On Which Microphysical Time Scales to Use in Studies of Entrainment-Mixing Mechanisms in Clouds

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Abstract

The commonly used time scales in entrainment-mixing studies are examined to seek the most appropriate one, based on aircraft observations of cumulus clouds from the RACORO campaign and numerical simulations with the Explicit Mixing Parcel Model. The time scales include: $\tau_{\text{evap}}$, the time for droplet complete evaporation; $\tau_{\text{phase}}$, the time for saturation ratio deficit ($S$) to reach $1/e$ of its initial value; $\tau_{\text{satu}}$, the time for $S$ to reach $-0.5\%$; $\tau_{\text{react}}$, the time for complete droplet evaporation or $S$ to reach $-0.5\%$.

It is found that the proper time scale to use depends on the specific objectives of entrainment-mixing studies. First, if the focus is on the variations of liquid water content (LWC) and $S$, then $\tau_{\text{react}}$ for saturation, $\tau_{\text{satu}}$ and $\tau_{\text{phase}}$ are almost equivalently appropriate, because they all represent the rate of dry air reaching saturation or of LWC decrease. Second, if one focuses on the variations of droplet size and number concentration, $\tau_{\text{react}}$ for complete evaporation and $\tau_{\text{evap}}$ are proper because they characterize how fast droplets evaporate and whether number concentration decreases. Moreover, $\tau_{\text{react}}$ for complete evaporation and $\tau_{\text{evap}}$ are always positively correlated with homogeneous mixing degree ($\psi$), thus the two time scales, especially $\tau_{\text{evap}}$, are recommended for developing parameterizations. However, $\psi$ and the other time scales can be negatively, positively, or not correlated, depending on the dominant factors of the entrained air (i.e., relative humidity or aerosols). Third, all time scales are proportional to each other under certain microphysical and thermodynamic conditions.
1. Introduction

Clouds play important roles in radiative transfer and climate [e.g., Betts and Harshvardhan, 1987; Guo et al., 2015; Randall et al., 2016; Yan et al., 2016; Li et al., 2017; Wu et al., 2017; Zhang et al., 2017]. Cloud-climate feedbacks are identified to be one of the largest uncertainties in simulations/projections of climate change [e.g., Bony et al., 2006]. There is still a general lack of knowledge about the mechanisms of cloud-climate feedbacks [Zhang et al., 2013]. One important reason is that understanding and parameterizations of physical processes related to clouds are far from complete [Lin et al., 2015; Zhang et al., 2018], e.g., turbulent entrainment-mixing processes [Donner et al., 2001; Liu et al., 2002]. Previous studies have shown that entrainment-mixing processes affect cloud dynamics (e.g., vertical velocity), thermodynamics (e.g., temperature and buoyancy) and cloud microphysics (e.g., number concentration, droplet size, spectral width), and further affect cloud-climate feedbacks, warm-rain initiation, evaluations of aerosol indirect effects, and radar retrieval of cloud microphysics [e.g., Blyth, 1993; Jiang et al., 2006; Xue and Feingold, 2006; Kim et al., 2008; Del Genio and Wu, 2010; Dawe and Austin, 2013; Lu et al., 2016; Yang et al., 2016].

Turbulent entrainment-mixing mechanisms have been studied for more than 50 years. Homogeneous/inhomogeneous entrainment-mixing mechanisms are the most studied concepts [e.g., Liu et al., 2002; Gerber et al., 2008; Small et al., 2013; Kumar
et al., 2014; Endo et al., 2015; Yum et al., 2015; Lu et al., 2018]. Mixing and droplet evaporation processes occur during entrainment-mixing. If mixing is much faster than droplet evaporation, the entrainment-mixing process is homogeneous whereby all droplets are exposed to the same subsaturated environment and evaporate at the same time, decreasing droplet radius \((r)\) and number concentration \((n)\) if complete evaporation occurs. If droplet evaporation is much faster than turbulent mixing, the entrainment-mixing process is extremely inhomogeneous, with some droplets evaporating completely to saturate the entrained dry air while other droplets are not affected at all. In the extremely inhomogeneous mixing, droplet size does not change but number concentration decreases significantly. The Damkohler number \((Da)\) defined as the ratio of two time scales is often used to quantify which process (mixing or evaporation) is faster,

\[
Da = \frac{\tau_{\text{mix}}}{\tau_r}, \tag{1}
\]

where \(\tau_{\text{mix}}\) is the turbulent mixing time, and \(\tau_r\) is the microphysical response time of droplet population [e.g., Lehmann et al., 2009]. Although there is little controversy with \(\tau_{\text{mix}}\), three microphysical time scales have been used to represent \(\tau_r\): Evaporation time \((\tau_{\text{evap}})\) [Baker et al., 1980; Burnet and Brenguier, 2007; Andrejczuk et al., 2009], phase relaxation time \((\tau_{\text{phase}})\) [Kumar et al., 2013] and reaction time \((\tau_{\text{react}})\) [Lehmann et al., 2009; Lu et al., 2011; 2013; 2014a]. Detailed discussion on all the time scales is deferred to Section 2.

Entrainment-mixing mechanisms have been also studied by employing microphysical mixing diagrams, such as \(r\) vs. \(n\) or liquid water content (LWC)
Homogeneous mixing degree ($\psi$) or similar quantities have been proposed to describe the types of mixing processes [Gerber et al., 2008; Andrejczuk et al., 2009; Lu et al., 2013; 2014b]. Theoretically, a larger $\psi$ corresponds to a smaller $Da$ [e.g., Lehmann et al., 2009], or a larger $\tau_r$ if $\tau_{mix}$ is fixed. $Da < 1$ means that mixing is faster than evaporation and entrainment-mixing processes are more homogeneous, according to the concept of homogeneous/inhomogeneous entrainment-mixing stated above. Andrejczuk et al. [2009] developed a parameterization of entrainment-mixing by relating the slope of the mixing diagram to $Da$. Jarecka et al. [2013] related the slope to the homogeneous mixing degree defined in Morrison and Grabowski [2008] and applied the parameterization in large eddy simulations. Lu et al. [2013; 2014a] developed parameterizations of entrainment-mixing mechanisms by relating homogeneous mixing degree to transition scale number (equivalent to $Da$). Gao et al. [2018] further compared different parameterizations based on the results of a particle-resolved direct numerical simulation (DNS) model.

Although much progress has been made, there are still disagreements about which time scales should be used for studying entrainment-mixing mechanisms. For example, when $\tau_{evap}$ and $\tau_{phase}$ are close to each other, both $\tau_{evap}$ and $\tau_{phase}$ are much smaller than the time for complete evaporation of a droplet or the time required to restore saturation [Lehmann et al., 2009]. To determine the proper time scale to use is critical in view of its central role in characterizing the entrainment-mixing processes.
[Andrejczuk et al., 2009; Lu et al., 2013]. Furthermore, because of the complexity of entrainment-mixing processes, it is possible that different time scales may be more appropriate than others for different scientific objectives and under different conditions. In this study, we theoretically analyze the definitions of time scales and \( \psi \), and compare the relationships between \( \psi \) and different time scales based on in situ measurements and numerical simulations. The objectives of this study are threefold:

1. Compare the relationships between \( \psi \) and the time scales, and reveal the factors affecting the relationships;
2. determine the most appropriate time scales for the objectives and conditions involved;
3. seek to unify the different time scales.

The rest of the paper is organized as follows. Section 2 introduces the definitions of the different time scales and \( \psi \). Section 3 presents the data from the Routine AAF [Atmospheric Radiation Measurement (ARM) Aerial Facility] Clouds with Low Optical Water Depths (CLOWD) Optical Radiative Observations (RACORO) field campaign [Vogelmann et al., 2012] and the relationships between \( \psi \) and time scales. Section 4 introduces the Explicit Mixing Parcel Model (EMPM) and presents the relationships from the model results and the factors affecting the relationships. Section 5 further discusses the proper time scales and the corresponding conditions. Concluding remarks are presented in Section 6.

2. Description of Time Scales and Homogeneous Mixing Degree

2.1. Microphysical Time Scales

2.1.1 Evaporation Time
The evaporation time $\tau_{\text{evap}}$ for a single droplet is defined as follows. In a subsaturated environment, droplet evaporation is described by

$$\frac{dr}{dt} = A \frac{S}{r},$$

(2a)

$$A = \frac{1}{\left( \frac{L_h}{R_v T} - 1 \right) \frac{L_a \rho_L}{K T} + \frac{\rho_l R_v T}{D e_s(T)}},$$

(2b)

where $S$ is saturation ratio deficit or supersaturation [relative humidity (RH) minus 1], $t$ is time, $L_h$ is latent heat, $R_v$ is the specific gas constant for water vapor, $T$ is air temperature, $\rho_L$ is density of liquid water, $K$ is coefficient of thermal conductivity of air, $D$ is coefficient of diffusion of water vapor in air, and $e_s(T)$ is saturation vapor pressure over a plane water surface at temperature $T$ [Rogers and Yau, 1989]. Under the conditions of constant $S$ (equal to initial saturation ratio deficit $S_0$) and $A$, integration of equation (2a) from the adiabatic droplet radius ($r_a$) to 0 yields the expression for the time of droplet of radius $r_a$ to completely evaporate (evaporation time):

$$\tau_{\text{evap}} = -\frac{r_a^2}{2 A S_0},$$

(3)

Note that the environmental air is sub-saturated with a negative $S_0$. Equation (3) indicates that $\tau_{\text{evap}}$ depends on $r_a$ and $S_0$ magnitude; an increase of $r_a$ or a decrease of $S_0$ magnitude prolongs $\tau_{\text{evap}}$. Baker and Latham [1979] and Baker et al. [1980] compared $\tau_{\text{evap}}$ with $\tau_{\text{mix}}$ to discuss spatial scales for different entrainment-mixing mechanisms; they found that entrainment-mixing was dominated by extremely inhomogeneous mechanism for spatial scales in excess of about one meter, except for
the highest levels of turbulence combined with low absolute value of $S_0$. Baker et al. [1984] calculated $\tau_{\text{evap}}$ and concluded that the extent to which $S_0$ was eliminated by total droplet evaporation before the Kolmogorov microscale was reached could be significant. Since then, $\tau_{\text{evap}}$ has been widely used to calculate Da and determine the dominant entrainment-mixing mechanisms [Burnet and Brenguier, 2007; Andrejczuk et al., 2009].

2.1.2. Phase Relaxation Time and Saturation Time

The phase relaxation time ($\tau_{\text{phase}}$) is defined as follows. The equation governing change of $S$ with $t$ is:

$$\frac{dS}{dt} = Q_1 \frac{dz}{dt} - Q_2 \frac{dLWC}{dt},$$

(4)

where $Q_1$ and $Q_2$ are functions of temperature and pressure [Rogers and Yau, 1989], and $z$ is height. In the case of isobaric mixing, the first term on the right hand side of equation (4) is omitted. Using equation (2a) and $LWC = \frac{4}{3} \pi \rho L_r n r^3$, equation (4) becomes

$$\frac{dS}{dt} = -Cn r S,$$

(5a)

$$C = 4 \pi \rho L_r \left[ \frac{RT}{\xi e(T)} + \frac{\varepsilon L_v^2}{PT_c p} \right]$$

$$\left[ \frac{L_h}{R T} - 1 \right] \frac{1}{\rho L_r p L}$$

(5b)

$$\left[ \frac{L_h}{R T} - 1 \right] \frac{1}{\rho L_r p L} \left[ \frac{1}{De(T)} + \frac{\rho L_r R T}{p} \right],$$

where $R'$ is the specific gas constant for dry air, $\varepsilon$ is 0.622, $\xi$ is $R'/R_v$, $P$ is air pressure, and $c_p$ is specific heat at constant pressure [Rogers and Yau, 1989; Kumar et al.,
2013]. Under the conditions of constant $n$ and $C$, taking $r = r_a$ as in equation (3) and integrating equation (5a) from $S_0$ to the final saturation ratio deficit ($S_f$) yields:

\[ \int_{S_0}^{S_f} \frac{dS}{S} = - \int_0^{t_f} Cnr_a dt. \]  

So the time ($t_f$) needed for $S_0$ to reach $S_f$ is

\[ t_f = - \frac{1}{Cnr_a} \ln \frac{S_f}{S_0}. \]  

Based on equation (7), $\tau_{\text{phase}}$ is defined as the e-folding time during which $S_0$ reaches $S_0/e$, such that

\[ \tau_{\text{phase}} = \frac{1}{Cnr_a}. \]  

Equation (8) indicates that $\tau_{\text{phase}}$ is a function of $n$ and $r_a$ (microphysical properties), but independent of $S_0$.

After $\tau_{\text{phase}}$ was first introduced to cloud physics by Squires [1952], $\tau_{\text{phase}}$ has been widely used in many aspects of cloud physics. First, quasi-steady supersaturation can be calculated from $\tau_{\text{phase}}$. Politovich and Cooper [1988] derived that quasi-steady supersaturation was proportional to the ratio of vertical velocity in clouds to $\tau_{\text{phase}}$. By comparing $\tau_{\text{phase}}$ with the characteristic time scale of vertical turbulent fluctuation, Korolev and Mazin [2003] concluded that the quasi-steady supersaturation gave a good estimate of supersaturation in liquid and mixed-phase clouds; but in ice clouds, estimation of supersaturation should be made with caution. Second, fluctuation and cloud droplet spectral broadening in clouds is related to $\tau_{\text{phase}}$. Ditas et al. [2012] found that $\tau_{\text{phase}}$ was a good time scale to explain different fluctuations of supersaturation under different conditions of number concentration and droplet size. Clark and Hall
[1979] found that the horizontal variability of all the thermodynamic fields increased significantly with $\tau_{\text{phase}}$. Similarly, Korolev [1995] found that an increase in $\tau_{\text{phase}}$ leads to an increase in fluctuation of liquid water content. Considering the different relationships between $\tau_{\text{phase}}$ and the characteristic Lagrangian time scale of turbulent fluctuations, Khvorostyanov and Curry [1999] developed a general kinetic equation of stochastic condensation to improving understanding of cloud droplet spectral broadening. Chandrakar et al. [2016] concluded that an increase in the width of the droplet size distribution for low aerosol concentration was consistent with larger variability of supersaturation due to the slow microphysical response, i.e., $\tau_{\text{phase}}$ is larger than turbulence correlation time scale. Austin et al. [1985] found that the fluctuation of the reciprocal of $\tau_{\text{phase}}$ was not related to the fluctuation of vertical velocity, but they claimed that such a behavior did not invalidate the general hypothesis that turbulent velocity fluctuations in a nonadiabatic region could broaden observed droplet size distribution. Third, many studies have used $\tau_{\text{phase}}$ in calculation of Damkohler number in studies of entrainment-mixing processes [e.g., Kumar et al., 2013; Pinsky et al., 2016a; Pinsky et al., 2016b]. Jensen and Baker [1989] pointed out that when $\tau_{\text{evap}} > \tau_{\text{phase}}$, entrained air rapidly saturates and the total evaporation of droplets is thus limited, whereas when $\tau_{\text{evap}} < \tau_{\text{phase}}$, some droplets evaporate completely before the entrained air is saturated. Based on Damkohler numbers calculated with $\tau_{\text{evap}}$ and $\tau_{\text{phase}}$ in six direct numerical simulation runs, Kumar et al. [2014] concluded that the response of the cloud system was governed by the smaller of $\tau_{\text{evap}}$ and $\tau_{\text{phase}}$. This is also illustrated in Fig. 1 of Tölle and Krueger [2014]. Pinsky
et al. [2016a; 2016b] claimed $\tau_{\text{phase}}$ was the right time scale to calculate Damkohler number to study evolutions of droplet size, number concentration and droplet size distribution during entrainment-mixing.

As mentioned above, $\tau_{\text{phase}}$ is independent of $S_0$ (initial RH). For example, $\tau_{\text{phase}}$ is the same for initial RH = 30% and 90%. However, after phase relaxation in the time of $\tau_{\text{phase}}$, $S_f$ is different. If initial RH is equal to 30%, $S_f = (\text{RH-1})/e$ is around -25%, which is far from saturation. If initial RH is equal to 90%, $S_f$ is around -4%, which is much closer to saturation than the previous case. It is well-known that RH is critical to entrainment-mixing processes [e.g., Lu et al., 2013], but $\tau_{\text{phase}}$ is not able to reflect the variation of RH. To overcome this weakness, we introduce saturation time scale ($\tau_{\text{sat}}$) that corresponds to $S_f = -0.5\%$ (RH = 99.5%)

$$
\tau_{\text{sat}} = -\frac{1}{Cn r_a} \ln \frac{-0.005}{S_0}
$$

(9)

Compared to $\tau_{\text{phase}}$, $\tau_{\text{sat}}$ depends on $S_0$ in addition to microphysical properties.

2.1.3. Reaction Time

Lehmann et al. [2009] discussed the disadvantages of $\tau_{\text{evap}}$ and $\tau_{\text{phase}}$, i.e., the restriction of constant $S$ in calculation of $\tau_{\text{evap}}$ and the restriction of constant $n$ and $r$ in calculation of $\tau_{\text{phase}}$, respectively. They argued that both $S$ and $r$ vary with time and thus equations (2a) and (5a) constitute a coupled system of differential equations. To consider the variations of $S$ and $r$ during the evaporation process, they defined reaction time $\tau_{\text{react}}$ as the time for droplet complete evaporation or RH to reach 99.5%. Here, if the first condition (droplet complete evaporation) is satisfied, $\tau_{\text{react}}$ is written
as $\tau_{\text{react}}$; if the second condition (RH to reach 99.5%) is satisfied, $\tau_{\text{react}}$ is written as $\tau_{\text{react}}$. Lehmann et al. [2009] applied $\tau_{\text{react}}$ to analyze entrainment-mixing mechanisms in cumulus clouds. Using $\tau_{\text{react}}$, Lu et al. [2011; 2013; 2014a] analyzed entrainment-mixing mechanisms in stratocumulus and cumulus clouds from in situ observations and model results, and developed parameterizations of entrainment-mixing mechanisms.

All four time scales are summarized in Table 1, including their definitions, assumptions, equations, etc. It is noteworthy that $\tau_{\text{phase}}$ is a relative parameter because it represents the time during which $S_0$ reaches $S_0/e$, whereas $\tau_{\text{evap}}$, $\tau_{\text{satu}}$ and $\tau_{\text{react}}$ are absolute times during which $r_a$ becomes 0 or $S_0$ reaches -0.5%. It would be preferable to use symbols to distinguish (e.g., symbol “$\tau$” and “$\tau$” for relative and absolute times, respectively); but the same notations, $\tau_{\text{phase}}$, $\tau_{\text{evap}}$, $\tau_{\text{satu}}$ and $\tau_{\text{react}}$, are used here for presentation convenience. The difference between relative and absolute definitions makes comparison of the numerical values difficult. Therefore, we do not compare the numerical values of the time scales, but focus on the relationships between the time scales and $\psi$.

In calculations of $\tau_{\text{phase}}$, $\tau_{\text{satu}}$, and $\tau_{\text{react}}$, $n$ can be either adiabatic droplet number concentration ($n_a$) or number concentration after entrainment but before mixing and evaporation ($n_0$). Only the time scales calculated with $n_0$ are hereafter included, because $n_0$ is a better choice than $n_a$ in entrainment-mixing studies [Kumar et al., 2013; Lu et al., 2013]. Our analysis shows that the results are similar if $n_a$ is used. The above discussion on the time scales is for monodisperse droplet size distributions.
where all different types of mean radius are equal. For polydisperse droplet size distributions in measurements and numerical simulations to be discussed next, volume-mean radius is used.

2.2. Homogeneous Mixing Degree

As introduced above, the homogeneous and extremely inhomogeneous entrainment-mixing mechanisms are two extreme types of entrainment-mixing processes. However, entrainment-mixing processes often fall between the above two extremes [Andrejczuk et al., 2009; Lu et al., 2011]. Lu et al. [2013; 2014a] proposed a microphysical measure — $\psi$, to unify the representation of the two limiting and intermediate mixing types with the so-called $r$-$n$ mixing diagram. The original figure that explained the concept in Lu et al. [2013] is reproduced in Fig. 1. The two lines starting from the point (1, 1) correspond to homogeneous mixing with RH of 66% in dry air and extremely inhomogeneous mixing, respectively. An entrainment-mixing event includes three states. State 1 is adiabatic. State 2 is just after entrainment but before evaporation. From State 2 to 3, mixing and evaporation occur. The final state should be located along the contour of $\gamma$ (the ratio of liquid water content in the cloud affected by entrainment-mixing (LWC$_c$) to its adiabatic value). In this example, $\gamma$ is assumed to be 0.2. States 3’ and 3’’ correspond to the extremely inhomogeneous and homogeneous mixing scenarios, respectively. The angle between the line 2-3’’ and the line 2-3’ is $\pi/2$ and the angle between the line 2-3 and the line 2-3’ is $\beta$. Briefly, $\beta$ and $\psi$ are given by
\[ \beta = \tan^{-1}\left(\frac{1 - \frac{r_c^3}{r_a^3}}{\frac{n_0 - n_c}{n_a}}\right), \quad (10a) \]

\[ \psi = \frac{\beta}{\pi/2}, \text{ for } n_c < n_0, \quad (10b) \]

or

\[ \psi = \frac{\beta}{\pi/2} + 2, \text{ for } n_c \geq n_0. \quad (10c) \]

where \( r_c \) and \( n_c \) are, respectively, droplet radius and number concentration in the cloud affected by entrainment-mixing. When \( \psi \) is larger, State 3 moves closer to 3'', i.e., mixing is more homogeneous. When \( \psi \) is smaller, State 3 moves closer to 3', i.e., mixing is more inhomogeneous.

3. Observational Analysis

3.1 RACORO Data

The Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft was used during RACORO, which operated over the ARM Southern Great Plains (SGP) site near Lamont, Oklahoma from 22 January to 30 June 2009 [Vogelmann et al., 2015]. The objectives were to obtain representative statistics of cloud microphysical properties needed to support process studies and model simulations of boundary layer clouds and to validate retrieval algorithms. The instruments for observing temperature and water vapor at 10 Hz (equivalent to 5 m spatial sampling) were a Rosemount probe and a Diode Laser Hygrometer (DLH) [Diskin et al., 2002], respectively. Aerosol and cloud droplet size distributions from 0.29 to 25 µm in radius were collected by the Cloud and Aerosol Spectrometer (CAS)
at 10 Hz. Only the cloud droplets with a bin-average radius bigger than 1 µm are included in calculations of microphysical properties, e.g., $n$. The sampling criteria for cloud droplet size distributions are $n$ larger than 10 cm$^{-3}$ and LWC larger than 0.001 g m$^{-3}$ [e.g., Deng et al., 2009]. Droplets in the range of 7.5 to 781 µm in radius were observed by a Cloud Imaging Probe (CIP) at 1 Hz. The criterion of the in-cloud drizzle water content (radius $> 25$ µm) averaged over the observation period of the flight smaller than 0.005 g m$^{-3}$ is used to select the non-drizzling cumulus clouds. In total, there are 186 non-drizzling growing cumulus clouds from eight flights (May 22, May 23, May 24, June 11, June 19, June 23, June 24, and June 26, 2009); the same dataset was used in Cheng et al. [2015] and Lu et al. [2014b]. As shown in Table 2, the total number of cloud droplet size distributions was 13652 from 186 clouds. The mean values of $r_c$, $n_c$ and LWC$_c$ are 924.7 cm$^{-3}$, 3.5 µm and 0.196 g m$^{-3}$, respectively.

The standard deviations of the three properties are also shown in Table 2.

### 3.2 Relationships in RACORO

The data from the 186 non-drizzling growing cumulus clouds are used to study the relationships between $\psi$ and different time scales, including $\tau_{\text{evap}}$, $\tau_{\text{react}}$, $\tau_{\text{satu}}$ and $\tau_{\text{phase}}$ (Fig. 2). The maximum number concentration in a cloud is assumed to be $n_a$ [Lu et al., 2014a]. $n_0$ is equal to $n_a \chi$, where $\chi$ is the mixing fraction of adiabatic cloud during the isobaric mixing between adiabatic cloud and dry air, and is calculated based on the conservations of total water and energy [Lu et al., 2012; Lu et al., 2014a]. The maximum liquid water content is assumed to be adiabatic liquid water content...
(LWCₐ) [Lu et al., 2012], and $r_a$ is calculated by:

$$r_a = \left(\frac{LWC}{4/3 \pi n_a}\right)^{1/3}.$$  \hfill (11)

The mean number concentration and volume-mean radius of cloud droplets measured in a cloud are taken to be $n_c$ and $r_c$, respectively. $n_a$, $n_0$, $n_c$, $r_a$ and $r_c$ are used to calculate $\psi$. $n_0$ and $r_a$ are used in calculations of $\tau_{evap}$, $\tau_{react}$, $\tau_{satu}$ and $\tau_{phase}$.

The clouds are classified into two groups according to the conditions in calculation of $\tau_{react}$ in Fig. 2. If the first condition (droplet complete evaporation) is satisfied, the cloud is labeled as “Evaporated” and shown in red; if the second condition (RH to reach 99.5%) is satisfied, the cloud is in the category of “Saturated” and marked in blue. The purpose of the classification is to see if the often claimed argument is correct or not that $\tau_{evap}$ is more appropriate in studies of entrainment-mixing processes when $\tau_{evap} < \tau_{phase}$ (the “Evaporated” group), whereas $\tau_{phase}$ is more appropriate when $\tau_{evap} > \tau_{phase}$ (the “Saturated” group), as mentioned above. In calculations of $\psi$, $\tau_{evap}$, $\tau_{react}$, $\tau_{satu}$ and $\tau_{phase}$, the properties of dry air entrained into clouds is taken from $d$ to $2d$ away from the edges of the cloud cores [Lu et al., 2012]. Lu et al. [2012] assumed many different dry air sources to estimate entrainment rate by setting $d = 10, 20, 30, 40, 50, 100, 300, 500$ m. Here $d = 10$ m is used because the dry air closer to the cloud cores is more likely to be entrained into the cores than the dry air farther away. The conclusions are similar even if other $d$ values are used. The edge of the cloud core is defined as the point along the direction from the cloud edge toward the interior of the cloud where vertical velocity changes from negative to positive for the first time (Figure 1a in Lu et al. [2012]).
Three points are noteworthy from Fig. 2. First, $\psi$ is positively correlated with $\tau_{\text{evap}}$ for both groups. Second, $\psi$ is positively correlated with $\tau_{\text{react}_e}$ for the “Evaporated” group; however, $\psi$ is negatively correlated with $\tau_{\text{react}_s}$ for the “Saturated” group. The positive $\psi$-$\tau_{\text{react}_e}$ correlation is easy to understand because $\tau_{\text{react}_e}$ is similar to $\tau_{\text{evap}}$. To understand the reasons for the negative correlation, $\psi$ is plotted against $\tau_{\text{satu}}$, because the criterion of RH being equal to 99.5% is the same in calculations of $\tau_{\text{react}_s}$ and $\tau_{\text{satu}}$.

Figure 2c shows that $\psi$ and $\tau_{\text{satu}}$ tend to be negatively correlated, although the $p$ values are larger than 0.05. The two data points with the smallest $\psi$ have the largest $r^3_c/a^3$ in the “Evaporated” group (not shown). Although it is somewhat arbitrary, removing these two data points leads to a negative correlation with correlation coefficient (R) = -0.21 and $p = 0.03$ in the “Evaporated” group. If all the data in the “Evaporated” and “Saturated” groups are put together, R is -0.31 with $p < 0.01$, and excluding the two points with the smallest $\psi$ changes R to -0.38 with $p < 0.01$. The negative correlation between $\psi$ and $\tau_{\text{satu}}$ explains why the correlation between $\psi$ and $\tau_{\text{react}_s}$ is negative.

When it takes shorter time to restore saturation, it is more likely that mixing is homogeneous. Third, $\psi$ does not appear to have any noted correlation with $\tau_{\text{phase}}$ for either group (“Evaporated” or “Saturated”).

As stated in the introduction, a larger $\psi$ corresponds to a smaller Da, or a larger $\tau_r$ if $\tau_{\text{mix}}$ is fixed. The results in Fig. 2 do not support the conclusion that $\tau_{\text{phase}}$ is more appropriate when $\tau_{\text{evap}} > \tau_{\text{phase}}$. See more discussions in Section 5.

4. Numerical Investigation
Section 3 shows the relationships between $\psi$ and time scales based on observations. Some relationships are complicated, which could be related to different factors that are difficult to distinguish based on observations. To dissect physical mechanisms underlying the observational relationships shown in Fig. 2, here the EMPM model is used to perform sensitivity studies to examine the effects of different factors on the relationships between $\psi$ and the time scales.

4.1. EMPM Model

Krueger et al. [1997] and Su et al. [1998] developed the EMPM model based on a linear eddy model developed by Kerstein [1988; 1992]. As summarized by Krueger [2008] and Tölle and Krueger [2014], the parcel is represented by a one-dimensional model domain that ascends adiabatically above cloud base; when entrainment occurs, the entrained dry air replaces a same-sized segment of the cloudy parcel; the dry air and cloudy air mix and droplets evaporate.

The setting of EMPM is similar to Lu et al. [2013]. The domain size is 20 m × 0.001 m × 0.001 m; the entrained dry air blob size is 2 m × 0.001 m × 0.001 m. The lengths of the horizontal sides are so different because the domain is one-dimensional, as stated above. The parcel moves upward at the speed of 2 m s$^{-1}$ from cloud base and stops at the entrainment level of 888.9 hPa. The pressure, water vapor mixing ratio and temperature at cloud base are 964.0 hPa, 15.7 g kg$^{-1}$ and 293.6 K, respectively.
Temperature and RH in environment at the entrainment level are 289.3 K and 11%, 22%, 44%, 66%, 77%, 88%, 95%, respectively. Different from Lu et al. [2013], RH = 77% and 95% are added to study the behaviors of time scales in entrainment-mixing when RH is high. The dissipation rate is $1 \times 10^{-3}$ m$^2$ s$^{-3}$ and the number of dry air blobs entrained during each entrainment event is 2, i.e., $\chi = 0.2$. Both dissipation rate and blob number are fixed to focus on the microphysical time scales, i.e., the denominator of equation (1). Setting dissipation rate to $1 \times 10^{-5}$ and $1 \times 10^{-2}$ m$^2$ s$^{-3}$ does not change the conclusions shown here although the values of $\psi$ are different for different dissipation rates. $n_0$ is 102.7, 205.4, 308.1, 410.8 and 513.5 cm$^{-3}$. $n_0$ is the number concentration at 375.75 s when dry air is entrained into clouds but before mixing and evaporation. $n_0$ calculated using this method is very close to $n_0\chi$. $r_a$ is the volume-mean radius at 375.75 s. $n_c$ and $r_c$ are, respectively, the droplet number concentration and volume-mean radius at the final states.

### 4.2 Relationships in EMPM

The EMPM results are shown in Fig. 3. Similar to Fig. 2a, $\psi$ and $\tau_{\text{evap}}$ are positively correlated for both groups: “Evaporated” and “Saturated” (Fig. 3a). It is noteworthy that $\tau_{\text{evap}}$ works well even when the entrained dry air is close to saturation (RH = 95%). In this case, $\psi$ is close to 1, the largest value. Correspondingly, $\tau_{\text{evap}}$ is also the largest, consistent with the theoretical analysis based on the Da (equation (1)). The correlations between $\psi$ and $\tau_{\text{react}}$ are generally similar to those in RACORO: $\psi$ is positively correlated with $\tau_{\text{react,es}}$ but tends to be negatively correlated with $\tau_{\text{react,s}}$ (Fig.
3b). However, the $\psi$-$\tau_{\text{satu}}$ relationship (Fig. 3c) is quite different from Fig. 2c; $\psi$ tends to be positively correlated with $\tau_{\text{satu}}$ for the “Evaporated” group, but virtually no correlation for the saturation group. Also different from the RACORO, the correlation between $\psi$ and $\tau_{\text{phase}}$ tends to be positive for both groups (Fig. 2d and 3d). As will be detailed in Section 4.3, the large but regular scatters in EMPM are likely related to aerosol concentrations in the environmental air and RH.

### 4.3 Effects of RH and Aerosols

RH and aerosols affect cloud microphysical properties [Fan et al., 2013; Wang et al., 2013] and further affect entrainment-mixing mechanisms. To reveal the roles of RH and aerosols (represented by $n_a$) in shaping the relationships between $\psi$ and the time scales, Fig. 3 is replotted in Fig. 4. Figure 4 shows the dependence of $\psi$ on different time scales with different RH values (color scheme) and aerosol concentrations (dot sizes). For a constant aerosol concentration, as shown in Fig. 4a, $\psi$ and $\tau_{\text{evap}}$ both increase with increasing RH, suggesting that RH variation leads to a positive correlation between $\psi$ and $\tau_{\text{evap}}$. The effect of RH on $\psi$ vs. $\tau_{\text{react}}$ exhibits two types of behaviors (Fig. 4b). For the “Evaporated” group, $\psi$ vs. $\tau_{\text{react, e}}$ is similar to $\psi$ vs. $\tau_{\text{evap}}$. However, for the “Saturated” group (the upper points), the RH-induced relationship between $\psi$ and $\tau_{\text{react, s}}$ is similar to that between $\psi$ and $\tau_{\text{satu}}$ (Fig. 4c). When RH is lower, $\tau_{\text{satu}}$ becomes larger based on equation (9), i.e., it takes a longer time to
reach RH = 99.5%. Thus, $\psi$ is negatively correlated with $\tau_{\text{react}}$ and $\tau_{\text{satu}}$ when RH varies. Different from the other time scales, $\tau_{\text{phase}}$ does not have any correlation with RH and therefore there is no correlation between $\psi$ and $\tau_{\text{phase}}$ resulting from variations of RH (Fig. 4d).

If RH is kept constant (e.g., the yellow points), variations of aerosol concentrations cause positive correlations between $\psi$ and all four time scales. Note that, in all the EMPM simulations, the entrainment level is the same (888.9 hPa). So LWC$_a$ is the same for the simulations. Mixing fraction of dry air is the same because the number of dry air blobs is 2 for all simulations; so liquid water content just after entrainment but before mixing and evaporation (LWC$_0$) is the same. When RH is fixed, LWC$_c$ is also the same. A larger $n_a$ corresponds to smaller droplet sizes and faster loss of droplets due to complete evaporation, so $n_c/n_a$ in the denominator of equation (10a) becomes smaller. Because

$$\frac{\text{LWC}_c}{\text{LWC}_a} = \frac{r_c^3 n_c}{r_a^3 n_a},$$

(12)

$r_c^3/r_a^3$ in the numerator of equation (10) becomes larger. Thus, $\psi$ decreases with increasing $n_a$. According to equation (3), $\tau_{\text{evap}}$ is smaller because $r_a$ is smaller. Therefore, $\psi$ and $\tau_{\text{evap}}$ are positively correlated. Why the other time scales are also positively correlated with $\psi$ in the EMPM when RH is fixed can be understood from the following analysis. The ratio of $\tau_{\text{evap}}$ to $\tau_{\text{phase}}$ can be expressed as

$$\frac{\tau_{\text{evap}}}{\tau_{\text{phase}}} = -\frac{C n_0 r_0^3}{2 A S_0} = \frac{\text{LWC}_0}{S_0} = \frac{\text{LWC}_0}{S_0} = B_1.$$  

(13)

When the ratio $B_1 = \text{LWC}_0/S_0$ is constant, $\tau_{\text{evap}}$ is proportional to $\tau_{\text{phase}}$. Baker et al.
[1984] also defined a similar quantity to study the influence of mixing on cloud droplet size distributions. The physical meaning of equation (13) is discussed in the next section. Similarly, the ratio of $\tau_{\text{evap}}$ to $\tau_{\text{satu}}$ can be expressed as

$$\frac{\tau_{\text{evap}}}{\tau_{\text{satu}}} = \frac{1}{\ln \frac{-0.005}{S_0}} \frac{Cn_0/r_a^3}{2AS_0} \sim \frac{1}{\ln \frac{-0.005}{S_0}} \frac{\text{LWC}_0}{S_0} = B_2. \quad (14)$$

When $B_2$ is constant, $\tau_{\text{evap}}$ is proportional to $\tau_{\text{satu}}$. Both $B_1$ and $B_2$ are constant if it is assumed that aerosol concentration is the only one that varies ($S_0$ and LWC$_0$ are both constant), so all the time scales are proportional to $\tau_{\text{evap}}$ and positively correlated with $\psi$.

To check if the RACORO data show a similar behavior, Fig. 5 examines the relationships between $\psi$ and the time scales for a narrow range of LWC$_0$/S$_0$ (-0.2 ~ -0.1 g m$^{-3}$). Consistent with theoretical expectation, $\psi$ is positively correlated with all time scales except for $\tau_{\text{satu}}$. The behavior of $\tau_{\text{satu}}$ is due to that, unlike in the EMPM simulations, a constant $B_1$ does not mean a constant $B_2$. In the EMPM results (Fig. 4), $S_0$ is constant for the data points with the same colors, and therefore

$$\frac{1}{\ln \frac{-0.005}{S_0}} \frac{\text{LWC}_0}{S_0}$$

is constant; so $\tau_{\text{evap}}$ is proportional to $\tau_{\text{satu}}$ and $\psi$ is positively correlated with $\tau_{\text{satu}}$ in EMPM. But in RACORO, $S_0$ varies from cloud to cloud, so $\psi$ is negatively correlated with $\tau_{\text{satu}}$.

These results suggest that variations in RH and aerosols exert similar effects on the variations of $\psi$, $\tau_{\text{react \_e}}$, and $\tau_{\text{evap}}$, so $\psi$ is positively correlated with the two time scales. But RH and aerosols play opposite roles in determining the relationship
between $\psi$ and $\tau_{\text{react,ss}}$, and also the relationship between $\psi$ and $\tau_{\text{satu}}$. Different combinations of the effects of RH and aerosols make the relationships complicated (Figs. 3b, 3c); many of them have low correlation coefficients and/or $p$ values larger than 0.05. As for $\tau_{\text{phase}}$, although it is affected by aerosols, not RH, the data points are scattered due to the effect of RH on $\psi$. Thus the correlation between $\psi$ and $\tau_{\text{phase}}$ tends to be positive (correlation coefficients larger than 0.43) but the $p$ value is larger than 0.05 for the “Saturated” group.

5. Discussions

There are two major objectives of using the various time scales: the variations of $S$ and LWC and the variations of $r$ and $n$. For the first objective, $\tau_{\text{react,ss}}$, $\tau_{\text{phase}}$, and $\tau_{\text{satu}}$ represent how fast the droplet system relaxes saturation, and thus the variations of $S$. Furthermore, neglecting the first term on the right hand side of equation (4) for isobaric mixing, the temporal variation of $S$ is then proportional to that of LWC. In other words, if the interest is in the variations of $S$ and LWC during isobaric mixing processes, then $\tau_{\text{react,ss}}$, $\tau_{\text{phase}}$, and $\tau_{\text{satu}}$ should be appropriate (Table 1). This may be the reason why $\tau_{\text{phase}}$ has been widely used in the studies on the evolution of $S$ in cloud physics [Kostinski, 2009; Chandrakar et al., 2017; Siebert and Shaw, 2017].

However, it is known that an infinite number of combinations of $n$ and $r$ can yield the same LWC ($\text{LWC} = \frac{4}{3} \pi \rho_l nr^3$), thus, these time scales do not seem to fit the
second objective that focuses on the variations of $n$ and $r$ and their relationships with entrainment-mixing mechanisms. For the second objective, it is critical to know if some droplets are completely evaporated and if number concentration decreases. Thus knowing how fast droplets evaporate (represented by $\tau_{\text{react},e}$ and $\tau_{\text{evap}}$) is critical. The variation of LWC is important, but the final LWC could be the same even if entrainment-mixing mechanisms are different. As mentioned above, $\tau_{\text{react},s}$, $\tau_{\text{phase}}$ and $\tau_{\text{satu}}$ represent how fast the cloud reaches saturation after entrainment-mixing. However, the process of increasing RH could be due to complete evaporation of some droplets with decreasing number concentration, or due to evaporation of all droplets at the same time with number concentration unchanged. Therefore, $\tau_{\text{react},s}$, $\tau_{\text{phase}}$, and $\tau_{\text{satu}}$ cannot be used to check whether some droplets are completely evaporated or not; but, $\tau_{\text{react},e}$ and $\tau_{\text{evap}}$ can. In addition, according to the definition of Da, when Da $>>$ 1, extremely inhomogeneous mixing dominates; when Da $<<$ 1, homogeneous mixing dominates [Burnet and Brenguier, 2007; Lehmann et al., 2009; Kumar et al., 2014; Yeom et al., 2017]. If $\tau_{\text{mix}}$ is fixed, larger $\tau_{r}$ corresponds to a greater degree of homogeneous mixing, i.e., a positive correlation between $\psi$ and $\tau_{r}$. Based on the results in RACORO and EMPM, $\psi$ and $\tau_{\text{react},e}$, and $\psi$ and $\tau_{\text{evap}}$ are always positively correlated, while the correlations between $\psi$ and $\tau_{\text{react},s}$, $\psi$ and $\tau_{\text{phase}}$, and $\psi$ and $\tau_{\text{satu}}$ vary, depending on whether RH or aerosols dominate the cloud microphysics. These results suggest that $\tau_{\text{react},e}$ and $\tau_{\text{evap}}$ are good choices (Table 1). Furthermore, $\tau_{\text{react},e}$ is better than $\tau_{\text{evap}}$ in principle because it considers the variation of $S$ whereas $\tau_{\text{evap}}$ assumes a constant $S$. This is evident from the larger correlation coefficients between
\( \psi \) and \( \tau_{\text{react},e} \) compared to those between \( \psi \) and \( \tau_{\text{evap}} \) for both RACORO (Fig. 2) and EMPM (Fig. 3) results. However, this is only true for the “Evaporated” group. For the “Saturated” group, \( \tau_{\text{react},s} \) is close to \( \tau_{\text{satu}} \) and the correlation between \( \psi \) and \( \tau_{\text{react},s} \) is negative. Importantly, \( \tau_{\text{evap}} \) covers all the different conditions, and thus is recommended in practice. Another reason for recommending \( \tau_{\text{evap}} \) is its simpler expression. It has been increasingly recognized that the effects of entrainment-mixing processes on the variations of \( n \) and \( r \) need to be parameterized in order to obtain realistic microphysical properties [Chosson et al., 2007; Hill et al., 2009; Jarecka et al., 2013; Endo et al., 2015]; therefore, \( \tau_{\text{evap}} \) or \( \tau_{\text{react},e} \), especially \( \tau_{\text{evap}} \), is recommended for developing parameterizations of entrainment-mixing processes. In addition, it is well-known that entrainment-mixing processes affect aerosol indirect effects [Kim et al., 2008]. As shown in Fig. 4, aerosols affect the degree of homogeneous mixing. Therefore, aerosols affect entrainment-mixing mechanisms and further affect aerosol indirect effects; the interactions between aerosol and entrainment-mixing processes need further investigations to improve aerosol-cloud parameterizations in models.

It is noteworthy that, as discussed above, the different time scales can be equivalent to one another under certain conditions, e.g., when \( \frac{\text{LWC}_0}{S_0} \) and/or \( \frac{1}{\ln \left( \frac{S_0}{S_0} \right)} \) are constant (Table 1). Rewriting equation (13), \( \tau_{\text{evap}} \) and \( \tau_{\text{phase}} \) are related to each other in the form:

\[
\frac{\text{LWC}_0}{\tau_{\text{evap}}} \sim \frac{S_0}{\tau_{\text{phase}}}. \tag{15}
\]

This equation quantitatively indicates that the time to decrease LWC due to droplet
evaporation (represented by $\tau_{\text{evap}}$) is proportional to the time to increase $S$ (represented by $\tau_{\text{phase}}$). The relationship between $\tau_{\text{evap}}$ and $\tau_{\text{satu}}$ is similar.

6. Concluding Remarks

The commonly used time scales in investigation of turbulent entrainment-mixing processes are examined by use of the in situ observations from the RACORO campaign, numerical simulations from the EMPM model, and theoretical analyses. It is found that the best time scales to use depend on the specific scientific objectives and conditions in question, and can be classified into three major categories.

First, if the scientific objective is to evaluate the variations of liquid water content (LWC) and supersaturation/saturation ratio deficit ($S$) in entrainment-mixing processes, reaction time scale ($\tau_{\text{react}}$) for the “Saturated” group, phase relaxation time scale ($\tau_{\text{phase}}$) and saturation time scale ($\tau_{\text{satu}}$) are the appropriate time scales. The reason is that these time scales represent how fast $S$ increases and LWC decreases and the variation rate of $S$ is proportional to that of LWC during isobaric mixing.

Second, if the scientific objective is to evaluate how droplet size and number concentration change during entrainment-mixing processes, the problem is more complicated than the first one. In RACORO, it is found that homogeneous mixing degree ($\psi$) is positively correlated with $\tau_{\text{evap}}$, but negatively correlated with $\tau_{\text{satu}}$ for both groups. For $\tau_{\text{react}}$, it is close to $\tau_{\text{evap}}$ and positively correlated with $\psi$ for the “Evaporated” group whereas for the “Saturated” group, $\tau_{\text{react}}$ is close to $\tau_{\text{satu}}$ and negatively correlated $\psi$. It is interesting that there is no correlation between $\psi$ and
\( \tau_{\text{phase}} \). In the EMPM simulations, the relationships between \( \psi \), \( \tau_{\text{evap}} \), and \( \tau_{\text{react}} \) are similar to those in RACORO. In contrast, \( \psi \) generally tends to be positively correlated with \( \tau_{\text{satu}} \) and \( \tau_{\text{phase}} \) (though some \( p \) values are larger than 0.05), different from those in RACORO. Further analysis shows that the relationships are significantly affected by relative humidity (RH) and aerosols in the environment. In the EMPM results, RH variation causes positive correlations between \( \psi \), \( \tau_{\text{react}} \) for the “Evaporated” group, and \( \tau_{\text{evap}} \), negative correlations between \( \psi \), \( \tau_{\text{react}} \) for the “Saturated” group, and \( \tau_{\text{satu}} \), and no correlation between \( \psi \) and \( \tau_{\text{phase}} \). Aerosol variation causes positive correlations between \( \psi \) and all time scales. The combination of the effects of RH and aerosols is responsible for weak and complicated relationships between \( \psi \) and some time scales as mentioned above.

Theoretical analysis based on the Damkohler number shows that \( \psi \) should be positively correlated with the microphysical time scales. \( \tau_{\text{react}} \) for the “Evaporated” group and \( \tau_{\text{evap}} \) are always positively correlated with \( \psi \). For the “Evaporated” group, we argue that \( \tau_{\text{react}} \) should be the best because variation of \( S \) during the process is considered. But not all cloud data are in the “Evaporated” group; instead, some could be in the “Saturated” group, where \( \tau_{\text{react}} \) is negatively correlated with \( \psi \). Thus, for practical use, \( \tau_{\text{evap}} \) is a better choice. The reason why \( \tau_{\text{react}} \) for the “Evaporated” group and \( \tau_{\text{evap}} \) are more appropriate than the other time scales is that \( \psi \) is affected by the variations of both number concentration and droplet size, i.e., \( \psi \) is closely related to how fast droplets are completely evaporated; the rate of droplet complete evaporation is represented by \( \tau_{\text{react}} \) for the “Evaporated” group and \( \tau_{\text{evap}} \). However, \( \tau_{\text{react}} \) for the
“Saturated” group, $\tau_{\text{sat}}$ and $\tau_{\text{phase}}$ are not good time scales for the second objective, because they only represent how fast dry air reaches saturation and LWC decreases and the same variations of $S$ and LWC could be due to complete evaporation of some droplets, or due to evaporation of all droplets. In addition, these time scales are negatively correlated with $\psi$. Therefore, $\tau_{\text{react}}$ for the “Evaporated” group or $\tau_{\text{evap}}$, especially $\tau_{\text{evap}}$, is recommended for developing parameterizations of entrainment-mixing.

Third, theoretical analysis shows that all time scales are proportional to one another if $\frac{\text{LWC}_0}{S_0}$ and/or $\frac{1}{\ln \frac{\text{LWC}_0}{S_0}}$ (LWC$_0$ is LWC after entrainment but before mixing) are constant. Under such conditions, all the time scales are equivalent in studies of entrainment-mixing processes, because the evaporation-induced decrease in LWC is proportional to the increase in $S$ due to phase relaxation. This is confirmed by the fact that $\psi$ is found to be positively correlated with $\tau_{\text{evap}}$, $\tau_{\text{react}}$ and $\tau_{\text{phase}}$, when the RACORO data are presented in a narrow range of $\frac{\text{LWC}_0}{S_0}$.

Two points are noteworthy. First, this study focuses on the commonly used $r$-$n$ mixing diagram that is highly relevant to representing the mixing mechanisms in the commonly used two-moment microphysics parameterization [Lu et al., 2013]. The effect of time scales on the spectral shape of cloud droplet size distribution deserves further study in view of their potentially significant impacts on parameterizations of effective radius [e.g., Liu and Daum, 2002; Liu et al., 2008] and development of precipitation [e.g., Wood, 2000; Liu et al., 2006; Xie and Liu, 2011]. Second, the
actual decaying time scales of $S$, LWC, droplet size and number concentration during entrainment-mixing processes could be different from the time scales calculated in section 2. Tölle and Krueger [2014] compared the two groups of time scales and discussed the best estimates of the actual time scales. Further studies regarding the relationships between the actual time scales and $\psi$ are needed.
Symbol List

A: equation (2b)

$B_1$: $\frac{\text{LWC}_0}{S_0}$

$B_2$: $\frac{1}{\ln \left( \frac{\text{LWC}_0}{S_0} \right)}$

$\beta$: An angle defined in Fig. 1 and equation (10a)

c$_p$: specific heat at constant pressure

D: coefficient of diffusion of water vapor in air

d: distance away from the edges of the cloud cores

Da: Damkohler number

$e_s(T)$: saturation vapor pressure over a plane water surface at temperature $T$

$\varepsilon$: 0.622

$\gamma$: ratio of liquid water content to its adiabatic value

K: coefficient of thermal conductivity of air

$\xi$: $R'/R_v$

$\chi$: mixing fraction of adiabatic cloud during the isobaric mixing between adiabatic cloud and dry air

$L_h$: latent heat

LWC: liquid water content

LWC$_0$: liquid water content just after entrainment but before mixing and evaporation

LWC$_a$: adiabatic liquid water content

LWC$_c$: liquid water content in the cloud affected by entrainment-mixing
$n$: number concentration

$n_0$: number concentration after entrainment but before mixing and evaporation

$n_a$: adiabatic droplet number concentration

$n_c$: number concentration in the cloud affected by entrainment-mixing

$R$: correlation coefficient

$R'$: individual gas constant for dry air

$r$: droplet radius

$r_a$: adiabatic droplet radius

$r_c$: droplet radius in the cloud affected by entrainment-mixing

$R_v$: specific gas constant for water vapor

$\rho_L$: density of liquid water

RH: relative humidity

$S$: supersaturation/saturation ratio deficit

$S_0$: initial saturation ratio deficit in dry air

$S_f$: final saturation ratio deficit

SD: standard deviation

$T$: air temperature

$t$: time

$\tau_{evap}$: time for droplet complete evaporation

$\tau_{mix}$: turbulent mixing time

$\tau_{phase}$: time for saturation ratio deficit to reach 1/e of its initial value
\( \tau_r \): microphysical response time of droplet population

\( \tau_{\text{react}} \): time for droplet complete evaporation or saturation ratio deficit to reach -0.5%

\( \tau_{\text{react}} e \): equal to \( \tau_{\text{react}} \) if the condition of droplet complete evaporation is satisfied.

\( \tau_{\text{react}} s \): equal to \( \tau_{\text{react}} \) if the condition of saturation ratio deficit to reach -0.5% is satisfied.

\( \tau_{\text{sat}} \): the time for saturation ratio deficit to reach -0.5%

\( p \): \( p \) value

\( P \): air pressure

\( \psi \): homogeneous mixing degree

\( Q_1 \): a property as a function of temperature and pressure

\( Q_2 \): a property as a function of temperature and pressure

\( z \): height
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**Caption List**

Figure 1 Mixing diagram illustrating the definition of homogeneous mixing degree. The three black solid lines correspond to extremely inhomogeneous mixing, homogeneous mixing (relative humidity of the dry air is 66%), and contour of $\gamma = 0.2$; $\gamma$ is the ratio of liquid water content to its adiabatic value. See text for the meanings of the other lines and symbols.

Figure 2 Homogeneous mixing degree ($\psi$) as a function of time scales in the cumulus clouds during RACORO: (a) Evaporation time scale ($\tau_{\text{evap}}$), (b) reaction time scale ($\tau_{\text{react}}$), (c) saturation time scale ($\tau_{\text{satu}}$), and (d) phase relaxation time scale ($\tau_{\text{phase}}$) with $n_0$, the number concentration just after entrainment but before evaporation. Each legend provides correlation coefficient ($R$) and $p$ value of the correlation. See text for details.

Figure 3 Homogeneous mixing degree ($\psi$) as a function of time scales based on the EMPM simulations: (a) Evaporation time scale ($\tau_{\text{evap}}$), (b) reaction time scale ($\tau_{\text{react}}$), (c) saturation time scale ($\tau_{\text{satu}}$), and (d) phase relaxation time scale ($\tau_{\text{phase}}$) with $n_0$, the number concentration just after entrainment but before evaporation. Each legend provides correlation coefficient ($R$) and $p$ value of the correlation. See text for details.

Figure 4 Homogeneous mixing degree ($\psi$) as a function of time scales binned according to relative humidity (RH) in environment based on the EMPM simulations; the sizes of data points represent adiabatic number concentrations of 102.7, 205.4,
308.1, 410.8 and 513.5 cm$^{-3}$ (larger-sized data points represent larger number concentrations). (a) Evaporation time scale ($\tau_{\text{evap}}$), (b) reaction time scale ($\tau_{\text{react}}$), (c) saturation time scale ($\tau_{\text{satu}}$), and (d) phase relaxation time scale ($\tau_{\text{phase}}$) with $n_0$, the number concentration just after entrainment but before evaporation. See text for details. The apparent “families” of points will be elaborated in Fig. 4.

Figure 5 Homogeneous mixing degree ($\psi$) as a function of time scales for LWC$_0$/S$_0$ in a narrow range of -0.2 ~ -0.1 g m$^{-3}$ in the cumulus clouds during RACORO; LWC$_0$ is liquid water content after entrainment but before evaporation, S$_0$ is initial saturation ratio deficit in dry air. (a) Evaporation time scale ($\tau_{\text{evap}}$), (b) reaction time scale ($\tau_{\text{react}}$), (c) saturation time scale ($\tau_{\text{satu}}$), and (d) phase relaxation time scale ($\tau_{\text{phase}}$) with $n_0$, the number concentration just after entrainment but before evaporation. Each legend provides correlation coefficient ($R$) and $p$ value of the correlation. See text for details.
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<td>Evaluate how fast droplet size and number concentration change.</td>
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<td>The time for ( S ) to reach ( S_0/e ) under the conditions of constant ( n ) and ( C ), and taking ( r = r_a )</td>
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<tr>
<td>Reaction Time</td>
<td>The time for droplet complete evaporation or ( S ) to reach -0.5%. ( \tau_{\text{react}} ) is calculated in the coupled system of equations (2a) and (5a)</td>
<td></td>
<td>( \tau_{\text{react}_e} ): Evaluate how fast droplet size and number concentration change.</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>( \tau_{\text{react}_s} ): Evaluate how fast liquid water content and supersaturation/saturation ratio deficit change.</td>
<td></td>
</tr>
</tbody>
</table>

Note: See text for the meanings of the symbols.
Number of data samples in the 186 cumulus clouds in the RACORO and their mean values and standard deviations (SD) of number concentration \( (n_c) \), volume-mean radius \( (r_c) \) and liquid water content \( (LWC_c) \).

<table>
<thead>
<tr>
<th>Data Sample Number</th>
<th>Mean ( n_c ) (cm(^{-3}))</th>
<th>SD of ( n_c ) (cm(^{-3}))</th>
<th>Mean ( r_c ) (µm)</th>
<th>SD of ( r_c ) (µm)</th>
<th>Mean ( LWC_c ) (g m(^{-3}))</th>
<th>SD of ( LWC_c ) (g m(^{-3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>13652</td>
<td>924.7</td>
<td>444.7</td>
<td>3.5</td>
<td>0.88</td>
<td>0.196</td>
<td>0.199</td>
</tr>
</tbody>
</table>
Figure 1 Mixing diagram illustrating the definition of homogeneous mixing degree. The three black solid lines correspond to extremely inhomogeneous mixing, homogeneous mixing (relative humidity of the dry air is 66%), and contour of $\gamma = 0.2$; $\gamma$ is the ratio of liquid water content to its adiabatic value. See text for the meanings of the other lines and symbols.
Figure 2 Homogeneous mixing degree ($\psi$) as a function of time scales in the cumulus clouds during RACORO: (a) Evaporation time scale ($\tau_{\text{evap}}$), (b) reaction time scale ($\tau_{\text{react}}$), (c) saturation time scale ($\tau_{\text{satu}}$), and (d) phase relaxation time scale ($\tau_{\text{phase}}$) with $n_0$, the number concentration just after entrainment but before evaporation. Each legend provides correlation coefficient ($R$) and $p$ value of the correlation. See text for details.
Figure 3 Homogeneous mixing degree ($\psi$) as a function of time scales based on the EMPM simulations: (a) Evaporation time scale ($\tau_{evap}$), (b) reaction time scale ($\tau_{react}$), (c) saturation time scale ($\tau_{satu}$), and (d) phase relaxation time scale ($\tau_{phase}$) with $n_0$, the number concentration just after entrainment but before evaporation. Each legend provides correlation coefficient ($R$) and $p$ value of the correlation. See text for details. The apparent “families” of points will be elaborated in Fig. 4.
Figure 4 Homogeneous mixing degree ($\psi$) as a function of time scales binned according to relative humidity (RH) in environment based on the EMPM simulations; the sizes of data points represent adiabatic number concentrations of 102.7, 205.4, 308.1, 410.8 and 513.5 cm$^{-3}$ (larger-sized data points represent larger number concentrations). (a) Evaporation time scale ($\tau_{\text{evap}}$), (b) reaction time scale ($\tau_{\text{react}}$), (c) saturation time scale ($\tau_{\text{sat}}$), and (d) phase relaxation time scale ($\tau_{\text{phase}}$) with $n_0$, the number concentration just after entrainment but before evaporation. See text for details.
Figure 5 Homogeneous mixing degree ($\psi$) as a function of time scales for LWC$_0$/S$_0$ in a narrow range of -0.2 ~ -0.1 g m$^{-3}$ in the cumulus clouds during RACORO; LWC$_0$ is liquid water content after entrainment but before evaporation, S$_0$ is initial saturation ratio deficit in dry air. (a) Evaporation time scale ($\tau_{\text{evap}}$), (b) reaction time scale ($\tau_{\text{react}}$), (c) saturation time scale ($\tau_{\text{satu}}$), and (d) phase relaxation time scale ($\tau_{\text{phase}}$) with $n_0$, the number concentration just after entrainment but before evaporation. Each legend provides correlation coefficient ($R$) and $p$ value of the correlation. See text for details.