

## **Abstract**:

 One of the challenges in representing warm rain processes in global climate models (GCM) is related to the representation of the subgrid variability of cloud properties, such as cloud water and cloud droplet number concertation (CDNC), and the effect thereof on individual precipitation processes such as autoconversion. This effect is conventionally treated by 35 multiplying the resolved-scale warm ran process rates by an enhancement factor  $(E_q)$  which is derived from integrating over an assumed subgrid cloud water distribution. The assumed subgrid cloud distribution remain highly uncertain. In this study, we derive the subgrid variations of liquid-phase cloud properties over the tropical ocean using the satellite remote sensing products from Moderate Resolution Imaging Spectroradiometer (MODIS) and investigate the corresponding enhancement factors for the GCM parameterization of autoconversion rate. We find that the conventional approach of using only subgrid variability of cloud water is insufficient, and that the subgrid variability of CDNC, as well as the correlation between the two, are also 43 important for the correctly simulating the autoconversion process in GCMs. Using the MODIS 44 data which has the near-global data coverage, we find that  $E_q$  shows a strong dependence on cloud regimes, due to the fact that the subgrid variability of cloud water and CDNC is regime-46 dependent. Our analysis shows a significant increase of  $E_a$  from the stratocumulus (Sc) to 47 cumulus (Cu) regions. Furthermore, the enhancement factor  $E_N$  due to the subgrid variation of CDNC is derived from satellite observation for the first time, and results reveal several regions downwind of biomass burning aerosols (e.g., Gulf of Guinea, East Coast of South Africa), air pollution (i.e., Eastern China Sea), and active volcanos (e.g., Kilauea Hawaii and Ambae Vanuatu), 51 where the  $E_N$  is comparable, or even larger than  $E_q$ , suggesting an important role of aerosol in 52 influencing the  $E_N$ . MODIS observations suggest that the subgrid variations of cloud liquid water path (LWP) and CDNC are generally positively correlated. As a result, the combined enhancement factor, including the effect of LWP and CDNC correlation, is significantly smaller than the simple 55 product of  $E_q \cdot E_N$ . Given the importance of warm rain processes in understanding the Earth system dynamics and water cycle, we conclude that more observational studies are needed to provide a better constraint on the warm rain processes in GCMs.

#### **1. Introduction**

 Marine boundary layer (MBL) clouds are a strong modulator of Earth's radiative energy budget (Klein and Hartmann, 1993; Trenberth et al., 2009). They can interact with other components of the climate system, such as aerosols and precipitations, in various ways. The feedback of MBL clouds to climate change remains one of the largest uncertainties in our understanding of the climate sensitivity (Bony and Dufresne, 2005; Soden and Held, 2006). Despite their importance in the climate system, simulating MBL clouds in general circulations models (GCM) has proved to be extremely challenging. A main difficulty is rooted in the fact the typical grid size of GCM (~100km) is much larger than the spatial scale of many cloud microphysical processes, and as a result these subgrid scale processes, as well as the subgrid cloud variations, have to be highly simplified and then parameterized as functions of resolved, grid-level variables.

 Of particular interest in this study is the warm rain processes in MBL clouds, which have fundamental impacts on the cloud water budget and lifetime. Although in reality it is highly complicated and involves multiple factors, warm rain formation in GCMsis usually parameterized as simple functions of only key cloud parameters. For example, the drizzle in MBL cloud is initialized by the so-called autoconversion process in which the collision-coalescence of cloud droplets gives birth to large drizzle drops (Pruppacher and Klett, 1997). In GCMs, for the sake of efficiency, this process is usually parameterized as a power function of liquid water content (LWC 77 or symbol  $q_c$ ) and cloud droplet number concentration (CDNC or symbol  $N_c$ ). One of the most widely used parameterization scheme is developed by Khairoutdinov and Kogan (2000) ("KK2000" hereafter), which has the form

$$
\frac{\partial q_r}{\partial t} = C(q_c)^{\beta_q} (N_c)^{\beta_N}, \tag{1}
$$

 where  $\frac{\partial q_r}{\partial t}$  is the rain water tendency due to the autoconversion process,  $q_c$  has the unit of kg/kg, 81 and  $N_c$  of cm<sup>-3</sup>. The three parameters  $C = 1350$ ,  $\beta_q = 2.47$  and  $\beta_N = -1.79$  are derived 82 through a simple least-square fitting of the autoconversion rate results from a large-eddy simulation with bin microphysics that can simulate the process-level physics. Even though this is highly simplified, the parametrization scheme still faces a great challenge. The calculation of grid-

85 mean autoconversion efficiency requires the knowledge of subgrid distributions of LWC and 86 CDNC, but in the GCMs only grid-mean quantities  $\langle q_c \rangle$  and  $\langle N_c \rangle$  are known and available for use 87 in the computation of autoconversion rate. As pointed out by Pincus and Klein (2000), for a 88 process  $f(x)$  such as autoconversion that is nonlinearly dependent on subgrid variables, x, the 89 grid-mean value  $\langle f(x) \rangle$  is not equal to the value estimated based on the grid-mean  $\langle x \rangle$ , i.e., 90  $\langle f(x) \rangle \neq f(\langle x \rangle)$ . Mathematically, if  $f(x)$  is convex, then  $f(\langle x \rangle) < \langle f(x) \rangle$  (Larson and Griffin, 91 2013; Larson et al., 2001). To take this effect into account, a parameter  $E$  is often introduced in 92 the GCM as part of the parameterization such that  $\langle f(x) \rangle = E \cdot f(\langle x \rangle)$ . It is referred to as the 93 "enhancement factor" in many studies and this study too because  $E > 1$  for a convex function. 94 Such a nonlinear effect is not just limited to the autoconversion process. Some other examples 95 are the plane-parallel albedo bias (Barker, 1996; Cahalan et al., 1994; Oreopoulos and Davies, 96 1998a), subgrid cloud droplet activation (Morales and Nenes, 2010) and accretion (Boutle et al., 97 2014; Lebsock et al., 2013).

98 The value of E is determined primarily by two factors: the nonlinearity of  $f(x)$  and the 99 subgrid probability density function (PDF)  $P(x)$ . Given the same subgrid variation of LWC, i.e., 100  $P(q_c)$ , the nonlinear effect impacts the autoconversion parameterization more than it does on 101 the accretion, because the former is a more nonlinear function of  $q_c$  than the latter. For the same 102  $f(x)$ , a grid box with a narrow and symmetric  $P(x)$  would require a smaller E than another grid 103 box with a broader and non-symmetric  $P(x)$ . Ideally, the value of the enhancement factor E 104 should be diagnosed from the subgrid cloud PDF  $P(x)$ . Unfortunately, because this is not possible 105 in most conventional GCMs, the value of  $E$  is usually assumed to be a constant for the lack of 106 better options. The  $E$  for autoconversion due to subgrid LWC variation is assumed to be 3.2 in 107 the two-moment cloud microphysics parameterization schemes by Morrison and Gettelman 108 (2008) (MG scheme hereafter), which is employed in the widely used Community Atmosphere 109 Model (CAM). This choice of  $E = 3.2$  is based on an early study by Barker et al. (1996), in which 110 the mesoscale variation of column-integrated optical thickness of the "overcast stratocumulus", 111 "broken stratocumulus" and "scattered stratocumulus" are studied. The value  $E = 3.2$  is derived 112 based on the mesoscale variation of the broken stratocumulus.

113 Clearly, a simple constant  $E$  is not adequate. The following is a list of attempts to better understand the subgrid cloud variations and the implications for warm rain simulations in GCMs. Several previous studies have shown that the mesoscale cloud water variation is a strong function of cloud regime—the subgrid cloud water variation of Sc cloud is much different from that of Cu clouds(Barker et al., 1996; Lee et al., 2010; Oreopoulos and Cahalan, 2005; Wood and Hartmann, 2006). As the first part of a two-part study, Larson and Griffin (2013) first laid out a systematic theoretical basis for understanding the effects of subgrid cloud property variations on simulating 120 various nonlinear processes in GCM, including not only the autoconversion but also the accretion, condensation, evaporation and sedimentation processes. In the second part, using cloud fields from a large-eddy simulation (LES), Griffin and Larson (2013) showed that inclusion of the 123 enhancement factor indeed leads to more rainwater at surface in single-column simulations and makes them agree better with high-resolution large-eddy simulations. Using a combination of in situ measurement and satellite remote sensing data, Boutle et al. (2014) analyzed the spatial variation of both cloud and rain water, as well as their covariation, and developed a simple 127 parameterization scheme to relate the subgrid cloud water variance to the grid-mean cloud fraction. Later, the study of Boutle et al. (2014) was extended by Hill et al. (2015) who developed a cloud regime dependent and scale-aware parameterization scheme for simulating subgrid cloud water variation. Recently, using the ground-based observations from three Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) sites, Xie and Zhang (2015) developed a scale-aware parameterization scheme for GCMs to account for subgrid cloud water variation. Also using ARM measurement, Ahlgrimm and Forbes (2016) analyzed the dependence of cloud water variability on cloud regime. Although these previous studies have shed important light on 135 subgrid cloud variation and the implications for GCM, they lack a global perspective because they are only based on limited data (e.g., LES cases, in situ and ground-based measurement). Currently, satellite remote sensing observation is the only way to achieve a global perspective. Using the observations from the space-borne radar CloudSat, Lebsock et al. (2013) showed that 139 the subgrid cloud water variance is smaller over the Sc region than over the Cu region, and as a result the enhancement factor shows an increasing trend from Sc to Cu region. They also highlighted importance of considering the subgrid co-variability of cloud water and rain water in 142 the computation of the accretion rate. On the modeling side, Guo et al. (2014) investigated the sensitivity of cloud simulation in the Geophysical Fluid Dynamics Laboratory (GFDL) Atmospheric General Circulation Model (AM) to the subgrid cloud water parameterization schemes. A similar 145 study was carried out by Bogenschutz et al. (2013) using the National Center of Atmospheric Research (NCAR) Community Atmospheric Model (CAM). Both studies show that the more sophisticated subgrid parameterization scheme— Cloud Layers Unified by Binormals (CLUBB) (Golaz et al., 2002a; 2002b; Larson et al., 2002)—lead to a better simulation of clouds in the model. However, a more recent study by Song et al. (2018b) reveals that the CLUBB in CAM version 5.3 (CAM5.3) overestimates the enhancement factor in the trade wind cumulus cloud region, which in turn leads to excessive drizzle in the model and "empty clouds" with near-zero cloud water. In addition to CLUBB, the so-called super-parameterization (a.k.a Multiscale Modeling Framework (MMF)), which uses cloud resolving model embeded in the GCM grids to diagnose sub-grid cloud variations (Randall et al., 2003), have also gained increasing popularity. Takahashi et al. (2017) compared the subgrid cloud water variations simulated by a CAM-MMF model with those derived from A-Train observations and found reasonable agreement.

 Despite these previous studies, many questions remain unanswered. First of all, all the previous studies, as far as we know, have focused on the impact of subgrid cloud water  $q_c$  variation. The potential impact of subgrid variation of  $N_c$  and the co-variability of  $N_c$  with  $q_c$ 160 have been overlooked so far. Given the same amount of  $q_c$ , a cloud with a smaller  $N_c$  would have 161 larger droplets and therefore larger precipitation efficiency than another cloud with a larger  $N_c$ . 162 For the same reason, other things equal, a grid with positive correlation of subgrid  $N_c$  and  $q_c$  would be less efficient in terms of autoconversion than a grid with negative correlation of the two. Secondly, most of previous studies are based on the assumption that the subgrid cloud 165 property variation follows certain well-behaved distributions, usually either Gamma (e.g., Barker, 1996; Morrison and Gettelman, 2008; Oreopoulos and Barker, 1999; Oreopoulos and Cahalan, 2005) or Lognormal (Boutle et al., 2014; Larson and Griffin, 2013; e.g., Lebsock et al., 2013). However, the validity and performance of the assumed PDF shape are seldom checked. Furthermore, although the study by Lebsock et al. (2013) has depicted a global picture of the  enhancement factor for the autoconversion modeling in GCM, the picture is far from clear due 171 to the small sampling rate of CloudSat observations.

172 In this study, we revisit the subgrid variations of liquid-phase cloud properties over the tropical ocean using 10 years of MODIS cloud observations, with the overarching goal to better understand the potential impacts of subgrid cloud variations on the warm rain processes in the conventional GCMs. Similar to previous studies, we will quantify the subgrid cloud water variations based on MODIS observations. Going one step further, we will also attempt to unveil 177 for the first time the subgrid  $N_c$  variation, as well as its correlation with cloud water, and investigate the implications for warm rain simulations in GCM. Moreover, we will take advantage of the wide spatial coverage of MODIS data to achieve a more detailed picture of the enhancement factor for the autoconversion simulation. Last but not least, we will evaluate the two widely used distributions, i.e., Lognormal and Gamma, in terms of their performance and limitations for simulating the enhancement factor. We will first explain the theoretical background in Section 2 and introduce the data and methodology in Section 3. The MODIS observations will be presented and discussed in Section 4. The implications for the autoconversion parameterization in the GCMs will be discussed in 5. The main findings will be 186 summarized in Section 6 with an outlook for future studies.

### **2. Theoretical Background**

## **2.1.Theoretical Distributions to describe subgrid cloud property variations**

 In previous studies, the spatial variations of cloud properties, such as cloud optical thickness (COT), cloud liquid water path (LWP) and cloud liquid water content (LWC), are often described using either of two theoretical distributions—the Gamma and Lognormal distribution. The probability density function (PDF) from a Gamma distribution is a two-parameter function as follows (Barker, 1996; Oreopoulos and Davies, 1998b):

$$
P_G(x) = \frac{1}{\Gamma(\nu)} \alpha^{\nu} x^{\nu - 1} \exp(-\alpha x),
$$
 (2)

194 where Γ is the Gamma function,  $v$  is the so-called inverse relative variance, and  $\alpha$  the so-called 195 rate parameter. If x follows the Gamma distribution, its mean value Is given by

$$
\langle x \rangle = \int_0^\infty x \, P_G(x) dx = \frac{v}{\alpha},\tag{3}
$$

196 and variance given by

$$
Var(x) = \int_0^{\infty} (x - \langle x \rangle)^2 P_G(x) dx = \frac{v}{\alpha^2}.
$$
 (4)

### 197 It follows from Eq. *(3)* and *(4)* that the so-called inverse relative variance is

$$
v = \frac{1}{\eta} = \frac{\langle x \rangle^2}{Var(x)},\tag{5}
$$

198 where  $\eta = \frac{Var(x)}{(x)^2}$  is the relative variance. If x follows the Gamma distribution, for a physical

199 process  $M(x)$  that is a power function of x,

$$
M(x) = Kx^{\beta}, \tag{6}
$$

200 then the expected value  $\langle M(x) \rangle$  is given by

$$
\langle M(x) \rangle_G = K \int_0^\infty x^\beta P_G(x) dx = \frac{\Gamma(\nu + \beta)}{\Gamma(\nu)\nu^\beta} K(x)^\beta, \ \beta > -\nu. \tag{7}
$$

201 As explained in the introduction, for a nonlinear process  $M(x)$ ,  $\langle M(x) \rangle \neq M(\langle x \rangle)$ . The ratio 202 between the two  $E$  is by definition the enhancement factor:

$$
E(P_G, v, \beta) = \frac{\langle Kx^{\beta} \rangle}{K(x)^{\beta}} = \frac{1}{\langle x \rangle^{\beta}} \int_0^{\infty} x^{\beta} P_G(x) dx = \frac{\Gamma(v+\beta)}{\Gamma(v)v^{\beta}},
$$
(8)

203 The PDF of a Lognormal distribution is given as follows (Larson and Griffin, 2013; 204 Lebsock et al., 2013):

$$
P_L(x) = \frac{1}{\sqrt{2\pi}x\sigma} \exp\left(-\frac{(\ln x - \mu)^2}{2\sigma^2}\right),\tag{9}
$$

205 where  $\mu = \langle \ln x \rangle$  and  $\sigma^2 = Var(lnx)$  correspond to the mean and variance of  $lnx$ , respectively.

206 The mean value of the Lognormal distribution is given by

$$
\langle x \rangle = \int_0^\infty x P_L(x) dx = e^{\mu + \frac{\sigma^2}{2}}, \tag{10}
$$

207 and the variance by

$$
Var(x) = \int_0^\infty (x - \langle x \rangle)^2 P_L(x) dx = e^{2\mu + \sigma^2} (e^{\sigma^2} - 1).
$$
 (11)

208 It follows from Eq. *(10)* and *(11)* that the inverse relative variance can be derived from the

209 following equation

$$
e^{\sigma^2} = 1 + \frac{Var(x)}{\langle x \rangle^2} = 1 + \frac{1}{v}.
$$
 (12)

210 If x follows the Lognormal distribution, the expected value of  $\langle M(x) \rangle$  is

$$
\langle M(x)\rangle_L = K \int_0^\infty x^\beta P_L(x) dx = \left(1 + \frac{1}{\nu}\right)^{\frac{\beta^2 - \beta}{2}} K\langle x \rangle^\beta.
$$
 (13)

211 Evidently, the corresponding enhancement factor is given by

$$
E(P_L, v, \beta) = \frac{\langle \kappa x^{\beta} \rangle}{\kappa \langle x \rangle^{\beta}} = \left(1 + \frac{1}{v}\right)^{\frac{\beta^2 - \beta}{2}}.
$$
 (14)

212 Note that Eq. (7) and (8) are only valid when  $\beta > -v$  because Gamma function  $\Gamma(v + \beta)$  can 213 run into singular values when  $v + \beta < 0$ . In contrast, Eq. (13) and (14) are valid for any real value 214  $\beta$ . This is one advantage of the Lognormal distribution over the Gamma distribution.

215 An example of the Gamma and Lognormal distributions for  $q_c$  is shown in Figure 1a. In 216 this example, both distributions have the same mean  $\langle q_c \rangle = 0.5g/kg$  and also the same inverse 217 relative variance  $v_q = 3$ . Although the general shapes of the two PDFs are similar, they differ 218 significantly at the two ends: the Gamma PDF is larger than Lognormal PDF over the small values 219 of  $q_c$ , and the opposite is true over the large values of  $q_c$ . The Gamma and Lognormal 220 distributions can also be used to describe the spatial variation of  $N_c$  (Gultepe and Isaac, 2004). 221 An example is given in Figure 1c, in which  $q_c$  is a constant of 0.5 $g/kg$ ,  $\langle N_c \rangle = 50$   $cm^{-3}$ , and  $v_N =$ 222 5.0. Figure 1 b shows the autoconversion rate based on the KK2000 parameterization scheme for 223 the Gamma  $P_G(q_c)$  and Lognormal  $P_L(q_c)$  that are shown in Figure 1a. Interestingly, although 224 the cumulative autoconversion rates based on the two types of PDFs are almost identical, the 225 contribution to the total autoconversion rate from the different LWC bins are quite different. As 226 show in Figure 1a, the  $P_L(q_c)$  has a longer tail than the  $P_G(q_c)$ , i.e., the occurrence probability of 227 large  $q_c$  (e.g.,  $q_c > 2.0g/kg$  ) is much higher in the Lognormal than in Gamma PDF. This 228 difference is further amplified in the autoconversion rate computation in Figure 1b because the 229 autoconversion rate is proportional to  $q_c^{2.47}$ .

230 The enhancement factors based on the Gamma (i.e.,  $E(P_G, \beta)$  in Eq. (8)) and Lognormal 231 (i.e.,  $E(P_L, \beta)$  in Eq. (14)) PDF for  $\beta_q = 2.47$  are plotted as a function of the inverse relative 232 variance v in Figure 2. When subgrid clouds are more homogenous i.e.,  $v > 1$ , the enhancement 233 factor based on the two PDFs are similar. However, for more inhomogeneous grids with i.e.,  $v <$  234 1, the  $E(P_L, \beta)$  is significantly larger than that  $E(P_G, \beta)$ , which is probably because of the longer 235 tail of  $P_L(q_c)$  as shown in Figure 1 a and b.

236

## 237 **2.2.Impacts of subgrid cloud variations on warm rain parameterization in GCM**

238 The warm rain process in MBL clouds involves many interacting microphysical processes. In this study, we only focus only on the simulation of autoconversion in GCM. Other nonlinear processes, such as accretion and evaporation have been investigated in previous studies (Boutle et al., 2014; Lebsock et al., 2013).

242 Ideally, if the subgrid variations of  $q_c$  and  $N_c$  are known, then the grid-mean in-cloud 243 autoconversion rate should be derived from the following integral

$$
\langle \frac{\partial q_r}{\partial t} \rangle = \int_0^\infty \int_0^\infty C(q_c)^{\beta_q} (N_c)^{\beta_N} P(q_c, N_c) dq_c dN_c, \tag{15}
$$

244 where  $P(q_c, N_c)$  is the joint PDF of  $q_c$  and  $N_c$ . Unfortunately, most conventional GCMs lack the 245 capability of predicting the subgrid variations of cloud properties, with only a couple of 246 exceptions (Thayer-Calder et al., 2015). What is known from the GCM is usually the in-cloud grid-247 mean values  $\langle q_c \rangle$  and  $\langle N_c \rangle$ . As a result, instead of using Eq. (15), the autoconversion rate in GCMs 248 is usually computed from the following equation

$$
\langle \frac{\partial q_r}{\partial t} \rangle = E \cdot C(\langle q_c \rangle)^{\beta_q} (\langle N_c \rangle)^{\beta_N}, \tag{16}
$$

249 where  $E$  is the enhancement factor defined as:

$$
E = \frac{\int_0^\infty \int_0^\infty (q_c)^{\beta q} (N_c)^{\beta N} P(q_c, N_c) dq_c dN_c}{\left( \langle q_c \rangle \right)^{\beta q} \left( \langle N_c \rangle \right)^{\beta N}}.
$$
\n(17)

250 The value of the enhancement factor depends on the subgrid variations of  $q_c$  and  $N_c$ . If clouds 251 are homogenous on the subgrid scale, then  $E \sim 1$ . The more inhomogeneous the clouds are, the 252 larger the E is. In the special case where  $q_c$  and  $N_c$  are independent, then the joint PDF  $P(q_c, N_c)$ 253 becomes  $P(q_c, N_c) = P(q_c)P(N_c)$ , where  $P(q_c)$  and  $P(N_c)$  are the PDF of the subgrid  $q_c$  and 254  $N_c$ . Consequently, Eq. (15) reduces to

$$
\langle \frac{\partial q_r}{\partial t} \rangle = C \int_0^\infty (q_c)^{\beta q} P(q_c) dq_c \int_0^\infty (N_c)^{\beta_N} P(N_c) dN_c,
$$
\n(18)

255 and Eq.(17) to

$$
E = E_q \cdot E_N,\tag{19}
$$

256 where  $E_q$  is the enhancement factor due to the subgrid variation of cloud water which has the 257 form,

$$
E_q = \frac{\int_0^\infty (q_c)^{\beta q} P(q_c) dq_c}{((q_c))^{\beta q}},
$$
\n(20)

258 and the  $E_N$  is the enhancement factor due to the subgrid variation of CDNC which has the form,

$$
E_N = \frac{\int_0^{\infty} (N_c)^{\beta_N} P(N_c) dN_c}{((N_c))^{\beta_N}}.
$$
 (21)

259 Obviously, if  $P(q_c)$  and  $P(N_c)$  follow either Gamma or Lognormal distribution, then the above 260 equations reduce to Eq. (*8*) or (*14*), respectively.

261 If  $q_c$  and  $N_c$  both have significant subgrid variations and they are not independent, the 262 enhancement factor should ideally be diagnosed from Eq. (17). However, the joint PDF  $P(q_c, N_c)$ 263 may not be known and the integration can be time-consuming. Some previous studies proposed 264 to approximate the  $P(q_c, N_c)$  as a bivariate lognormal distribution as follows:

$$
P(q_c, N_c) = \frac{1}{2\pi q_c N_c \sigma_q \sigma_N \sqrt{1 - \rho^2}} \exp\left(-\frac{\zeta}{2}\right)
$$
  

$$
\zeta = \frac{1}{1 - \rho^2} \left[ \left(\frac{\ln q_c - \mu_q}{\sigma_q}\right)^2 - 2\rho \left(\frac{\ln q_c - \mu_q}{\sigma_q}\right) \left(\frac{\ln N_c - \mu_N}{\sigma_N}\right) + \left(\frac{\ln N_c - \mu_N}{\sigma_N}\right)^2 \right],
$$
(22)

265 where  $\rho$  is the correlation coefficient between  $q_c$  and  $N_c$  (Larson and Griffin, 2013; Lebsock et 266 al., 2013). As such, both  $q_c$  and  $N_c$  follow a marginal lognormal distribution in Eq. (9). Substituting 267 Eq. (*22*) into Eq. (*17*), we obtain the enhancement factor for the bivariate lognormal distribution 268 that consists of three terms

$$
E = E_q(P_L, v_q, \beta_q) \cdot E_N(P_L, v_N, \beta_N) \cdot E_{COV}(\rho, \beta_q, \beta_N v_q, v_N), \qquad (23)
$$

where  $E_q\big(P_L, \nu_q, \beta_q\big) = \left(1 + \frac{1}{\nu_q}\right)$  $\beta_q^2 - \beta_q$ <sup>2</sup> and  $E_N(P_L, v_N, \beta_N) = \left(1 + \frac{1}{v_N}\right)$  $\beta_N^2 - \beta_N$ 269 where  $E_q(P_L, v_q, \beta_q) = \left(1 + \frac{1}{r}\right)^{-2}$  and  $E_N(P_L, v_N, \beta_N) = \left(1 + \frac{1}{r}\right)^{-2}$  correspond to the

270 impacts of subgrid  $q_c$  and  $N_c$  variance, respectively (i.e., Eq.  $(14)$ ), and the third term

$$
E_{cov}(\rho, \beta_q, \beta_N, \nu_q, \nu_N) = \exp(\rho \beta_q \beta_N \sigma_q \sigma_N), \qquad (24)
$$

271 corresponds to the impact of the co-variation of  $q_c$  and  $N_c$  on the enhancement factor. Obviously, 272 Eq. (23) reduces to Eq. (19) when  $q_c$  and  $N_c$  are uncorrelated (i.e.,  $\rho = 0$ ,  $E_{COV} = 1$ ). If  $q_c$  and  $N_c$ 273 are negatively correlated (i.e.,  $\rho$  < 0 and  $E_{COV}$  > 1), clouds with larger  $q_c$  would tend to have 274 smaller  $N_c$ . The autoconversion rate in such a case would be larger than that in the case where 275  $q_c$  and  $N_c$  are positively correlated (i.e., i.e.,  $\rho > 0$  and  $E_{COV} < 1$ ). A positive correlation would 276 exist, for instance, if all droplets in cloud were the same size, but some parcels had more droplets 277 than other parcels.

278 Most current GCMs do not have the capability to simulate the subgrid cloud property 279 variations. They usually have to use pre-defined subgrid cloud variations in the computation of 280 grid-mean autoconversion rate instead of using prognostic values. For example, in the MG 281 scheme for the CAM5.3, the subgrid  $q_c$  is assumed to follow the Gamma distribution in Eq. *(2)* 282 with a fixed  $v_q = 1$  and as a result constant  $E_q = 3.2$ . Lately, advanced subgrid parameterization 283 schemes, such as CLUBB, have been implemented in several GCMs, including CAM6 and GFDL 284 AM model (Bogenschutz et al., 2017; Guo et al., 2015; 2014), which provides information on the 285 subgrid  $q_c$  variation to the host model. The information can then be used to dynamically diagnose 286 the enhancement factor  $E_q$ , which will help the model simulate the cloud regime dependence of 287  $E_q$  (Guo et al., 2010; 2014).

288 However, as explained above, not only the subgrid variation of  $q_c$  but the subgrid 289 variation of  $N_c$  can also influence the enhancement factor. Unfortunately, this aspect has been 290 ignored by almost all GCMs, even the latest CAM6 with CLUBB. Physically, provided the same  $q_c$ , 291 a cloud with smaller  $N_c$  would have larger droplet size and therefore larger precipitation 292 efficiency than the cloud with larger  $N_c$ . Because the autoconversion rate depends nonlinearly 293 on  $N_c$ , the grid-mean autoconversion rate computed based on a skewed PDF of  $N_c$  (i.e.,  $\int_0^\infty (N_c)^{\beta_N}P(N_c)dN_c)$  would be different from that computed based on the mean of  $N_c$  (i.e., 295  $((N_c))^{\beta_N}$ ). The autoconversion enhancement factor based on the Lognormal PDF  $E(P_L, \beta)$  for 296  $\beta_N = -1.79$  is given in Figure 2. Interestingly, at the same inverse relative variance v, the 297 enhancement factor based on the same Lognormal PDF  $E(P_L, \beta)$  for  $\beta_N = -1.79$  is actually 298 Iarger than that for  $\beta_q = 2.47$  because of the formula of the exponent in Eq. (14) (i.e.,  $\frac{\beta^2-\beta}{2}$ ). 299 Moreover, the correlation between  $N_c$  and  $q_c$  can also be important. Going back to Eq.(23),

300 evidently,  $E > E_q$  if and only if  $E_N \cdot E_{COV} > 1$ . After some manipulation, we can show that if 301  $\beta_N < 0$  and  $\sigma_N > 0$ , then

$$
E_N \cdot E_{COV} > 1, \text{ if } \rho < \frac{\sigma_N}{\sigma_q} \cdot \frac{(1 - \beta_N)}{2\beta_q}.\tag{25}
$$

302 This equation reveals that when  $q_c$  and  $N_c$  are weakly or negatively correlated ( $\rho \le 0$ ), 303 considering only  $E_q$  would tend to underestimate E. On the other hand, however, if  $q_c$  and  $N_c$ 304 are highly positively correlated ( $\rho$   $\sim$  1) then considering  $E_q$  only would tend to overestimate E.

## 305 **3. Data and Methodology**

306 To derive the above-mentioned enhancement factors, we will use 10 years (2007  $\approx$  2016) of the latest collection 6 (C6) *daily mean* level-3 cloud retrieval product from the Aqua-MODIS instrument (product name "MYD08\_D3"), which contains the gridded statistics of cloud properties computed from pixel-level (i.e., level-2) retrievals. As summarized in Platnick et al. (2003; 2017), the operational level-2 MODIS cloud product provides cloud masking (Ackerman et al., 1998), cloud top height (Menzel et al., 1983), cloud top thermodynamic phase determination (Menzel et al., 2006), and COT, cloud effective radius (CER) and LWP retrievals based on the bi- spectral solar reflectance method (Nakajima and King, 1990). All MODIS level-2 atmosphere 314 products, including the cloud, aerosol and water vapor products, are aggregated to  $1^{\circ} \times 1^{\circ}$  spatial resolution on a daily, eight-day, and monthly basis. Aggregations include a variety of scalar statistical information, including mean, standard deviation, max/min occurrences, as well as histograms including both marginal and joint histograms. For COT, CER and LWP, the MODIS 318 level-3 product provides both their "in-cloud" grid-mean values  $(\langle x \rangle)$  and subgrid standard 319 deviations  $(\sigma_x)$ . The inverse relative variance v can then be derived from Eq. *(5)*, i.e.,  $v =$  $\langle x \rangle^2 / \sigma_x^2$ . Note that the operational MODIS product provides two CER retrievals, one based on 321 the observation from the band 7 centered around 2.1  $\mu$ m and the other from band 20 at 3.7  $\mu$ m. As discussed in several previous studies (Cho et al., 2015; Zhang and Platnick, 2011; Zhang et al., 323 2012; 2016), the 3.7 µm band CER retrieval is more resilient to the 3-D effects and retrieval failure 324 than the 2.1 µm band retrievals. For these reasons, it is used as the observational reference in this study.



326 Given the COT and CER retrieval, the operational MODIS product estimates the LWP of cloud

using

$$
LWP = \frac{2}{3}\rho_w COT \cdot CER,\tag{26}
$$

328 where  $\rho_w$  is the density of water. Several studies have argued that a smaller coefficient of 5/9, instead of 2/3, should be used in estimation of LWP (Lebsock et al., 2011; Seethala and Horváth, 2010; Wood and Hartmann, 2006). The choice of coefficient does not matter in this study because 331 it is a common factor in the calculation of  $v$ . The choice of the coefficient has no impact on our 332 study, because we are interested in the relative inverse variance  $v = \langle x \rangle^2 / \sigma_x^2$ . We note here that 333 it is the LWC  $q_c$ , instead of the LWP, that is used in the KK2000 scheme. So, the spatial variability of LWC is what is most relevant. However, the remote sensing of cloud water vertical profile from satellite sensor for liquid-phase clouds is extremely challenging even with active sensors. It is why most previous studies using the satellite observations analyzed the spatial variation of LWP, rather than LWC. In fact, even Lebsock et al. (2013), who used the level-2 CloudSat observations, had to use the vertical averaged LWC in their analysis. Airborne in situ measurement faces similar challenge. For example, Boutle et al. (2014) use the LWC observation along "horizontal flight 340 tracks" to study the spatial variability of cloud water, which only samples the LWC at certain levels of MBL clouds. Ground-based observations are much better than satellite and airborne observation in this regard. Recently, Xie and Zhang (2015) analyzed the cloud water profiles retrieved using ground-based radars from the three ARM sites and found no obvious in-cloud vertical dependence of the spatial variability of LWC. Following these previous studies, we assume that the horizontal subgrid variation of LWC is *not* strongly dependent on height and its value can be inferred from the spatial variability of the vertical integrated quantity LWP. The uncertainty caused by this assumption will be assessed in future studies.

 The current MODIS level-3 cloud product does *not* provide CDNC retrievals. Following previous studies (Bennartz, 2007; Bennartz and Rausch, 2017; Grosvenor and Wood, 2014; 350 McCoy et al., 2017a), we estimate  $N_c$  of liquid-phase clouds from the MODIS retrieved COT ( $\tau$ ) 351 and CER  $(r_e)$  based on the classic adiabatic cloud model

$$
N_c(\tau, r_e) = \frac{\sqrt{5}}{2\pi k} \frac{\sqrt{f_{ad} \Gamma_w}}{\sqrt{\rho_w Q_e}} \tau^{\frac{1}{2}} r_e^{-\frac{5}{2}} = \frac{\sqrt{15}}{2\pi k} \frac{\sqrt{f_{ad} \Gamma_w}}{\rho_w \sqrt{2Q_e}} LWP^{\frac{1}{2}} r_e^{-3},
$$
(27)

352 where  $\rho_w$  is the density of water;  $Q_e \approx 2$  is the extinction efficiency of cloud droplets; k is the 353 ratio of  $r_e$  to mean volume-equivalent radius;  $f_{ad}$  is the adiabaticity of the cloud;  $\Gamma_w$  is the LWC 354 lapse rate. Following previous studies, we assume  $k = 0.8$  and  $f_{ad} = 1.0$  to be constant and 355 compute  $\Gamma_W$  from the grid mean liquid cloud top temperature and pressure. The theoretical 356 basis and main uncertainty sources of the CDNC estimation based on the adiabatic cloud model 357 from MODIS-like passive cloud retrievals are nicely reviewed by Grosvenor et al. (2018).

358 Ideally, the values of LWP and CDNC should be estimated on pixel-by-pixel basis from the level-2 MODIS product. However, pixel-by-pixel estimation is highly time consuming, which makes it difficult to achieve a global perspective. Using an alternative method, many previous studies estimate the grid-level CDNC statistics from the joint histogram of COT vs. CER provided in the level-3 MODIS cloud products (Bennartz, 2007; McCoy et al., 2017a; 2017b). For a given 1°×1° grid-box, the liquid-phase COT-CER joint histogram provides the counts of successful cloud property retrievals with respect to 108 joint COT-CER bins that are bounded by 13 COT bin 365 boundaries, ranging from 0 to 150, and 10 CER bin boundaries, ranging from 4  $\mu$ m to 30  $\mu$ m. With 366 the joint histogram, which is essentially the joint PDF of COT and CER  $P(\tau, r_e)$ , we can estimate the grid mean and variance of CDNC from the following equations

$$
\langle x \rangle = \int \int x(\tau, r_e) P(\tau, r_e) d\tau dr_e, \qquad (28)
$$

$$
Var(x) = \int \int (x(\tau, r_e) - \langle N_c \rangle)^2 P(\tau, r_e) d\tau dr_e, \qquad (29)
$$

368 where x can be either LWP or CDNC. Figure 3a shows the LWP in Eq.  $(26)$  as a function of the 13 369 COT bins and 10 CER bins from the MODIS level-3 product. As expected, the largest LWP values 370 are found when both COT and CER are large. Figure 3b shows the CDNC in Eq. *(27)* as a function 371 of the COT and CER bins. As expected, the largest CDNC values are found when both COT is large 372 and CER is small. Figure 3c shows an example of the COT-CER joint histogram from the Aqua-373 MODIS daily level-3 product "MYD08\_D3" on January 09<sup>th</sup>, 2007 at the grid box 1°S and 1°W. In 374 this particular grid box, a combination of  $\sim$ 2-4 COT and  $\sim$ 10-12  $\mu$ m CER is the most frequently

 observed cloud value. Using the joint histogram in Figure 3c, we can derive the mean and variance of both LWP and COT using the Eqs. *(28)* and *(29)*.

 The efficiency of using the level-3 MODIS product is accompanied by three important limitations. First of all, as mentioned earlier MODIS provides only LWP retrievals while LWC is needed in the KK2000 scheme. Second, the current level-3 MODIS cloud product has a fixed 1°x1° spatial resolution. Although this resolution is highly relevant to the current generation of GCMs, i.e., Coupled Model Intercomparison Project Phase 6 (CMIP5) (Eyring et al., 2016), future GCMs may have significantly finer resolution. Third, it is difficult to sub-sample the pixels with the best retrieval quality. These limitations will have to be addressed in future studies.

## **4. Grid-mean and subgrid variations of liquid-phase cloud properties**

 In this study, we limit our analysis to tropical oceans only where warm rain is frequent and MODIS cloud retrievals have a relatively better quality than over land or over high latitude. The 387 annual mean total cloud fraction ( $f_{tot}$ ), liquid-phase cloud fraction ( $f_{liq}$ ), in-cloud COT, CER from the 3.7 µm band, LWP and estimated CDNC over the tropical oceans based on 10 years Aqua-389 MODIS retrievals are shown in Figure 4. The highest  $f_{liq}$  in the tropics is usually found in the stratocumulus (Sc) decks over the Eastern boundary of the ocean, e.g., SE Pacific off coast of Peru, NE Pacific off the coast of California and SE Atlantic off the coast of Namibia. The liquid-cloud fraction reduces significantly toward the open ocean trade wind regions, where the dominant cloud types are broken cumulus (Cu). Close to the continents, the Sc decks are susceptible to the influence of continental air mass with higher loading of aerosols in comparison with pristine ocean environment, which is probably the reason the SC decks have smaller CER and higher CDNC than the open-ocean trade cumulus (Figure 4 d and f). The in-cloud COT (Figure 4 c) and LWP (Figure 4 e) generally increase from the Sc decks to the open-ocean Cu regime, although less dramatically than the transition of cloud fraction. The Sc decks and the Sc-to-Cu transition are the most prominent features of liquid-phase clouds in the tropics. However, as mentioned in the 400 introduction, simulating these features in the GCMs proves to be an extremely challenging task, 401 and most GCMs suffer from some common problems, such as the "too few too bright" problem and the abrupt Sc-to-Cu transition problem (Kubar et al., 2014; Nam et al., 2012; Song et al., 2018a).

404 Switching the focus now from grid-mean values to subgrid variability, we will show the 405 grid-level inverse relative variances  $v = \langle x \rangle^2 /Var(x)$  for several key cloud properties. Here, we 406 first derive the daily mean  $v$  and then aggregate the result to monthly mean values. Therefore, 407 for each grid box we have 120 samples (i.e., 10 years x 12 months) of monthly mean  $v$  for analysis 408 and visualization. Because the value of  $v$  can be ill-behaved when  $Var(x)$  approaches zero, 409 instead of the mean value, we plot the median value of  $\tilde{v}$  based on 120 months of MODIS 410 observations in Figure 5. There are several interesting and important features in Figure 5. First of 411 all, the  $\tilde{v}$  of all four sets of cloud properties (i.e., COT, CER, LWP and CDNC) all exhibits a clear 412 and similar Sc-to-Cu transition, with larger values in the Sc region and smaller value in the broken 413 Cu regions. This indicates that cloud properties, including both optical and microphysical 414 properties, are more homogenous, in terms of spatial distribution within the grid, in the Sc region 415 than in the Cu region. Secondly, the value of  $\tilde{v}$  of CER (i.e., 10~100 in Figure 5b) is larger than 416 that of the other properties (i.e.,  $1^{\sim}10$ ) by almost an order of magnitude, indicating that the 417 subgrid variability of CER is very small. On the other hand, however, it is important to note that 418 the  $\tilde{v}$  of CDNC (Figure 5d) is comparable with that of COT (Figure 5a) and LWP (Figure 5c). The 419 reason is probably in part because the highly nonlinear relationship between CDNC and CER (i.e.,

 $N_c \sim r_e$  $-\frac{5}{3}$  $1420$   $N_c \sim r_e^{-2}$  ) leads to a stronger variability of CDNC than CER, and also in part because the variability 421 of CDNC is also contributed by the subgrid variation of COT. In some regions, the Gulf of Guinea, 422 East and South China Sea, and Bay of Bengal for example, the  $\tilde{v}$  of CDNC is close to unity, 423 indicating the subgrid standard deviation of CDNC is comparable to the grid-mean values in these 424 regions. As discussed in the next section, the significant subgrid variability of CDNC in these 425 regions should be taken into account when modeling the nonlinear processes, such as the 426 autoconversion, in GCM to avoid systematic biases due to the nonlinearity effect.

427 The values of  $\tilde{v}$  in Figure 5 from this study are in reasonable agreement with previous 428 studies. Barker (1996) selected a few dozens of cloud scenes, each about 100 ~ 200 km in size, 429 from the Landsat observation and analyzed their spatial variability of COT. It is found that the 430 typical value of  $v$  for "overcast stratocumulus", "broken stratocumulus" and "scattered cumulus" 431 is 7.9, 1.2, and 0.7, respectively (see their Table 3), which is consistent with the Sc-to-Cu transition 432 pattern seen in Figure 5. Oreopoulos and Cahalan (2005) derived the subgrid inhomogeneity of

 COT on a global scale from the level-3 Terra-MODIS retrievals. Although using a different metric 434 (i.e., their inhomogeneity parameter is defined as  $\chi = \exp(\ln(\tau))/\langle \tau \rangle$ ), they also found 435 systematic increase of inhomogeneity (decreasing value of  $\chi$ ) from the Sc region to cu region. Also using the MODIS cloud property retrievals, Wood and Hartmann(2006) investigated the 437 meso-scale spatial variability of LWP in the NE Pacific and SE Pacific region. The  $v$  of LWP is found to increase systematically with meso-scale cloud fraction and the relationship between the two can be reasonably explained by a simple PDF cloud thickness model in Considine et al. (1997). See also Kawai and Teixeira (2010).

 As explained in section 2, the correlation between cloud water and CDNC can also 442 influence the computation of enhancement factor and thereby the grid-mean autoconversion 443 rate. Figure 5e shows the median value of the LWP and CDNC correlation coefficient  $\tilde{\rho}$ . Similar to 444 the derivation of median  $\tilde{v}$ , we first compute the monthly mean  $\rho$  from daily MODIS observations 445 and then derive the median value of  $\tilde{\rho}$  for each grid from the 120 months of observation. As shown in Figure 5e, at the subgrid level, the LWP and CDNC tend to be positively correlated almost over all tropical oceans. Mathematically, this is not surprising because as shown in Figure 448 5b and c, the subgrid variability of  $r_e$  is order of magnitude smaller than that of LWP. Since CDNC 449 is proportional to  $LWP^{\frac{1}{2}}r_{e}^{-3}$  according to Eq. (27), the subgrid variability of CDNC is mainly determined by the variability of LWP, leading to the positive correlation. Physically, the correlation can be explained by several mechanisms. For example, Wood et al. {\*Wood:2018cx} and O et al. {\*O:2018to} found that a large amount of low-level water clouds over the stratocumulus to cumulus transition are "optically thin veil clouds". These clouds are usually associated with low LWP and low CDNC (therefore positive correlation) and probably caused by 455 the strong precipitation scavenging process in the active cumulus. Note that our definition of  $\rho$  is the subgrid spatial correlation of LWP and CDNC. It may be different from the definition used in many aerosol indirect effect studies where the temporal correlation of monthly mean LWP and CDNC is more interested.

#### 460 **5. Implications for warm-rain simulations in GCM**

### 461 **5.1.Influence of subgrid variation of cloud water**

462 As discussed in Section 2.2, most current GCMs only considers the impact of subgrid cloud 463 water variation on autoconversion rate but ignore the impact of subgrid CDNC variation. To make 464 our analysis relevant to the current GCMs, we first analyze  $E_q$  in Eq. (20) based on observation. 465 The impacts of subgrid CDNC variation (i.e.,  $E_N$ ) and its correlation with cloud water (i.e.,  $E_{COV}$ ) 466 will be analyzed in the next section.

467 We derive  $E_q$  using two approaches. First, we derive it from the observed LWP PDF based on 468 Eq. *(20)*. As such, we do not have to make any assumption about the shape of LWP PDF although 469 solving the integration in Eq. (20) is time-consuming. In the second approach, we first derive the 470 relative inverse relative variance  $v$  of LWP and then derive the enhancement factor by assuming 471 the subgrid PDF to be either Gamma or Lognormal. This approach is more efficient, but it may be 472 subject to error if the true PDF deviates from the assumed PDF shape. Figure 6a shows the annual 473 mean enhancement factor  $E_a$  in the tropical region derived based on Eq. (20) (i.e., the first 474 approach) from 10 years of MODIS observation. Figure 6 b and c show the annual mean 475 enhancement factor  $E_a$  derived by assuming the subgrid cloud water follows the Lognormal (i.e., 476 Eq. (14)) and Gamma distribution (i.e., Eq. *(8)*), respectively. There are a couple of interesting and 477 important points to note. First of all, similar to the grid-mean quantities in Figure 4, the 478 enhancement factor  $E_q$  also shows a clear Sc-to-Cu transition. Over the Sc decks, because clouds 479 are more homogeneous ( $\tilde{v} > 5$ ), the enhancement factor  $E_q$  is only around 1  $\sim$  2.5, while over 480 the Cu regions, the more inhomogeneous clouds with  $\tilde{v} < 1$  leads to a larger enhancement 481 factor  $E_q$  around 3~5. As aforementioned, in the current CAM5.3,  $E_q$  is assumed to be a constant 482 of 3.2. While this value is within the observational range, it obviously cannot capture the Sc-to-483 Cu transition. In fact, the constant value 3.2 overestimates the  $E_q$  over the Sc region and 484 underestimates the  $E_q$  over the Cu region, which could lead to unrealistic drizzle production in 485 both regions and to consequential impacts on cloud water budget, radiation and even aerosol 486 indirect effects on the model. The second point to note is that the  $E_q$  based on the Lognormal 487 PDF assumption in Figure 6 b agrees well with the results in Figure 6 a derived directly from the 488 observation. In contrast, the  $E_q$  based on the Gamma PDF assumption in Figure 6 c tends to be

489 smaller, especially in the Cu regions. This result seems to suggest that the Lognormal distribution 490 provides a better fit to the observed subgrid cloud water variation than the Gamma distribution, 491 which has rarely been noted and reported in the previous studies.

492 A flexible, cloud-regime dependent  $E_q$  could help improve the simulation of Sc-to-Cu 493 transition in the GCM. If a GCM employs an advanced cloud parameterization scheme, such as 494 CLUBB, that is able to provide regime-dependent information on subgrid cloud variation, i.e.,  $v$ , 495 then the enhancement factor  $E_q$  could be diagnosed from  $v$ . However, most traditional cloud 496 parameterization schemes do not provide information on subgrid cloud variation. In such case, if 497 one does not wish to use a constant  $E_q$ , but a varying regime-dependent scheme, then either  $v$ 498 or  $E_a$  need to be parameterized as a function of some grid-mean cloud properties resolved by 499 the GCM. In fact, several attempts have been made along this line. Based on the combination air-500 borne in situ measurement and satellite remote sensing product, Boutle et al. (2014) 501 parameterized the "fractional standard deviation" (which is equivalent to  $1/\sqrt{\nu}$  in our definition) 502 of liquid-phase cloud as a function of grid-mean cloud fraction. This scheme was later updated 503 and tested in a host GCM in Hill et al. (2015), and was found to reduce the shortwave cloud 504 radiative forcing biases in the model. In a recent study, Xie and Zhang (2015) derived the subgrid 505 cloud variations from the ground-based observations from three Department of Energy (DOE) 506 Atmospheric Radiation Measurement (ARM) sites, and then parameterize the inverse relative 507 variance  $v$  as a function of the atmospheric stability.

508 Figure 7a shows the variation of inverse relative variance  $v$  as a function of the grid-mean 509 liquid-phase cloud fraction  $f_{liq}$ . In general, the value of  $v$  increases with the increasing  $f_{liq}$ , which 510 is expected from the Sc-to-Cu increase of  $f_{liq}$  in Figure 4b and the Sc-to-Cu decrease of  $v$  in Figure 511 5c. The  $v(f_{liq})$  pattern in Figure 7a is also consistent with the results reported in Wood and 512 Hartmann (2006) and Lebsock et al. (2013). In the hope of obtaining a simple parameterization 513 scheme for  $v(f_{liq})$  that can be used in GCMs, we fit the median value of  $v$  as a simple 3<sup>rd</sup> order 514 polynomial of  $f_{liq}$  as follows:

$$
v(f_{liq}) = 2.38 - 4.95f_{liq} + 8.74f_{liq}^2 - 0.49f_{liq}^3, f_{liq} \in [0,1].
$$
 (30)

515 To test the performance of this simple parameterization, we first substitute the  $f_{liq}$  from MODIS 516 daily mean level-3 product into the above equation and then use the resultant  $v$  to compute the 517 enhancement factor  $E_q$ . Unfortunately, the enhancement factor  $E_q$  computed based on the 518 parameterized  $v(f_{liq})$  as shown in Figure 8a substantially underestimate the observation-based 519 results in Figure 6, especially over the Cu regions. The deviation is probably because the 520 relationship between  $E_q$  and  $v$  is highly nonlinear (e.g., Eq. *(8)* and *(14)*) and therefore the above 521 parameterization scheme that only fits the = value of  $v$  is not able to capture the variability of  $522$   $E_a$ . Based on this consideration, we tried an alternative approach. Instead of parameterization 523 of v, we directly parameterize the enhancement factor  $E_q$  as a function of  $f_{liq}$ . Figure 7b shows 524 the variation of  $E_q$  as a function of  $f_{liq}$ . As expected,  $E_q$  generally decreases with increasing  $f_{liq}$ . 525 The median value of  $E_q$  is fitted with the following 3<sup>rd</sup> order polynomial of  $f_{liq}$ 

$$
E_q(f_{liq}) = 2.72 + 7.33f_{liq} - 19.17f_{liq}^2 + 10.69f_{liq}^3, f_{liq} \in [0,1].
$$
 (31)

526 As shown in Figure 8b, the value of  $E_q$  based on the above equation clearly agrees with the 527 observation-based values in Figure 6 better than that based on the parameterization of  $v(f_{lia})$ . 528 The elimination of the middle step indeed improves the parameterization results. While this is 529 encouraging, it should be kept in mind that the Eq. *(31)* has very limited application, i.e., it is only 530 useful for the autoconversion rate computation for a particular value of the autoconversion 531 exponent beta, i.e.,  $\beta_q = 2.47$ . A good parameterization of v could be useful for not only 532 autoconversion, but also for accretion and radiation computations. Another caution is that, if 533 applied to a GCM, the performance of the  $E_q(f_{liq})$  parameterization in Eq. *(31)* will be dependent 534 on the simulated accuracy of  $f_{liq}$  in the model.

535

## 536 **5.2.Influence of subgrid variance of CDNC**

537 Now we will investigate the impacts of subgrid CDNC variation on the autoconversion rate 538 simulation. For the moment, we will consider  $E_N$  only. The impact of CDNC and cloud water 539 correlation will be discussed in the next section. Similar to  $E_q$  we first derive  $E_N$  from the CDNC 540 PDF based on Eq. *(21)*. The annual mean result based on 10 years of MODIS observationsis shown 541 in Figure 9a. There are several intriguing points to note. First of all, the value of  $E_N$  is actually

542 larger than  $E_q$  in Figure 9 such that we even have to use a different color scale for this plot. 543 Secondly,  $E_N$  the regions with escalated  $E_N$  seem to coincide with the downwind regions of 544 biomass burning aerosols (e.g., Gulf of Guinea, East Coast of South Africa), air pollution (i.e., 545 Eastern China Sea), and, most interestingly, active volcanos (e.g., Kilauea Hawaii and Ambae 546 Vanuatu). We have also checked the seasonal variation of the  $E_N$  and the results also support 547 this observation. Another interesting feature to note is that, although the dust outflow regions 548 such as Tropical East Atlantic and Arabian Sea, have heavy aerosol loading, the value of  $E_N$  there 549 is only moderate. Figure 9b shows the value of  $E_N$  computed based on Eq. (14) from the inverse 550 relative variance of  $v$ , assuming that the subgrid CDNC follows a Lognormal PDF. Although the 551 overall pattern is consistent with Figure 9a, the assumption of Lognormal PDF seems to 552 underestimate  $E_N$ . A closer examination indicates that the Lognormal PDF tend to underestimate 553 the population of clouds with small CDNC, and therefore underestimate the variance of CDNC as 554 well as  $E_N$ . We did not compute the  $E_N$  based on the Gamma distribution because of the singular 555 value problem aforementioned in Section2.1.

556 We could not find any previous observation-based study on the global pattern of the 557 subgrid variation of CDNC and the corresponding  $E_N$ . So, it is difficult for us to corroborate our 558 results. On one hand, the magnitude of  $E_N$  is surprisingly large. As explained in Section 3, the 559 CDNC is estimated based on Eq. *(27)* from the MODIS retrieval of COT and CER. Several previous 560 studies have shown that the sub-pixel level surface contamination, subpixel cloud 561 inhomogeneity, and three-dimensional radiative transfer effects, can cause significant errors in 562 the MODIS CER retrievals especially over broken cloud regions (Zhang and Platnick, 2011; Zhang 563 et al., 2012; 2016). Given the fact that the CDNC retrieval is highly sensitive to CER error as a result of  $N_d \sim r_e$  $-\frac{5}{2}$ 564 result of  $N_d \sim r_e^{-2}$ , the influence of retrieval uncertainty on subgrid CDNC variation cannot be 565 ruled out. On the other hand, the pattern of  $E_N$  in Figure 9a seems to suggest that there are some 566 underlying physical mechanisms controlling the subgrid variation of CDNC, in which aerosols 567 seem to play an important role. To achieve a better understanding, we analyzed the dependence 568 of  $E_N$  on liquid cloud fraction and grid-mean CDNC in Figure 10, which reveals that  $E_N$  has a 569 stronger dependence on CDNC than cloud fraction. This result seems to indicate that the pattern 570 of  $E_N$  in Figure 9 is largely determined by physical mechanisms rather than retrieval

571 uncertainties. Interestingly, the largest  $E_N$  is usually found when liquid cloud fraction is small and CDNC is large and decreases with decreasing CDNC and increasing cloud fraction. This pattern leads us to the following hypothesis: In the regions where aerosol is limited, even weak updraft can activate most cloud condensation nuclei (CCN). As a result, even if there is significant subgrid variation of turbulence at cloud base, the subgrid variation of CDNC remains small. In contrast, in regions where aerosol is abundant, the subgrid variation of turbulence becomes important. 577 The subgrid variation of updraft leads to subgrid variation CDNC and thereby large  $E_N$ .

578 As far as we know, the results in Figure 9 and Figure 10 mark the first attempt based on 579 satellite observations to unveil the global pattern of the subgrid variations of CDNC and 580 investigate the consequential impacts on warm rain simulations in GCMs. Although obscured by 581 satellite retrieval uncertainties, the results still provide valuable insights. First of all, the 582 enhancement factor  $E_N$  due to the subgrid variations of CDNC is nonnegligible, even comparable 583 the effect of subgrid cloud water variation (i.e.,  $E_q$ ). Second, the global pattern of  $E_N$  in Figure 9 584 provides a valuable map for future studies.

## 585 **5.3.The combined effect of subgrid variations of cloud water and CDNC**

586 Finally, in this section we examine the combined effect of subgrid variations of cloud 587 water and CDNC, as well as their correlation, on the autoconversion rate simulation. The annual 588 mean combined enhancement factor E derived based on Eq. (17) from 10 years of MODIS COT 589 and CER observation is shown in Figure 11a. Comparing to the  $E_q$  in Figure 6 and  $E_N$  in Figure 9, 590 the combined enhancement factor is generally larger. It is easy to see that the in some regions 591 (e.g., Gulf of Guinea, East Coast of South Africa and Eastern China Sea) the combined 592 enhancement factor E resembles the  $E_N$  while in other regions (i.e., trade wind cumulus regions 593 over open ocean) it resembles more of  $E_q$ . Interestingly, because both  $E_q$  and  $E_N$  are small over 594 the Sc decks, those regions have the smallest combined enhancement factor  $E$ . As discussed in 595 Section 2.2, only when the subgrid variation of cloud water is uncorrelated with the subgrid 596 variation of CDNC can the combined enhancement factor  $E$  be decomposed into the simple 597 product of  $E_q$  and  $E_N$  (i.e., Eq. (19)). Figure 11b shows the annual mean value of the simple 598 product  $E_q \cdot E_N$ , without considering the correlation between cloud water and CDNC. Evidently, 599 the simple product substantially overestimates the combined enhancement factor derived from  the joint PDF of LWP and CDNC. This result can be explained by the mostly positive subgrid correlation between LWP and CDNC in Figure 5e. As explained in section 2.2, the positive correlation means that clouds with more water also tend to have more CDNC. The autoconversion rate of such configuration is lower than that when LWP and CDNC have no correlation.

605 Together, the  $E_q$  in Figure 6,  $E_N$  in Figure 9 and the combined enhancement factor in Figure 11 lead us to the following important conclusion. It is not sufficient to consider only the 607 impact of subgrid variation of cloud water (i.e.,  $E_q$ ) on the autoconversion rate simulation. The influences of subgrid CDNC variation, as well as the correlation between cloud water and CDNC, must also be taken into account to avoid significant error.

 Finally, the combined enhancement factor derived based on Eq. (*23*) assuming that the LWP and CDNC follow the bi-variate lognormal distribution is shown in Figure 11c. Despite the tendency of overestimation, the result agrees reasonably well with that based on observed joint 613 PDF in Figure 11a, clearly better than the simple product  $E_q \cdot E_N$ . This is encouraging as it suggests that the bi-variate lognormal distribution can be used in the future to model the combined effect of cloud water and CDNC on autoconversion rate simulation in GCMs.

### **6. Summary and Outlook**

 One of the difficulties in GCM simulation of the warm rain parameterization is how to account for the impact of subgrid variations of cloud properties, such as cloud water and CDCN, on nonlinear precipitation processes such as autoconversion. In practice, this impact is often treated by adding the enhancement factor term to the parameterization scheme. In this study, we derived the subgrid variations of liquid-phase cloud properties over the tropical ocean using the satellite remote sensing products from MODIS and investigated the corresponding enhancement factors for parameterizations of autoconversion rate. In comparison with previous work, our study is able to shed some new light on this problem in the following regards:

 1. A theoretical framework is presented to explain the importance of the subgrid variation of CDNC and its correlation with cloud water on the autoconversion rate simulation in GCMs.

- 2. The wide spatial coverage of the Level-3 MODIS product enables us to depict a 630 detailed quantitative picture of the enhancement factor  $E_a$ , which shows a clear 631 cloud regime dependence, i.e., a Sc-to-Cu increase. The constant  $E_q = 3.2$  used in 632 the current CAM5.3 model overestimates and estimates the observed  $E_q$  in the Sc and Cu regions, respectively.
- 634 3. The  $E_q$  based on the Lognormal PDF assumption performs significantly better than that based on the Gamma PDF assumption. A simple parameterization scheme is 636 **provided to relate**  $E_q$  **to the grid-mean liquid cloud fraction, which can be readily** used in GCMs.
- 638 4. For the first time, the enhancement factor  $E_N$  due to the subgrid variation of CDNC is derived from satellite observation, and the results reveal several regions downwind of biomass burning aerosols (e.g., Gulf of Guinea, East Coast of South Africa), air pollution (i.e., Eastern China Sea), and active volcanos (e.g., Kilauea 642 **Hawaii and Ambae Vanuatu). The largest**  $E_N$  **is usually found where CDNC is large**  and liquid cloud fraction is small and decreases with decreasing CDNC and increasing cloud fraction.
- 5. MODIS observations suggest that the subgrid LWP and CDNC are mostly positively correlated. As a result, the combined enhancement factor is significantly smaller 647 than the simple product of  $E_q \cdot E_N$  (i.e., assuming no correlation). The combined enhancement factor derived assuming LWP and CDNC to follow the bi-variate lognormal distribution agree with the observation-based results reasonably well.
- As noted in the previous sections, this study has several important limitations, most of which are a result of using the level-3 MODIS observations. The fixed 1°x1° spatial resolution of MODIS level-3 product makes it impossible for us to investigate the scale-dependence of subgrid cloud variation. Similar to previous studies, we have to make several assumptions when estimating the CDNC from level-3 MODIS product. Furthermore, the retrieval uncertainties associated with the optically thin clouds in MODIS product pose a challenging obstacle for the quantification of subgrid cloud property variations and the corresponding enhancement factors. These limitations have to be addressed using additional independent observations from, for

 example, ground based remote sensing product and/or in situ measurement from air-borne field campaigns. Recently, a few novel methods have been developed to provide certain information on the subgrid cloud property variations to the host GCM. Most noticeable examples are the super-parameterization method (a.k.a. multi-scale modeling framework) (Wang et al., 2015) and the PDF-based higher-order turbulence closure methods (e.g., Cloud Layer Unified By Binormals, CLUBB (Golaz et al., 2002a; Guo et al., 2015; Larson et al., 2002) and Eddy-Diffusivity Mass-Flux (EDMF) (Sušelj et al., 2013)). The subgrid cloud property variations derived in this study provide the valuable observational basis for the evaluation and improvement of these schemes. 

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687 Figures:



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689 *Figure 1* a) The probability density function (PDF) and cumulative distribution function (CDF) of 690 cloud LWC ( $q_c$ ) that follow the Gamma (dashed) and Lognormal (solid) distribution. For the both 691 distributions,  $\langle q_c \rangle = 0.5g/kg$  and  $v_q = 3.0$ . b) The PDF and CDF of autoconversion rate 692 computed based on the KK2000 scheme in Eq.(15) and the PDF of  $q_c$ . In the computation, the  $N_c$ 693 is kept at a constant of 50  $cm^{-1}$ . c) The PDF and CDF of  $N_c$  that follow the Gamma (dashed) and 694 Lognormal (solid) distribution. For the both distributions,  $\langle N_c \rangle = 50 cm^{-3}$  and  $v_N = 5.0$ . d) the 695 PDF and CDF of the autoconversion rate computed based on the KK2000 scheme in Eq. (15) and 696 the PDF of  $N_c$ . The  $q_c$  is kept at 0.5 $g/kg$  in the computation.



 Figure 2 Enhancement factors based on Lognormal  $E(P_L, \beta)$  and Gamma  $E(P_G, \beta)$  subgrid PDF

700 *for different*  $\beta$  *as a function of the inverse relative variance*  $\nu$ *.* 



 *Figure 3 The (a) LWP and (b) CDNC as a function of COT and CER. (c) An exmaple of the COT-CER Joint histogram observed by Aqua-MODIS on Jan. 09<sup>th</sup>, 2007 at 1°S and 1°W.* 



*Figure 4* 10-year (2007~2016) averaged annual mean a) total cloud fraction, b) liquid cloud

711 fraction, c) cloud optical thickness, d) cloud effective radius retrieved from the 3.7  $\mu$ m band, e)

cloud wather path and f) cloud droplet concentration retrievals from Aqua-MODIS over the

713 tropical (30° S-30° N) oceans. All quantaties are "in-cloud" mean that are averaged over the

cloudy-part of the grid only.



*Figure 5* Median value of the inverse relative variance (i.e.,  $v = \langle x \rangle^2 / Var(x)$ ) for a) COT, b)

CER, c) LWP and d) CDNC, and e) median value of the correlation coefficient between LWP and

 CDNC derived from 10 years of MODIS observations. Note that the color scale of CER is different from others'.



723 Figure 6 The annual mean factor for the KK2000 scheme due to subgrid variation of LWP 725 computed a) directly from observation, i.e.,  $E_q$  in Eq.(20), b) from relative variance assuming 726 Lognormal PDF of LWP, i.e.,  $E_q$  in Eq.(14) and c) from relative variance assuming the Gamma

727 PDF of LWP i.e.,  $E_q$  in Eq.(8).







LWP subgrid variability assuming Log-normal PDF as a function of grid-mean liquid cloud

734 fraction, where the solid line, dark shaded area, and light shaded area correspond to the<br>735 median value, 25%~75% percentiles, and 10~90% percentiles, respectively. The dotted lin

median value, 25%~75% percentiles, and 10~90% percentiles, respectively. The dotted lines

736 correspond to simple 3-rd order polynomial fitting.



  $2.38-4.95f_{liq} + 8.74f_{liq}^2 - 0.49f_{liq}^3$  parameterization scheme in Eq. (30) and b)  $E_q(f_{liq}) =$ 

 $2.72+7.33f_{liq} - 19.17f_{liq}^2 + 10.69f_{liq}^3$  parameterization scheme in Eq. (31).







747 *Figure 9 Annual mean value of the enhancement factor*  $E_N$  *derived from a) observation based*  $748$  *on Eq.* (21) and b) from Eq. (14) assuming Lognormal subgrid CDNC distribution. *on Eq.* (21) *and b) from Eq.* (14) *assuming Lognormal subgrid CDNC distribution.* 



 Figure 10 Dependence of  $E_N$  on  $f_{liq}$  and  $N_d$ . The color map corresponds to the mean value of  $E_N$ 

752 *for a given N<sub>d</sub> and*  $f_{liq}$  *bin. The white contour lines correspond to the relative sampling* 

*frequency of*  $N_d$  and  $f_{liq}$  bins (i.e., the most frequently observed combination is  $N_d \sim 50 cm^{-3}$ 

**and**  $f_{liq}$  ~0.1 ).



 *Figure 11 The combined enhancement factor derived a) based on Eq. (17) from the observed joint PDF of LWP and CDNC, b) assuming that subgrid variations of LWP and CDNC are*  761 *uncorrelated, i.e.,*  $E_q \cdot E_N$  *only and c) based on Eq.* (23) *assuming that the subgrid LWP and* 

*CDNC following the bi-variate lognormal distribution.*

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