with height. In general, the N_d profile remains relatively constant with a slight increase and decrease near base and top, respectively (Fig. S1), which is consistent with results in other VOCALS-REx observations (Painemal and Zuidema, 2011). Fig. 2f reveals that the effective diameter of aerosol particles (D_a) below cloud is larger than that above cloud, which is probably attributable~~attributed~~ to the different chemical composition and sources of aerosols. The profile of *CCN/CN* is similar to that of D_a (Fig. 2g), suggesting that aerosols with large size are more likely to become CCN (Dusek et al., 2006; Zhang et al., 2011). Larger D_a and *CCN/CN* 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. Most studies based on satellite data employed aerosol optical depth or aerosol index as 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. 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, the impact of CCN number concentration near cloud layer, e.g. below and above cloud respectively, on cloud properties is assessed.

The relationships between sub-cloud CCN number concentration (*sub-CCN*) and cloud properties during all flights are shown in Fig. 3. The red dots signify the ten flights with typical well mixed boundary layer and non-drizzling cases, which have relatively 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 mixed boundary layer 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 the case of typical well mixed boundary with non-drizzling, both *LWC* (Fig. 3a) and N_d (Fig. 3b) exhibit the positive relationships with *sub-CCN*, with correlation coefficients of 0.60 and 0.79, respectively, while R_e has no evident correlation with *sub-CCN* (Fig. 3c). 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. In other cases (i.e. blue dots), the impacts of aerosols on the cloud is not evident due to large difference in the meteorological conditions and the boundary layer structure.

Compared to sub-cloud CCN, the influence of above-cloud CCN on cloud properties is very weak. The absolute values of correlation coefficient between above-cloud CCN number concentration (*abv-CCN*) and cloud properties are all less than 0.4 (not shown~~figure omitted~~), none of which pass the significance test (α = 0.05). In this study, above-cloud aerosol number concentration is very low (129.8 ± 60.1 cm^{-3}) and the inversion capped the cloud top is extremely strong, which weakens the mixing of the aerosol with cloud layer and hence the effects of aerosol on cloud properties. Some previous studies based on aircraft observation 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).

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, 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. 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). On the contrary, there are enough particles which may potentially activated into cloud droplets under the polluted case, thus N_d increases rapidly with *LWC*.~~ As the certain amount water is shared by large amount particles,~~ However, due to a large number of aerosols competing for limited water vapor, the increase of R_e is not significant~~limited~~. It is suggested that the increase of *LWC* is mainly controlled~~contributed~~ 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 cloud formation between clean and polluted conditions is less evident, which is probably attributable~~attributed~~ to the much lower aerosol concentration difference between clean and polluted cases in this study (about 400 cm^{-3}) than that in Zhang et al. (2011) (about 7000 cm^{-3}).

3.3 Cloud droplet formation and its controlling factors

Sub-cloud CCN is considered as a good proxy for the aerosol entering cloud. However, during actual flight, it is difficult to collect enough samples of sub-cloud CCN and cloud droplets simultaneously, which may result in uncertainty in statistical analysis. This limitation can be overcome by employing interstitial aerosols. Interstitial aerosols are particles observed in-cloud that either never activate into cloud droplets or have been activated but then return into aerosols after evaporation of cloud droplet. Kleinman et al. (2012) pointed out that the number concentration of interstitial aerosol (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 cloud. By employing aircraft observations over both land and ocean, Gultepe et al. (1996) found that the difference of the number concentration between total in-cloud particles ($N_d + N_i$) measured directly and sub-cloud aerosols is very small. It is thus assumed that total in-cloud particles can characterize the overall level of in-cloud aerosol concentration before activation. The flight on Oct. 18 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 other flights. The average ratio of $N_d + N_i$ to 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 during VOCALS-REx. Therefore, the observation of interstitial aerosols in this study is unlikely to be significantly interfered 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 16 non-drizzling flights are shown in Fig. 6, in which the colors represent in-cloud vertical velocities. Positive 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 in Fig. 6, data are close to the 1:1 line when vertical velocity is relatively large, namely in-cloud aerosols are almost entirely activated into cloud droplets. However, data deviate from the 1:1 line when 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). This is possibly attributable~~attributed~~ to high supersaturation caused by the adiabatic uplift under conditions with large vertical velocity. 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 dynamical conditions, aerosol microphysical properties, such as size distribution and chemical components, also 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 ~~aerosols~~ aerosol population ~~in the cloud have~~has activated~~ to cloud droplets~~, it is difficult to obtain the information of aerosol size before activation. According to Köhler theory, the critical supersaturation of aerosol with large size is relatively low, and thus they activate preferentially, i.e. the effective diameter of interstitial aerosol (D_i) is smaller than that of initial aerosols before activation. Li et al. (2011) compared the difference of size distribution between interstitial aerosol 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 increase of D_i, though the quantitative relation 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 interstitial aerosol can still represent the size of initial aerosols before activation to some extent. As indicated in Fig. 7, the larger D_i is, the closer the data 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 is larger than 1.0 μm, but only 0.64 ± 0.23 when D_i is less than 0.5 μm. It is because ~~that~~ those aerosols with large size 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 "dispersion effect") and thereby 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., 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 aerosol number concentration (N_a). However, the relationship between ε and N_a still remains large uncertainty. Table 1 shows that the observed correlations between ε and N_d (or N_a) can be positive, negative, or not evident. Different relations are indicative of the fact that the effect of aerosol on ε is often intertwined with 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, 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 vertical velocity (w, m s^{-1}) as a proxy for cloud dynamical condition. As shown in Fig. 8, the correlations between ε and N_d based on in-flight data is significantly negative during all 16 non-drizzling flights, which is mainly modulated by w, i.e., larger w corresponds to a smaller ε but larger N_d. High supersaturation leads to more cloud droplets to activate and grow to the same size (i.e., narrow the droplet spectrum) when w is relative large, but a portion of cloud droplets may evaporate into smaller size and even deactivate into interstitial aerosols when w is small or even negative, resulting in the 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 one based on the flight-averaged data could be either positive, negative, or even uncorrelated. The uncertain relationships of the later may result from variations of the strength of cloud dynamic between flights, which would disrupt or even cancel the real influence of aerosol on relative dispersion (Peng et al., 2007; Lu et al., 2012). However, many previous studies did not take the difference of cloud dynamics in 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, data in all flights were sampled over the same location, i.e., Point Alpha, which can reduce the difference of dynamic conditions caused by variations of horizontal sampling location. In addition, we also distinguish the flights of typical mixed boundary layer and the others to ensure relatively similar meteorological conditions (see section 3.2). Fig. 9 shows the probability distribution function of w with mean values and standard deviations for 16 non-drizzling flights. The related statistics are shown in Table 2. It can be found that, except for other cases (~~gray shadow~~crosses; especially Oct. 24, Oct. 29, Nov. 8, and Nov. 13), the difference of in-cloud dynamics between typical well mixed boundary flights is very small, which confirms the assumption of similar meteorological conditions. As indicated in Fig. 10a, ε and N_d are positively correlated (correlation coefficient of 0.29 and the slope of 1.9×10^{-4}) in the case of the typical well mixed boundary, indicating that aerosol increases ε and N_d at the same time. However, correlation coefficient and slope reduce to 0.11 and 7.7×10^{-5}, respectively in the all cases (i.e., not to constrain w), implying that the influence of aerosol on ε-N_d relationship tends to be weaker after intertwined with effects of cloud dynamics. Although the perturbations of cloud dynamics have been eliminated as far as possible, N_d is still likely determined by both aerosols number concentrations and updraft velocity together. Therefore, similar statistical analysis are also conducted for sub-cloud CCN. The relationship between ε and sub-cloud CCN is similar to that between ε and N_d, but, as expected, the correlation coefficient (slope) in the case of typical well mixed 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 in the clouds, which plays an important role in the formation and evolution of clouds and the change of droplet spectrum, as well as 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 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 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 showed that the vertical profile of LWC is essentially same as the adiabatic profile, i.e. the cloud is almost adiabatic (Keil and Haywood, 2003).

In order to explore the entrainment in stratocumulus during VOCALS-REx, we firstly compared the differences of cloud microphysics between entrainment and non-entrainment zone near the cloud top. Here, entrainment and non-entrainment zone are defined as the regions within 20 m above and below the height of maximal LWC, respectively. As anticipated, adiabatic fraction (AF, the ratio of the measured LWC to its adiabatic value) in entrainment zone (AF_{ent}) is generally lower than that in non-entrainment zone ($AF_{non\text{-}ent}$), with the mean values of all flights of 0.64 and 0.77 respectively (Table 2), which further confirms the rationality in dividing the two zones. Compared with non-entrainment zone, the peak diameters of cloud droplets in entrainment zone has little change (Fig. 11), and the effective diameters of cloud droplet (D_e) increases only by 1.8 % (Table 2). However, N_d and LWC decrease significantly by 28.9 % and 24.8% respectively on average (Table 2), especially during flights on Oct. 18, Nov. 04, Nov. 09 and Nov. 13, N_d decreases by 60.1 %, 56.3 %, 56.1 % and 59.2 %, and LWC decreases by 55.7 %, 62.1 %, 55.8 % and 58.7 %, respectively (Table 2). It is suggested that dry and warm air entrained from cloud top dilutes N_d and LWC by a similar amount, while the size of droplets is relatively unaffected, which is thought as extreme inhomogeneous entrainment-mixing process. Moreover, both P_{LWC} and P_{Nd} are negatively correlated with $AF_{ent}/AF_{non\text{-}ent}$, with correlation coefficients of -0.60 and -0.47, respectively, indicating the dependence of the changes in LWC and N_d on the changes in adiabatic fraction (Fig. S2), where P_{LWC} and P_N are the percentages of reduction in LWC and N_d within entrainment zone relative to non-entrainment zone. ~~Although i~~It is still unclear whether the entrainment-mixing mechanism is predominantly homogeneous, inhomogeneous, or in between (Andrejczuk et al., 2009; Lehmann et al., 2009)~~,~~. ~~s~~Some 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). ~~Furthermore,~~ By employing a different vertical description in characterizing the region near cloud top (Malinowski et al., 2013), Gerber et al. (2016) pointed out that both extreme inhomogeneous mixing and homogenous mixing play a role in unbroken stratocumulus, but the reduction in cloud droplet effective radius appears secondary in comparison to the dilution process that preserves the relative shape of the droplet spectrum.

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. 12b, dry and warm air entrained from the top reduces the relative humidity in entrainment zone by 8.8 % on average, and hence acts to accelerate the cloud droplets evaporation. As a consequence, $N_d/(N_d + N_i)$ in entrainment zone (0.56 ± 0.22) is much lower than that in non-entrainment zone (0.73 ± 0.13) (Fig. 12c). Moreover, the relative dispersion in entrainment zone is overall larger than that in non-entrainment zone (Fig. 12d), implying that drier air entrained from the top could broaden cloud droplet spectrum by promoting the 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). It is noted that the probability of D_i 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 (Fig. 12a). This result suggests that, in addition to dry and warm air, small particles are also entrained into cloud from the top (Fig. 2f) and large particles are detrained out cloud at the same time. However, 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 overall tend towards zero with only about 4% of updrafts 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 by inhibiting activation due to the negligible cloud nucleation here. 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, 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 presented that the BL is well mixed and capped by a sharp inversion during 16 non-drizzling flights. Cloud variables, such as LWC, N_d, and cloud depth, are all positively correlated with sub-cloud CCN number concentration, having the correlation coefficients of 0.60, 0.79 and 0.71, respectively. No evident correlation was found between cloud properties with above-cloud CCN number concentrations. This is mainly due to low aerosol number concentrations above-cloud (129.8 ± 60.1 cm^{-3}) and the extremely strong inversion capped the cloud top, which inhibits the mixing of the above-cloud aerosol with cloud layer. Therefore, the influence of above-cloud CCN on cloud properties is very weak compared to sub-cloud CCN. Additionally, the comparison of cloud formation under different aerosol number concentrations conditions suggested that the increase of LWC is probably controlled~~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 condition 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 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. Although chemical components of aerosol is 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 was not discussed in this study due to unavailable measurements.

The correlations between ε and N_d based on the in-flight data, used to represent w-induced correlation, is significantly negative, while the correlations derived from flight-averaged data (i.e., aerosol-induced correlation) is positive. This implies that an increase in aerosol concentration tends to 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 with theoretical analysis (Liu et al., 2006). After constraining the differences of cloud dynamics between flights, positive correlation between ε and N_d become stronger, indicating that perturbations of w could weaken the influence of aerosol on ε, and hence may result in an underestimation of aerosol dispersion effect. Thus, it requires more attention to isolate the response of relative dispersion to aerosol perturbations from dynamical effects when investigating aerosol dispersion effect and estimating aerosol indirect forcing.

The entrainment in stratocumulus is overall quite weak, and close to adiabatic in some case. In this study, the difference of cloud microphysics between entrainment and non-entrainment zone 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 28.9 % and 24.8 %, respectively, while had little effect on D_e (only increases by 1.8 %). During flights on Oct. 18, Nov. 04, Nov. 09 and Nov. 13, the entrainment is relatively strong and dilutes N_d and LWC by about 50 %. In entrainment zone, the ~~smaller aerosols and~~ drier air entrained from the top result in the smaller $N_d/(N_d + N_i)$ (0.56 ± 0.22) than that in non-entrainment zone (0.73 ± 0.13). This implies that entrainment may significantly influence cloud droplet formation and hence cloud properties near the top by ~~both inhibiting aerosol activation and~~ promoting cloud droplets evaporation. Furthermore, we also found that the relative dispersion in entrainment zone is larger than that in non-entrainment zone. In addition to the dry and warm air, aerosols with smaller size are also entrained into entrainment zone, but due to the negligible droplet nucleation near the top of stratocumulus, these aerosols seem unlikely to influence cloud droplet formation by inhibiting activation. That is, the effect of entrainment mixing on stratocumulus is mainly determined by the entrained dry air instead of aerosols with different properties from those near the cloud base. But for cumulus, things may be different. 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. By 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. Thus, it might be an interesting topic that how and to what extent the entrained aerosols with different properties from sub-cloud aerosols can affect the formation and evolution of clouds. ~~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	55.7	33.4	24.8	24.6	29.3	-2.7	11.2	3.1
P_{Nd}[d~~b~~]	32.1	60.1	30.1	38.6	28.2	34.4	4.9	19.6	6.4
P_{De}[f~~e~~]	-1.9	-5.7	0.9	-6.7	-1.9	-0.1	-4.1	-2.4	-1.8
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	2.5	55.8	2.9	-1.8	58.7	24.8
P_{Nd}	7.6	56.3	24.0	56.1	-1.6	7.5	59.2	28.9
P_{De}	0.2	4.4	-8.4	-2.1	3.4	-2.5	-1.2	-1.8
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~~, ~~c~~b, f~~e~~] P_{LWC}, P_{Nd}, and P_{De} are the percentages of reduction in LWC, N_d and 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.

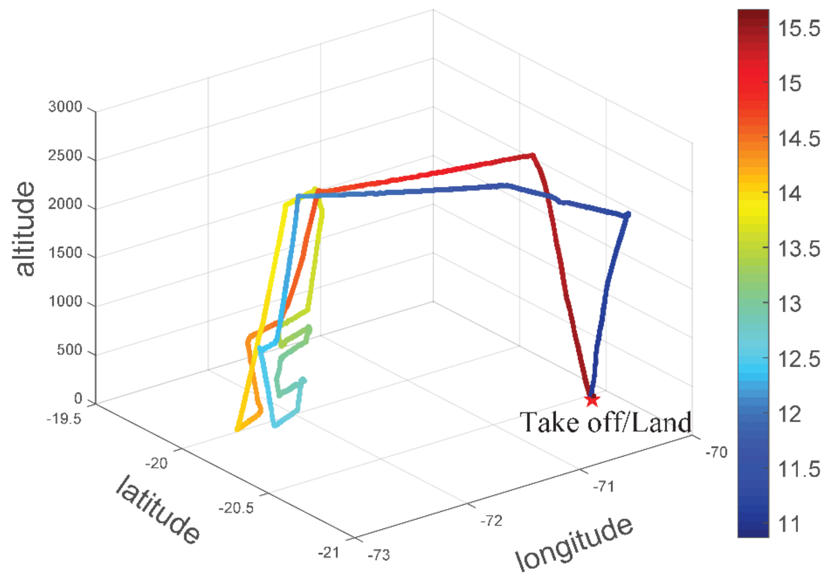
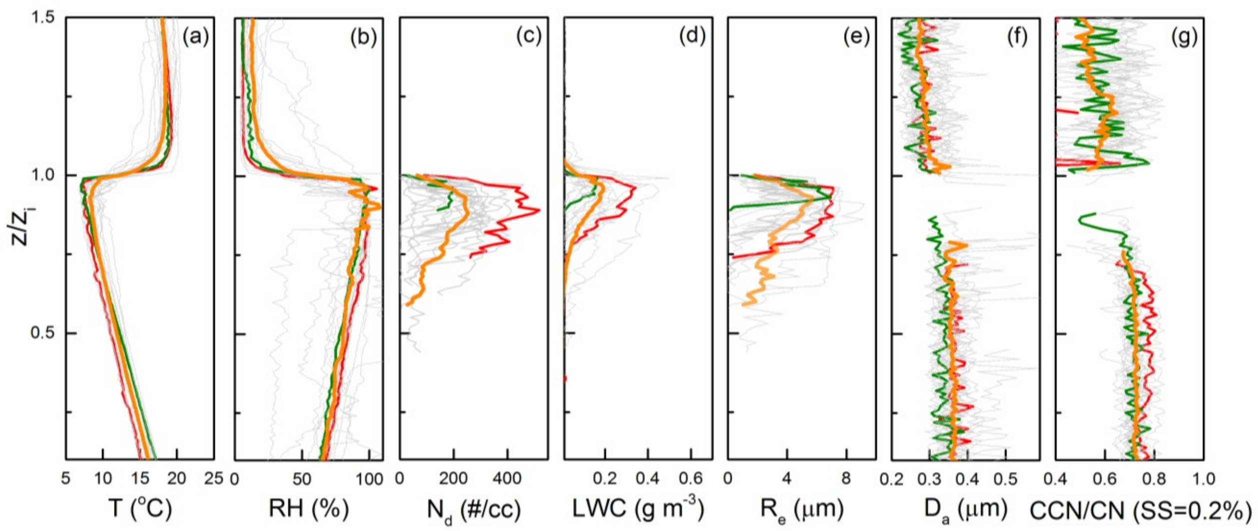


Fig. 1. The flight track in Oct. 18, and the colors represent flight time in hour (UTC).



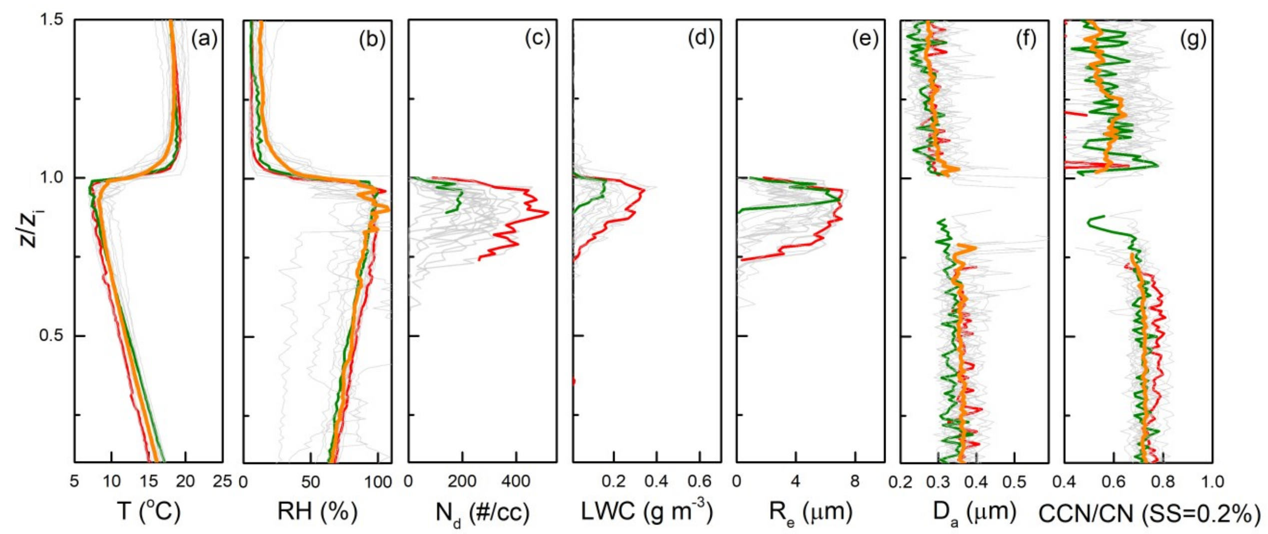
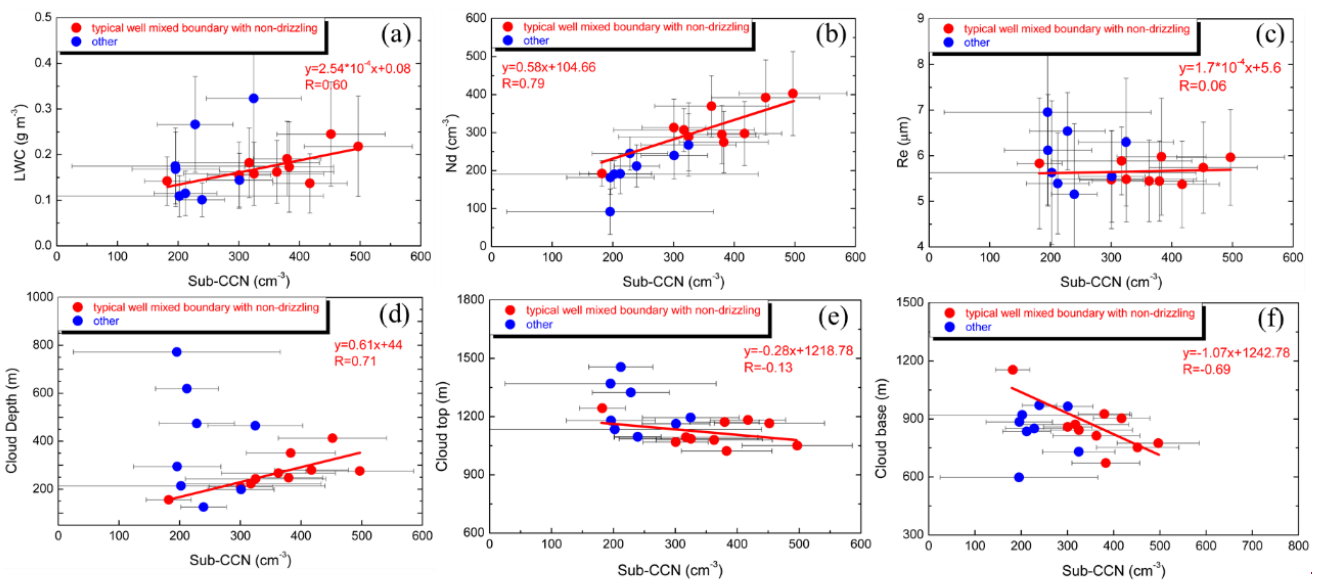


Fig. 2. Vertical profiles scaled by the inversion height. (a) temperature (K); (b) relative humidity (%); (c) cloud droplet number concentration (cm $^{-3}$); (d) liquid water content (g m $^{-3}$); (e) effective radius of cloud droplets (μm); (f) effective diameter of aerosols (μm), and (g) the number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



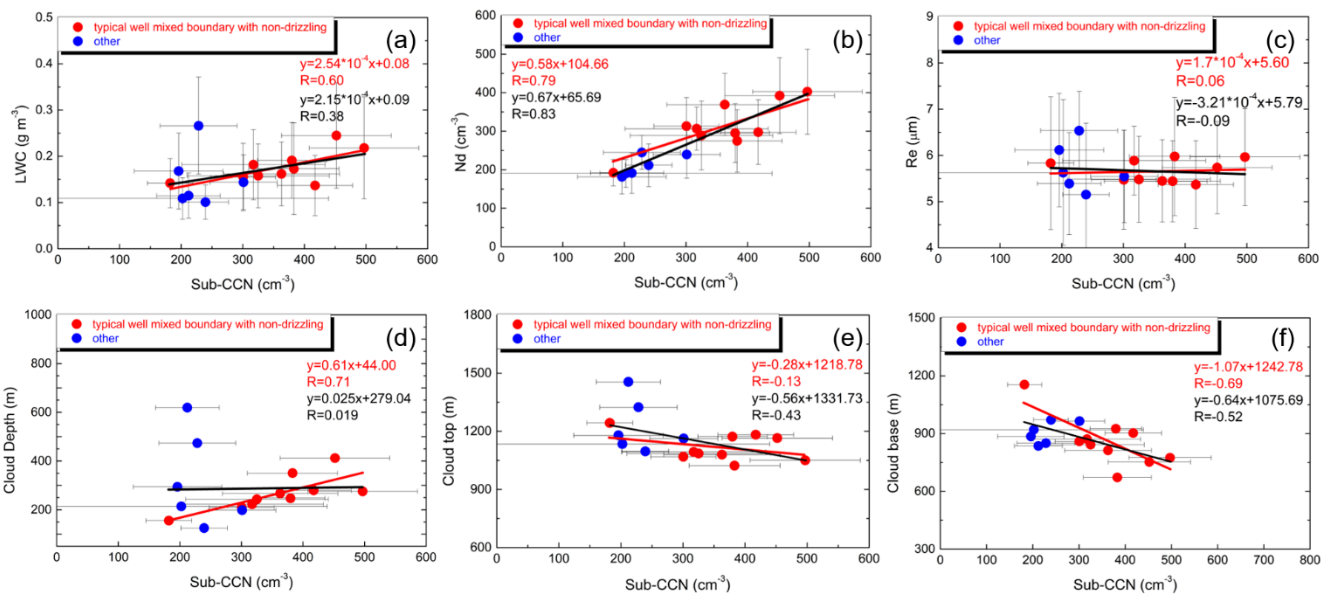


Fig. 3. (a) *LWC* (g cm $^{-3}$); (b) N_d (cm $^{-3}$); (c) R_e (μm); (d) cloud depth (m); (e) cloud top height (m); (f) cloud base height (m) as a function of sub-cloud CCN concentrations (SS=0.2%) for all 16 non-drizzling flights. The error bars through these symbols indicate the standard deviation. Red symbols are the typical well mixed boundary 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).

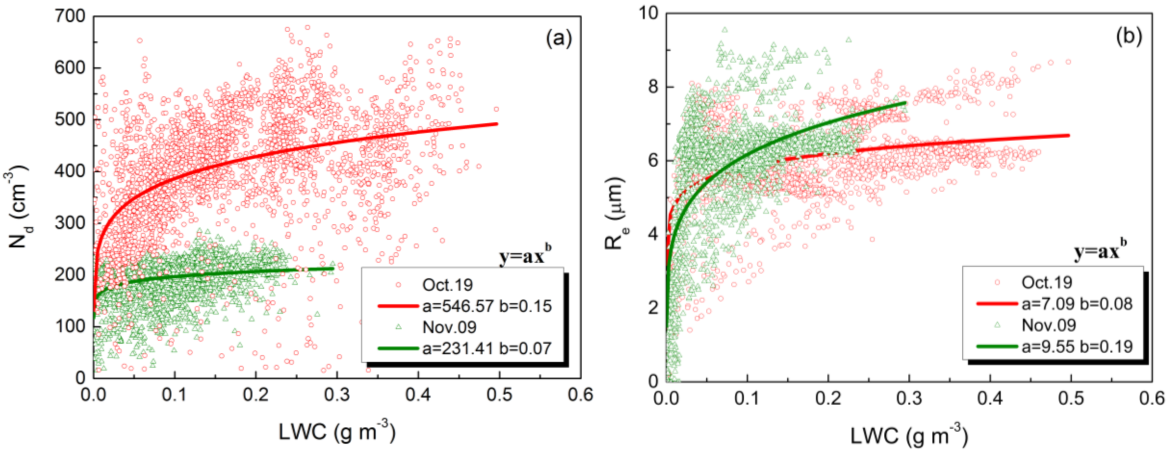


Fig. 4. Correlations between (a) N_d (cm^{-3}), (b) R_e (μm) and LWC (g m^{-3}) for clean (green) and polluted (red) cases, respectively.

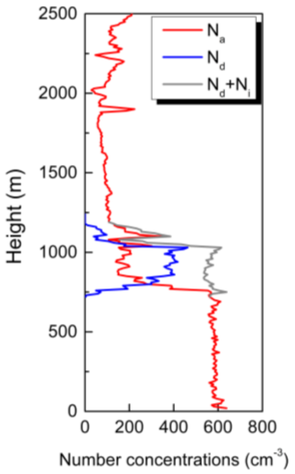
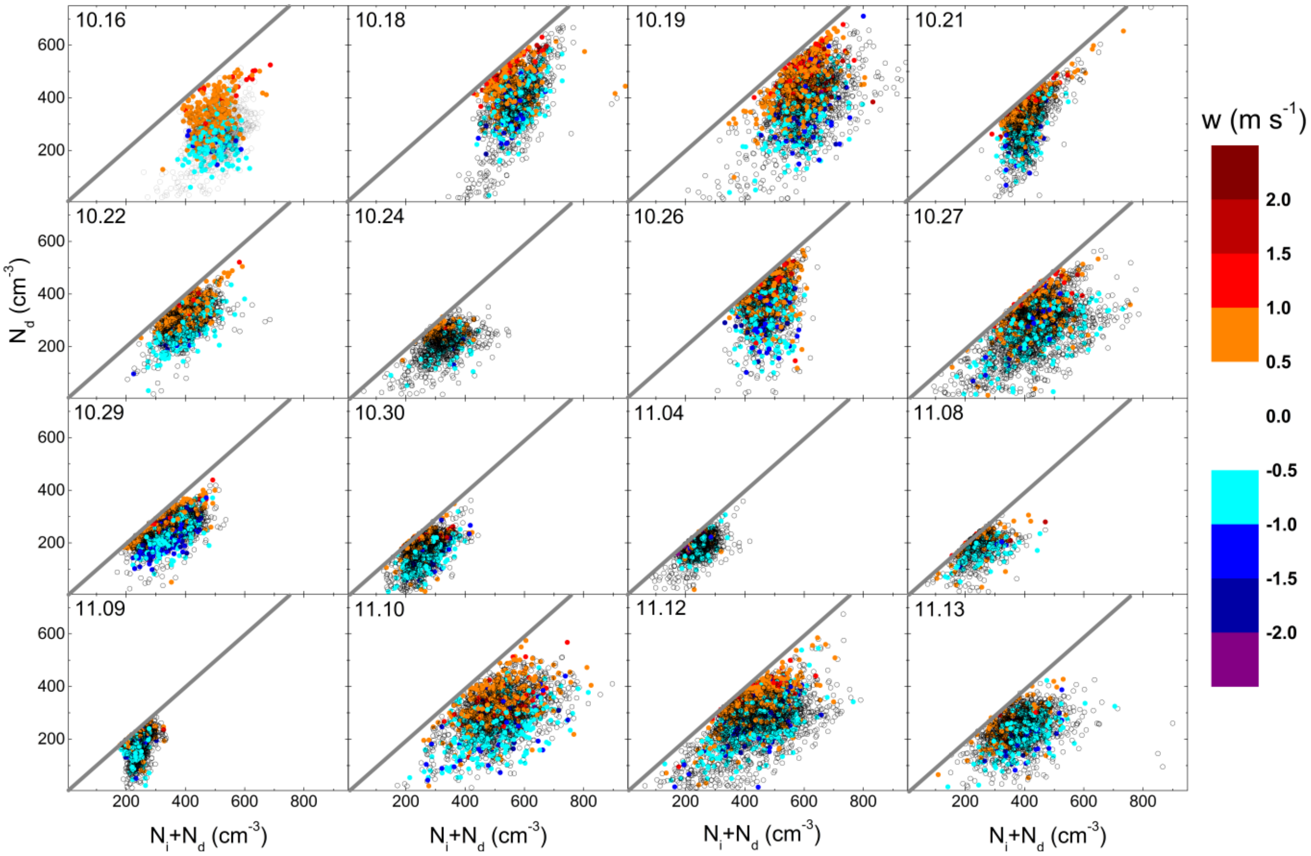


Fig. 5. Vertical profiles of number concentrations of aerosols (N_a), cloud droplets (N_d) and total in-cloud particles ($N_d + N_i$) during the flight on Oct. 18.



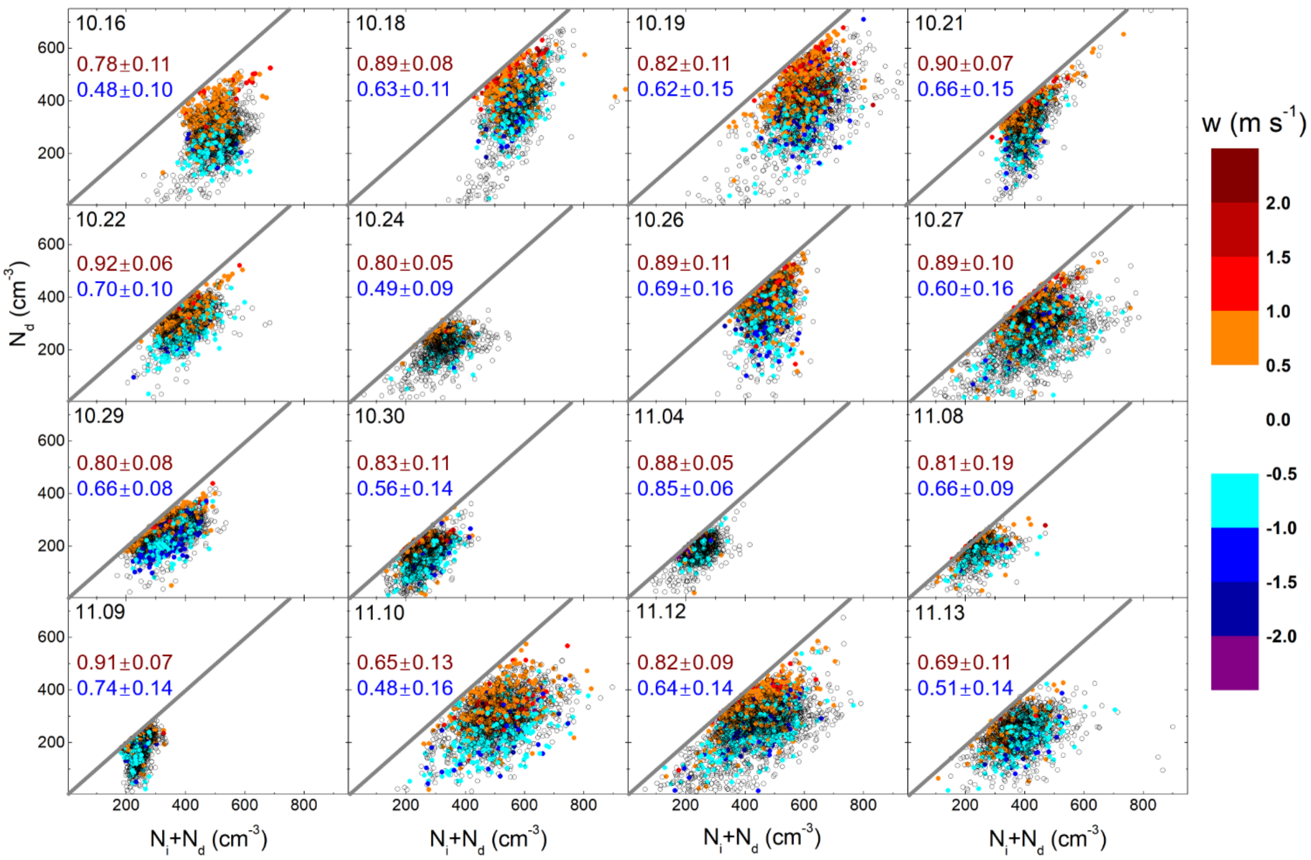


Fig. 6. Relationships between N_d and $N_i + N_d$ during all 16 non-drizzling flights. The colors represent in-cloud vertical velocities (m s^{-1}), and gray line is 1:1 line. **The mean and standard deviation of Nd/(Nd+Ni) for vertical velocity greater than 1 m s^{-1} (red) and less**

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

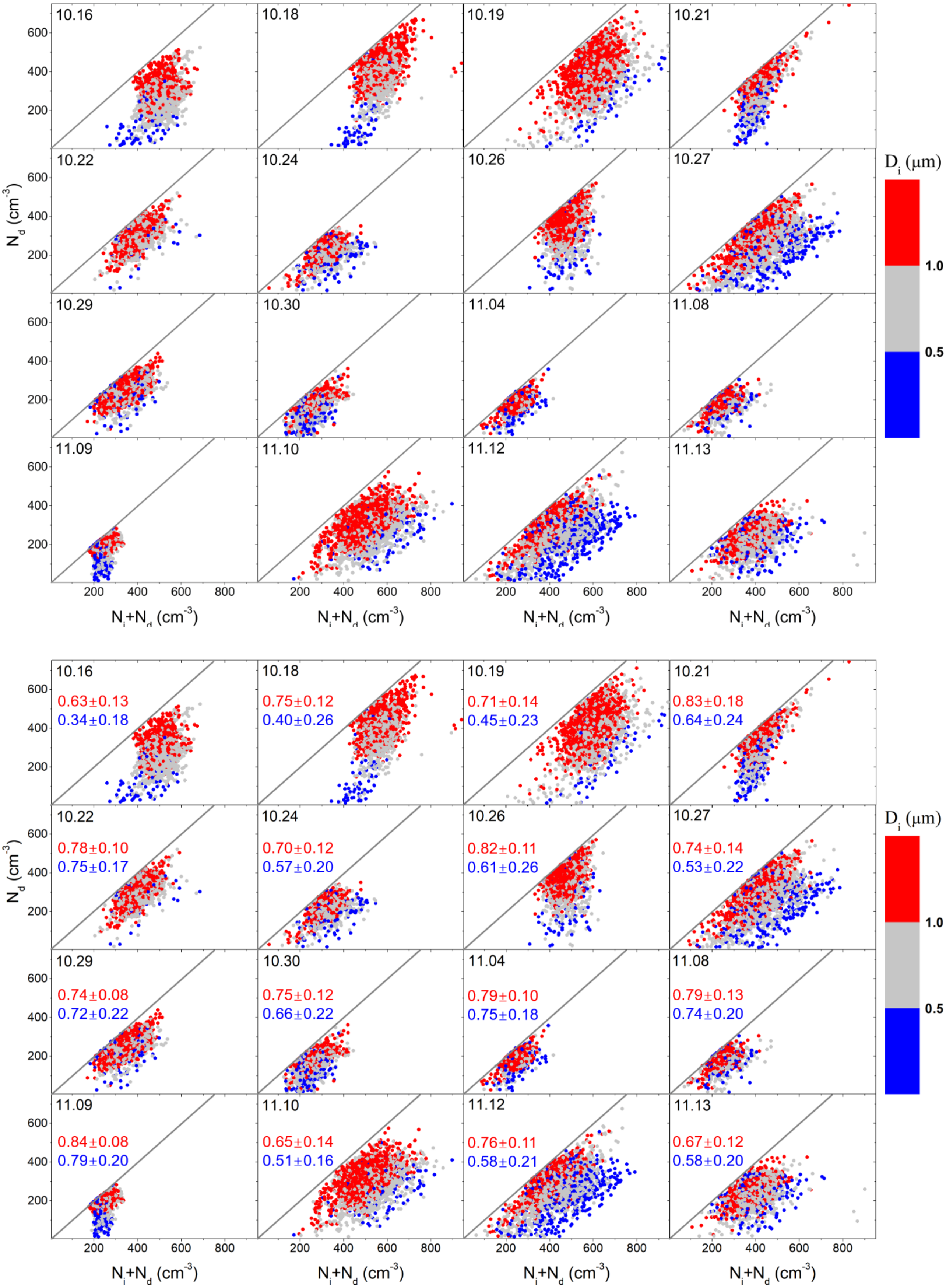


Fig. 7. Same as Fig. 6, but the colors represent the effective diameter of interstitial aerosol (D_i) (μm). The mean and standard deviation of $N_d/(N_d+N_i)$ for D_i greater than 1 μm (red) and less than 0.5 μm (blue) are shown.

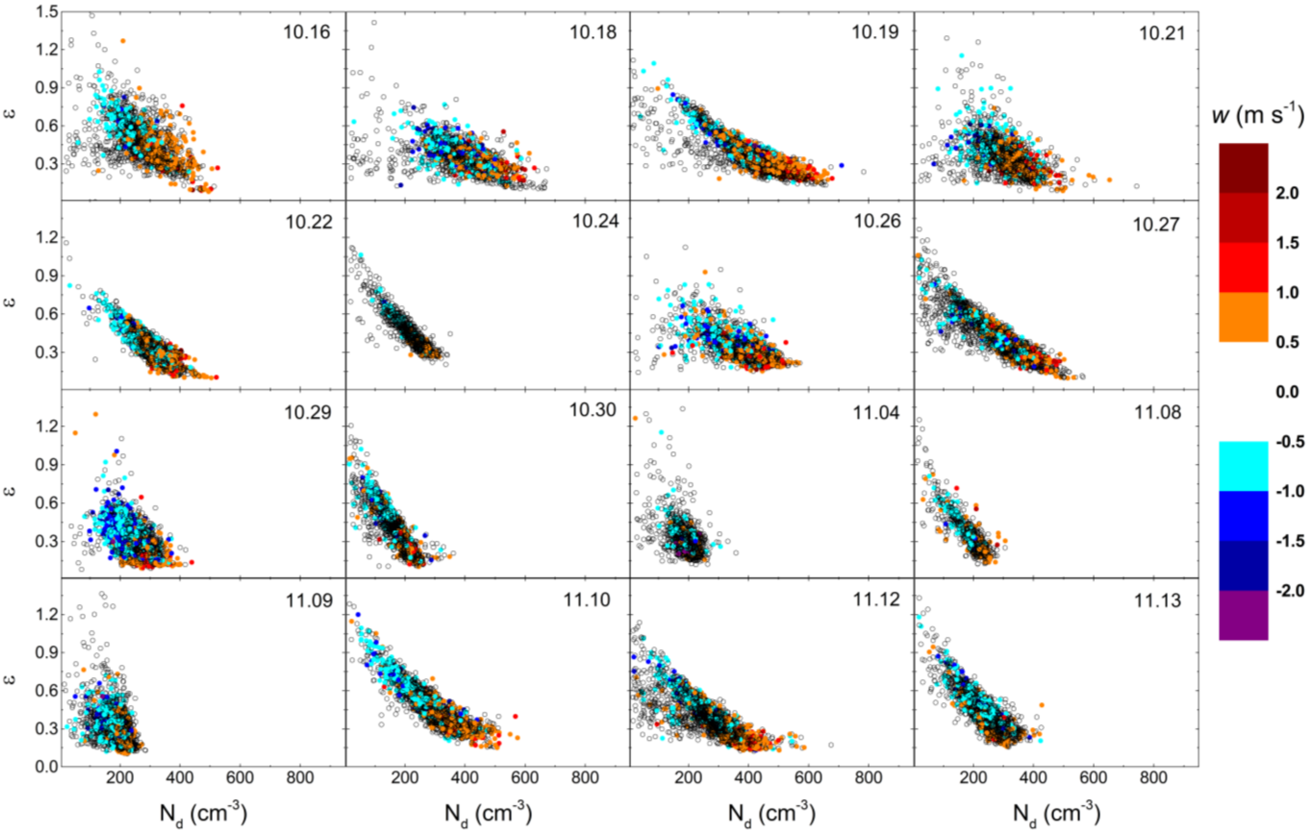
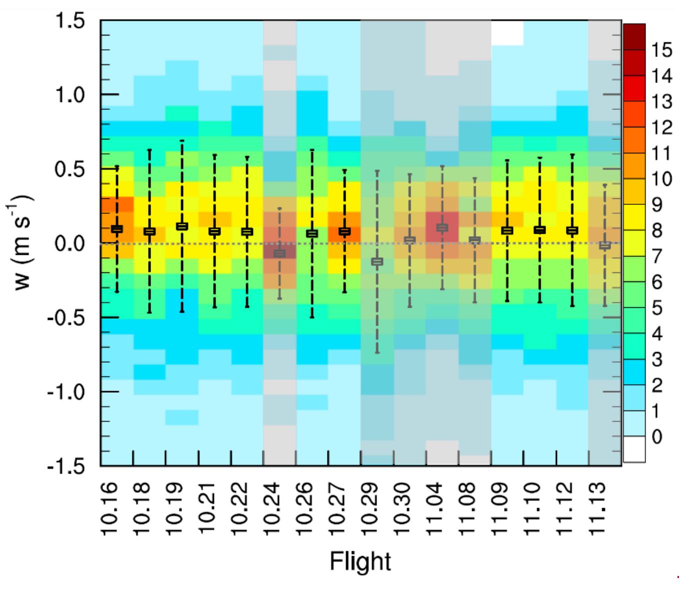


Fig. 8. Relationships between relative dispersion (ε) and N_d during all 16 non-drizzling flights, in which the colors representing in-cloud vertical velocities ($m\ s^{-1}$).



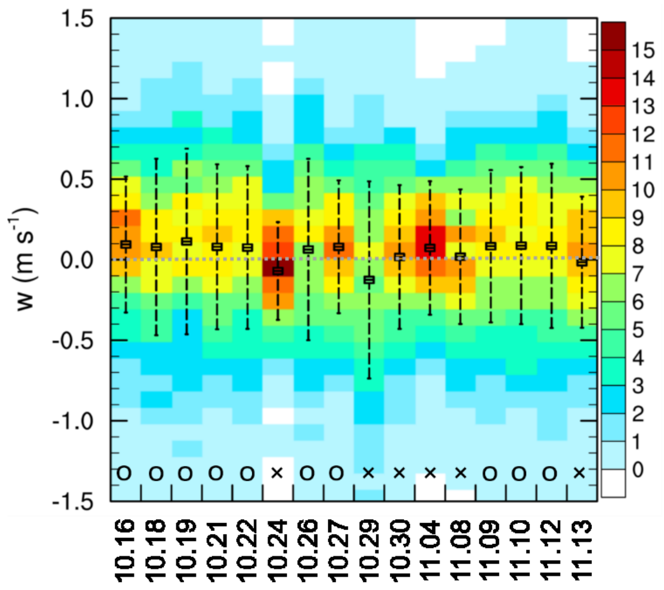


Fig. 9. Probability distribution function (units: %) of vertical velocity (*w*) for 16 non-drizzling flights. Black symbols are mean values of *w*, and error bars through these symbols indicate the standard deviation. ~~Gray shadow represents the flights other than typical well mixed boundary with non-drizzling.~~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

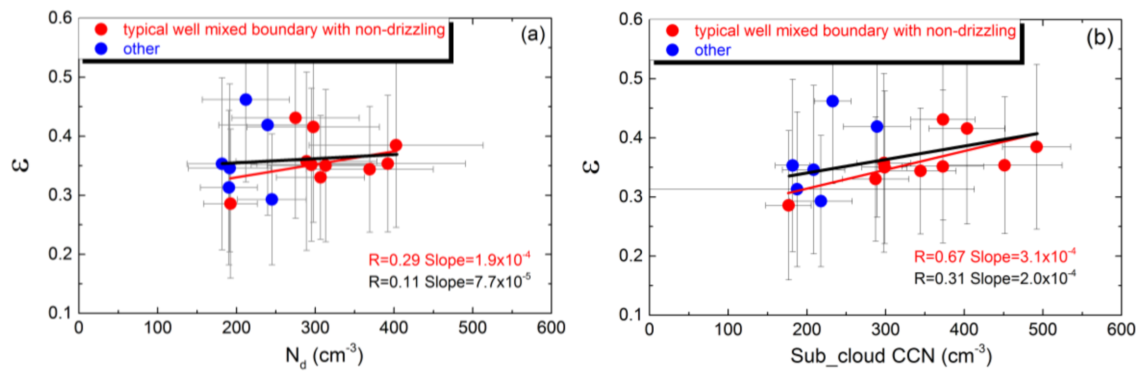


Fig. 10. Relative dispersion (*ε*) as a function of (a) N_d and (b) sub-cloud CCN concentrations (*SS*=0.2%) for all flights. The error bars through these symbols indicate the standard deviation. Red symbols are the typical well mixed boundary with non-drizzling, and blue symbols for others. Red (black)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

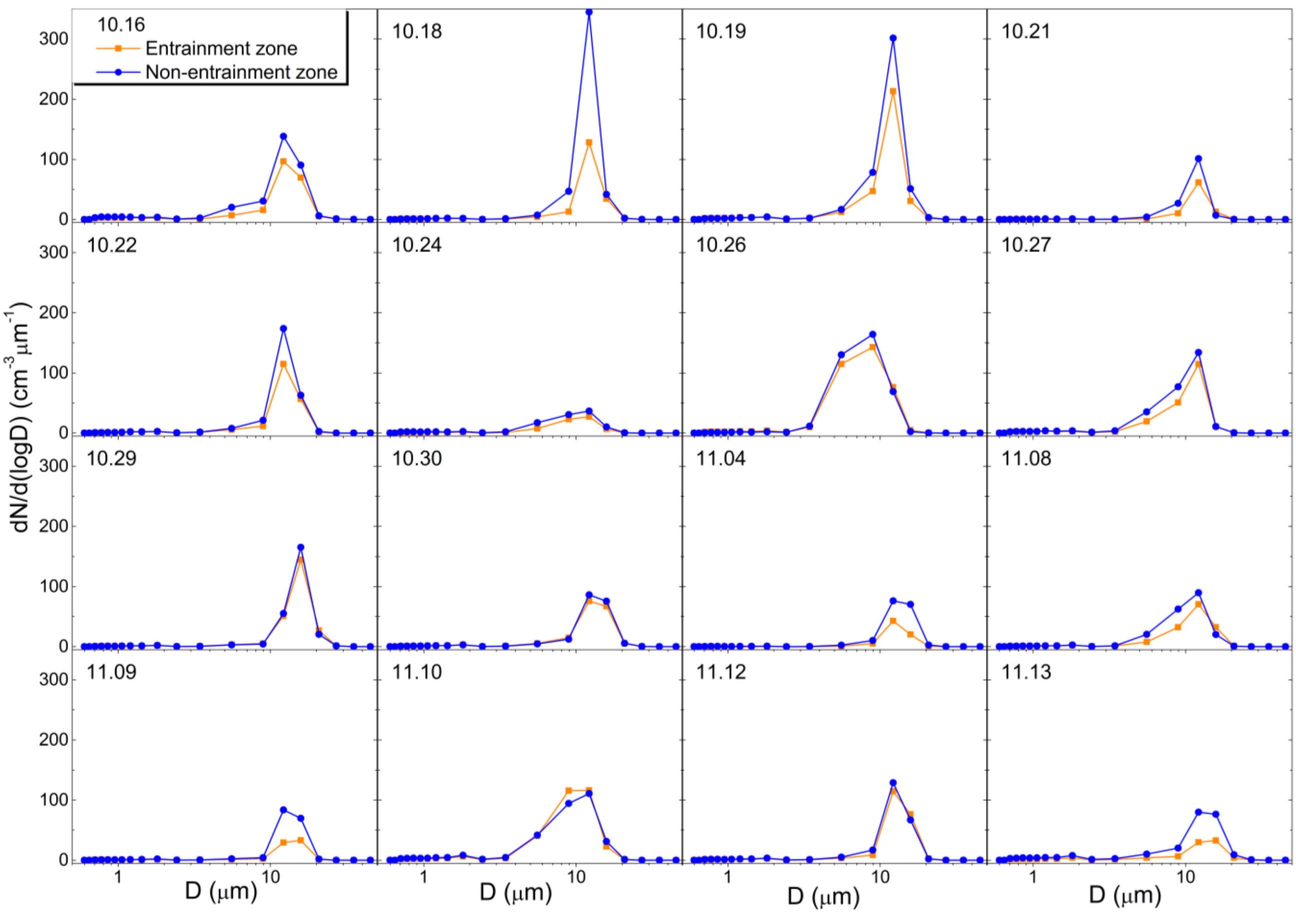


Fig. 11. Number size distributions of cloud droplets in entrainment (yellow) and non-entrainment zone (blue) during all 16 non-drizzling flights.

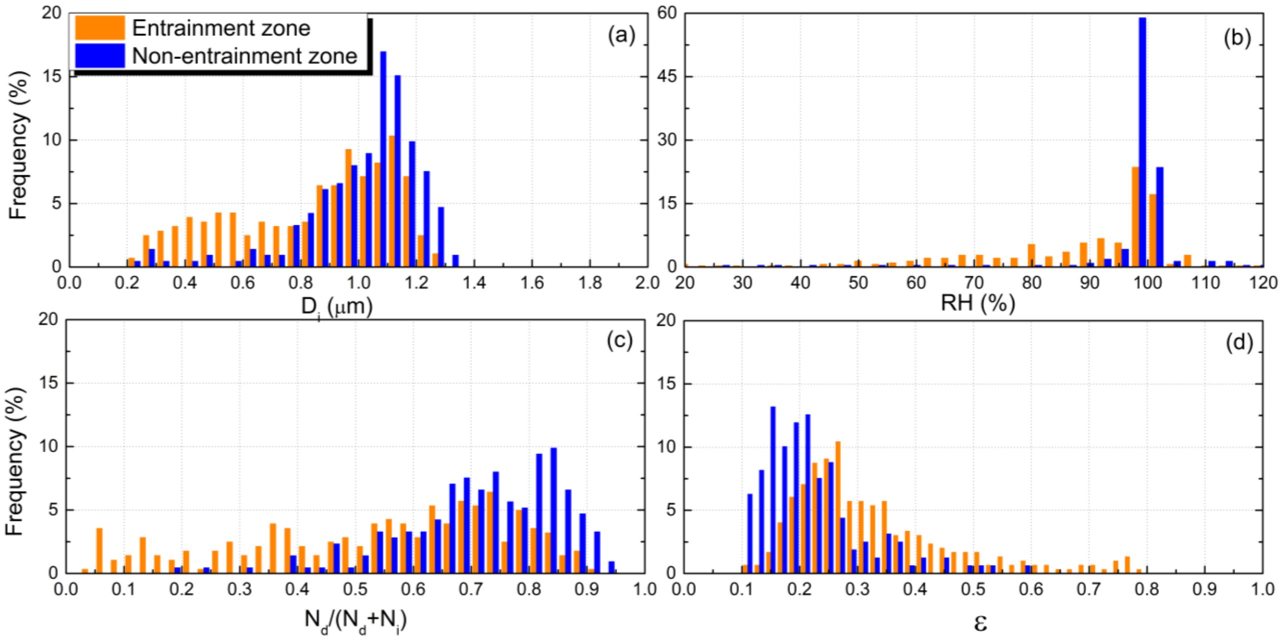


Fig. 12. Probability density functions of (a) D_i (μm), (b) *RH* (%), (c) $N_d/(N_d + N_i)$, and (d) ε in entrainment (yellow) and non-entrainment zone (blue) during the flight on Oct. 18.

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

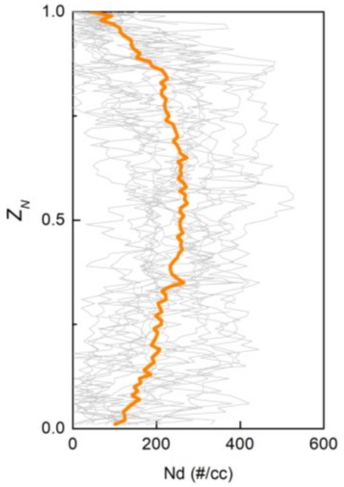

Figure S2