Ultraclean Layers and Optically Thin Clouds in the Stratocumulus-to-Cumulus Transition. Part II: Depletion of Cloud Droplets and Cloud Condensation Nuclei through Collision–Coalescence

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ABSTRACT

In Part I, aircraft observations are used to show that ultraclean layers (UCLs) in the marine boundary layer (MBL) are a common feature of the stratocumulus-to-cumulus transition (SCT) region over the northeast Pacific. The ultraclean layers are defined as layers of either cloud or clear air in which the concentration of particles with diameter larger than $0.1\ \mu\text{m}$ is below $10\ \text{cm}^{-3}$. Here, idealized microphysical parcel modeling shows that in the cumulus regime, collision–coalescence can strongly deplete cloud droplet concentration in cumulus (Cu) updrafts, thereby removing cloud condensation nuclei (CCN) from the atmosphere, suggesting that collision scavenging is likely the key process causing the low particle concentration in UCLs. Furthermore, the model results suggest that the stratocumulus regime is typically not favorable for UCL formation, because condensate amounts are generally not large enough to deplete drops in the time it takes to loft air to the upper planetary boundary layer (PBL). A bulk parameterization of the coalescence-scavenging rate is derived based on in situ measurements. The fractional coalescence-scavenging rate is found to be strongly dependent upon liquid water content (LWC) and, hence, the height above cloud base, indicating that a higher cloud top and thus a greater cloud thickness in a Cu updraft is an important factor accounting for the observed sharp rise of UCL coverage in the SCT region. An important implication is that PBL height, which controls maximum cloud thickness, and therefore LWC in updrafts, could be a crucial factor constraining coalescence scavenging and thus the formation of UCLs in the MBL.

1. Introduction

The number concentration of water droplets N_d and cloud condensation nuclei (CCN) N_{ccn} continue to be variables of importance in marine boundary layer (MBL) clouds because they help determine cloud albedo (i.e., first indirect effect; Twomey 1977) and its perturbation due to aerosol can modulate the ability to form precipitation (Albrecht 1989). The N_d is limited by N_{ccn} through the process of cloud droplet activation in which CCN serve as nuclei for cloud droplets. Changes in precipitation due to variations in N_d and N_{ccn} can enhance the change in cloud albedo associated with the Twomey effect. The smaller droplet size (i.e., smaller effective radius r_e) resulting from increasing N_d and N_{ccn} (for a given cloud liquid water content q_L) can hinder collision–coalescence and result in a lower precipitation rate at cloud base R_{cb} , which could potentially increase cloud lifetime and hence cloud albedo, often called the

second indirect effect (Albrecht 1989). Several observational studies have found a consistent relationship between R_{cb} and N_d in MBL clouds and suggest that R_{cb} generally decreases as N_d increases (Hudson and Li 1995; Ferek et al. 2000; Bretherton et al. 2004; Wood 2005; Lu et al. 2007; Terai et al. 2012). However, there remain discrepancies in the degree of precipitation susceptibility to N_d and its dependence on liquid water path (LWP) and cloud thickness H_t across modeling and observational studies (Feingold and Siebert 2009; Sorooshian et al. 2009; Terai et al. 2015; Jung et al. 2016). Several studies have suggested that the changes in macrophysical processes such as entrainment rate in the MBL induced by aerosol perturbations can counter the second indirect effect and, in some cases, may overcome it (Ackerman et al. 2004; Wood 2007; Bretherton et al. 2007; Small et al. 2009; Lee et al. 2012), emphasizing the difficulty in establishing a simple susceptibility metric.

Previous studies have focused upon variability of N_d and N_{ccn} as a driver of variability in precipitation. On the other hand, precipitation can also act as the driver of

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variability in N_d and N_{ccn} through collision–coalescence. As pointed out by [Hudson and Frisbie \(1991\)](#), collision–coalescence can efficiently remove CCN within droplets from the atmosphere even with light precipitation rate ([Feingold et al. 1996](#); [Wood 2006](#)). An evaporated coalesced droplet only leaves behind one residual haze particle that is made up of the nuclei from many droplets, the so-called coalescence-scavenging process, which is an irreversible process. Recently, by using a simplified aerosol budget model, driven by observations from aircraft and satellites, [Wood et al. \(2012\)](#) showed that the geographical and temporal variability of global MBL N_d and N_{ccn} are highly impacted by coalescence scavenging. This poses a potential positive feedback in the aerosol–cloud–precipitation interaction as suggested by recent modeling studies ([Yang et al. 2012](#); [Bernier et al. 2013](#)) such that the increase in N_{ccn} can reduce the efficiency of precipitation sink of N_{ccn} , which hence amplify the initial perturbation of increasing N_{ccn} .

In situ measurements showing the coalescence-scavenging driving of the vertical variability in N_d and N_{ccn} have been presented in a mounting body of previous studies. Early in situ measurements of marine cumulus have reported that there is a substantial decrease in N_d with increasing height ([Weickmann and Aufm Kampe 1953](#); [Squires 1958](#)), suggesting the efficient depletion of N_d by collision–coalescence. Measurements of N_{ccn} in cumulus and stratocumulus decks over the northeastern Pacific Ocean ([Hudson and Frisbie 1991](#); [Hudson 1993](#)) have shown there is vertical decrease of N_{ccn} , which the authors attributed to coalescence scavenging. Aircraft observations of MBL clouds over the southern Pacific Ocean ([Stephens and Platt 1987](#); [Boers and Krummel 1998](#); [Boers et al. 1996](#)) have demonstrated that N_d generally decreases with increasing altitude while r_e shows the reverse trend, which is consistent with the occurrence of droplet growth by collision–coalescence. Similar vertical variations of N_d are also observed in midlatitude warm clouds and Arctic warm clouds as summarized by [Gultepe and Isaac \(2004\)](#). In recent observations over the southeastern Pacific Ocean, [Terai et al. \(2014\)](#) found that there is major vertical stratification of N_{ccn} such that measurements at the surface can be an order of magnitude higher than those at the level of the main cloud deck within the pockets of open cells (POCs). For observations near the top of the planetary boundary layer (PBL), N_d scaled strongly with vertical motion within active cumulus (Cu; [Wood et al. 2011](#)). Further, N_d and N_{ccn} within POCs were found to be extremely low ($\sim 5\text{ cm}^{-3}$ in the mean), demonstrating that in-cloud collision–coalescence can be efficient in depleting N_d and N_{ccn} , especially in the upper MBL and prompting

characterization of these layers as ultraclean layers (UCLs; [Sharon et al. 2006](#); [Wood et al. 2011](#)).

In [Wood et al. \(2018, hereafter Part I\)](#), we demonstrated that during the Cloud System Evolution in the Trades (CSET) field program (July–August 2015; subtropical northeastern Pacific), the NSF–NCAR Gulfstream V (G-V) aircraft frequently encountered UCLs, which were therein defined as layers of either cloud or clear air in which the concentration of particles larger than $0.1\text{ }\mu\text{m}$ is below 10 cm^{-3} (see [Part I](#) for details regarding the definition of UCLs). We showed that the clouds in the UCLs are typically geometrically thin and were commonly the veil clouds associated with aggregated Cu clusters. Visible satellite images indicate that a large fraction of clouds observed in the UCLs were optically thin ($\tau < 3$). In part, this is caused by the extremely low N_d in the UCL clouds (see discussion in [Part I](#)). Aircraft in situ measurements showed that (i) UCL coverage in CSET was as high as 40%–60% between 135° and 155°W [i.e., stratocumulus-to-cumulus transition (SCT) region with deep PBL height] but that UCLs occurred very infrequently ($<10\%$) east of 130°W (i.e., the low-topped near-coastal stratocumulus region); (ii) UCLs are most commonly found at a height of 1.5–2 km, typically close to top of the PBL; and (iii) r_e in the UCL clouds is $\sim 20\text{ }\mu\text{m}$ in the mean, which is twice as large as the mean r_e in the non-UCL clouds (i.e., $N_d > 10\text{ cm}^{-3}$; see [Fig. 1](#)).

Based on the analysis of the aircraft measurements summarized in [Part I](#), we hypothesize that the observed UCLs most likely result from efficient collision–coalescence in the upper MBL over the SCT region. The deep PBL height ($\sim 1500\text{ m}$) in the SCT region gives high q_L in the active trade Cu towers. It provides an opportunity for collision–coalescence to sufficiently deplete N_d and hence could result in the formation of UCL clouds with extremely low N_d and large r_e in the upper MBL. Detrainment of cloudy air from the Cu towers forms horizontally extensive, thin cloud layers in the upper MBL with extremely low N_d and low cloud optical depth as indicated by the satellite data (see [Part I](#)). Upon evaporation of UCL clouds, only a few accumulation mode particles are returned to the clear sky, which could leave the upper MBL strongly depleted in N_{ccn} .

In this work, we quantitatively test the proposed hypothesis for the formation process of the UCLs using idealized microphysical parcel model. The focus is on the depletion of N_d and N_{ccn} through in-cloud collision–coalescence. An adiabatic two-dimensional (i.e., spanning droplet and aerosol dry size) bin parcel model is chosen as our main tool to test the hypothesis. The details of the model formulation are furnished in [section 2](#).

FIG. 1. Cloud droplet effective radius r_e of UCL clouds (red) and non-UCL clouds (blue) measured during CSET. The blue and red lines are the mean r_e for non-UCL clouds and UCL clouds, respectively. The calculation of r_e is based on the measured DSDs of the CDP and 2DC probes. UCL cloud is defined as the cloudy layer with $N_d < 10 \text{ cm}^{-3}$, and non-UCL cloud is defined as the cloudy layer with $N_d > 10 \text{ cm}^{-3}$. The criterion used to separate the saturated cloudy samples from the unsaturated clear-air samples can be found in [Part I](#).

[Section 3](#) gives the parcel model results showing the temporal evolution of cloud microphysical processes in an idealized rising parcel in typical cumulus and stratocumulus regimes. In [section 4](#), by obtaining the best fit with the in situ measurements of cloud droplet size distributions (DSDs) from CSET, a bulk parameterization of the droplet coalescence-scavenging rate $[\dot{N}_d]_{cs}$ is derived from the stochastic collection equation as a function of N_d and q_L . With the parameterization, the relative importance of updraft, aerosol concentration, cloud thickness, and adiabaticity in coalescence-scavenging process are accessed. [Section 5](#) offers a conclusion.

2. Parcel model

a. Basic model formulations

A Lagrangian adiabatic parcel model with explicit two-dimensional bin microphysics spanning aerosol and water droplet sizes is formulated to simulate the evolution of cloud microphysical processes in a rising parcel. First, the trajectory of the parcel follows an adiabatic ascent with a constant vertical velocity U_z . After the parcel reaches the assumed height of cloud top z_{top} , we include a period where the parcel moves horizontally at the cloud-top layer with $U_z = 0$ (see [Fig. 2](#)). Cloud microphysical processes are considered by holding the particles on a two-dimensional grid, and each grid point is characterized by the sizes of the aerosol and the water droplet. Grid spacing for the aerosol size (i.e., dry size) and the water droplet size (i.e., wet size) are logarithmically

spaced. The grid of the droplet size starts and ends at $D_d = 0.02$ and 0.7 cm . The aerosol size grid spans from $D_a = 0.016$ to 0.4 cm . Both liquid water and aerosol grids have 40 bins resulting in total 1600 bins in the 2D grid space. The model is integrated with the time step of 0.05 s .

The prognostic thermodynamics variables are temperature and saturation ratio, and the microphysical processes that are explicitly simulated in the model are activation of aerosol, condensation–evaporation, interstitial scavenging, collision–coalescence, and droplet loss from the parcel by sedimentation (the details regarding the treatments of droplet sedimentation process in the parcel model are given below). Details of the equations describing condensation and the changes of the thermodynamic variables applied in the model can be found in the appendix of [Korolev and Mazin \(2003\)](#). There is no artificial separation between unactivated aerosols, activated cloud droplets, and drizzle. The nucleation of unactivated aerosol (i.e., haze particles) is explicitly calculated by solving the droplet growth equations with κ –Kohler parameterization for aerosol hygroscopicity developed by [Petters and Kreidenweis \(2007\)](#); the details regarding the size distribution and κ value of aerosol are given later).

Collision–coalescence increases not only the size of water droplets but also size of the aerosol inside. The advantage of the 2D bin framework is that the resulting aerosol mass can be saved after collision–coalescence takes place, which may be important for the temporal evolution of DSDs. The hybrid bin scheme developed by

FIG. 2. Conceptual diagram of the formation of UCLs in the Sc–Cu transition.

Chen and Lamb (1994; CL scheme) is applied to treat condensation and evaporation processes. The two-dimensional flux method developed by Bott (2000; Bott scheme) is used to treat collision–coalescence in the 2D bin framework. Both the CL scheme and the Bott scheme are mass conservative and numerically efficient. The CL scheme is a two-moment explicit scheme designed for the evolution of the DSDs by condensation–evaporation, which produces little numerical diffusion and can be implemented into the 2D bin framework. The Bott scheme is an advanced method introducing the flux method for the stochastic collection equations developed by Bott (1998) into the 2D bin framework with low artificial broadening of DSDs. The gravitational collection kernel is applied here and can be given by $K(r, r') = E_c \pi(r + r')^2 |V_T(r) - V_T(r')|$ (where E_c is the collision efficiency, r and r' are the radius of droplets, and V_T is the terminal velocity of droplet). The collision efficiency E_c used in the collection kernel is based on the combination of data from Hall (1980) and Rogers and Yau (1989). The coalescence efficiency is set to unity. The inertial scavenging is considered in the model with the collection kernel for inertial scavenging developed by Berner et al. (2013). It is shown later that aerosol activation is the dominant process depleting N_{ccn} , and inertial scavenging does not contribute significantly to the depletion of N_{ccn} in the cloud regime investigated here. The terminal particle velocity V_T is based on the relationship of V_T with D_d given in Pruppacher and Klett (2012).

b. Initial conditions and aerosol distribution

A set of initial conditions must be specified to run the model. The initial temperature T_0 , the initial saturation ratio S_0 , and the initial pressure P_0 are specified to be 300 K, 0.8, and 1013 hPa, respectively, which are typical of the lower MBL and give the cloud-base z_{base} [i.e., lifting condensation level (LCL)] ~ 500 m. The average normalized aerosol size distribution from the surface legs ($z < 170$ m) of 16 flights during CSET is used to specify the initial aerosol dry size spectrum in the model.

The average aerosol size distribution shown in Fig. 3 is the combination of data from two probes, an ultra-high-sensitivity aerosol spectrometer (UHSAS; $0.06 < D_a < 1 \mu\text{m}$) and a cloud droplet probe (CDP; $D_a > 1 \mu\text{m}$). The CDP can quantify giant CCN (GCCN). The ambient size given by CDP is corrected to the dry size by using a growth factor for sea salt of 1.8 as reported in Zhang et al. (2014). The sensitivity of the model results to GCCN is tested by removing GCCN from the initial aerosol input. The results (not shown) indicate that for the cloud regimes investigated here (i.e., in terms of number concentration of aerosol, aerosol size distribution, cloud-top height, and updraft speed), removal of GCCN from the initial aerosol input does not significantly change the temporal evolution of N_d and N_{ccn} in a rising parcel. GCCN could be relatively more important in the regime with more aerosol and lower cloud-top height. To thoroughly explore the importance of GCCN for the droplet coalescence-scavenging rate, a more comprehensive study is needed to test the sensitivity to GCCN in various cloud regimes, which is not our focus in this study. Clarke et al. (2006) reported the mean aerosol size distribution over the coastal regions of Hawaii (i.e., the northeastern Pacific, which is the same area investigated in this study) with average 10-m wind speed of $\sim 7 \text{ m s}^{-1}$. Note that the measurements in CSET only provide the aerosol size distribution with $D_a > 0.06 \mu\text{m}$. To completely include the Aitken mode aerosol ($D_a < 0.1 \mu\text{m}$), the aerosol concentration in size range $0.016 < D_a < 0.06 \mu\text{m}$ from Clarke et al. (2006) is included in the initial aerosol size distribution. The total initial aerosol concentration assigned in the parcel model will be discussed in section 3. The κ value of aerosols is specified to be 0.8, which is typical at the surface over the northeastern Pacific according to a recent modeling study [see Figs. 3 and 5 in Pringle et al. (2010)]. When the initial aerosol size distribution is introduced into the 2D aerosol and cloud bin spectrum, the initial equilibrium wet size of the aerosol is calculated based on the κ –Köhler equation with the specified κ value and the initial conditions of S_0 , P_0 , and T_0 .

FIG. 3. Average normalized aerosol size distribution from CSET surface legs ($z < 170$ m) of 16 flights (colors with circles) based on the UHSAS and CDP probes. The black line is the average normalized aerosol size distribution from all flights.

c. Treatment of droplet sedimentation in the parcel model

A significant deficiency of the Lagrangian parcel model is that droplet loss from the parcel by sedimentation is typically not taken into account. This may lead to unrealistic rates of collision–coalescence leading to the formation of millimeter-size drops with fall speeds substantially faster than the updrafts. Such deficiency in the Lagrangian approach can be avoided in Eulerian modeling frameworks like large-eddy simulation (LES), which explicitly calculate droplet loss rate by sedimentation. On the other hand, in the Eulerian framework, the complexity of microphysical schemes is usually limited by computational resources, and cloud microphysical processes must typically be parameterized. With a focus in this study on microphysical processes for UCLs formation, we choose the 2D bin microphysics parcel model as the tool for our modeling study. However, we also wish to consider the droplet loss due to sedimentation in the Lagrangian parcel model, and to achieve this, a simple treatment is selected and tested.

According to Houze (2014), with an assumption that the parcel has thickness h_p , the sedimentation loss rate of given size droplets in a parcel can be approximated by the sedimentation flux

$$[\dot{N}_{ij}]_{\text{sedi}} = -N_{ij}V_{Tj}/h_p, \quad (1)$$

where i and j denote the aerosol dry size bin and droplet wet bin on the two-dimensional grid. A two-step process is applied in the sedimentation flux method and is similar to the treatment in the 2D collision–coalescence scheme proposed by Bott (2000). We first integrate the 2D particle spectrum over the aerosol dry size, which yields a one-dimensional (1D) droplet spectrum as a function of the droplet size. In the next step, the flux loss

rate of droplet given in Eq. (1) is applied to each grid box of this intermediate spectrum. In the last step, the resulting new droplet spectrum is redistributed into the 2D bin grids based on the fraction of droplets over the aerosol size grid. The parcel thickness h_p is specified to be 100 m and is also used to specify the depth of the cloud-top layer (i.e., cloud-top detrainment layer) in the simulation.

The most uncertain factor in the sedimentation flux method is h_p , which requires an assumption of the parcel thickness. It is shown later that there is only a weak sensitivity to h_p assumed in the model. In general, there is no perfect treatment for the droplet sedimentation process in the Lagrangian parcel model, so we will perform sensitivity tests to ensure our conclusion on the microphysical process of the UCLs is consistent among the parcel model runs with different assumed values of h_p .

3. Parcel modeling results

In Part I, analysis of the aircraft measurements in CSET found that UCLs are commonly found between 135° and 155°W over the northeastern Pacific, the typical SCT region, but occur infrequently east of 130°W over the northeastern Pacific, the overcast stratocumulus region. We showed that the UCL clouds are commonly the veil clouds associated with the aggregated Cu clusters. This implies that the cumulus regime is favorable for the formation of UCLs. Here, two sets of idealized modeling experiments representing the temporal evolution of cloud microphysics in a rising parcel in stratocumulus (Sc) and Cu are presented. According to Eastman et al. (2017), retrievals of PBL height from Moderate Resolution Imaging Spectroradiometer (MODIS) CALIPSO and COSMIC GPS radio occultation show that the typical cloud-topped PBL heights at 20°–40°N are 1500 m between 135° and 155°W (i.e., Cu regime) and 800 m east of 130°W (i.e., Sc regime) over the northeastern Pacific. These values are used to specify z_{top} in the parcel modeling for the Cu and Sc regimes, respectively. The updraft speeds U_z in MBL clouds are typically 0.1–3 m s^{−1}, and U_z in cumulus is typically more vigorous than in stratocumulus (Comstock et al. 2007; Gerber et al. 2008; Ghate et al. 2011). Updraft velocity can influence both in-cloud times available for air parcels rising through a cloud and the maximum supersaturation ratio in a cloud updraft. The updraft speed U_z in the *base cases* for the Cu and Sc regimes are specified to be 0.75 and 0.25 m s^{−1}, respectively. We will explore the sensitivity to updraft in both regimes by considering updraft velocities of 0.1 and 3 m s^{−1}. In the following discussion, N_d is defined as the number concentration of the particles with diameter ≥ 1 μm, and N_{ccn} is defined

FIG. 4. Average accumulation mode aerosol concentration from CSET surface legs ($z < 170$ m) as a function of longitude based on the UHSAS probe.

as the number concentration of the particles with diameter $< 1 \mu\text{m}$. The initial aerosol concentration with $D_a > 0.06 \mu\text{m}$ is based on the mean near-surface (i.e., $z < 170$ m) aerosol concentration measured by the UHSAS probe ($1 > D_a > 0.06 \mu\text{m}$) during CSET (Fig. 4) and is specified to be 75 cm^{-3} (i.e., the mean value at $135^\circ\text{--}155^\circ\text{W}$, the typical cumulus regime) in the Cu regime and 200 cm^{-3} (i.e., the mean value at $125^\circ\text{--}132.5^\circ\text{W}$, the typical stratocumulus regime) in the Sc regime. Sensitivity tests on the initial aerosol condition by swapping input aerosol concentration in the Cu and Sc base cases are performed and will be discussed. Note that the near-surface concentration of GCCN with $D_a > 1 \mu\text{m}$ measured by CDP is mostly $< 1 \text{ cm}^{-3}$ in CSET. The aerosol concentration ($\sim 7 \text{ cm}^{-3}$) and size distribution with $D_a < 0.06 \mu\text{m}$ from Clarke et al. (2006) is included in the initial aerosol distribution in both regimes to completely include the Aitken mode aerosol. A summary of the setups and the results of the Cu and Sc cases simulated in this study is given in Tables 1 and 2. The discussion regarding liquid water content q_L in the parcel modeling will be given in section 4.

a. Cumulus regime

Figure 5 shows the base case for the Cu regime, which suggests the Cu regime is favorable for the formation of UCLs and supports our hypothesis. The modeling result shows that N_d is strongly depleted in the cumulus updraft ($> 80\%$ depletion of N_d), leading to the occurrence of UCL clouds at the cloud-top layer. Figure 5d shows the fraction of droplet loss caused by collision-coalescence, droplet sedimentation, and evaporation. It indicates that collision-coalescence within the parcel is the dominant process in depleting N_d and the formation of UCLs in the Cu regime. The temporal evolution of r_e is shown in Fig. 5b with the black line, indicating

TABLE 1. Summary of the setups and the results of the cumulus cases simulated in this study. Initial N_{ccn} includes both Aitken mode and accumulation mode aerosol measured during CSET and from Clarke et al. (2006), where z_{top} is the cloud-top height, U_z is the parcel updraft, and $\bar{r}_e$ is the average effective radius.

Parameters	Cu cases			
Figure	Fig. 5	Figs. 7a–e	Figs. 7f–j	Figs. 11a–e
Initial N_{ccn} (cm^{-3})	77	77	77	207
z_{top} (m)	1500	1500	1500	1500
U_z (m s^{-1})	0.75	0.1	0.3	0.75
$\bar{r}_e$ of UCL clouds (μm)	20	19	21	17
UCL formation	Yes	Yes	Yes	Yes

considerable growth of droplet size in the Cu updraft by collision-coalescence. Cloud droplets can grow through condensation and collision-coalescence. By using an analytical expression for diffusional droplet growth, Pinsky et al. (2014) has shown that without including the collision-coalescence process, r_e of droplets in warm liquid clouds can hardly reach $15 \mu\text{m}$ by condensation alone [see Fig. 6 in Pinsky et al. (2014)]. Thus, the substantial growth of r_e shown in the parcel modeling and the large r_e in UCL clouds observed during CSET ($20 \mu\text{m}$ in the mean; see Fig. 1) must be due to collision-coalescence. The modeling results show that r_e can be larger than $\sim 30 \mu\text{m}$ in the UCL clouds (black line in Fig. 5b), and such large droplet size in the UCL clouds is also observed during CSET, as shown in Fig. 1. After the parcel has reached its assumed maximum height, because of the relatively large droplet loss caused by droplet sedimentation (black line in Fig. 5c), r_e in the UCL clouds decreases gradually. This may explain why the UCL cloudy samples observed during CSET have relatively large spread in r_e (see Fig. 1). After the parcel stops lifting, droplet sedimentation gradually becomes the primary process depleting N_d (see Fig. 5d) with very low droplet loss rate and very low droplet concentration ($< 5 \text{ cm}^{-3}$). The results presented here suggest that there

TABLE 2. As in Table 1, but for the stratocumulus cases.

Parameters	Sc cases			
Figure	Fig. 9	Figs. 10a–e	Figs. 10f–j	Figs. 11f–j
Initial N_{ccn} (cm^{-3})	207	207	207	77
z_{top} (m)	800	800	800	800
U_z (m s^{-1})	0.25	0.1	3	0.25
$\bar{r}_e$ of UCL clouds (μm)	—	10	—	—
$\bar{r}_e$ of non-UCL clouds (μm)	8	9	10	10
UCL formation	No	Yes	No	No

FIG. 5. Cumulus base case with U_z of 0.75 m s^{-1} . (a) Height (orange) and supersaturation ratio (blue) of the parcel; (b) temporal evolution of number concentration of cloud condensation nuclei N_{ccn} (red), droplet sedimentation N_{sed} (green), cloud droplet N_d (blue), and effective radius of droplet r_e (black) in the parcel, and the black dashed lines indicates a particle concentration of 10 cm^{-3} ; (c) total droplet loss rate (green) and the droplet loss rate by sedimentation (black), collision-coalescence (blue), and evaporation (red); (d) fraction of droplet loss rate by sedimentation (black), collision-coalescence (blue), and evaporation (red); and (e) temporal evolution of DSDs in the parcel.

is a transition of dominant cloud microphysics process from the updraft to the cloud-top layer such that the depletion of N_d is dominated by collision-coalescence resulting in the increase of r_e in the updraft, while at the cloud-top layer, sedimentation loss is the main process depleting N_d and thus causes the decrease of r_e . MODIS retrievals of r_e suggests that such behavior of r_e is typical in the SCT region. Figure 6 shows that in the Cu clusters, r_e (from $3.7\text{-}\mu\text{m}$ MODIS channel) reduces from $\sim 30 \mu\text{m}$

at the updraft region to $\sim 10 \mu\text{m}$ at the cloud edge, which is consistent with the sedimentation process at the cloud top shown by the parcel modeling presented here. The retrieval of r_e using $2.1\text{-}\mu\text{m}$ MODIS channel shows the similar pattern (not shown) for all these selected cases. The schematic evolution of cloud microphysics in the cumulus updraft described here is summarized in Fig. 2.

The left column in Fig. 7 shows the parcel modeling results for the Cu regime but with an artificially small U_z

FIG. 6. MODIS retrieval of effective radius of Cu clusters in the Sc-Cu transition (3.7- μm MODIS channel).

of 0.1 m s^{-1} . The cumulus case with U_z of 0.1 m s^{-1} has low S_{max} ($\sim 0.2\%$) in the updraft, resulting in fewer particles activated at the cloud base than in the cases with U_z of 0.75 and 3 m s^{-1} (Figs. 5 and 7f–j). The N_d in the Cu updraft is strongly depleted by collision-coalescence, which eventually causes the formation of UCLs in the updraft. In the case with low updraft of 0.1 m s^{-1} , UCLs are formed at fairly low altitude ($\sim 1100 \text{ m}$) even before the parcel reaches its assumed max height (i.e., 1500 m). Note that in the Cu cases with U_z of 0.75 and 3 m s^{-1} , UCLs are formed after the parcel reaches the assumed cloud-top height. This indicates that slower U_z can help the formation of UCLs at lower altitude, which can be linked to the updraft time scale τ_{up} (i.e., a parcel to ascend from base to top) as discussed in Ovchinnikov et al. (2013). A longer τ_{up} (i.e., slower U_z , given same cloud thickness) can provide longer in-cloud time in updraft for collision-coalescence to act to deplete N_d , so the formation of UCLs is favored by slower updrafts [see detailed discussion in Ovchinnikov et al. (2013)]. In addition to the maximum supersaturation S_{max} at the cloud base, there is a second maximum in S_w occurring near the cloud top because of the substantial decrease of N_d (i.e., depletion term for supersaturation ratio S_w) and constant U_z in the Cu updraft [i.e., source term for S_w ; see Eq. (2) in Pinsky et al. 2013], leading to multiple secondary activation events of small aerosols (see Fig. 7e)

that cannot be activated at the cloud base. Secondary activation of aerosols not only increases N_d but also hence slightly increases the droplet loss rate by collision-coalescence [droplet loss rate by collision-coalescence is positively correlated with N_d ; see Eq. (3) in section 4], which results in the blue jagged line of fraction of droplet loss by collision-coalescence shown in Fig. 7d.

The right column in Fig. 7 shows the parcel modeling result for the Cu regime with U_z of 3 m s^{-1} . Because of the high U_z (i.e., 3 m s^{-1}) assumed in this case, the available time for collision-coalescence to act to deplete N_d in the updraft is much shorter than in the other two cases (i.e., $U_z = 0.1$ and 0.75 m s^{-1}). The fractional loss of N_d from the cloud base to the cloud-top $f_{\text{top-loss}}$ is $\sim 50\%$, which is lower than in the cases with U_z of 0 and 0.75 m s^{-1} ($f_{\text{top-loss}} > 80\%$). In this case with high U_z , the maximum coalescence-scavenging rate (i.e., blue line in Fig. 7h) is at the cloud top. On the contrary, in the previous cases with relatively lower updraft speed (i.e., Fig. 5 and left column of Fig. 7), the maximum coalescence-scavenging rates are typically not at the cloud top and have much lower values (see blue lines in Figs. 5c, 7c, and 7h). This is because the coalescence-scavenging rate is positively correlated with N_d and q_L (see section 4 for details). In the cases with relatively low updraft (i.e., $U_z = 0.1$ and 0.75 m s^{-1}), most of N_d is depleted before the parcel reaches the cloud top. When

FIG. 7. Cu regime as in Fig. 5, but for $U_z =$ (a)–(e) 0.1 and (f)–(j) 3 m s^{-1} .

the parcel reaches the cloud top, because of very low N_d ($< 10 \text{ cm}^{-3}$), the coalescence-scavenging rate is very low even though maximum q_L in the adiabatic updraft is at the cloud top. In contrast, in the case with fast U_z , more than 50% of N_d still remain in the parcel at the cloud top where adiabatic q_L reaches its maximum in the updraft, resulting in the maximum coalescence-scavenging rate at the cloud top. Regarding the droplet size in the UCL clouds, both of the modeling results shown in Fig. 7 suggest that r_e is $\sim 20 \mu\text{m}$ in the UCL clouds, consistent with the observations shown in Fig. 1. The parcel modeling results for the Cu regime also suggest that UCL clouds contain a monomodal droplet size distribution, which is consistent with the observations summarized in Part I. This indicates that the bin model can produce a droplet spectrum in UCL clouds that is realistic compared with the observations. With the sensitivity test presented in Figs. 5 and 7 by considering a wide range of updraft speed in the Cu regime, we conclude that the parcel modeling results for the Cu regime agree well with the hypothesized microphysical process we proposed for UCL formation. The results indicate that in the Cu regime, collision-coalescence can cause the

formation of UCL clouds by rapidly depleting N_d . Once the UCL clouds evaporate, this efficient coalescence-scavenging process can result in very low N_{ccn} in the upper MBL.

Figure 8 shows a sensitivity test of the Cu base case to the choice of h_p applied in the sedimentation flux. The model runs with h_p of 100, 300, and 500 m are demonstrated here. The results shown in Fig. 8 all suggest that collision-coalescence is the key physical process controlling the depletion of N_d and droplet loss by sedimentation is of secondary importance. The comparison between three model runs in Fig. 8 suggests that in the parcel, there is a compensating feedback in the parcel modeling between collision-coalescence and sedimentation processes in depleting N_d . When the parcel is assumed to be thinner (e.g., $h_p = 100 \text{ m}$), large drizzle-sized droplets tend to fall out of the parcel faster by sedimentation (i.e., depleting N_d), but this lowers the efficiency of collision-coalescence by removing those drops that are the most efficient collectors, and vice versa. As a result of compensation between collision-coalescence and droplet sedimentation in the parcel model, the choice

FIG. 8. Sensitivity test to the value of h_p used in the Cu base case ($U_z = 0.75 \text{ m s}^{-1}$) for $h_p = 100$ (solid), 300 (dotted), and 500 m (dashed). (a) Temporal evolution of cloud droplet concentration N_d in the parcel; (b) total droplet loss rate (green), droplet loss rate by collision-coalescence (blue), and sedimentation (black); and (c) fraction of droplet loss rate by sedimentation (black) and collision-coalescence (blue).

of h_p applied in the parcel model does not result in major differences in the temporal evolution of N_d . The parcel modeling runs for the Sc regime and the Cu regime with different U_z suggest the same compensating feedback (not shown). Thus, based on the sensitivity test presented in Fig. 8, we conclude that the sedimentation treatment applied in the parcel model does not affect our conclusion that collision-coalescence is the process governing the formation of UCLs.

b. Stratocumulus regime

One important feature in stratocumulus is that usually parcels would be forced to descend by longwave cooling in a relatively shorter period of time than in cumulus, and the parcels could potentially pass stratocumulus multiple times. Here, we only allow the air parcel in Sc case to pass cloud layer only once in order to fairly compare the potential of UCLs formation in the stratocumulus cases and the cumulus cases (section 3a). The effect of multiple air parcel cycling on droplet depletion and its associated potential to produce UCLs in stratocumulus cloud will be discussed in section 4c. By using LES, Kogan (2006) showed that the residence time of air parcels at the cloud top is typically <30 min in stratocumulus. Thus, in the parcel modeling runs for the Sc regime, the time for which parcels stay in the cloud-top layer is simply assumed to be 30 min.

The modeling result of the Sc base case indicates that Sc regime is not favorable for the formation of UCLs (Fig. 9) as compared to the Cu regime. Although there is also an appreciable decrease of N_d caused largely by collision-coalescence, the fractional loss of N_d in the stratocumulus updraft is smaller than in the Cu updraft. The value of N_d is 126 cm^{-3} when first reaching the cloud top, giving the fractional loss of 37.5% from the cloud base (N_d is $\sim 200 \text{ cm}^{-3}$ at the cloud base). After the parcel stops lifting, there is a remarkable ripening effect (i.e., large droplets with positive growth rates continue to grow at the expense of small droplets with negative growth rates; Celik and Marwitz 1999; Wood et al. 2002), which results in the evaporation of small droplets and thus an increase of interstitial N_{ccn} (see Fig. 9e) at the cloud-top level. Droplet evaporation can contribute about 10% of the droplet loss at the cloud-top level in the Sc regime. On the contrary, in the Cu case, there is nearly no evaporation because droplets in the Cu updraft can grow to larger sizes (which have saturation vapor pressure $\sim 100\%$) because of higher S_{max} and more efficient collision-coalescence process in the Cu updrafts. At cloud-top level in the Sc base case, when there is still a considerable concentration of droplets and aerosols, the droplet depletion rate by collision-coalescence becomes very low, preventing the formation of UCLs at the cloud-top layer. The mean r_e in the non-UCL clouds in the Sc base case is $\sim 10 \mu\text{m}$, which is consistent with the in situ measurements summarized in Fig. 1.

The left column in Fig. 10 shows the modeling results for the Sc regime with U_z of 0.1 m s^{-1} . Because of low S_{max} caused by low U_z , a large fraction of aerosols are not activated at the cloud base. Because of long τ_{up} (because of low U_z) in the updraft and relatively small amount of activated aerosols compared with the Sc base case, UCL cloud ($N_d < 10 \text{ cm}^{-3}$) is formed at the cloud-top layer mainly caused by collision-coalescence. This suggests that in the Sc regime, if U_z is low enough, formation of UCL cloud is possible after the air parcel passes the stratocumulus layer only once, but because of low S_{max} and thus a large amount of unactivated aerosols remaining in the parcel, formation of a clear UCL (i.e., $N_{\text{ccn}} < 10 \text{ cm}^{-3}$) is difficult. Such low U_z ($\sim 0.1 \text{ m s}^{-1}$) in stratocumulus rarely occurs over the northeastern Pacific according to a recent observational study. Zheng et al. (2016) showed that observed cloud-base updraft in stratocumulus over the northeastern Pacific are typically in the range of $0.2\text{--}1 \text{ m s}^{-1}$, which may explain why the observed occurrence of UCL clouds is fairly low ($<10\%$) in the stratocumulus decks observed in CSET, as we showed in Part I. The right column in Fig. 10 shows the Sc case with U_z of 3 m s^{-1} , suggesting that high U_z in updraft can possibly result in a nearly constant vertical profile of N_d in

FIG. 9. As in Fig. 5, but for the stratocumulus regime.

stratocumulus, which is consistent with the previous discussion regarding fractional droplet loss and τ_{up} in the updraft. For both cases, r_e in the non-UCL clouds is $\sim 10 \mu\text{m}$, consistent with the in situ measurements shown in Fig. 1. Based on the parcel modeling results for the Sc regime shown in Figs. 9 and 10 with the sensitivity test on U_z , we conclude that the Sc regime is typically not favorable for UCL formation as compared to the Cu regime.

Although there are some major advantages to the adoption of a Lagrangian parcel model approach, especially for the study focus on cloud microphysical processes, there are two major caveats that should be addressed and discussed here: 1) the droplets falling into the parcel from above and 2) the entrainment of aerosol

from free troposphere are not taken into account, which could be a significant source of droplet concentration in the rising parcel. However, large droplets falling from above could also increase the efficiency of collision-coalescence by adding those drops that are the most efficient collectors, which can result in the compensating feedback as shown in Fig. 8. In addition, as shown in Part I, veil clouds in UCLs tend to be much less turbulent than non-UCL clouds in the same altitude, suggesting that the entrainment rate of aerosol from above into the veil clouds may be much lower than in the active cumulus updraft. Therefore, these arguments support the notion that the processes of sedimentation and entrainment do not affect our conclusion that strong collision-coalescence in the precipitating cumulus clusters is the dominant

FIG. 10. Sc regime as in Fig. 8, but for $U_z =$ (a)–(e) 0.1 and (f)–(j) 3 m s^{-1} .

process contributing to the prevailing UCLs feature throughout SCT regions.

c. Sensitivity to the initial aerosol condition

In this section, we aim to test the sensitivity of the modeling results to the initial aerosol condition assumed in the parcel model by swapping the initial N_{ccn} assumed in the Sc and Cu base cases. The left column in Fig. 11 shows the Cu case (i.e., cloud top = 1500 m and $U_z = 0.75 \text{ m s}^{-1}$) with an initial N_{ccn} of 207 cm^{-3} . With relatively high initial N_{ccn} , the Cu case still shows the formation of UCLs (i.e., low N_d and large r_e) caused by dominant collision-coalescence process shortly after the parcel reaches the assumed cloud-top height (1500 m). This result indicates that the initial N_{ccn} plays only a minor role in determining the formation of UCLs in the Cu regime. On the other hand, with initial N_{ccn} of 77 cm^{-3} assumed in the Sc case (Fig. 11, right column; i.e., cloud top = 800 m and $U_z = 0.25 \text{ m s}^{-1}$), N_d is depleted to 17 cm^{-3} after the parcel stays for 30 min at the cloud top, indicating that even with artificially low aerosol concentration, the Sc regime is still rather unfavorable for the formation of UCLs (i.e., $N_d < 10 \text{ cm}^{-3}$) as compared to the Cu regime. These

two sensitivity experiments show that cloud thickness and liquid water amount are much more crucial factors than initial aerosol concentration for the formation of UCLs.

4. Parameterization of droplet coalescence-scavenging rate

a. Derivation

The stochastic collection equation (SCE) gives an expression for the droplet loss rate through coalescence-scavenging $[\dot{N}_d]_{\text{cs}}$ (e.g., Wood 2006):

$$[\dot{N}_d]_{\text{cs-SCE}} = -\frac{1}{2} \int_0^\infty \int_0^\infty n(r)n(r')K(r,r')drdr'. \quad (2)$$

Given that each droplet coalescence event reduces the number of CCN by one, Eq. (2) can also be interpreted as being a loss rate for the CCN. To explicitly solve Eq. (2), a bin microphysics scheme and the full DSD is required. However, deployment of bin microphysics in a numerical model needs considerable computational expense, so the microphysical schemes in LES and general circulation models (GCMs) are mostly of the

FIG. 11. Sensitivity tests to the initial N_{ccn} assumed in the Cu and Sc cases by the swapping initial aerosol condition in the Cu and Sc base cases. (a)–(e) Updraft = 0.75 m s^{-1} , initial $N_{ccn} = 207 \text{ cm}^{-3}$, and cloud top = 1500 m. (f)–(j) Updraft = 0.25 m s^{-1} , initial $N_{ccn} = 77 \text{ cm}^{-3}$, and cloud top = 800 m.

bulk form, which solve the prognostic equations only for the bulk parameters such as N_d and q_L . Further, by using Eq. (2) and the parcel modeling, the time scale of droplet depletion and its dependence on N_d and q_L cannot be easily explored and examined. Thus, we use CSET observations to derive a parameterization for $[\dot{N}_d]_{cs-SCE}$ as a function of bulk parameters, which may provide more insight into the time scale of droplet depletion by collision–coalescence. In addition, with the parameterization of droplet coalescence–scavenging rate, we will explore the role of adiabaticity f_{ad} in the collision–scavenging process. With the in situ measurements of DSDs [i.e., droplet size distribution from CDP and 2D cloud (2DC) probes of only the cloudy samples] from CSET (the details of the measurements used here and the criteria for choosing the cloudy samples are summarized in Part I), we fit the following formula to the droplet loss rate calculated from the SCE [Eq. (2)]:

$$[\dot{N}_d]_{cs-PARAM} = -a(N_d)^b(q_L)^c. \quad (3)$$

Note that units of $[\dot{N}_d]_{cs-PARAM}$, N_d , and q_L in Eq. (3) are $\text{m}^{-3} \text{s}^{-1}$, m^{-3} , and kg m^{-3} . The values of a , b , and c

from the best fitting are 8×10^4 (7×10^4 , 9×10^4), 0.55 (0.545, 0.555), and 1.37 (1.36, 1.375), where the values in parentheses are the lower and upper confidence bounds of the 95% confidence interval. Figure 12 shows the comparison of Eq. (3) with the values calculated by SCE, indicating that the bulk parameterization of Eq. (3) agrees fairly well with the SCE (correlation coefficient of 0.84). It should be noted that most of the bulk microphysics models divide condensate into cloud and rain and calculate the loss of N_d in terms of the sum of autoconversion and accretion processes. The derived droplet loss rate from SCE [Eq. (2)] combines both of these effects and describes them in terms of a single q_L , which is simpler than one would use in a bulk parameterization and is useful for our purpose here. To further examine the accuracy of the parameterization, $[\dot{N}_d]_{cs-BIN}$ calculated from the bin microphysics parcel model (i.e., blue lines in Figs. 5c and 9c) can serve as the benchmark for the parameterized formula of Eq. (3).

Figures 13a and 13d show the temporal evolution of N_d and q_L calculated from the parcel model in the Cu and Sc base cases (same as Figs. 5 and 9). Note that in Fig. 13, $q_L[\text{parcel}]$ is the liquid water content of the cloud

droplets inside the parcel, and q_L [parcel + sedimentation] is q_L [parcel] plus the liquid water content lost to sedimentation. In both cases, the comparison of adiabatic q_L [calculated based on Eq. (19) from Pinsky et al. (2013)] with the output of q_L from the parcel model (i.e., $q_L[N_d + \text{sedimentation}]$) shows good agreement, indicating that the mass of the condensate is well conserved in the modeled adiabatic rising parcel. With the outputs of N_d and $q_L[N_d]$ from the parcel model, $[\dot{N}_d]_{\text{cs-PARAM}}$ can be calculated based on Eq. (3). Figures 13b and 13e show the comparison of $[\dot{N}_d]_{\text{cs-PARAM}}$ with $[\dot{N}_d]_{\text{cs-BIN}}$ in the Cu and Sc base cases. The droplet loss rate calculated from the bin microphysics scheme and the parameterization are quite similar, suggesting that Eq. (3) could be a useful parameterization for $[\dot{N}_d]_{\text{cs}}$.

The maximum q_L in the Cu base case is 3 times as high as in the Sc base case because of the higher z_{top} assigned in the Cu base case. Interestingly, as the product of N_d and q_L , the values of $[\dot{N}_d]_{\text{cs}}$ are actually of the same order in two cases (N_d is higher in the Sc base case). Because the Cu and Sc base cases have different initial aerosol concentration assigned in the simulation, a better way to compare the efficiency of collision-coalescence is to use the fractional droplet loss rate given by

$$\frac{[\dot{N}_d]_{\text{cs-PARAM}}}{N_d} = -8 \times 10^4 (N_d)^{-0.45} (q_L)^{1.37}. \quad (4)$$

Figures 13c and 13f show the fractional loss rate of droplets in the Cu and Sc cases. The results indicate that the maximum fractional loss rate in the Sc base case is nearly 5 times smaller than in the Cu base case. In the Cu base case, the fractional loss rate can be higher than $20\% \text{ min}^{-1}$. On the contrary, the maximum fractional loss rate in the Sc base case is lower than $5\% \text{ min}^{-1}$. Given that the initial aerosol concentration in the Sc base case is twice as large as in the Cu base case, this further illustrates that the formation of UCLs is very unfavorable in the Sc regime.

With Eq. (4), the exponential time scale of droplet depletion by collision-coalescence can be written as

$$\tau_{\text{cc}}[s] = \frac{(N_d)^{0.45} (q_L)^{-1.37}}{8 \times 10^4}. \quad (5)$$

Figure 14 shows τ_{cc} calculated based on Eq. (5), indicating that the time scale of droplet depletion by collision-coalescence has strong dependence upon q_L but weak dependence upon N_d . This finding is consistent with the modeling results presented in Feingold et al. (1996), which suggest that the fractional droplet depletion rate by collision-coalescence is a strongly convex function of q_L and is almost independent of N_d [see

FIG. 12. Drop coalescence-scavenging rates estimated by integration of the SCE of Eq. (2) using observed DSDs from CSET against the parameterization given by Eq. (3). The collection kernel used here is described in section 2.

Fig. 9 in Feingold et al. (1996)]. Because q_L is proportional to the height above cloud base, it implies that the cloud-thickness H_t is a more important factor constraining the coalescence-scavenging time scale in the MBL than is the aerosol concentration, as suggested in Wood [2006; see Eq. (18) in Wood (2006)].

b. Fractional loss of droplets $f_{\text{top-loss}}$ and its dependence on U_z , N_d , H_t , and f_{ad}

In this section, we derive a simple formula from Eq. (3) relating $f_{\text{top-loss}}$ with U_z , N_d , H_t , and f_{ad} in order to access the relative importance of different factors in the coalescence-scavenging process. By making an assumption that q_L increases linearly with the height z above cloud base, we can write

$$q_L = f_{\text{ad}} \Gamma_{\text{ad}} z, \quad (6)$$

where Γ_{ad} is the adiabatic rate of increase of liquid water content with respect to height and f_{ad} is the degree of adiabaticity with the range between 0 and 1. In stratocumulus, f_{ad} is often observed to be close to unity, particularly for nondrizzling stratocumulus (Albrecht et al. 1990; Zuidema et al. 2005). However, in cumulus, f_{ad} can be significantly lower because of entrainment and precipitation (Hobbs and Rangno 1996; Rauber et al. 2007). Several observational studies have shown that f_{ad} typically decreases with increasing altitude in cumulus (Hobbs and Rangno 1996; Rauber et al. 2007). Wood et al. (2009) showed that observed f_{ad} in cumulus can be parameterized by $z_0/(z_0 + z)$, where z_0 is a scaling parameter, set to 500 m. Our main purpose here is to derive a simple formula relating $f_{\text{top-loss}}$ with U_z , N_d , H_t ,

FIG. 13. Comparisons of the droplet coalescence-scavenging rates calculated based on the parameterization and the bin microphysics scheme. (a)–(c) The cumulus base case; (d)–(f) the stratocumulus base case. (a),(d) The temporal evolution of droplet concentration N_d (blue), liquid water content inside the parcel q_L [parcel] (black dot-dashed), liquid water content inside the parcel plus liquid water content lost to sedimentation q_L [parcel + sedimentation] (black dotted), and adiabatic liquid water content q_L [ad] (black solid); (b),(e) droplet coalescence-scavenging rates from the parameterization of Eq. (3) and the parcel model; (c),(f) fractional droplet coalescence-scavenging rates from the parameterization of Eq. (4) and the parcel model.

and f_{ad} , which is no longer possible if we retain the height dependence of f_{ad} in the derivation. Therefore, in this study, we assume f_{ad} is constant with height for simplicity. Also, Γ_{ad} is assumed to have a fixed value of $2.1 \text{ g m}^{-3} \text{ km}^{-1}$, which is typical for the MBL clouds in the subtropics and tropics (Albrecht et al. 1990). With Eq. (6), integrated liquid water path from the cloud base to z can be written as

$$\text{LWP}(z) = \frac{f_{ad} \Gamma_{ad} z^2}{2}. \quad (7)$$

By substituting q_L from Eq. (6) into Eq. (3) and dividing Eq. (3) by U_z , the decrease in N_d with increasing altitude can be given by

$$\frac{dN_d}{dz} = \frac{-a(N_d)^{0.55} (f_{ad} \Gamma_{ad} z)^{1.37}}{U_z}. \quad (8)$$

The integration of Eq. (8) yields

$$\frac{(N_d)_z^{0.45} - (N_d)_{\text{base}}^{0.45}}{0.45} = \frac{-a(f_{ad} \Gamma_{ad})^{1.37} z^{2.37}}{2.37 U_z}, \quad (9)$$

where $(N_d)_z$ is the number concentration of droplets at the altitude z and $(N_d)_{\text{base}}$ is the number concentration of droplets activated at the cloud base. Then, by Eq. (9),

the fractional loss of N_d from the cloud base to the cloud top $f_{\text{top-loss}}$ by collision-coalescence can be given by

$$f_{\text{top-loss}} = 1 - \frac{(N_d)_{\text{top}}}{(N_d)_{\text{base}}} = 1 - (1 - \chi_{cc})^{2.2} \quad (10)$$

$$\chi_{cc} = k \frac{(f_{ad})^{1.37} H_t^{2.37}}{(N_d)_{\text{base}}^{0.45} U_z}$$

$$k = 0.19a(\Gamma_{ad})^{1.37}$$

where $(N_d)_{\text{top}}$ is the droplet concentration at the cloud top and χ_{cc} is termed the collision-coalescence efficiency factor. The expression in Eq. (10) shows that $f_{\text{top-loss}}$ increases with the increase in f_{ad} and H_t but decreases with the increase in U_z and $(N_d)_{\text{base}}$. The positive correlation of $f_{\text{top-loss}}$ with f_{ad} and H_t is straightforward. The droplet coalescence-scavenging rate is strongly dependent on q_L (see section 4a), so higher adiabaticity and deeper cloud depth (i.e., higher LWP) both increase the depletion of N_d through collision-coalescence. Regarding the negative correlation of $f_{\text{top-loss}}$ with U_z and $(N_d)_{\text{base}}$, the illustration is already given in sections 3 and 4a. Figure 14 shows contour plots of $f_{\text{top-loss}}$ against U_z , N_d , H_t , and f_{ad} by assuming a reference $U_z = 1 \text{ m s}^{-1}$, $(N_d)_{\text{base}} = 75 \text{ cm}^{-3}$, $H_t = 1000 \text{ m}$, and $f_{ad} = 0.6$. These values are used when they are not the variables in the contour plots (e.g., $U_z = 1 \text{ m s}^{-1}$ and

FIG. 14. Time scale of droplet loss by collision-coalescence calculated by Eq. (5) as a function of liquid water content.

$f_{ad} = 0.6$ are assumed in Fig. 15a). To explore the sensitivity to the assumed reference values, another set of reference values is used [i.e., $U_z = 0.25 \text{ m s}^{-1}$, $(N_d)_{base} = 200 \text{ cm}^{-3}$, $H_t = 300 \text{ m}$, and $f_{ad} = 1$], and the results are shown in Fig. 16. The results calculated using two different sets of reference values are qualitatively very similar, which indicate that H_t is the most dominant factor determining $f_{top-loss}$, and $f_{top-loss}$ is found to be very insensitive to $(N_d)_{base}$. We found that U_z is found to play a more important role in determining $f_{top-loss}$ than does f_{ad} . Overall, the relative importance of these factors determining the fraction loss of droplets in the updraft can be summarized as $H_t > U_z > f_{ad} > (N_d)_{base}$. This finding implies that H_t is a critical factor governing the efficiency of the coalescence-scavenging process. It should be noted that the order of importance of the factors discussed in this section is for the fraction loss in the updraft $f_{top-loss}$, which is different from the indicator for the UCLs, the actual absolute number of droplets and aerosols ($< 10 \text{ cm}^{-3}$). However, the discussion here gives an important implication for the UCLs such that the formation of UCLs could be strongly susceptible to the variability of cloud thickness, which will be the focus of a future study.

c. Effect of parcel cycling on droplet depletion in stratocumulus

Air parcels in stratocumulus could cycle more than one time, and multiple cycles through cloud could have an effect on depletion of aerosol concentration and the associated potential to produce UCLs. The parameterization of droplet fraction loss in the updraft through collision-coalescence [i.e., Eq. (10)] is a useful tool to address this question. In the typical Sc regime, the background aerosol concentration N_{ccn} in the surface mixed layer ranges from 200 cm^{-3} (CSET; see section 2c for details) to $> 400 \text{ cm}^{-3}$ [VAMOS Ocean-Cloud-Atmosphere-Land Study (VOCALS); southeastern Pacific; see Allen et al. 2011]. A recent model study

(Chen et al. 2016) suggested that the number concentration of activated droplet is approximately equal to the initial N_{ccn} in the Sc regime. Here, we simply assumed that the typical initial activated N_d (i.e., before first parcel cycling) in the Sc regime is 300 cm^{-3} . Based on Eq. (10), fraction loss of N_d after one parcel cycling (i.e., $f_{top-loss}$) in the Sc regime (i.e., cloud thickness = 300 m , updraft = 0.25 m s^{-1}) is approximately 30% (see Fig. 16a). Note that the dependence of $f_{top-loss}$ on N_d is relatively small and is neglected in the discussion here. If there is no new aerosol source while parcel cycling, then the number of parcel cycling required for the formation of UCLs in the Sc regime would be 10 (i.e., $300 \text{ cm}^{-3} \times 0.7^{10} = 8.4 \text{ cm}^{-3}$). This indicates that a relatively large number of cycles through cloud is necessary for the formation of UCLs in the typical Sc regime as opposed to the Cu regime, where only one parcel cycle is needed, as shown in section 3. We should also note that a large number of cloud cycles also implies more possibility for aerosol entrainment, which could suppress the formation of UCLs. By using in situ aircraft observations, Terai et al. (2014) found that the POCs of stratocumulus clouds are typically aerosol-depleted UCLs. Based on the discussion given here, we postulate that formation of POCs is strongly associated with aerosol depletion that results from multiple cycles of air through a stratocumulus cloud, a hypothesis that is worthy of investigation in future modeling studies.

5. Conclusions

In Part I, based on the aircraft measurements from CSET, we found that ultraclean layers (UCLs) are common features in the SCT region over the north-eastern subtropical Pacific Ocean and their fractional coverage can exceed $\sim 50\%$ where the PBL height exceeds 1 km . In this paper, the main goal is to test a hypothesis for the formation of UCLs in the MBL, namely, that the deep PBL height over the SCT region can provide high q_L in the active Cu for collision-coalescence to sufficiently deplete N_d , resulting in low N_d and large r_e in clouds that are frequently observed in the upper MBL over the SCT region. Because of the efficient coalescence-scavenging process, after the evaporation of UCL clouds, only a few accumulation mode particles are returned to the clear sky, leaving very low N_{ccn} in the upper MBL.

A rising adiabatic parcel model with a 2D bin microphysics scheme is formulated to test the hypothesis. The results suggest that in the cumulus regime, collision-coalescence can efficiently deplete N_d , grow droplets, and remove N_{ccn} from the atmosphere in Cu updrafts, leading to the occurrence of UCLs in the upper MBL. In

FIG. 15. Contour plots of fractional loss of N_d at the cloud top from the cloud base $f_{\text{top-loss}}$ and its dependence on U_z , $(N_d)_{\text{base}}$, H_t , and f_{ad} calculated using Eq. (10). A reference $U_z = 1 \text{ m s}^{-1}$, $(N_d)_{\text{base}} = 75 \text{ cm}^{-3}$, $H_t = 1000 \text{ m}$, and $f_{\text{ad}} = 0.6$ are assumed when they are not the variables in the plot [e.g., (a) $U_z = 1 \text{ m s}^{-1}$ and $f_{\text{ad}} = 0.6$ are assumed].

Part I, a case study with the geostationary satellite data has revealed that the UCL clouds are often associated with considerable amount of optically thin MBL clouds ($\tau < 0.3$). We postulate that the observed optically thin clouds are often layers detraining from the active Cu towers in the upper MBL. The shallow depth and low N_d in these cloudy detraining layers both contribute to the observed low cloud optical depth. The maintenance

of these optically thin cloudy detraining layers involves a balance between radiation, mesoscale ascent, and cloud microphysics, which is certainly important but beyond the scope of our current paper. We leave that for a future LES study. On the other hand, the parcel modeling suggests that the formation of UCLs is typically not favored in the Sc regime as compared to the Cu regime. In summary, the idealized cloud parcel simulation

FIG. 16. As in Fig. 15, but a reference $U_z = 0.25 \text{ m s}^{-1}$, $(N_d)_{\text{base}} = 200 \text{ cm}^{-3}$, $H_t = 300 \text{ m}$, and $f_{\text{ad}} = 1$ are assumed when they are not the variables in the plot [e.g., (a) $U_z = 0.25 \text{ m s}^{-1}$ and $f_{\text{ad}} = 1$ are assumed].

in this work is able to explain the observed sharp rise of UCL coverage in the SCT region.

A parameterized expression for the droplet coalescence-scavenging rate $[\dot{N}_d]_{\text{cs-PARAM}}$ is derived as a function of bulk parameters N_d and q_L based on the in situ measurements of DSDs from CSET. The comparison of $[\dot{N}_d]_{\text{cs-PARAM}}$ with the droplet loss rate derived from the SCE and the bin microphysics parcel model generally shows good agreement, suggesting that

$[\dot{N}_d]_{\text{cs-PARAM}}$ is a useful parameterization. The derived parameters in $[\dot{N}_d]_{\text{cs-PARAM}}$ imply that the time scale of droplet depletion by collision-coalescence is strongly dependent upon q_L but is weakly dependent upon N_d . An expression for the fractional droplet loss from the cloud base to the cloud top $f_{\text{top-loss}}$ by collision-coalescence is formulated as a function of a collision-coalescence efficiency factor χ_{cc} that depends on the cloud-layer thickness, adiabaticity, updraft velocity, and

cloud-base droplet concentration. Of these factors, cloud thickness is the most dominant factor determining $f_{\text{top-loss}}$, and such finding is consistent with the results presented in Feingold et al. (1996) and Wood (2006). In the SCT region, the variation of cloud thickness is controlled mainly by cloud-top height (i.e., cloud-base heights do not vary as much as cloud-top heights), which is constrained by PBL height. The implication is that PBL depth could play a crucial role governing the temporal and spatial variation of N_d and thus UCLs in the transition region, an issue that will be thoroughly explored in future work.

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