

Dear Co-Editor,

We thank the reviewers for their thoughtful comments, which have allowed us to clarify and improve the manuscript. We have carefully revised the manuscript according to the reviewer's suggestions and comments.

In particular, we have made substantial changes to the abstract and introduction to clarify the novelty and contribution of this study. Although previous studies published on VOCALS measurements have certainly improved our understanding of some aspects related to aerosols, clouds and boundary layer (BL) properties over the southeast Pacific Ocean (SEP), several important factors remain understudied or unexplored. First, the effect of aerosols on clouds is often intertwined with the effects of other factors, especially meteorological conditions. Currently, the impact of aerosols on the shape of the cloud droplet size spectrum (i.e., dispersion effect) is reported to remain largely uncertain, which may be mostly attributable to the coincidentally changing cloud dynamics. Thus, it is necessary to isolate the response of relative dispersion to aerosol perturbations from meteorological effects, which to our knowledge, has not been the focus of attention in many previous studies. Second, applying different assumptions to the entrainment-mixing mechanism has a significant impact on the cloud albedo. However, it remains unclear whether the entrainment-mixing mechanism is predominantly homogeneous, inhomogeneous, or in between, which could lead to inaccurate assessments of aerosol indirect effects. Thus, more attention should be given to this topic.

In this manuscript, we examine the detailed processes of aerosol-cloud interactions over the SEP, with a focus on the understudied topics (separation of aerosol effects from dynamic effects, dispersion effects, and turbulent entrainment-mixing processes). This study is helpful for reducing the uncertainty in dispersion effects and entrainment mixing in stratocumulus clouds, and the result of this study may benefit cloud parameterizations in global climate models to more accurately assess the aerosol indirect effects.

Listed below are our responses to the comments from the reviewers. For clarity and visual distinction, the reviewer's comments are listed in black and the authors' responses are in blue.

Sincerely,

Xiaoyan Ma, Hailing Jia, and Yangang Liu

Response to Referee #1

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 103 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 N_d 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 154-160 in the revised manuscript).

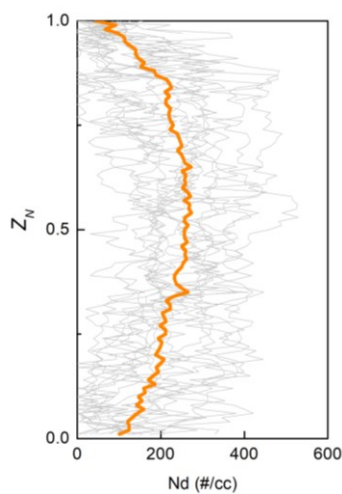


Figure R1. Normalized profiles of N_d . Values of $Z_N=0$ indicates the cloud base whereas $Z_N=1$ the cloud top. Orange line indicates the average profiles.

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 D_i (Fig. R3). It is found that, without strong supersaturation, $Nd/(Nd+Ni)$ still tend to increases with D_i , so it seems saturated environment might not influence our conclusion significantly.

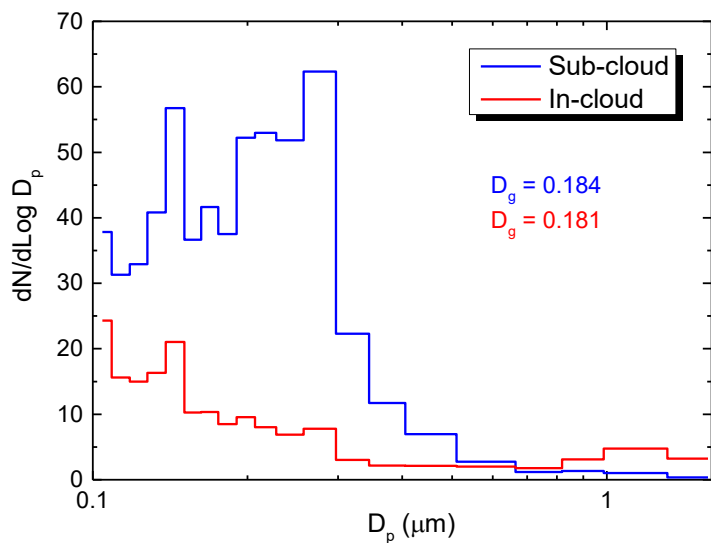


Figure R2. The size distributions for the sub-cloud aerosol (blue) and the in-cloud aerosol (red) during the flight on 18th Oct.

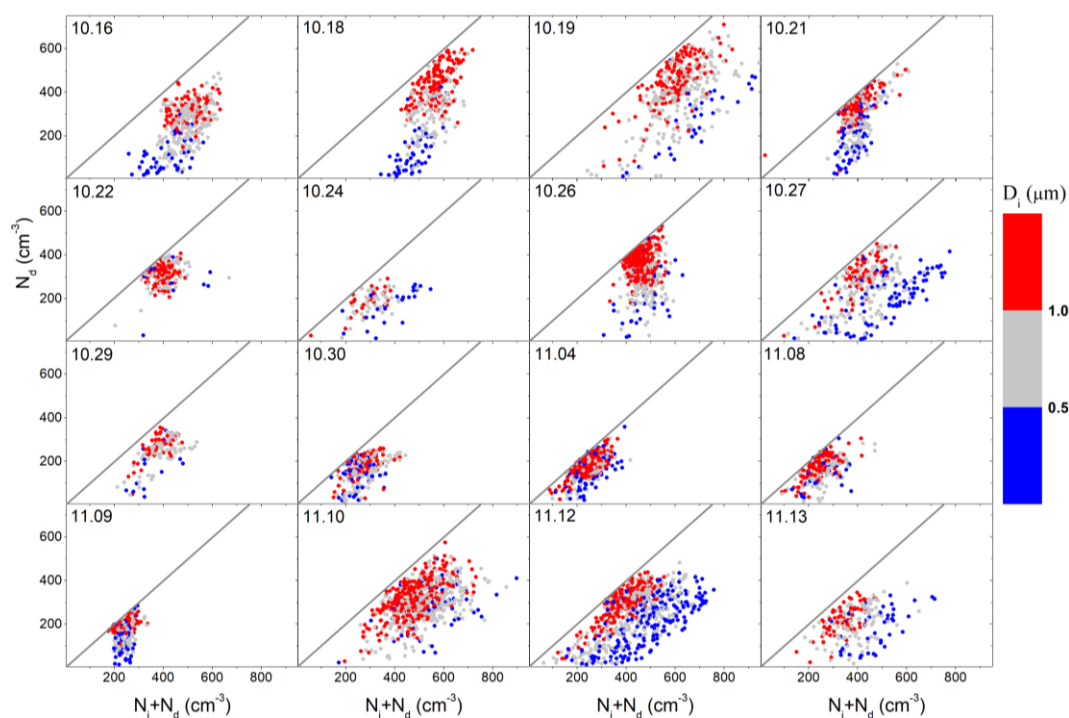


Figure R3. Relationships between N_d and $N_i + N_d$ during all 16 non-drizzling flights when RH is larger than 97%. The colors represent the effective diameter of interstitial aerosol (D_i) (μm), and gray line is 1:1 line.

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.

As seen 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.

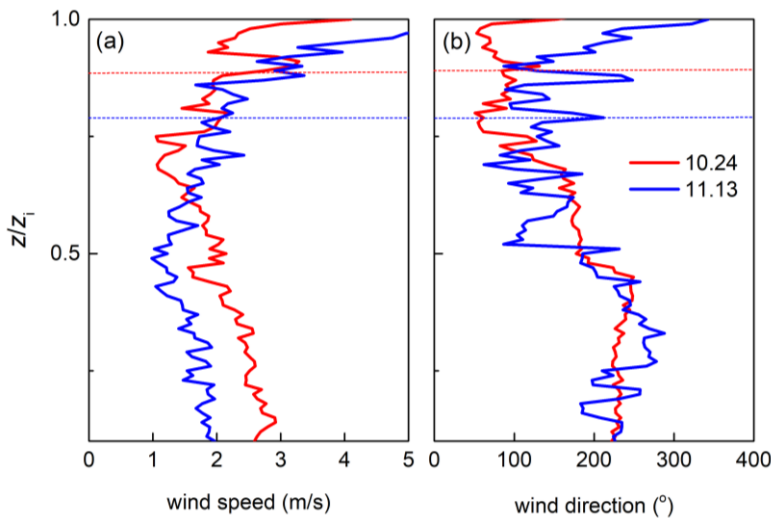


Figure R4. Vertical profiles of (a) horizontal wind speed and (b) wind direction during flights on 24th Oct (red) and 13th Nov. Dashed lines indicate the height of the cloud base.

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 AF_{dent} 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 $AF_{\text{ent}}/AF_{\text{non-ent}}$ with P_{LWC} and P_{Nd} , respectively, where $AF_{\text{ent}}/AF_{\text{non-ent}}$ 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 $AF_{\text{ent}}/AF_{\text{non-ent}}$, 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 346-349 in the revised manuscript).

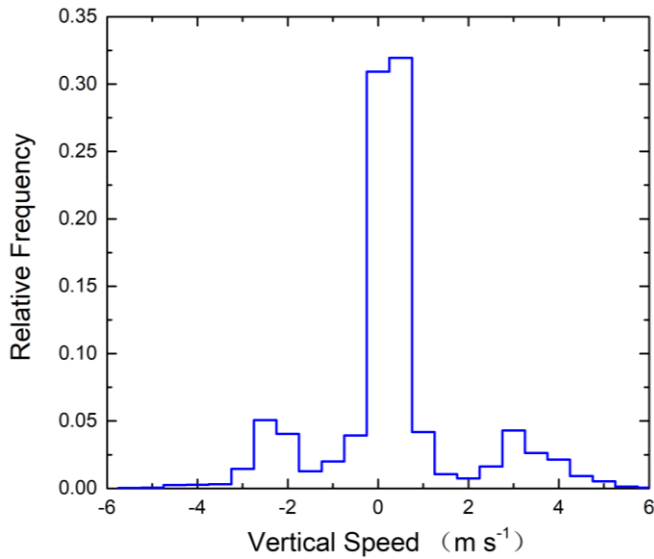


Figure R5. Probability density functions of the vertical speed of the CIRPAS Twin Otter aircraft during the flight on 18th Oct.

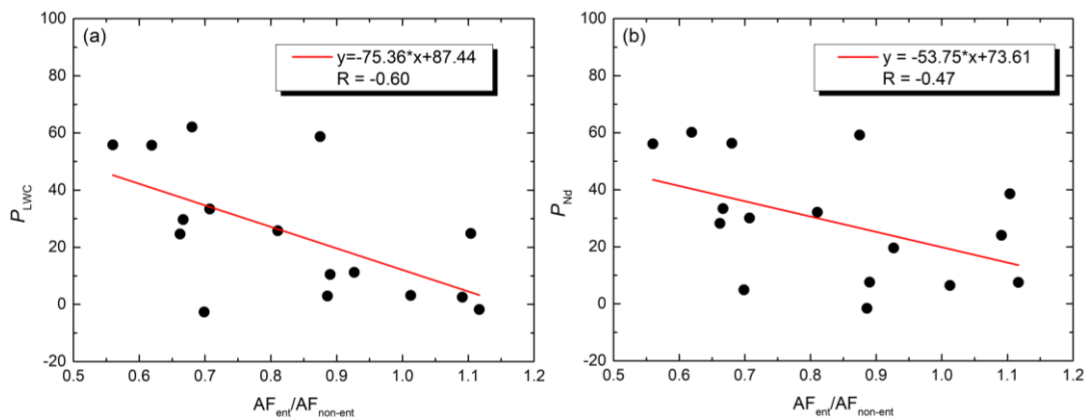


Figure R6. (a) P_{LWC} and (b) P_{Nd} as a function of $AF_{ent}/AF_{non-ent}$ for all 16 non-drizzling flights.

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 D_i .

Line 288 onwards - drier air would also cause 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 N_d instead of R_e , 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.

References

- Allen, G., Coe, H., Clarke, A., Bretherton, C., Wood, R., Abel, S. J., Barrett, P., Brown, P., George, R., Freitag, S., McNaughton, C., Howell, S., Shank, L., Kapustin, V., Brekhovskikh, V., Kleinman, L., Lee, Y.-N., Springston, S., Toniazzi, T., Krejci, R., Fochesatto, J., Shaw, G., Krecl, P., Brooks, B., McMeeking, G., Bower, K. N., Williams, P. I., Crosier, J., Crawford, I., Connolly, P., Allan, J. D., Covert, D., Bandy, A. R., Russell, L. M., Trembath, J., Bart, M., McQuaid, J. B., Wang, J., and Chand, D.: South East Pacific atmospheric composition and variability sampled along 20° S during VOCALS-REx, *Atmos. Chem. Phys.*, 11, 5237–5262, doi:10.5194/acp-11-5237-2011, 2011.
- Painemal, D., and Zuidema, P. (2011). Assessment of MODIS cloud effective radius and optical thickness retrievals over the Southeast Pacific with VOCALS-REx in situ measurements. *Journal of Geophysical Research: Atmospheres*, 116(D24).
- Costantino, L. and Bréon, F.-M.: Analysis of aerosol-cloud interaction from multi-sensor satellite observations, *Geophys. Res. Lett.*, 37, L11801, 2010.
- Ghate, V. P., Albrecht, B. A., and Kollias, P.: Vertical velocity structure of nonprecipitating continental boundary layer stratocumulus clouds, *J. Geophys. Res.*, 115, D13204, doi:10.1029/2009JD013091, 2010.
- Keil, A. and Haywood, J. M.: Solar radiative forcing by biomass burning aerosol particles during safari 2000: a case study based on measured aerosol and cloud properties, *J. Geophys. Res.*, 108(D13), 2003.
- Malinowski, S. P., Gerber, H., Jen-La Plante, I., Kopec, M. K., Kumala, W., Nurowska, K., Chuang, P. Y., Khelif, D., and Haman, K. E.: Physics of Stratocumulus Top (POST): turbulent mixing across capping inversion, *Atmos. Chem. Phys.*, 13, 12171-12186, <https://doi.org/10.5194/acp-13-12171->

2013, 2013.

- Painemal, D. and Zuidema, P.: Assessment of MODIS cloud effective radius and optical thickness retrievals over the Southeast Pacific with VOCALS-REx in situ measurements, *J. Geophys. Res.*, 116, D24206, doi:10.1029/2011jd016155, 2011.
- Stevens, B.: Entrainment in stratocumulus-topped mixed layers, *Q. J. Roy. Meteorol. Soc.*, 128, 2663–2690, doi:10.1256/qj.01.202, 2002
- Shank, L. M., Howell, S., Clarke, A. D., Freitag, S., Brekhovskikh, V., Kapustin, V., McNaughton, C., Campos, T., and Wood, R.: Organic matter and non-refractory aerosol over the remote Southeast Pacific: oceanic and combustion sources, *Atmos. Chem. Phys.*, 12, 557–576, doi:10.5194/acp-12-557-2012, 2012
- Twohy, C. H., Anderson, J. R., Toohey, D. W., Andrejczuk, M., Adams, A., Lytle, M., George, R. C., Wood, R., Saide, P., Spak, S., Zuidema, P., and Leon, D.: Impacts of aerosol particles on the microphysical and radiative properties of stratocumulus clouds over the southeast Pacific Ocean, *Atmos. Chem. Phys.*, 13, 2541–2562, 2013.
- Wood, R.: Stratocumulus clouds, *Mon. Weather Rev.*, 140, 2373–2423, doi:10.1175/MWR-D-11-00121.1, 2012.
- Zhang, Q., Quan, J., Tie, X., Huang, M., and Ma, X.: Impact Aerosol Particles on Cloud Formation: Aircraft Measurements in Beijing, China, *Atmos. Environ.*, 45, 665–672, 2011.
- Zheng, Y., Rosenfeld, D., and Li, Z.: Estimating the decoupling degree of subtropical marine stratocumulus decks from satellite, *Geophysical Research Letters*, 45(22), 12-560, 2018.

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 reexamination 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 (N_a). However, as shown in Table 1, the relationship between ϵ and N_a 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 interstitial aerosol. First, the PCASP-100 misses the large fraction of sub-0.1- μm aerosols that are unlikely to act as CCN and would therefore remain as interstitial 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- μm 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 ($> 0.1 \mu\text{m}$) measured by PCASP-100 is less than the concentration of all un-activated aerosols, i.e., the interstitial aerosols with diameters smaller than $0.1 \mu\text{m}$ 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 ($\sim 50 \text{ nm}$), 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 $0.1\text{-}0.15 \mu\text{m}$ (Kleinman et al., 2012). Therefore, treating aerosols larger than $0.1 \mu\text{m}$ 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- $0.1\text{-}\mu\text{m}$ aerosol concentration to the super- $0.1\text{-}\mu\text{m}$ aerosol concentration during flights on Oct. 18, where sub- $0.1\text{-}\mu\text{m}$ aerosol concentration is derived from the concentration measured by CPC (size range: $> 15 \text{ nm}$) minus that measured by PCASP-100 (size range: $0.1\text{-}2.0 \mu\text{m}$), and the super- $0.1\text{-}\mu\text{m}$ 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 $0.1 \mu\text{m}$ 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 interstitial aerosol (D_i) and cloud droplet sizes (D_e) 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.

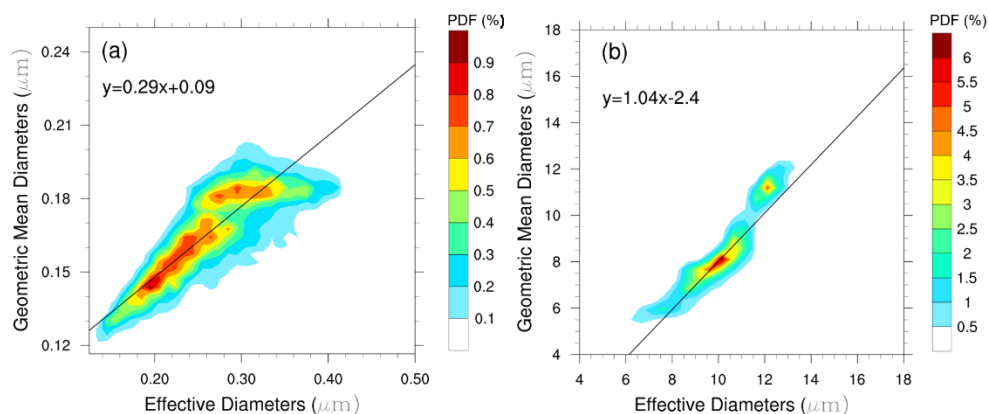


Figure R1. Correlation between geometric mean diameters and effective diameters of (a) aerosols and (b) cloud droplets, respectively. Colors represent the frequency (units: %).

- 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 example, 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 N_d . 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 (N_d) and size (R_e) of cloud droplets, i.e., both N_d and R_e contribute to the change in LWC. The objective of the analysis on the relationships between LWC and N_d/R_e (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 N_d with LWC, which promotes the rapid increase in R_e with LWC. In contrast, there are enough particles that may potentially be activated into cloud droplets under polluted conditions; thus, N_d increases rapidly with LWC, while the increase in R_e 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.

References

- Andrejczuk, M., Grabowski, W. W., Malinowski, S. P., and Smolarkiewicz, P. K.: Numerical simulation of cloud-clear air interfacial mixing: Homogeneous versus inhomogeneous mixing, *J. Atmos. Sci.*, 66, 2493–2500, 2009
- Andersen, H. and Cermak, J.: How thermodynamic environments control stratocumulus microphysics and interactions with aerosols, *Environ. Res. Lett.*, 10, 24004, 2015.
- Chen, Y.-C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H.: Satellite-based estimate of global aerosol-cloud radiative forcing by marine warm clouds, *Nat. Geosci.*, 7, 643–646, 2014.
- Chen, Y.-C., Christensen, M. W., Diner, D. J., and Garay, M. J.: Aerosol-cloud interactions in ship tracks using Terra/MODIS/MISR, *J. Geophys. Res.-Atmos.*, 120, 2819–2833, 2015.
- Chosson, F., Brenguier, J.-L., and Schuller, L.: Entrainment-mixing and radiative transfer simulation in boundary layer clouds, *J. Atmos. Sci.*, 64, 2670–2682, 2007
- Clarke, A. D., Uehara, T., and Porter, J. N.: Atmospheric nuclei and related aerosol fields over the Atlantic: Clean subsiding air and continental pollution during ASTEX, *J. Geophys. Res.*, 102, 25281–25292, 1997.
- Gerber, H., Frick, G., Jensen, J. B., and Hudson, J. G.: Entrainment, mixing and microphysics in trade-wind cumulus, *J. Meteorol. Soc. Jpn.*, 86A, 87–106, 2008.
- Gerber, H., Frick, G., Malinowski, S. P., Jonsson, H., Khelif, D., and Krueger, S. K.: Entrainment rates and microphysics in POST stratocumulus, *J. Geophys. Res.-Atmos.*, 118, 12094–12109, doi:10.1002/jgrd.50878, 2013.
- Gerber, H., Malinowski, S. P., and Jonsson, H.: Evaporative and radiative cooling in POST stratocumulus, *J. Atmos. Sci.*, 73, 3877–3884, <https://doi.org/10.1175/Jas-D-16-0023.1>, 2016.

- Grabowski, W. W.: Indirect impact of atmospheric aerosols in idealized simulations of convective-radiative equilibrium, *J. Climate*, 19, 4664–4682, 2006.
- Hallar, A. G., Strawa, A. W., Schmid, B., Andrews, E., Ogren, J., Sheridan, P., Ferrare, R., Covert, D., Elleman, R., Jonsson, H., Bokarius, K., and Luu, A.: Atmospheric Radiation Measurements Aerosol Intensive Operating Period: Comparison of aerosol scattering during coordinated flights, *J. Geophys. Res.*, 111, D05S09, doi:10.1029/2005JD006250, 2006.
- Hudson, J. G. and Frisbie, P. R.: Cloud condensation nuclei near marine stratus, *J. Geophys. Res.*, 96, 20795–20808, 1991.
- Kleinman, L. I., Daum, P. H., Lee, Y.-N., Lewis, E. R., Sedlacek III, A. J., Senum, G. I., Springston, S. R., Wang, J., Hubbe, J., Jayne, J., Min, Q., Yum, S. S., and Allen, G.: Aerosol concentration and size distribution measured below, in, and above cloud from the DOE G-1 during VOCALS-REx, *Atmos. Chem. Phys.*, 12, 207–223, 2012
- Lehmann, K., Siebert, H., and Shaw, R. A.: Homogeneous and Inhomogeneous Mixing in Cumulus Clouds: Dependence on Local Turbulence Structure, *J. Atmos. Sci.* 66, 3641–3659, 2009.
- Liu, Y. G. and Daum, P. H.: Anthropogenic aerosols – Indirect warming effect from dispersion forcing, *Nature*, 419, 580–581, 2002.
- Peng, Y., Lohmann, U., Leaitch, R., Banic, C., and Couture, M.: The cloud albedo-cloud droplet effective radius relationship for clean and polluted clouds from RACE and FIRE.ACE, *J. Geophys. Res.*, 107, doi:10.029/2000JD000 281, 2002.
- Pontikis, C. A., and Hicks, E. M.: The influence of clear air entrainment on the droplet effective radius of warm maritime convective clouds, *J. Atmos. Sci.*, 50, 2889–2900, 1993.
- Slawinska, J., W. W. Grabowski, H. Pawlowska, and A. A. Wyszogrodzki (2008), Optical properties of shallow convective clouds diagnosed from a bulk-microphysics large-eddy simulation, *J. Clim.*, 21(7), 1639–1647.
- Snider, J. R. and Petters, M. D., Optical particle counter measurement of marine aerosol hygroscopic growth, *Atmos. Chem. Phys.*, 8, 1949–1962, doi:10.5194/acp-8-1949-2008, 2008
- Strapp, J. W., Leaitch, W. R., and Liu, P. S. K.: Hydrated and dried aerosol-size distribution measurements from the Particle Measuring Systems FSSP-300 Probe and the deiced PCASP-100XProbe, *J. Atmos. Oceanic Technol.*, 9, 548–555, 1992.
- Vogelmann, A. M., McFarquhar, G. M., Ogren, J. A., Turner, D. D., Comstock, J. M., Feingold, G., Long, C. N., Jonsson, H. H., Bucholtz, A., Collins, D. R., Diskin, G. S., Gerber, H., Lawson, R. P., Woods, R. K., Andrews, E., Yang, H. J., Chiu, J. C., Hartsock, D., Hubbe, J. M., Lo, C. M., Marshak, A., Monroe, J. W., McFarlane, S. A., Schmid, B., Tomlinson, J. M., and Toto, T.: Racoro

extended-term aircraft observations of boundary layer clouds, *B. Am. Meteorol. Soc.*, 93, 861–878, doi:10.1175/Bams-D-11-00189.1, 2012.

Weber, R. J., Clarke, A. D., Litchy, M., Li, J., Kok, G., Schillawski, R. D., and McMurry, P. H.: Spurious aerosol measurements when sampling from aircraft in the vicinity of clouds, *J. Geophys. Res.*, 103, 28337–28346, 1998.

Yang, Y., Zhao, C., Dong, X., Fan, G., Zhou, Y., Wang, Y., and Yan, F.: Toward understanding the process-level impacts of aerosols on microphysical properties of shallow cumulus cloud using aircraft observations, *Atmospheric Research*, 2019.

Yum, S. S., Wang, J., Liu, Y., Senum, G., Springston, S., McGraw, R., and Yeom, J. M.: Cloud microphysical relationships and their implication on entrainment and mixing mechanism for the stratocumulus clouds measured during the VOCALS project, *J. Geophys. Res.-Atmos.*, 120, 5047–5069, doi:10.1002/2014JD022802, 2015

Zhang, Q., Quan, J., Tie, X., Huang, M., and Ma, X.: Impact Aerosol Particles on Cloud Formation: Aircraft Measurements in Beijing, China, *Atmos. Environ.*, 45, 665–672, 2011.

Zhao, C. F., Qiu, Y. M., Dong, X. B., Wang, Z. E., Peng, Y. R., Li, B. D., Wu, Z. H., and Wang, Y.: Negative Aerosol-Cloud $r(e)$ Relationship From Aircraft Observations Over Hebei, China, *Earth Space Sci.*, 5, 19–29, <https://doi.org/10.1002/2017ea000346>, 2018.

Zhao, C., Zhao, L., & Dong, X.: A Case Study of Stratus Cloud Properties Using In Situ Aircraft Observations over Huanghua, China, *Atmosphere*, 10(1), 19, 2019.

Exploring aerosol cloud interaction using VOCALS-REx aircraft measurements

Hailing Jia¹, Xiaoyan Ma¹ and Yangang Liu²

¹Key Laboratory of Meteorological Disaster, Ministry of Education (KLME)/Joint International Research Laboratory of Climate and Environment Change (ILCEC)/Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters (CIC-FEMD)/Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science & Technology, Nanjing 210044, China

²Environmental and Climate Sciences Department, Brookhaven National Laboratory, Upton, NY, USA

Correspondence to: Xiaoyan Ma (xma@nuist.edu.cn)

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 s 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 tThe 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 s less cloud droplets s 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.

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~~ plays a key role in the radiative energy budget of the Earth by reflecting incoming shortwave radiation and thus cools the ~~planet~~ surface ~~of the planet~~ and offsets the warming by greenhouse gases (Hartmann et al., 1992).

35 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. Huneus 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 ~~at~~ of northern Chile and southern Peru to the remote ocean in the SE-Pacifie 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 this~~ ~~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).

65 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
70 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
75 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
80 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
85 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)~~
90 ~~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

95 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
100 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~~The 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
105 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
110 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
115 measurements close to Point Alpha (20° S, 72° W) are analysed. ~~Only t~~The ~~occurrence of clouds is defined by the following criterion, i.e., with~~ $LWC > 0.05 \text{ g m}^{-3}$ and $N_d > 15 \text{ cm}^{-3}$ ~~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
120 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:

$$\overline{DRa} = \sum n_i \overline{dr}_i^3 / \sum n_i \overline{dr}_i^2 \quad (1)$$

where n_i is the aerosol number concentration in the i th bin of PCASP, and \overline{dr}_i represents the arithmetic mean diameter-radius of i th 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 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 sealed– 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). This normalization could minimize 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 sharply with the height within the BL, which is close to the dry adiabatic lapse rate. A strong inversion occurred 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 rapidly with the increasing height (Fig. 2b). T and RH reached the minimum and maximum, respectively, when z/z_i was 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 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 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 $0.9 < z/z_i$

155 < 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/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 $z/z_i = 0.9$ and then decreased with further increasing height when $0.9 < z/z_i < 1.0$. 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_{a,R_d}) below cloud is larger than that above cloud, which is probably attributable to the differences in aerosol sources and aerosol properties (e.g., chemical composition) + chemical composition and sources of aerosols (Fig. 2f). The profile of CCN/CN is similar to that of D_{a,R_d} (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_d} and CCN/CN values are also found in polluted case than clean cases.

3.2 Relationships between aerosol and cloud properties

170 ~~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.

180 Figure 3 shows ~~the 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-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-mixed boundary

185 ~~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. 190 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 195 $sub-CCN$ 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 200 significance test ($\alpha = 0.05$). In this study, ~~the~~ above-cloud aerosol number concentration is very low ($129.8 \pm 60.1 \text{ cm}^{-3}$), and the inversion ~~eapped-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).

205 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 $647.78 \pm 60.47 \text{ cm}^{-3}$ and $268.97 \pm 35.67 \text{ cm}^{-3}$, 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 210 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

215 large amount particles, the increase of R_e is 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 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~~ ~~eases~~ 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

225 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 aerosols ~~concentration inside the clouds~~. Interstitial aerosols are particles observed ~~inside~~ -clouds that either ~~have~~ never activated ~~into~~ cloud droplets or have been ~~de~~activated ~~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 $583.7 \pm 55.4 \text{ cm}^{-3}$ and $567.4 \pm 59.1 \text{ cm}^{-3}$, 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 aerosols ~~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.

245 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 colors represents ~~the~~ in-cloud vertical velocity ~~ies~~. ~~All flights exhibited p~~ 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 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 aerosols 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 ($0.45 \mu\text{m}$) was much smaller than that of the latter ($0.8 \mu\text{m}$). 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 1.00.5 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_d). However, the ~~dependence of relationship between ε and on N_d still is much less studied and~~ remains ~~even more large uncertain uncertainty than that of N_e~~ . Table 1 ~~summarize shows 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~~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 (Pawłowska 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 ~~we 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~~ ~~increased~~ 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~~ ~~is~~), 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 aerosols number concentrations and updraft velocity together. Therefore, ~~a~~ similar statistical analysis ~~isare~~ 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 ~~approximatelyabout~~ 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 to~~ explore ~~the~~ entrainment in stratocumulus during VOCALS-REx, we ~~firstly~~ compared 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 ($AF_{non-ent}$), 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 ~~diametersradius~~ 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~~ 1.82 % (Table 2). However, N_d and LWC decrease significantly ~~on average~~ by 28.929 % and 24.825 %, 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 56.7 %, 62.1 %, 56.8 % and 58.79 %, 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_{N_d} 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_{N_d} are the reduction percentages in LWC and N_d within the entrainment zone relative to the non-entrainment zone. Although it is still unclear whether the entrainment mixing mechanism is predominantly homogeneous, inhomogeneous, or in between (Andrejezuk et al., 2009; Lehmann et al., 2009), some previous studies showed that stratocumulus is, in general, dominated by the inhomogeneous (Pawłowska 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 (Pawłowska 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 $R_i < 0.38 \mu\text{m}$, but the opposite is true when $R_i > 0.5 \mu\text{m}$ (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 $D_i < 0.75 \mu\text{m}$, but the opposite is true when $D_i > 1.1 \mu\text{m}$. 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 $0.18 \mu\text{m}$ 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 ($129.8 \pm 60.1 \text{ cm}^{-3}$) 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 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 dynamical 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 ~~298.9~~ % and ~~254.8~~ %, 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

430 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 droplets 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.col.ucar.edu/master_list/?project=VOCALS.

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

Acknowledgements. We are grateful for the dedicated efforts of several support staff and scientists in making the observations from the CIRPAS Twin Otter during VOCALS-REx. We also thank Prof. Bruce Albrecht in University of Miami for kindly providing the aerosol, cloud and meteorological variables observations, which are the basis of this manuscript. This study is supported by the National Natural Science Foundation of China grants (41475005 and 41675004).

References

- Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245, 1227–1230, 1989.
- Allen, G., Coe, H., Clarke, A. D., Bretherton, C., Wood, R., Abel, S. J., Barrett, P., Brown, P., George, R., Freitag, S.,

- McNaughton, C., Howell, S., Shank, L., Kapustin, V., Brekhovskikh, V., Kleinman, L., Lee, Y.-N., Springston, S.,
460 Toniazzo, T., Krejci, R., Fochesatto, J., Shaw, G., Krecl, P., Brooks, B., McMeeking, G., Bower, K. N., Williams, P. I.,
Crosier, J., Crawford, I., Connolly, P., Allan, J. D., Covert, D., Bandy, A. R., Russell, L. M., Trembath, J., Bart, M.,
McQuaid, J. B., Wang, J., and Chand, D.: South East Pacific atmospheric composition and variability sampled along 20°
S during VOCALS-REx, *Atmos. Chem. Phys.*, 11, 5237–5262, 2011.
- Almeida, G. P., Brito, J., Morales, C. A., Andrade, M. F., and Artaxo, P.: Measured and modelled cloud condensation nuclei
465 (CCN) concentration in São Paulo, Brazil: the importance of aerosol size-resolved chemical composition on CCN
concentration prediction, *Atmos. Chem. Phys.*, 14, 7559–7572, 2014.
- Andersen, H. and Cermak, J.: How thermodynamic environments control stratocumulus microphysics and interactions with
aerosols, *Environ. Res. Lett.*, 10, 24004, 2015.
- Andrejczuk, M., Grabowski, W. W., Malinowski, S. P., and Smolarkiewicz, P. K.: Numerical simulation of cloud-clear air
470 interfacial mixing: Homogeneous versus inhomogeneous mixing, *J. Atmos. Sci.*, 66, 2493–2500, 2009
- ~~Betts, A. K.: Non-precipitating cumulus convection and its parameterization, *Quart. J. R. Met. Soc.*, 99, 178–196, 1973.~~
- Blyth, A. M., Cooper, W. A., and Jensen, J. B.: A Study of the Source of Entrained Air in Montana Cumuli, *J. Atmos. Sci.*, 45,
3944–3964, 1988.
- Bretherton, C. S., Uttal, T., Fairall, C. W., Yuter, S. E., Weller, R. A., Baumgardner, D., Comstock, K., Wood, R., and Raga,
475 G. B.: The Epic 2001 Stratocumulus Study, *B. Am. Meteorol. Soc.*, 85, 967–977, 2004
- Bretherton, C. S., Wood, R., George, R. C., Leon, D., Allen, G., and Zheng, X.: Southeast Pacific stratocumulus clouds,
precipitation and boundary layer structure sampled along 20° S during VOCALS-Rex, *Atmos. Chem. Phys.*, 10, 10639–
10654, 2010.
- Burnet, F. and Brenguier, J. L.: Observational study of the entrainment-mixing process in warm convective clouds, *J. Atmos.*
480 *Sci.*, 64, 1995–2011, 2007.
- Cai, Y., Snider, J. R., & Wechsler, P. (2013). Calibration of the passive cavity aerosol spectrometer probe for airborne
determination of the size distribution. *Atmospheric Measurement Techniques*, 6(9), 2349.
- Carslaw, K. S., Lee, L. A., Reddington, C. L., Pringle, K. J., Rap, A., Forster, P. M., Mann, G. W., Spracklen, D. V.,
Woodhouse, M. T., Regayre, L. A., and Pierce, J. R.: Large contribution of natural aerosols to uncertainty in indirect
485 forcing, *Nature*, 503, 67–71, 2013.
- Chand, D., Hegg, D. A., Wood, R., Shaw, G. E., Wallace, D., and Covert, D. S.: Source attribution of climatically important
aerosol properties measured at Paposo (Chile) during VOCALS, *Atmos. Chem. Phys.*, 10, 10789–10801, 2010.
- ~~Chen, J., Liu, Y., Zhang, M., and Peng, Y.: New understanding and quantification of the regime dependence of aerosol-cloud
interaction for studying aerosol indirect effects, *Geophys. Res. Lett.*, 43, 1780–1787, doi:10.1002/2016GL067683, 2016.~~
- 490 Chen, Y. and Penner, J. E.: Uncertainty analysis for estimates of the first indirect aerosol effect, *Atmos. Chem. Phys.*, 5,
2935–2948, 2005.
- Chen, Y.-C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H.: Satellite-based estimate of global aerosol-cloud
radiative forcing by marine warm clouds, *Nat. Geosci.*, 7, 643–646, 2014.
- Chen, Y.-C., Christensen, M. W., Diner, D. J., and Garay, M. J.: Aerosol-cloud interactions in ship tracks using
495 TerraMODIS/MISR, *J. Geophys. Res.-Atmos.*, 120, 2819–2833, 2015.
- ~~Chosson, F., Brenguier, J.-L., and Schuller, L.: Entrainment-mixing and radiative transfer simulation in boundary layer~~

[clouds, J. Atmos. Sci., 64, 2670–2682, 2007](#)

[Clarke, A. D., Uehara, T., and Porter, J. N.: Atmospheric nuclei and related aerosol fields over the Atlantic: Clean subsiding air and continental pollution during ASTEX, J. Geophys. Res., 102, 25281–25292, 1997.](#)

500 Costantino, L. and Bréon, F.-M.: Analysis of aerosol-cloud interaction from multi-sensor satellite observations, Geophys. Res. Lett., 37, L11801, 2010.

Costantino, L. and Bréon, F.-M.: Aerosol indirect effect on warm clouds over South-East Atlantic, from co-located MODIS and CALIPSO observations, Atmos. Chem. Phys., 13, 69–88, 2013.

505 [de Roode, S. R. and Wang, Q.: Do stratocumulus clouds detrain? FIRE I data revisited, Bound.-Lay. Meteorol., 122, 479–491, doi:10.1007/s10546-006-9113-1, 2007.](#)

[de Rooy, W. C., Bechtold, P., Frohlich, K., Hohenegger, C., Jonker, H., Mironov, D., Siebesma, A. P., Teixeira, J., Yano, J. I.: Entrainment and detrainment in cumulus convection: an overview, Q. J. Roy. Meteorol. Soc., 139, 1–19, 2013.](#)

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., and Andreae, M. O.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, 510 Science, 312, 1375–1378, 2006.

Ervens, B., Feingold, G., and Kreidenweis, S. M.: Influence of water-soluble organic carbon on cloud drop number concentration, J. Geophys. Res., 110(D18), 18 211, 2005.

515 [Fan, J., Yuan, T., Comstock, J. M., Ghan, S., Khain, A., Leung, L. R., Li, Z., Martins, J. V., and Ovchinnikov, M.: Dominant role by vertical wind shear in regulating aerosol effects on deep convective clouds, J. Geophys. Res., 114, D22206, doi:10.1029/2009JD012352, 2009.](#)

~~[Ghan, S. J., Abdul-Razzak, H., Nenes, A., Ming, Y., Liu, X., Ovchinnikov, M., Shipway, B., Mekhidze, N., Xu, J., and Shi, X.: Droplet nucleation: Physically-based parameterizations and comparative evaluation, J. Adv. Model. Earth Syst., 3, M10001, 2011.](#)~~

520 George, R. C. and Wood, R.: Subseasonal variability of low cloud radiative properties over the southeast Pacific Ocean, Atmos. Chem. Phys., 10, 4047–4063, 2010.

Gerber, H., Arends, B. G., and Ackerman, A. S.: A new microphysics sensor for aircraft use, Atmos. Res., 31, 235–252, 1994.

Gerber, H., Frick, G., Malinowski, S. P., Brenguier J.-L., and Burnet, F.: Holes and entrainment in stratocumulus, J. Atmos. Sci., 62, 443–459, 2005.

525 Gerber, H., Frick, G., Jensen, J. B., and Hudson, J. G.: Entrainment, mixing and microphysics in trade-wind cumulus, J. Meteorol. Soc. Jpn., 86A, 87–106, 2008.

[Gerber, H., Malinowski, S. P., and Jonsson, H.: Evaporative and radiative cooling in POST stratocumulus, J. Atmos. Sci., 73, 3877–3884, https://doi.org/10.1175/Jas-D-16-0023.1, 2016.](#)

530 ~~[Ghan, S. J., Abdul-Razzak, H., Nenes, A., Ming, Y., Liu, X., Ovchinnikov, M., Shipway, B., Mekhidze, N., Xu, J., and Shi, X.: Droplet nucleation: Physically-based parameterizations and comparative evaluation, J. Adv. Model. Earth Syst., 3, M10001, 2011.](#)~~

[Ghate, V. P., Albrecht, B. A., and Kollias, P.: Vertical velocity structure of nonprecipitating continental boundary layer stratocumulus clouds, J. Geophys. Res., 115, D13204, doi:10.1029/2009JD013091, 2010.](#)

[Grabowski, W. W.: Indirect impact of atmospheric aerosols in idealized simulations of convective-radiative equilibrium, J. Climate, 19, 4664–4682, 2006.](#)

- 535 Gultepe, I. and Isaac, G.: The relationship between cloud droplet and aerosol number concentrations for climate models, *Int. J. Climatol.*, 16, 941–946, 1996.
- Haman, K. E., Malinowski, S. P., Kurowski, M. J., Gerber, H., and Brenguier, J.-L.: Small scale mixing processes at the top of a marine stratocumulus – A case study, *Q. J. Roy. Meteor. Soc.*, 133, 213–226, 2007.
- Hartmann, D. L., Ockert-Bell, M. E., and Michelsen, M. L.: The effect of cloud type on Earth's energy balance: Global analysis, *J. Climate*, 5, 1281–1304, 1992.
- 540 Hawkins, L. N., Russell, L. M., Covert, D. S., Quinn, P. K., and Bates, T. S.: Carboxylic acids, sulfates, and organosulfates in processed continental organic aerosol over the southeast Pacific Ocean during VOCALS-Rex 2008, *J. Geophys. Res.*, 115, D13201, 2010.
- Hegg, D. A., Covert, D. S., Jonsson, H. H., and Woods, R. K.: A simple relationship between cloud drop number concentration and precursor aerosol concentration for the regions of Earth's large marine stratocumulus decks, *Atmos. Chem. Phys.*, 12, 1229–1238, 2012.
- 545 Hoffmann, F., Raasch, S., and Noh, Y.: Entrainment of aerosols and their activation in a shallow cumulus cloud studied with a coupled LCM–LES approach, *Atmos. Res.*, 156, 43–57, 2015.
- [Hudson, J. G. and Frisbie, P. R.: Cloud condensation nuclei near marine stratus, *J. Geophys. Res.*, 96, 20795–20808, 1991.](#)
- 550 Hudson, J. G., Noble, S., and Jha, V.: Stratus cloud supersaturations, *Geophys. Res. Lett.*, 37, 21813, 2010.
- Hudson, J. G., Noble, S., and Jha, V.: Cloud droplet spectral width relationship to CCN spectra and vertical velocity, *J. Geophys. Res.*, 117, D11211, 2012.
- Huneus, N., Gallardo, L., and Rutllant, J. A.: Offshore transport episodes of anthropogenic sulfur in northern Chile: Potential impact on the stratocumulus cloud deck, *Geophys. Res. Lett.*, 33, L19819, 2006.
- 555 IPCC, *Climate Change: The Scientific Basis: contributions of Working Group I to the Third Assessment Report of the IPCC*, edited by: Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., Dai, X., Maskell, K., and Johnson, C. A., Cambridge University Press, New York, 881 pp., 2001.
- IPCC, *Climate Change: The Physical Science Basis: contribution of Working Group 1 to the Fourth Assessment Report of the IPCC*, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H.
- 560 L., Cambridge University Press, New York, 996 pp., 2007.
- IPCC, *Climate change: The Physical Science Basis: contribution of Working group I to the Fifth Assessment Report of the IPCC*, edited by: Stocker, T. F., Dahe, Q., Plattner, G. K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P. M., Cambridge University Press, Cambridge, 1535 pp., 2013.
- [Jia, H., Ma, X., Quaas, J., Yin, Y., and Qiu, T.: Is Positive Correlation between Cloud Droplet Effective Radius and Aerosol Index over Land Due to Retrieval Artifacts or Real Physical Processes?, *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2019-47>, in review, 2019.](#)
- 565 Keil, A. and Haywood, J. M.: Solar radiative forcing by biomass burning aerosol particles during safari 2000: a case study based on measured aerosol and cloud properties, *J. Geophys. Res.*, 108(D13), 2003.
- Klein, S. A. and Hartmann, D. L.: The Seasonal Cycle of Low Stratiform Clouds, *J. Climate*, 6, 1587–1606, 1993.
- 570 Kleinman, L. I., Daum, P. H., Lee, Y.-N., Lewis, E. R., Sedlacek III, A. J., Senum, G. I., Springston, S. R., Wang, J., Hubbe, J., Jayne, J., Min, Q., Yum, S. S., and Allen, G.: Aerosol concentration and size distribution measured below, in, and above cloud from the DOE G-1 during VOCALS-REx, *Atmos. Chem. Phys.*, 12, 207–223, 2012.

- Koren, I., Kaufman, Y. J., Rosenfeld, D., Remer, L. A., and Rudich, Y.: Aerosol invigoration and restructuring of Atlantic convective clouds, *Geophys. Res. Lett.*, 32, L14828, 2005.
- 575 Koren, I., Feingold, G., and Remer, L. A.: The invigoration of deep convective clouds over the Atlantic: aerosol effect, meteorology or retrieval artifact?, *Atmos. Chem. Phys.*, 10, 8855–8872, 2010.
- Kumar, V. A., Pandithurai, G., Leena, P. P., Dani, K. K., Murugavel, P., Sonbawne, S. M., Patil, R. D., and Maheskumar, R. S.: Investigation of aerosol indirect effects on monsoon clouds using ground-based measurements over a high-altitude site in Western Ghats, *Atmos. Chem. Phys.*, 16(13), 8423–8430, 2016.
- 580 Lance, S., Nenes, A., and Rissman, T. A.: Chemical and dynamical effects on cloud droplet number: Implications for estimates of the aerosol indirect effect, *J. Geophys. Res.*, 109, 2004.
- Lehmann, K., Siebert, H., and Shaw, R. A.: Homogeneous and Inhomogeneous Mixing in Cumulus Clouds: Dependence on Local Turbulence Structure, *J. Atmos. Sci.* 66, 3641–3659, 2009.
- Leck, C. and Svensson, E.: Importance of aerosol composition and mixing state for cloud droplet activation over the Arctic pack ice in summer, *Atmos. Chem. Phys.*, 15, 2545–2568, 2015.
- 585 Lee, Y. N., Springston, S., Jayne, J., Wang, J., Hubbe, J., Senum, G., Kleinman, L., and Daum, P. H.: Chemical composition and sources of coastal marine aerosol particles during the 2008 VOCALS-REx campaign, *Atmos. Chem. Phys.*, 14, 5057–5072, 2014.
- Li, W., Li, P., Sun, G., Zhou, S., Yuan, Q., and Wang, W.: Cloud residues and interstitial aerosols from non-precipitating clouds over an industrial and urban area in northern China, *Atmos. Environ.*, 45, 2488–2495, 2011.
- 590 Liu, Y., Leeuw, G. D., Kerminen, V. M., Zhang, J., Zhou, P., Nie, W., Qi, X., Hong, J., Wang, Y., Ding, A., Guo, H., Krüger, O., Kulmala, M., and Petäjä, T.: Analysis of aerosol effects on warm clouds over the Yangtze River Delta from multi-sensor satellite observations, *Atmos. Chem. Phys.*, 17, 5623–5641, 2017.
- Liu, Y. G. and Daum, P. H.: Anthropogenic aerosols – Indirect warming effect from dispersion forcing, *Nature*, 419, 580–581, 2002.
- 595 [Liu, Y., Daum, P. H., and Yum, S. S.: Analytical expression for the relative dispersion of the cloud droplet size distribution. *Geophys. Res. Lett.*, 33, L02810, doi:10.1029/2005GL024052, 2006.](#)
- Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, *Atmos. Chem. Phys.*, 5, 715–737, 2005.
- 600 Lu, C., Liu, Y., and Niu, S.: Examination of turbulent entrainment-mixing mechanisms using a combined approach, *J. Geophys. Res.*, 116, D20207, 2011.
- Lu, C., Liu, Y., Niu, S., and Vogelmann, A. M.: Observed impacts of vertical velocity on cloud microphysics and implications for aerosol indirect effects, *Geophys. Res. Lett.*, 39, L21808, 2012.
- 605 Lu, M.-L., Conant, W. C., Jonsson, H. H., Varutbangkul, V., Flagan, R. C., and Seinfeld, J. H.: The Marine Stratus/Stratocumulus Experiment (MASE): Aerosol-cloud relationships in marine stratocumulus, *J. Geophys. Res.*, 112, D10209, 2007.
- Lu, M.-L., Sorooshian, A., Jonsson, H. H., Feingold, G., Flagan, R. C., and Seinfeld, J. H.: Marine stratocumulus aerosol-cloud relationships in the MASE-II experiment: Precipitation susceptibility in eastern Pacific marine stratocumulus, *J. Geophys. Res.*, 114, D24203, 2009.
- 610 Ma, J., Chen, Y., Wang, W., Yan, P., Liu, H., Yang, S., Hu, Z., and Lelieveld, J.: Strong air pollution causes widespread haze-clouds over China, *J. Geophys. Res.*, 115, D18204, 2010.

Ma, X., Yu, F., and Quaas, J.: Reassessment of satellite-based estimate of aerosol climate forcing, *J. Geophys. Res.-Atmos.*, 119, 10394–10409, 2014.

Ma, X., Jia, H., Yu, F., and Quaas, J.: Opposite aerosol index-cloud droplet effective radius correlations over major industrial regions and their adjacent oceans, *Geophys. Res. Lett.*, 45, 5771–5778, 2018.

615 [Malinowski, S. P., Gerber, H., Jen-La Plante, I., Kopec, M. K., Kumala, W., Nurowska, K., Chuang, P. Y., Khelif, D., and Haman, K. E.: Physics of Stratocumulus Top \(POST\): turbulent mixing across capping inversion, *Atmos. Chem. Phys.*, 13, 12171-12186, <https://doi.org/10.5194/acp-13-12171-2013>, 2013.](#)

Martin, G. M., Johnson, D. W., and Spice, A.: The measurement and parameterization of effective radius of droplets in warm stratocumulus clouds, *J. Atmos. Sci.*, 51, 1823–1842, 1994.

620 McFiggans, G., Artaxo, P., Baltensperger, U., Coe, H., Facchini, M. C., Feingold, G., Fuzzi, S., Gysel, M., Laaksonen, A., Lohmann, U., Mentel, T. F., Murphy, D. M., O’Dowd, C. D., Snider, J. R., and Weingartner, E.: The effect of physical and chemical aerosol properties on warm cloud droplet activation, *Atmos. Chem. Phys.*, 6, 2593–2649, 2006.

McCoy, D. T., Bender, F. M., Mohrmann, J. K. C., Hartmann, D. L., Wood, R., and Grosvenor, D. P.: The global aerosol-cloud first indirect effect estimated using MODIS, MERRA, and AeroCom, *J. Geophys. Res.-Atmos.*, 122, 1779–1796, 2017.

625 Nenes, A., Charlson, R. J., Facchini, M. C., Kulmala, M., Laaksonen, A., and Seinfeld, J. H.: Can chemical effects on cloud droplet number rival the first indirect effect?, *Geophys. Res. Lett.*, 29, 2002.

[Painemal, D. and Zuidema, P.: Assessment of MODIS cloud effective radius and optical thickness retrievals over the Southeast Pacific with VOCALS-REx in situ measurements, *J. Geophys. Res.*, 116, D24206, doi:10.1029/2011jd016155, 2011.](#)

630 Pandithurai, G., Dipu, S., Prabha, T. V., Mahes Kumar, R. S., Kulkarni, J. R., and Goswami, B. N.: Aerosol effect on droplet spectral dispersion in warm continental cumuli, *J. Geophys. Res.*, 117, 2012.

Pawlowska, H., Brenguier, J. L., and Burnet, F.: Microphysical properties of stratocumulus clouds, *Atmos. Res.*, 55, 15–33, 2000.

635 Pawlowska, H., Grabowski, W. W., and Brenguier, J.-L.: Observations of the width of cloud droplet spectra in stratocumulus, *Geophys. Res. Lett.*, 33, L19810, 2006.

Peng, Y. and Lohmann, U.: Sensitivity study of the spectral dispersion of the cloud droplet size distribution on the indirect aerosol effect, *Geophys. Res. Lett.*, 30, 1507, 2003.

640 Peng, Y., Lohmann, U., Leaitch, R., and Kulmala, M.: An investigation into the aerosol dispersion effect through the activation process in marine stratus clouds, *J. Geophys. Res.*, 112, D11117, 2007.

Pinsky, M. B. and Khain, A. P.: Effects of in-cloud nucleation and turbulence on droplet spectrum formation in cumulus clouds, *Q. J. Roy. Meteor. Soc.*, 128, 501–533, 2002.

[Reutter, P., Trentmann, J., Su, H., Simmel, M., Rose, D., Gunthe, S. S., Wernli, H., Andreae, M. O., and Poschl, U.: Aerosol- and updraft-limited regimes of cloud droplet formation: influence of particle number, size and hygroscopicity on the activation of cloud condensation nuclei \(CCN\), *Atmos. Chem. Phys.*, 9, 7067–7080, 2009, <http://www.atmos-chem-phys.net/9/7067/2009/>.](#)

645 Saponaro, G., Kolmonen, P., Sogacheva, L., Rodriguez, E., Virtanen, T., and de Leeuw, G.: Estimates of the aerosol indirect effect over the Baltic Sea region derived from 12 years of MODIS observations, *Atmos. Chem. Phys.*, 17, 3133–3143,

2017.

- 650 [Slawinska, J., W. W. Grabowski, H. Pawlowska, and A. A. Wyszogrodzki \(2008\), Optical properties of shallow convective clouds diagnosed from a bulk-microphysics large-eddy simulation, *J. Clim.*, 21\(7\), 1639–1647.](#)
- Slawinska, J., Grabowski, W. W., Pawlowska, H., and Morrison, H.: Droplet activation and mixing in large-eddy simulation of a shallow cumulus field, *J. Atmos. Sci.*, 69, 444–462, 2012.
- 655 [Stevens, B.: Entrainment in stratocumulus-topped mixed layers, *Q. J. Roy. Meteorol. Soc.*, 128, 2663–2690, doi:10.1256/qj.01.202.2002](#)
- Su, W., N. G. Loeb, K.-M. Xu, G. L. Schuster, and Z. A. Eitzen: An estimate of aerosol indirect effect from satellite measurements with concurrent meteorological analysis, *J. Geophys. Res.*, 115, D18219, 2010.
- Tang, J., Wang, P., Mickley, L. J., Xia, X., Liao, H., Yue, X., Sun, L., and Xia, J.: Positive relationship between liquid cloud droplet effective radius and aerosol optical depth over Eastern China from satellite data, *Atmos. Environ.*, 84, 244–253, 660 2014.
- Twohy, C. H., Austin, P. H., and Charlson, R. J.: Chemical consequences of the initial diffusional growth of cloud droplets: a clean marine case, *Tellus B*, 41, 51–60, 1989.
- Twohy, C. H., Anderson, J. R., Toohey, D. W., Andrejczuk, M., Adams, A., Lytle, M., George, R. C., Wood, R., Saide, P., Spak, S., Zuidema, P., and Leon, D.: Impacts of aerosol particles on the microphysical and radiative properties of 665 stratocumulus clouds over the southeast Pacific Ocean, *Atmos. Chem. Phys.*, 13, 2541–2562, 2013.
- Twomey, S.: Pollution and Planetary Albedo, *Atmos. Environ.*, 8, 1251–1256, 1974.
- Wang, F., Guo, J., Wu, Y., Zhang, X., Deng, M., Li, X., Zhang, J., and Zhao, J.: Satellite observed aerosol-induced variability in warm cloud properties under different meteorological conditions over eastern China, *Atmos. Environ.*, 84, 122–132, 2014.
- 670 Wang, F., Guo, J., Zhang, J., Huang, J., Min, M., Chen, T., Liu, H., Deng, M., and Li, X.: Multi-sensor quantification of aerosol-induced variability in warm clouds over eastern China, *Atmos. Environ.*, 113, 1–9, 2015.
- [Wang, Q. and Albrecht, B. A.: Observations of cloud-top entrainment in marine stratocumulus clouds, *J. Atmos. Sci.*, 51, 1530–1547, 1994.](#)
- 675 Warner, J.: The water content of cumuliform cloud, *Tellus*, 7, 449–457, 1955.
- Warner, J.: The Microstructure of Cumulus Cloud. Part I. General Features of the Droplet Spectrum, *J. Atmos. Sci.*, 26, 1049–1059, 1969a.
- Warner, J.: The microstructure of cumulus clouds. Part II. The effect on droplet size distribution of cloud nucleus spectrum and updraft velocity, *J. Atmos. Sci.*, 26, 1272–1282, 1969b.
- 680 [Weber, R. J., Clarke, A. D., Litchy, M., Li, J., Kok, G., Schillawski, R. D., and McMurry, P. H.: Spurious aerosol measurements when sampling from aircraft in the vicinity of clouds, *J. Geophys. Res.*, 103, 28337–28346, 1998.](#)
- [Wood, R.: Stratocumulus clouds, *Mon. Weather Rev.*, 140, 2373–2423, doi:10.1175/MWR-D-11-00121.1, 2012.](#)
- Wood, R., Bretherton, C., Huebert, B., Mechoso, C. R., and Weller, R.: VOCALS-SouthEast Pacific Regional Experiment (REx), Scientific program overview, 2006.
- 685 Wood, R., Comstock, K. K., Bretherton, C. S., Cornish, C., Tomlinson, J., Collins, D. R., and Fairall, C.: Open cellular structure in marine stratocumulus sheets, *J. Geophys. Res.*, 113, D12207, 2008.

- Wood, R., Mechoso, C. R., Bretherton, C. S., Weller, R. A., Huebert, B., Straneo, F., Albrecht, B. A., Coe, H., Allen, G., Vaughan, G., Daum, P., Fairall, C., Chand, D., Gallardo Klenner, L., Garreaud, R., Grados, C., Covert, D. S., Bates, T. S., Krejci, R., Russell, L. M., de Szoeko, S., Brewer, A., Yuter, S. E., Springston, S. R., Chaigneau, A., Toniazzi, T., Minnis, P., Palikonda, R., Abel, S. J., Brown, W. O. J., Williams, S., Fochesatto, J., Brioude, J., and Bower, K. N.: The VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx): goals, platforms, and field operations, *Atmos. Chem. Phys.*, 11, 627–654, 2011.
- 690
- Yum, S. S., Wang, J., Liu, Y., Senum, G., Springston, S., McGraw, R., and Yeom, J. M.: Cloud microphysical relationships and their implication on entrainment and mixing mechanism for the stratocumulus clouds measured during the VOCALS project, *J. Geophys. Res.-Atmos.*, 120, 5047–5069, 2015.
- 695
- Zhao, C., Tie, X., Brasseur, G., Noone, K. J., Nakajima, T., Zhang, Q., Zhang, R., Huang, M., Duan, Y., Li, G., and Ishizaka, Y.: Aircraft measurements of cloud droplet spectral dispersion and implications for indirect aerosol radiative forcing, *Geophys. Res. Lett.*, 33, L16809, 2006.
- Zhang, Q., Quan, J., Tie, X., Huang, M., and Ma, X.: Impact Aerosol Particles on Cloud Formation: Aircraft Measurements in Beijing, China, *Atmos. Environ.*, 45, 665–672, 2011.
- 700
- Zheng, X., Albrecht, B. A., Minnis, P., Ayers, K., and Jonson, H. H.: Observed aerosol and liquid water path relationships in marine stratocumulus, *Geophys. Res. Lett.*, 37, L17803, 2010.
- Zheng, X., Albrecht, B., Jonsson, H. H., Khelif, D., Feingold, G., Minnis, P., Ayers, K., Chuang, P., Donaher, S., Rossiter, D., Ghate, V., Ruiz-Plancarte, J., and Sun-Mack, S.: Observations of the boundary layer, cloud, and aerosol variability in the southeast Pacific near-coastal marine stratocumulus during VOCALSREx, *Atmos. Chem. Phys.*, 11, 9943–9959.
- 705

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	Typical	Typical	Other
						Wind shear			Decoupled
w_{ave}^a	<u>0.09</u>	<u>0.08</u>	<u>0.11</u>	<u>0.08</u>	<u>0.08</u>	<u>-0.06</u>	<u>0.06</u>	<u>0.08</u>	<u>-0.13</u>
w_{std}^b	<u>0.42</u>	<u>0.55</u>	<u>0.58</u>	<u>0.51</u>	<u>0.51</u>	<u>0.30</u>	<u>0.56</u>	<u>0.41</u>	<u>0.61</u>
w_{skew}^c	<u>-0.38</u>	<u>-0.16</u>	<u>-0.27</u>	<u>-0.21</u>	<u>-0.27</u>	<u>0.00</u>	<u>-0.23</u>	<u>0.08</u>	<u>-0.27</u>
P_{LWC}^{da}	25.826	55.756	33.4	24.825	24.625	29.3	-2.73	11.2	3.1
P_{Nd}^{db}	32.1	60.1	30.1	38.639	28.2	34.4	4.95	19.620	6.4
P_{RDe}^{fe}	-21.9	-5.76	<u>0.91</u>	-6.77	-1.92	<u>0.01</u>	-4.1	-2.4	-1.82
AF_{ent}^{gd}	0.77	0.52	0.58	0.85	0.49	0.52	0.51	0.76	0.81
$AF_{non-ent}^{he}$	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	Other	Other	Typical	Typical	Typical	Other		
	Wind shear	Wind shear, Decoupled	Decoupled				Wind Shear		
w_{ave}	<u>0.02</u>	<u>0.07</u>	<u>0.02</u>	<u>0.08</u>	<u>0.09</u>	<u>0.09</u>	<u>-0.02</u>		
w_{std}	<u>0.45</u>	<u>0.41</u>	<u>0.42</u>	<u>0.47</u>	<u>0.49</u>	<u>0.51</u>	<u>0.41</u>		
w_{skew}	<u>-0.13</u>	<u>-0.48</u>	<u>-0.03</u>	<u>-0.48</u>	<u>-0.26</u>	<u>-0.27</u>	<u>-0.42</u>		
P_{LWC}	10.5	62.1	32.5	55.856	2.93	-1.82	58.759	254.8	
P_{Nd}	7.68	56.3	24.0	56.1	-21.6	87.5	59.2	28.9	
P_{RDe}	0.2	4.4	-8.4	-2.1	3.4	-32.5	-1.2	-1.82	
AF_{ent}	0.73	0.66	0.84	0.28	0.70	0.67	0.56	0.64	
$AF_{non-ent}$	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.

^{da, db, fe} P_{LWC} , P_{Nd} , and P_{RDe} are the percentages of reduction in LWC , N_d and RDe within entrainment zone relative to non-entrainment zone.(unit: %)

^{gd, he} AF_{ent} and AF_{non-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.

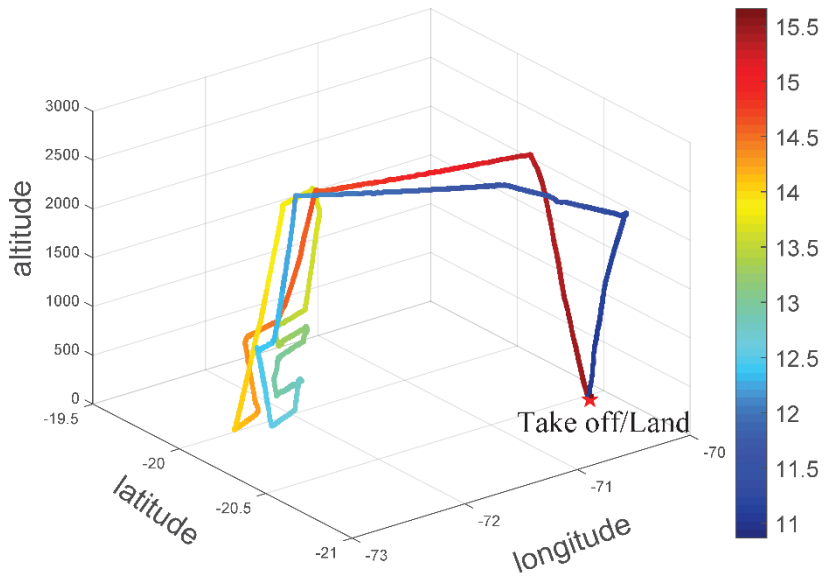


Fig. 1. The flight track in Oct. 18, and the colors represent flight time in hour (UTC).

720

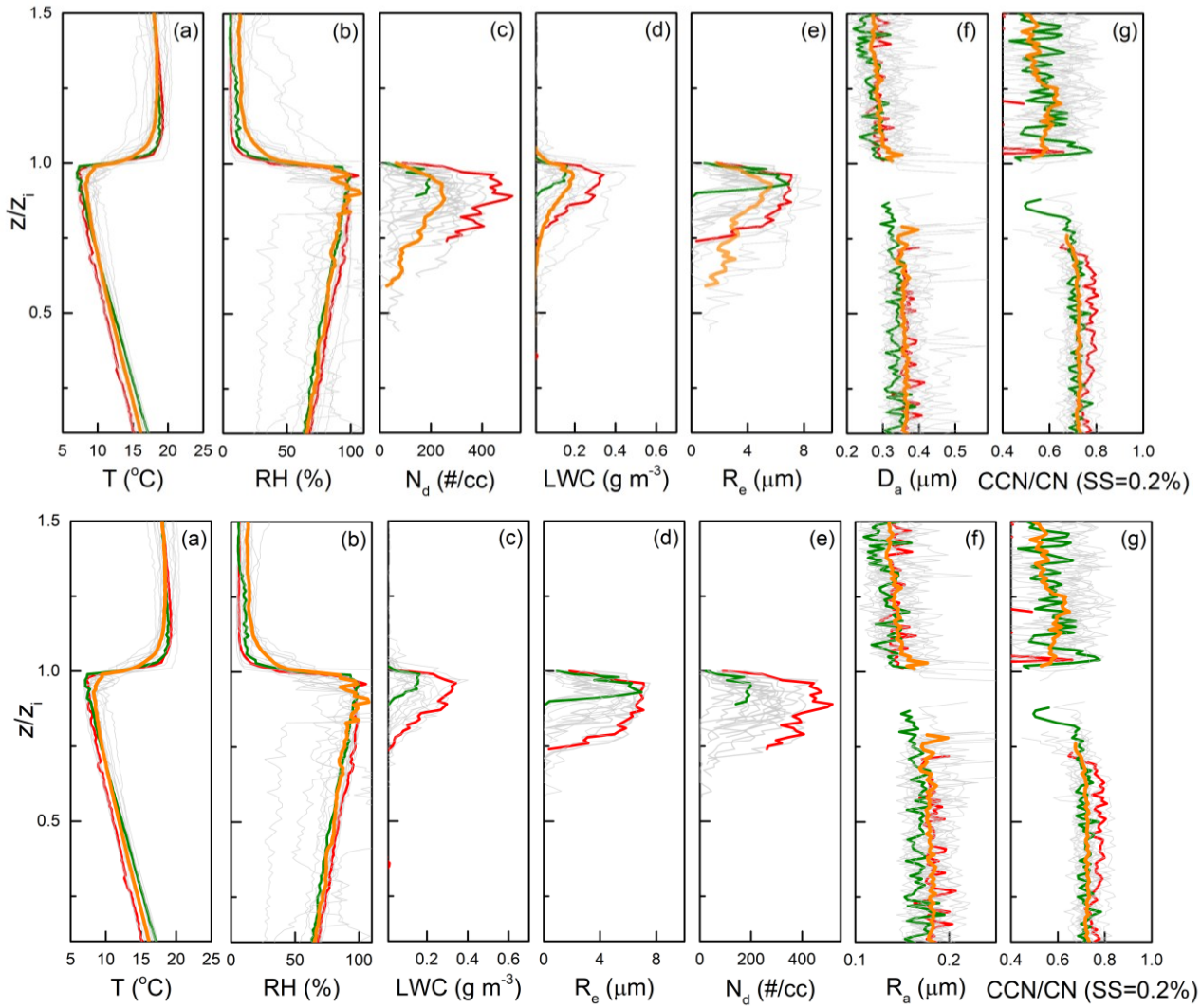


Fig. 2. Vertical profiles scaled by the inversion height. (a) temperature (K); (b) relative humidity (%); (c) liquid water content (g m^{-3}); (d) cloud droplet number concentration (cm^{-3}); (e) cloud droplet effective radius (μm); (f) cloud droplet diameter (μm); (g) CCN/CN (SS=0.2%)

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.

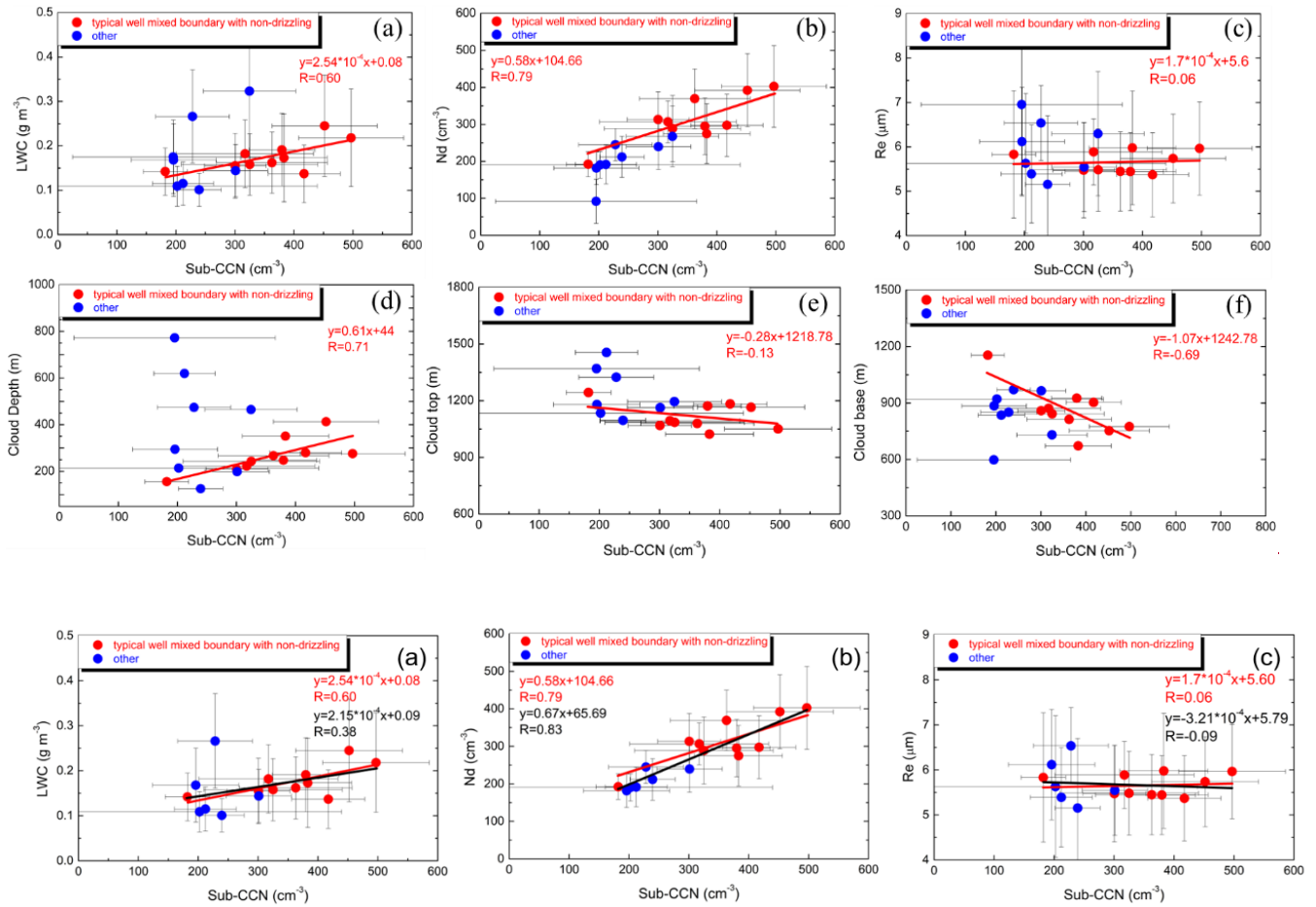
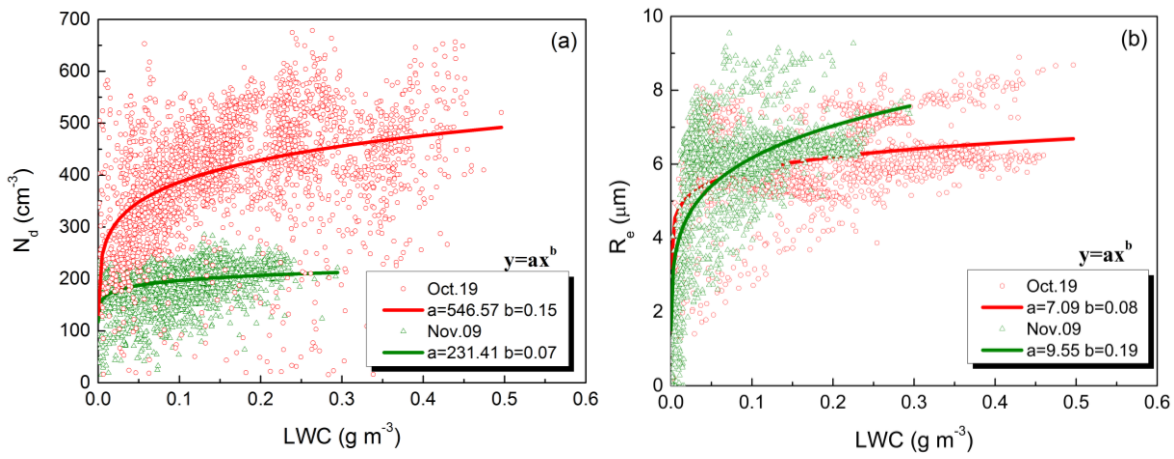
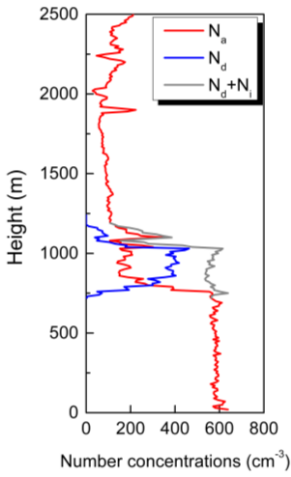


Fig. 3. (a) LWC (g cm^{-3}); (b) N_d (cm^{-3}); (c) R_e (μm); (d) cloud depth (m); (e) cloud top height (m); (f) cloud base height (m) as a function of sub-cloud CCN concentrations ($SS=0.2\%$) for all 16 non-drizzling flights. The error bars through these symbols indicate the standard deviation. Red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



740 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.



745 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.

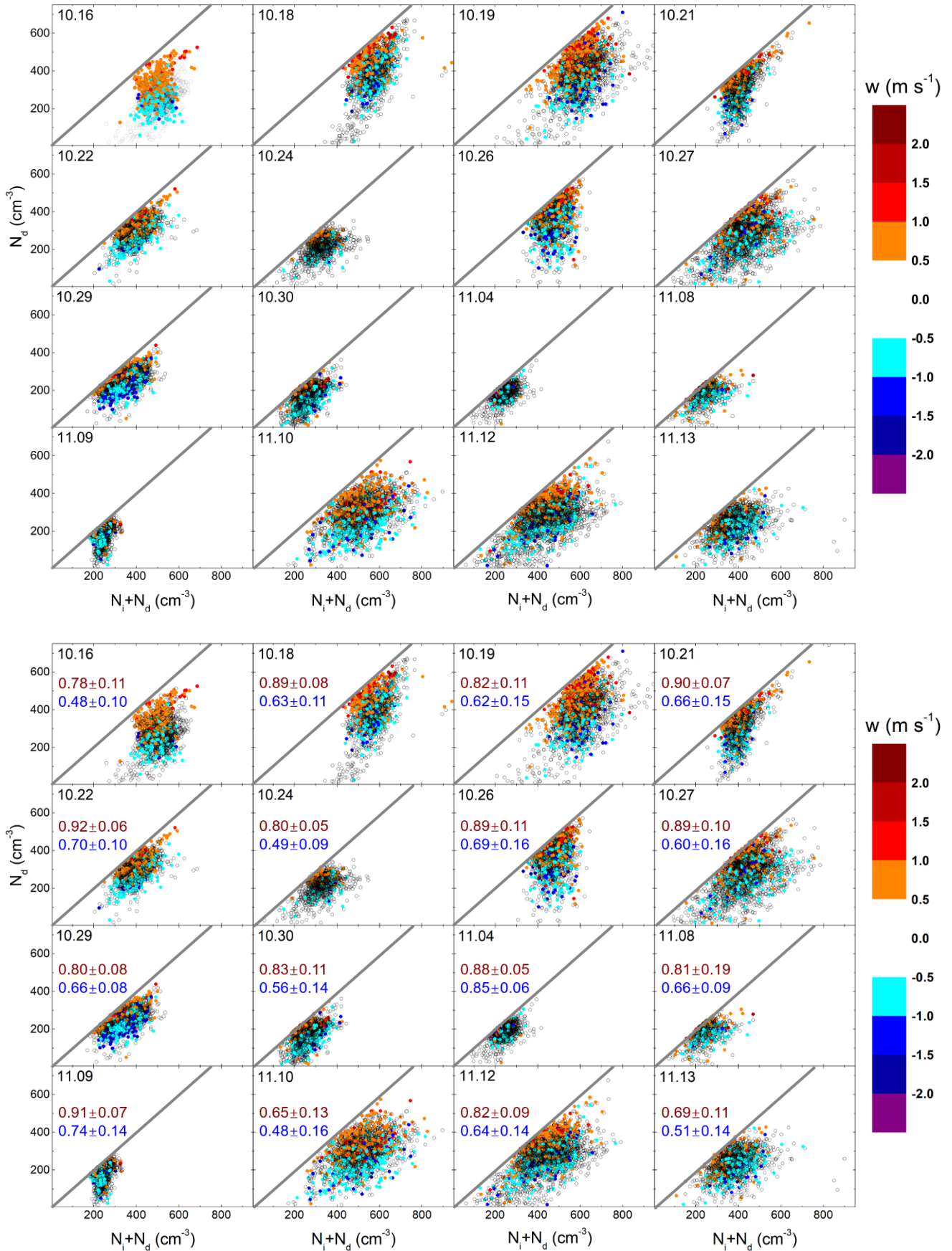
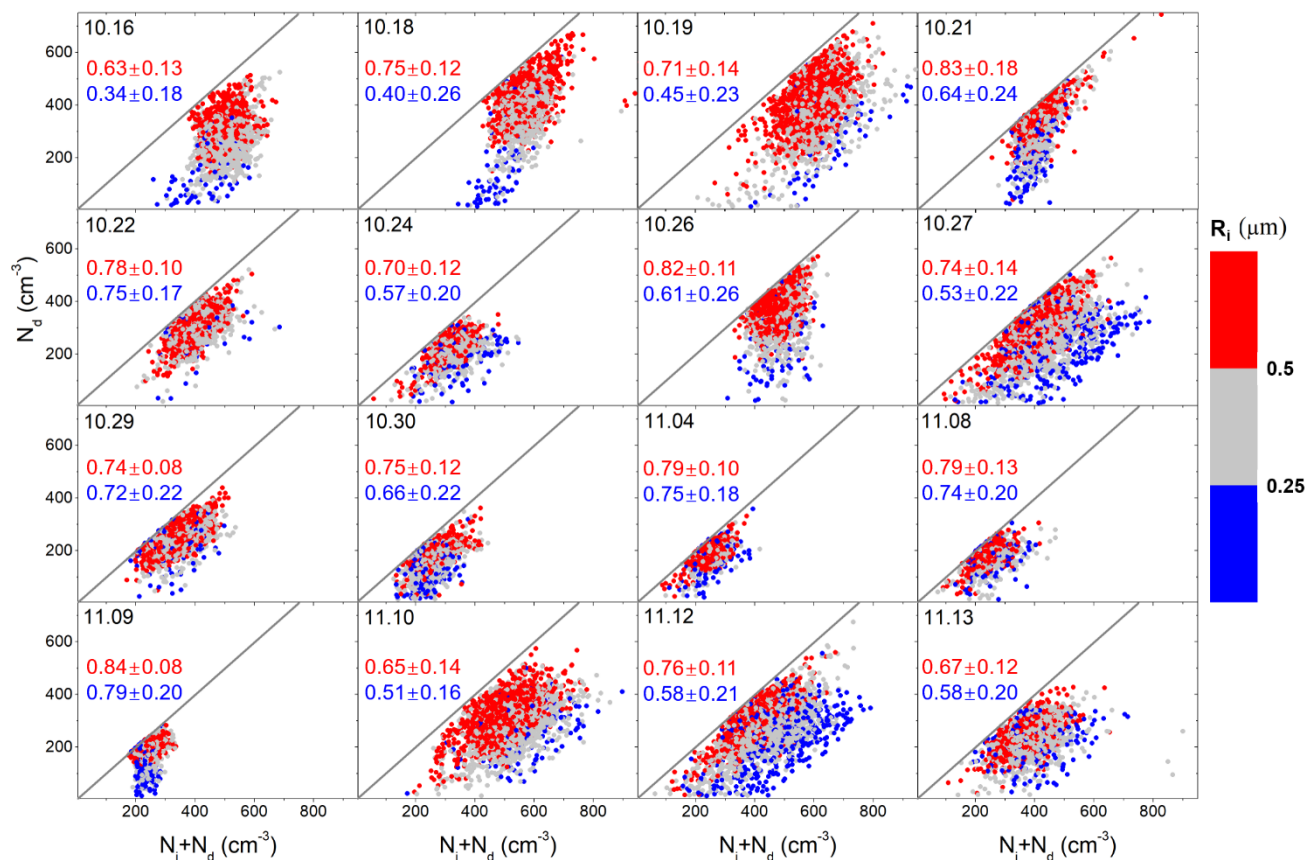
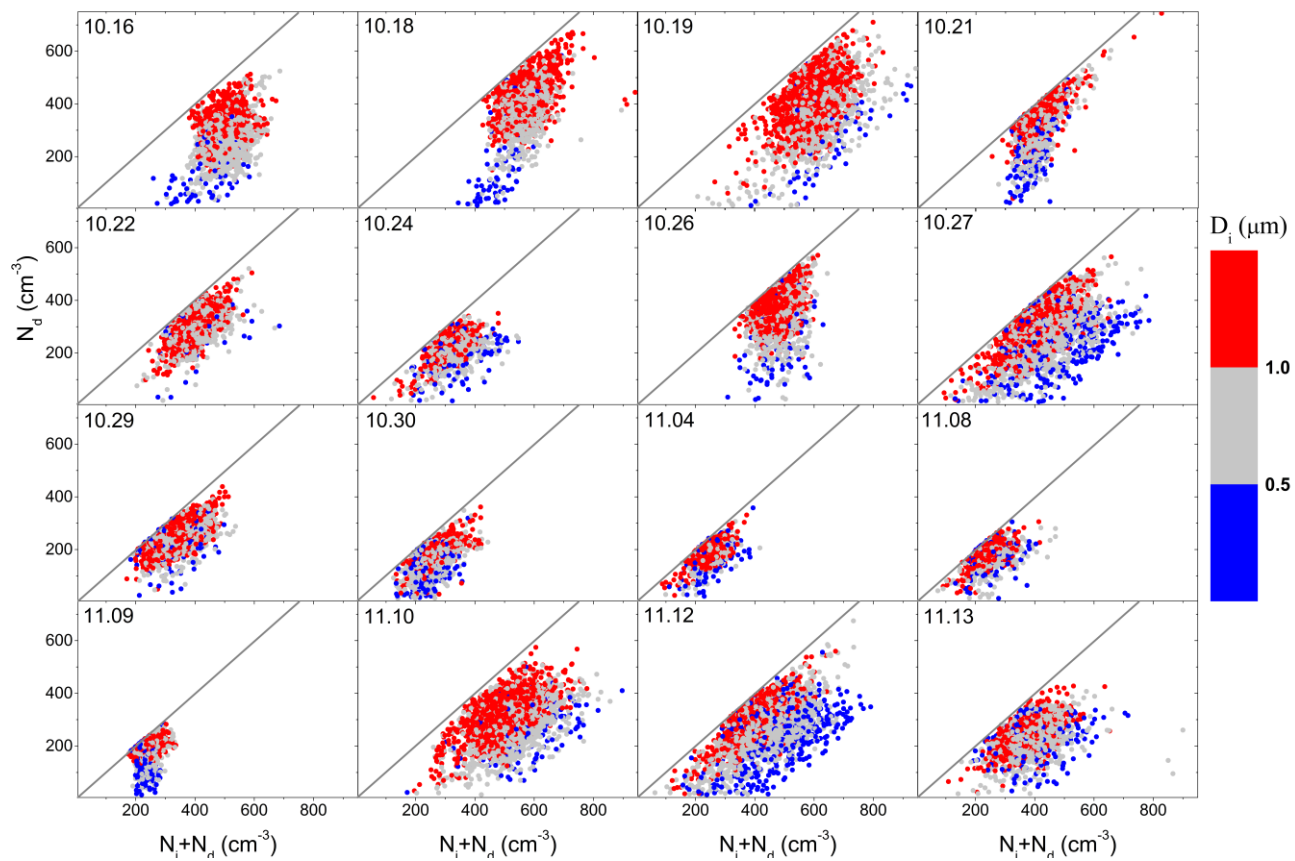


Fig. 6. Relationships between N_d and $N_i + N_d$ during all 16 non-drizzling flights. The colors represents in-cloud vertical velocities (m s^{-1}), and gray line is 1:1 line. The mean and standard deviation of $N_d/(N_d+N_i)$ for vertical velocity greater than 1 m s^{-1} (red) and less

750

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



755

Fig. 7. Same as Fig. 6, but the colors 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 $0.5 \mu\text{m}$ (red) and less than $0.25 \mu\text{m}$ (blue) are shown.

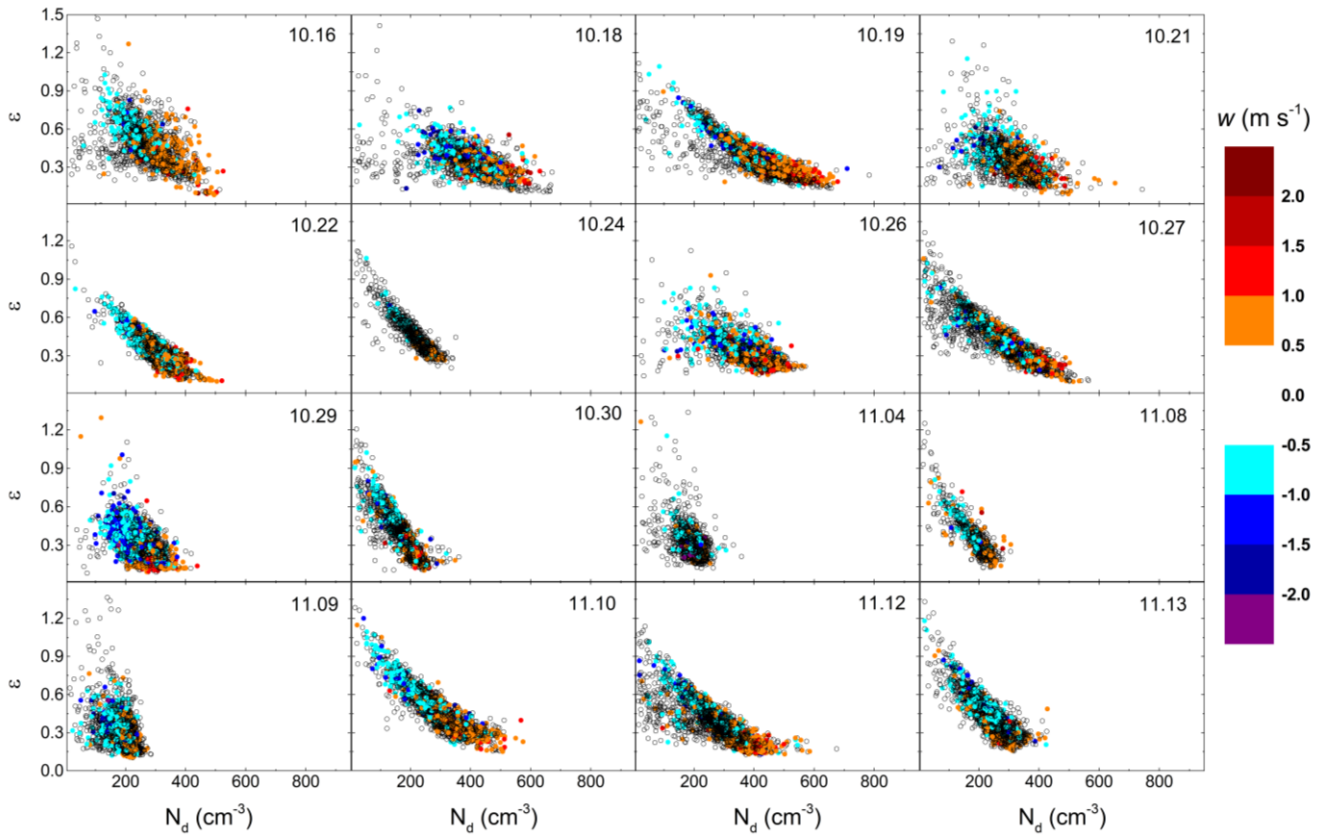
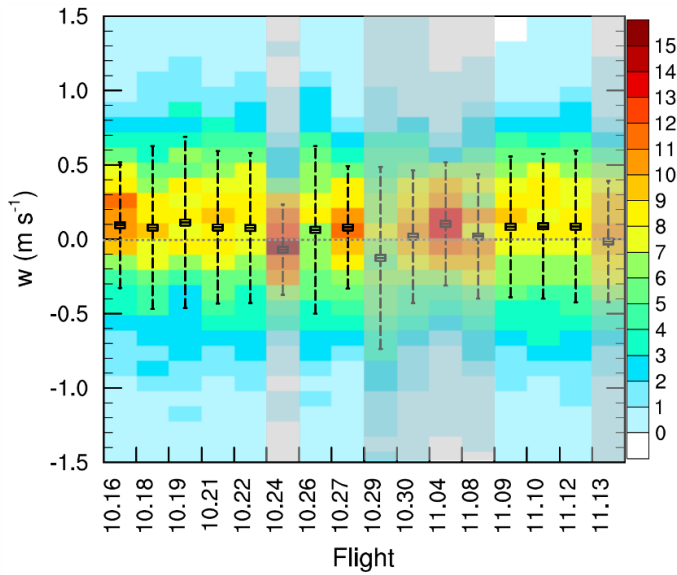


Fig. 8. Relationships between relative dispersion (ϵ) and N_d during all 16 non-drizzling flights, in which the colors **representing** **represents** in-cloud vertical velocities (m s^{-1}).

760



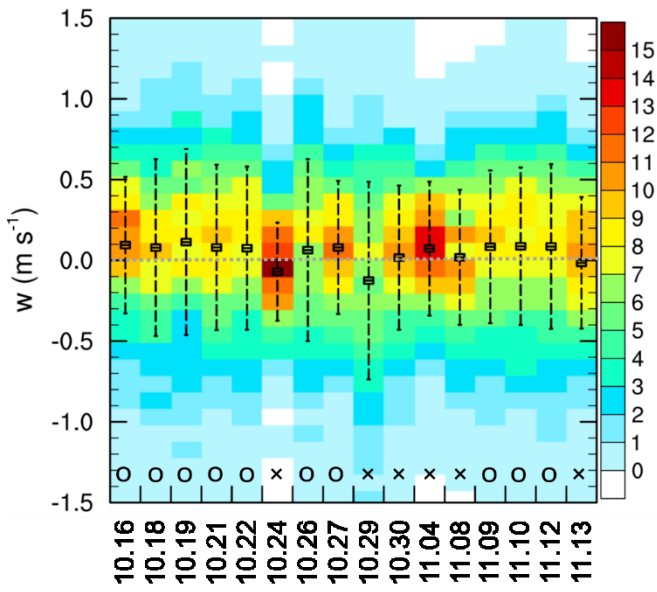


Fig. 9. Probability distribution function (units: %) of vertical velocity (w) for 16 non-drizzling flights. Black symbols are mean values of w , and error bars through these symbols indicate the standard deviation. ~~Gray shadow represents the flights other than typical well-mixed boundary with non-drizzling.~~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

765

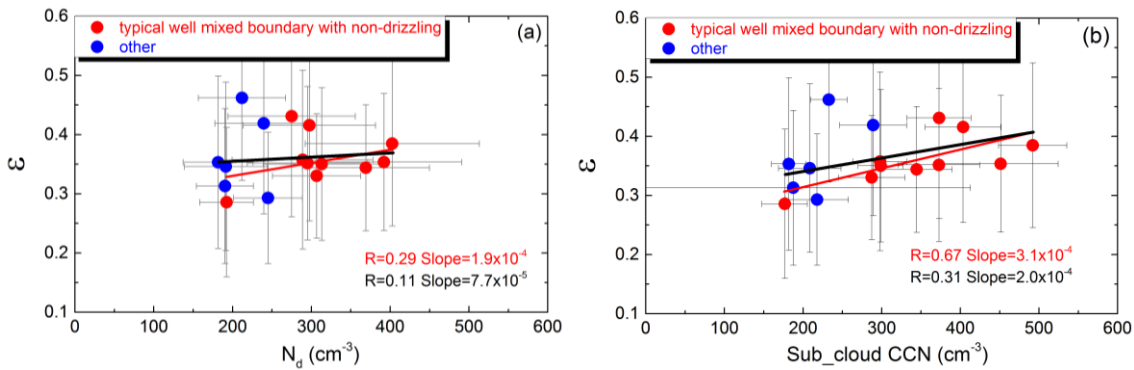


Fig. 10. Relative dispersion (ϵ) as a function of (a) N_d and (b) sub-cloud CCN concentrations ($SS=0.2\%$) for all flights. The error bars through these symbols indicate the standard deviation. Red symbols are the cases with typical well-mixed boundary BL with non-drizzling, and blue symbols for others.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

770

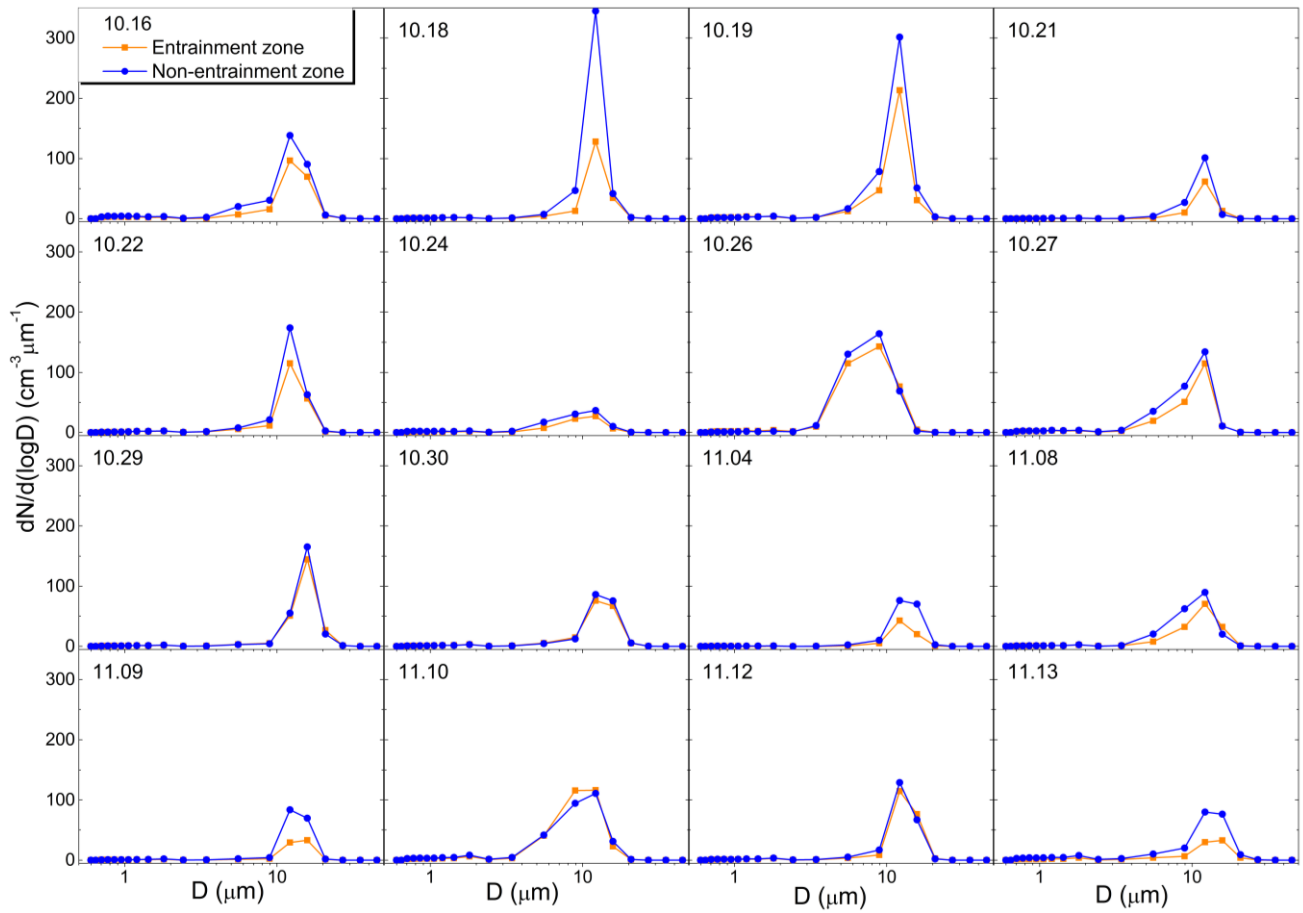
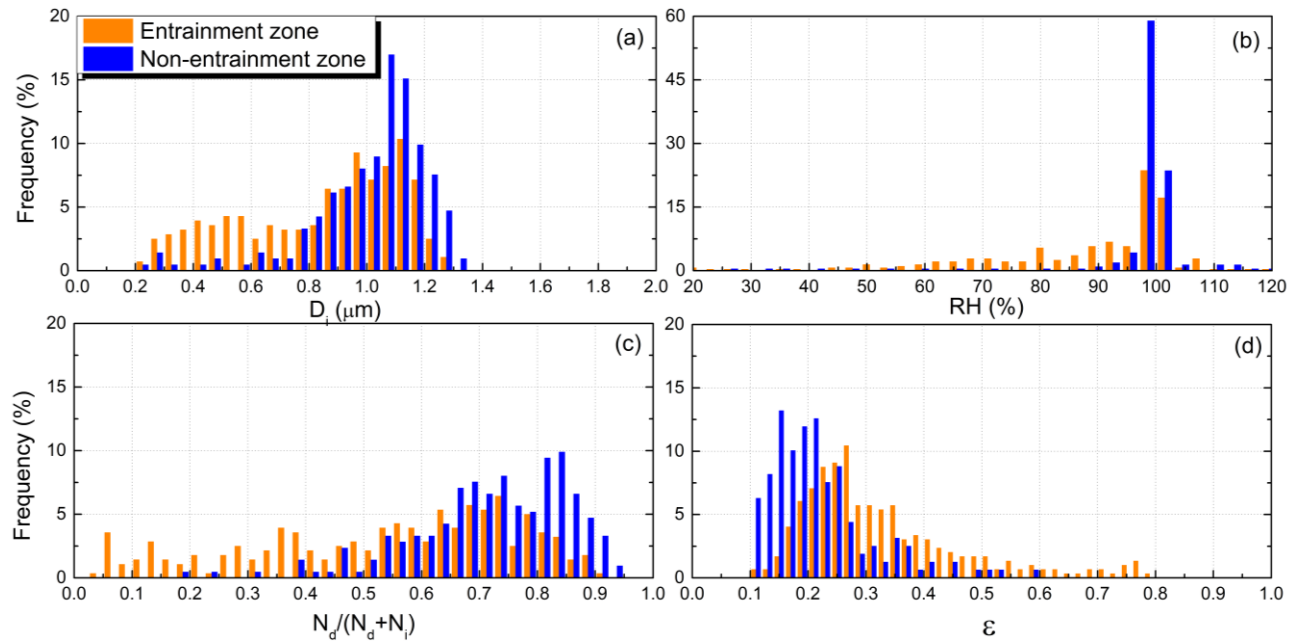
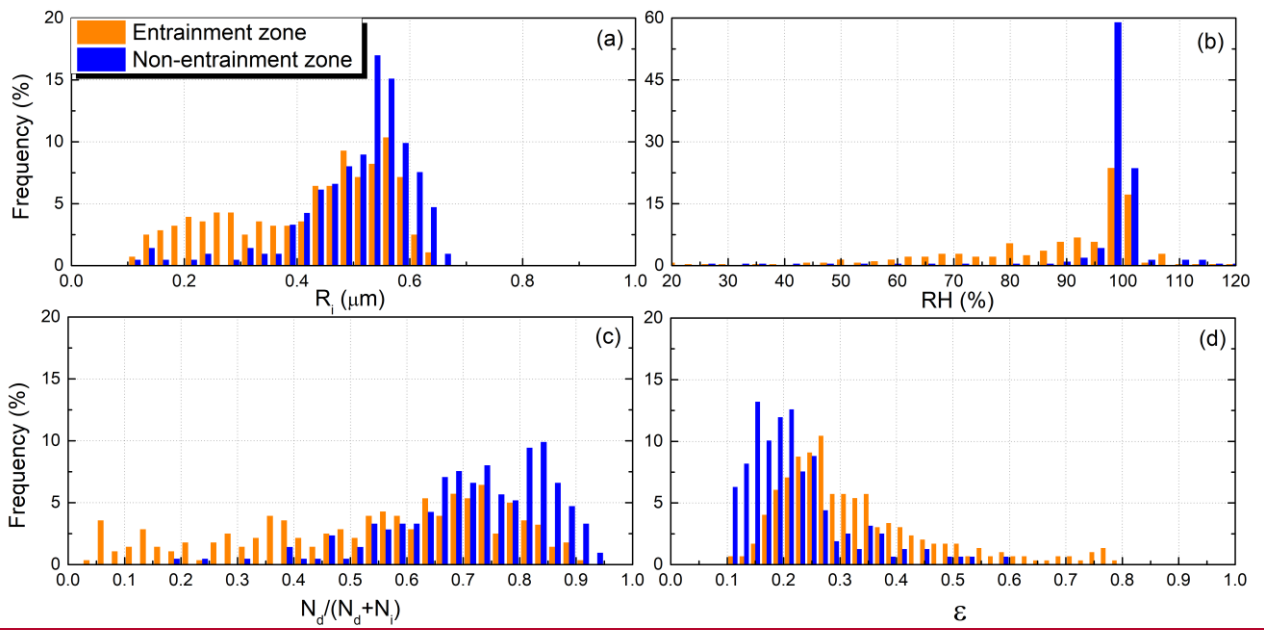


Fig. 11. Number size distributions of cloud droplets in the entrainment (yellow) and non-entrainment zones (blue) during all 16 non-drizzling flights.





780

Fig. 12. Probability density functions of (a) $D_r R_i$ (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**

Hailing Jia, Xiaoyan Ma and Yangang Liu

Correspondence to: Xiaoyan Ma (xma@nuist.edu.cn)

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_{N_d} as a function of $AF_{ent}/AF_{non-ent}$ for all 16 non-drizzling flights.

Figure S1

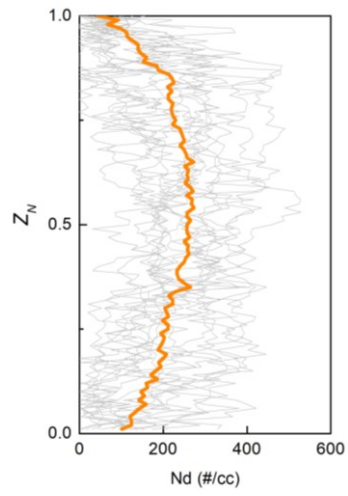


Figure S2

