## Response to Reviewers

# Response to Reviewer #1 Matthew Lebsock

I would like to thank Dr. Matthew Lebsock for his insightful and suggestive comments that helped us substantially improve the manuscript. Point-to-point replies to the comments are provided below (reviewer's comments in italic blue font).

### General comments:

This paper uses Daily gridded Level-3 histograms of MODIS cloud retrievals to derive the small-scale variability in liquid cloud properties, specifically cloud liquid water path (lwp) and droplet number concentration(cdnc). This is the first study to address the variance of cdnc from satellite data. The variability is then used to diagnose the expected enhancement of the autoconversion process due to sub-grid scale distribution of cloud fields in global models. The regional variation of the enhancement are shown. Surprisingly the enhancement due to variability in the cdnc is shown to be often larger than that due to lwp.

The largest enhancement due to number concentration variability is correlated with number concentration itself. This correlation is largely unexplained and a major result of the paper. There is a limited attempt to attempt to explain the unexpectedly large cdnc enhancement factor based on retrieval uncertainty in broken cloud scenes but the authors should consider physical mechanisms as well. I would suggest that thin detrained veil clouds near precipitating cumulus could be a physical mechanism for seeing this variability in the observations.

The science focus of this paper is novel and timely, the methodology is appropriate, and the presentation is generally good. I've included some additional references to add and specific comments below. In terms of additional analysis I would advocate quantifying the correlation between E\_n and other cloud properties on various scales (correlate 1 degree grids (super pixel), correlate spatial patterns) to identify the controlling factors. This will help us better understand what variables might be influencing the high E\_n (i.e. cloud fraction, low optical depth, CDNC, LWP, etc. A Table might work well to present these results.

Reply: Thanks for the review and helpful comments. Following your suggestions, we made significant revisions to the paper. Major changes include:

- We added more discussions on the correlation between LWP and CDNC and its implications for enhancement factor.
- We also provide some possible physical explanation on the large  $E_N$ . Please see details below.
- Figure 5, 7, 10, 11 are updated.

## Specific comments:

Lines 123-128: add Ahlgrimm et al., 2016 (https://doi.org/10.1002/qj.2783). They also use DOE data and create a parameterization of E based on cloud fraction.

Reply: Thanks. The paper is added to the citation list.

Line 135: Add citation to Takahashi et al., 2017 (https://doi.org/10.1002/2016JD026404). They have shown that more advance parameterization, specifically a version of the Multiscale Modeling Framework model is able to produce reasonable distributions of regional distributions of the cloud water heterogeneity when compared against the satellite observations (their figure 2).

Reply: Thanks. The paper is added to the citation list.

Line 137: Somewhere in here you should point out that the estimate of variance depends on the spatial resolution of the observations. With satellite observations (even MODIS) we are using relatively coarse observations and therefore we cannot resolve variance on the smallest scales. So satellite observations will necessarily underestimate variance because of this effect, however, they should provide an accurate assessment of regional distributions of the microphysical process enhancement factors.

Reply: Good point. Some discussions on the limitations of satellite observations are added after the Lebsock (2013) study.

Line 146: I wouldn't say that the 'empty cloud' problem is a well defined term. I can guess what this means but I would state explicitly a diagnosis of the problem. Probably there is too much rain and clouds with very low or zero liquid water path at the end of the time step?

Reply: You are right. "empty clouds" have near-zero cloud water which is caused by excessive rain (Song et al. 2018). This sentence is revised.

Line 246: I think that E q should be E N here and cloud water should be CDNC.

Reply: Thanks for catching this. It is revised.

Figure 1B/Line 262/Line 263, and elsewhere: What is plotted here is not the rain rate. It is rate of conversion cloud water to precipitation water (or the autoconversion process rate). Rain rate is the integral of over the precipitation drop size distribution multiplied by the density-dependent fall velocity for each drop radius. This should be corrected throughout the manuscript.

Reply: Thanks for pointing this out. We should be more careful. In the revised manuscript, we use "autoconversion rate", instead of "rain rate" throughout the paper.

Line 318: You should point out that calculating the nu parameter in this way can be very sensitive to outliers as the sample size gets small (i.e. low cloud fraction) and there are other methods to calculate nu from the data (e.g. Oreopoulos and Cahalan, 2005) that will give different answers.

Reply: Thanks for pointing this out. Indeed, the method we used in this study is the method of moment (MOM). The inverse relative variance can also be estimated using the maximum likelihood estimate (MLE). We pointed this out in the revised manuscript.

The MODIS level 3 product reports the logarithm mean of cloud optical thickness which enables us to use the MLE method to estimate the  $\nu_{MLE}$  from Eq. 6 of Oreopoulos and Cahalan (2005). The results are shown below compared with the value from the MOM  $\nu_{MOM}$ . Apparently,  $\nu_{MLE}$  tends to be larger than  $\nu_{MOM}$  especially over regions with low water cloud fraction, although the spatial pattern is similar. This is probably because, as you pointed out, the MOM is more prone to the impact of extreme values when cloud fraction is small. Nevertheless, the difference does not change any conclusions.

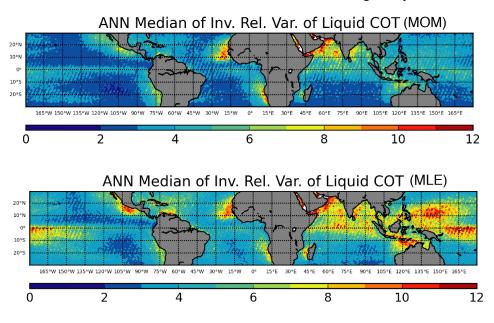


Figure 5: The caption says these are means, as does panel b. But the other panels say median as does the paper text. Which is it? Median I think. . ..

Reply: It's a typo and should be "Median". Corrected.

Line 327: Lebsock et al., 2011 (https://doi.org/10.1175/2010JAMC2494.1) also argue this about 3.7 micron re.

Reply: Thanks. This paper is cited in the revised version. Of course, the choice of coefficient for LWP computation does not matter in this study because it is a common

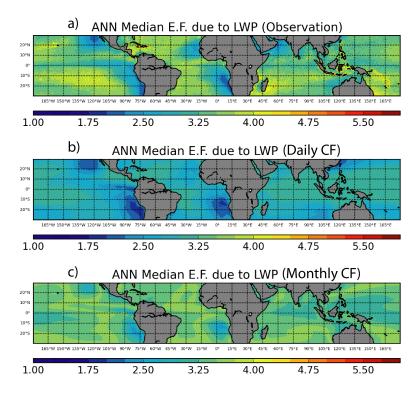
factor in both numerator and denominator in the calculation of v.

Figure 8: you should describe in the caption the difference in the fitting so the reader doesn't have to go to the text and equations.

Reply: Good suggestion. We added the information.

Figure 8: I do think it is useful to show that the parameterization of nu based on cloud fraction does not work because of the non-linearity in the process. However, can you explain why even the parameterization of the enhancement factor directly on cloud fraction under-predicts the direct calculation? That relationship show in 7b is fairly normally distributed so I can't understand why the parameterization would not get the median about right.

Reply: This is a very good question. To answer it, first let us explain how  $E_q$  in Figure 6 and Fig. 8 are obtained. The parameterization scheme in Eq. (27) and Fig. 7 are developed based on the relation between monthly-mean observation-based  $E_q$  and monthly-mean  $f_{liq}$  in the tropics (i.e., 10 years x 12months x 360 longitude x 60 latitude x fraction of ocean). The sample size would be too large if daily products were used. After we obtained the parameterization scheme (i.e., Eq. 27), we then used it to compute the daily  $E_q$  based on daily  $CF_{liq}$ . The daily  $E_q$  values are then temporally aggregated, weighted by daily  $f_{liq}$ , to first obtain monthly and then annual  $E_q$  in Fig. 8b in the same way as we obtain observed  $E_q$  in Fig. 6. Going back to your question, we think the underestimation of parameterized  $E_q$  (Figure 8b compared to Figure 6a) is due to the fact that the parameterization is developed based on monthly data but applied to daily  $f_{liq}$ . To test this, we applied the parameterization scheme to monthly  $f_{liq}$ . The results are significantly better. See below.



The lesson learned is that the simple parameterization scheme developed based on monthly  $f_{liq}$  cannot capture the day-to-day variation of  $E_q$ , which is not surprising. In our view, the parameterization scheme is only better than assuming a constant  $E_q$  in the sense that it can capture the cloud regime dependence. However, it would be unrealistic to hope that it can simulate the dramatic instantaneous variation. For that, we would have to rely on advanced scheme like CLUBB or MMF.

Eqs. 26/27 and related discussion: I don't like this parameterization of nu based on cloud fraction because it isn't well justified physically. Ideally both the cloud fraction and nu could be calculated from either prognostic or diagnostic distribution of the subgrid co-variability of total water and temperature. CLUBB in fact can do this so there should be no need to for such an ad-hoc representation. It is true that such relationships have been advocated in the past but they strike me as very unphysical. I wouldn't advocate this in the context of CLUBB, which is heavily referenced here.

Reply: We agree with your point about the parameterization of  $\nu$ . The highly non-linear relation between  $\nu$  and the enhancement factor makes the parameterization not so useful. It is shown here simply because some previous studies, e.g. Boulte et al. (2014), Xie and Zhang (2015), had tried to parameterize the  $\nu$  directly. The unsatisfying results motived us to parameterize the enhancement factor directly.

On the other hand, we think the direct parameterization of enhancement factor is meaningful. It provides with a simple way for those GCMs without advanced sub-grid parameterization scheme to account for the impacts of cloud inhomogeneity on precipitation simulation. We agree that CLUBB presumably would do a better job than simple parameterization. Nevertheless, the results from this study, including the parameterization of enhancement factor, provide observational basis for evaluating the results from CLUBB.

Line 603: One physical interpretation of the MODIS retrievals of high effective radii in these broken cloud scenes is that they could be 'optically thin veil' clouds as described by O et al. (https://doi.org/10.1029/2018GL077084) to be extensive detrained anvil cloud from shallow cumulus with low liquid water content and very low CDNC -> thus potentially large radius. Indeed they are often seen by cloud radar (Wood et al., 2018). Now if this is the case in reality these clouds might contribute quite a bit to the variance in CDNC but shouldn't lead to any substantial increase in the autoconversion because the low CDNC pixels should also have very low liquid water path -> so the correlation should matter. In fact you show this exact correlation later on.

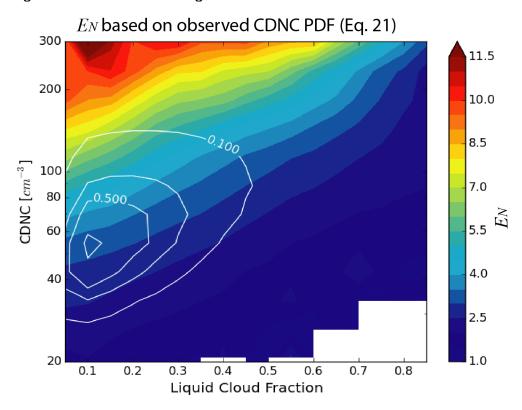
Reply: This is a very insightful comment and thanks for the references (we are aware of Wood et al. 2018 but not O et al.). Following your suggestions, we have de-emphasized the influence of retrieval error and focused more on the potential physical processes that lead to the large subgrid CDNC variance. These papers are now in Section 4 when discuss the new Figure 5 e about the correlation between LWP and CDNC and the its implications.

Line 603: I think it is important to explore and show some correlations between the E\_N and various other parameters, such as CDNC, cloud fraction, number of pixels with cloud optical depth < 4. Clearly if some factor is influencing E\_N (Like cloud fraction) you would expect to see some correlation between the variables. You could show either the regional correlations, or do this for individual 1 degree grids. It seems quite clear that there is not a good correlation between liquid cloud fraction and E\_N which doesn't support the idea that cloud-fraction related retrieval artifacts have much to do with these results. If on the other hand the E\_N mostly correlates with large CDNC, which I suspect it does, then there is a mystery yet to be explained.

Reply: Thanks for the great suggestions! We made several significant changes to the paper accordingly. We replaced original Figure 10 (which focuses on the retrieval artifacts) with an analysis of the dependence of  $E_N$  on liquid cloud fraction and CDNC. See below. As you suspected,  $E_N$  shows a stronger dependence on CDNC than cloud fraction, which seems to suggest that the dependence is largely due to some underlying physical mechanisms rather than retrieval artifacts. The largest  $E_N$  is usually found where CDNC is large and cloud fraction is small and it decreases with decreasing CDNC and to a less extent also with increasing cloud fraction. The strong dependence of  $E_N$  on CDNC might be explained by the following mechanism in which aerosol plays an important role: when aerosol loading is small, even weak updraft can activate most CCN. As a result, the subgrid turbulence and variance of thermodynamical conditions are not importance leading to small  $E_N$ . In contrast, when aerosol loading is large, subgrid variations of updraft and thermodynamical conditions could lead to significant subgrid variations of CDNC, leading to large  $E_N$ .

In addition to the analysis  $E_N$ , we also added some more in-depth explanation of the importance of LWP and CDNC correlation on enhancement factor simulation at the end of Section 2.2. First, a formula for *combined* enhancement based on the bi-variate lognormal distribution is presented (Eq. 22). Second, we pointed out that the current GCMS, even those with advanced sub-grid parameterization such as CLUBB, only consider the enhancement factor due to LWP  $E_q$ , the effect of  $E_N$  and the correlation term  $E_{COV}$  are ignored. Moreover, an equation is added (Eq. 25) to explain under what circumstances would  $E_q$  underestimate or overestimate the combined effect  $E_q \cdot E_N \cdot E_{COV}$ . In addition, Figure 5 e is added to show the subgrid correlation coefficient of LWP and CDNC and in Figure 11 we discussed the importance of considering  $E_{COV}$  in computing the combined enhancement factor.

We feel that these revisions, based on your suggestions, had made the paper more insightful and more revealing.



Dependence of  $E_N$  on  $f_{liq}$  and  $N_d$ . The color map corresponds to the mean value of  $E_N$  for a given  $N_d$  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  $f_{liq} \sim 0.1$ ).

Line 646: Again, I think that there may be a physical explanation for this correlation. Specifically that there are a lot of these low water, low N veil clouds around shallow convection.

Reply: See our reply above.

Line 665: I would argue significantly better.

Reply: agree and revised.

Line 683: Another example of parameterization that includes subgrid information is the EDMF approach (e.g. Sušelj et al., 2013, https://doi.org/10.1175/JAS-D-12-0106.1), variants of which are used in a number of models.

Reply: we added the EDMF as another example of "advanced subgrid cloud parameterization scheme". Thanks for pointing it out.

## Technical comments:

Line 41 The phrasing 'clear cloud' might be confusing. Consider 'obvious' or 'demonstrable' instead of clear.

Line 94: superfluous 'on'

Line 369: the 2~4 notation seems odd to me. I would use ~2-4 COT and ~10-12 um.

Line 402: 'dominate' -> 'dominant'

Line 458: missing 'of'

Line 480: grammar, missing word after more. Line

493: 'product' -> 'production'

Line 496: second 6b should be 6a. Line 497: 'tend' -> 'tends'

Line 508: 'facts' -> 'fact'

Eqs. 26/27: parenthesis don't match.

Reply: Thanks a lot for catching these typos and mistakes. They are all corrected.

I would like to thank the reviewer for the comments and suggestions. Point-to-point replies to the comments are provided below (reviewer's comments in italic blue font).

Authors derived the subgrid variations of liquid-phase cloud properties over the tropical ocean and investigated the autoconversion enhancement factors using MODIS product. This paper is well written, and of relevance to a broad audience. It is worthy of publication subject to the following issue.

(1) Authors assumed that subgrid variation of LWC could be inferred from the spatial variability of LWP. LWP is the vertical integrated LWC over cloud depth, so its subgrid variations include cloud depth variations. But LWC's variations does not. Please justify this assumption.

Reply: Indeed, MODIS retrievals only provide the LWP instead of the vertically resolved LWC retrieval. This is an important limitation of this study which we pointed out clearly in Section 3.

However, as we also pointed out, other techniques face more or less similar challenge. "We note here that it is the LWC  $q_{\rm C}$ , 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 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."

Typos: (1) Lines 359 ": : :any type of data quality-based data", Should be ": : :any type of quality-based data".

- (2) Lines 396 ": : :On the hand ", Should be ": : :On the other hand".
- (3) Lines 466-467 "... Figure 6 b derived directly from the observation", Should be
- ": :: Figure 6 a derived directly from the observation."

Reply: thanks for catching these typos. They are all corrected

I would like to the reviewer for the insightful and suggestive comments that helped us substantially improve the manuscript. Point-to-point replies to the comments are provided below (reviewer's comments in italic blue font).

This paper discusses the GCM sub-grid scale variability of cloud water content and droplet number observed by MODIS, and the consequences this variability has for autoconversion parametrization in GCMs. This has become a popular topic in recent years with many papers discussing the cloud water content variability, although the attempts to discuss droplet number variability are particularly novel and welcome in this study. The paper is well written and interesting. I have compiled a list of relatively minor comments or suggestions that the authors may wish to consider.

## General comment -

the paper is very long, I'd encourage the authors to look for opportunities to be more concise in their descriptions and refrain from repetition of points.

Reply: The theoretical background part is longer than we hoped but necessary so the readers to understand the studies that followed. The length of the revised version is reduced by one page. It is not trivial considering that we extend the scope of the research significantly.

L57 - the reference here should be Boutle et al. (2014, QJ) not Boutle & Abel (2012) Reply: we updated the references.

L60-62 - would be good to clarify a couple of things in these lines. Firstly, I think it would be better to refer to autoconversion and accretion "parametrizations" rather than "processes" - we shouldn't confuse the way we parametrize these things with physical reality, as there is not much overlap! Secondly, you should also clarify that you are ignoring variability in rain water content (qr) or Nc, as the nonlinearity of these (and correlations with qc) could strongly influence the result.

Reply: Agree, the KK2000 is simply a parameterization based on the least-square fitting to the LES results. We change the wording from "process" to "parameterization" throughout the text whenever appropriate.

We pointed out at the beginning of section 2.2 that we will only focus on the simulation of autoconversion while other processes such as accretion have been investigated in previous studies.

L92-95, 99-106 - this reads a little harshly on Boutle et al. (2014), who also used CloudSat data in their analysis to give a global perspective (and discussed the increase in variability from Sc to Cu and importance of co-variability on accretion). It might be worth mentioning the study of Hill et al. (2015, QJ) here as well, who extended this work to explicitly build in the regime dependence to the parametrizations. Also there is a typo on L104/5, which should say "cloud water variance is larger over the Cu region than over the Sc region".

Reply: Agree, we revised the discussion, added the Hill et al. (2015) and also corrected the typo.

L117-118 - again, might be good to clarify here - Boutle et al. (2014) and Lebsock et al. (2013) discuss the variation in rain water (which is distinct from cloud water). But you are correct that I'm also unaware of any studies looking at CDNC variability.

Reply: Following your suggestion, we pointed out again that *Boutle et al.* (2014) and *Lebsock et al.* (2013) have investigated the variation of subgrid cloud water as well as rain water.

L194 - would be good to clarify here - it's not the LES that was important in KK2000, but the fact that they used a bin-resolved microphysics scheme, which accurately represented the physical processes of collision-coalescence, to derive the simple parametrizations

Reply: Agree. In the revised version, we pointed out after the introduction of KK2000 parameterization scheme that the KK2000 is "derived through a least-square fitting of the autoconversion rate results from a large-eddy simulation with bin microphysics that can simulate the process-level physics."

Figure 1d - I cannot see this referred to at all in the text, yet it shows something interesting/puzzling to me, namely a significantly different CDF of rain rate for the gamma and lognormal distributions of CDNC - can you explain why this is?

Reply: As shown in Figure 1, provided the same mean value and same inverse relative variance v, the lognormal distribution  $P_L(x)$  is generally larger than the Gamma distribution  $P_G(x)$ . The difference is clearly visible when x > 2.0 in Figure 1 b. This differences in PDF gives rise to the difference in the CDF of autoconversion rate.

L256-260 - I'd always thought part of the argument for ignoring Nc is that its value is typically linked to the underlying aerosol distribution, which varies on much larger spatial scales than qc, therefore the amount of Nc variability that would be 'sub-grid' is

## expected to be small/negligible.

Reply: Aerosol loading is only on part of the story. CDNC is not only determined by aerosol loading but also critically be the subgrid turbulence (i.e., updraft). Many previous studies have pointed out the importance of subgrid variations of updrafts in simulating cloud microphysics in GCM (e.g., (Morales and Nenes, 2010)). However, most GCMs lack the capability of simulating the subgrid variations of updrafts until very recently the advanced parameterization schemes, such as CLUBB and MMF, became available.

L274 - please define CER as this has not been previously defined

Reply: It is clarified.

L278 - brackets should be around the years only of Platnick et al. (2013,2017)

Reply: Corrected.

L344 - I'd say the current generation of GCMs are those being used for CMIP6, so perhaps update this and the reference (although 1x1degree still doesn't seem unreasonable for what many models are running)

Reply: Updated and added the new reference (Eyring et al., 2016)

L372 - should say "dominant" cloud types

Reply: Changed.

L450 - I think there is something missing from this sentence - "this approach is more although it may be..."

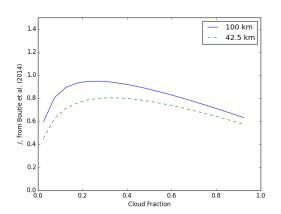
Reply: It should be more "efficient". Corrected.

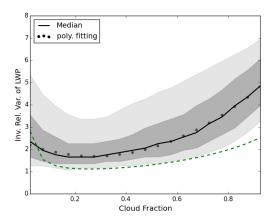
Figure 6d - does not appear to be referred to in the text. It should either be discussed why it is relevant, or not shown.

Reply: It is removed. Thanks for catching this.

L487-501 - given there is already a parametrization of v(f\_liq) in existence, namely that of Boutle et al. (2014), it would be interesting and very easy to see how well their parametrization compares to the independent MODIS dataset generated in this study. It's probably too much work to investigate the Hill et al (2015) parametrization as that would require a way of determining from MODIS whether cloud is convective or not, but that would also be interesting.

Reply: We first replicate the Figure 4 in Boutle et al. (2014) to confirm our code works consistently with the original result.





Then, we compared the parameterization scheme from *Boutle et al. (2014)* for grid size ~ 100km to out Figure 7a (the green dashed line). Apparently, there are some differences between the two especially for large cloud fraction, probably because the two studies are based on different data. Since the difference between the two studies are out of the scope of this paper. These figures are not shown in the paper.

L506-516 - it would also be worth noting that these fits are only applicable to a single model resolution, and so not as useful as existing parametrizations with inbuilt scale adaptiveness.

Reply: Agree and we already mentioned that this parameterization is only valid for 1x1 degree model resolution when we list the important limitation of this study.

# L559 - there is reference to supplementary materials, yet I cannot find any?

Reply: We originally planned to add the seasonal plots (e.g., DJF and JJA) in the supplementary materials, but we found that seasonal plots do not really add any additional insights. So, we simply removed them. Sorry for the confusion.

Boutle, I. A., Abel, S. J., Hill, P. G. and Morcrette, C. J.: Spatial variability of liquid cloud and rain: observations and microphysical effects, Quarterly Journal of the Royal Meteorological Society, 140(679), 583–594, doi:10.1002/qj.2140, 2014.

## Reference:

Boutle, I. A., Abel, S. J., Hill, P. G. and Morcrette, C. J.: Spatial variability of liquid cloud and rain: observations and microphysical effects, Quarterly Journal of the Royal Meteorological Society, 140(679), 583–594, doi:10.1002/qj.2140, 2014.

Eyring, V., Bony, S., Meehl, G. A., Senior, C. A., Stevens, B., Stouffer, R. J. and Taylor, K. E.: Overview of the Coupled Model Intercomparison Project Phase 6 (CMIP6) experimental design and organization, Geosci. Model Dev., 9(5), 1937–1958, doi:10.5194/gmd-9-1937-2016, 2016.

Morales, R. and Nenes, A.: Characteristic updrafts for computing distribution-averaged cloud droplet number and stratocumulus cloud properties, J. Geophys. Res., 115(D18), 1227, 2010.

Satellite Observations and Implications for Warm Rain Simulation in  Climate Models  Zhibo Zhang <sup>1,2*</sup> , Hua Song <sup>2</sup> , Po-Lun Ma <sup>3</sup> , Vincent E. Larson <sup>4</sup> , Minghuai Wang <sup>5</sup> , Xiquan Dong <sup>6</sup> ,  Jianwu Wang <sup>7</sup> Physics Department, University of Maryland Baltimore County (UMBC), Baltimore, MD, USA  Joint Center for Earth Systems Technology, UMBC, Baltimore, MD, USA  Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA, USA  Department of Mathematical Sciences, University of Wisconsin—Milwaukee, Milwaukee, Wi, USA  Department of Climate and Global Change Research & School of Atmospheric Sciences, Nanjing University, Nanjing, China  Department of Hydrology & Atmospheric Sciences, University of Arizona, Tucson, AZ, USA  Department of Information Systems, UMBC, Baltimore, MD, USA  Deleted: System Formatted: Font: +Body (Ca	Font: +Body (Calibri), Section: Hidden
Zhibo Zhang <sup>1,2*</sup> , Hua Song <sup>2</sup> , Po-Lun Ma <sup>3</sup> , Vincent E. Larson <sup>4</sup> , Minghuai Wang <sup>5</sup> , Xiquan Dong <sup>6</sup> ,  Jianwu Wang <sup>7</sup> 1. Physics Department, University of Maryland Baltimore County (UMBC), Baltimore, MD, USA  2. Joint Center for Earth Systems Technology, UMBC, Baltimore, MD, USA  3. Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA, USA  4. Department of Mathematical Sciences, University of Wisconsin—Milwaukee, Milwaukee, WI, USA  5. Institute for Climate and Global Change Research & School of Atmospheric Sciences, Nanjing University, Nanjing, China  19 6. Department of Hydrology & Atmospheric Sciences, University of Arizona, Tucson, AZ, USA  20 7. Department of Information Systems, UMBC, Baltimore, MD, USA  Deleted: System Formatted: Font: +Body (Ca	
Zhibo Zhang <sup>1,2*</sup> , Hua Song <sup>2</sup> , Po-Lun Ma <sup>3</sup> , Vincent E. Larson <sup>4</sup> , Minghuai Wang <sup>5</sup> , Xiquan Dong <sup>6</sup> ,  Jianwu Wang <sup>7</sup> 1. Physics Department, University of Maryland Baltimore County (UMBC), Baltimore, MD, USA  2. Joint Center for Earth Systems Technology, UMBC, Baltimore, MD, USA  3. Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA, USA  4. Department of Mathematical Sciences, University of Wisconsin—Milwaukee, Milwaukee, WI, USA  5. Institute for Climate and Global Change Research & School of Atmospheric Sciences, Nanjing University, Nanjing, China  6. Department of Hydrology & Atmospheric Sciences, University of Arizona, Tucson, AZ, USA  Deleted: System  Formatted: Font: +Body (Ca	
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### Abstract:

33 One of the challenges in representing warm rain processes in global climate models 34 (GCM) is related to the representation of the subgrid variability of cloud properties, such as cloud 35 water and cloud droplet number concertation (CDNC), and the effect thereof on individual 36 precipitation processes such as autoconversion. This effect is conventionally treated by 37 multiplying the resolved-scale warm ran process rates by an enhancement factor  $(E_a)$  which is 38 derived from integrating over an assumed subgrid cloud water distribution. The assumed subgrid 39 cloud distribution remain highly uncertain. In this study, we derive the subgrid variations of 40 liquid-phase cloud properties over the tropical ocean using the satellite remote sensing products 41 from Moderate Resolution Imaging Spectroradiometer (MODIS) and investigate the 42 corresponding enhancement factors for the GCM parameterization of autoconversion rate. We 43 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 44 45 important for the correctly simulating the autoconversion process in GCMs. Using the MODIS 46 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-47 48 <u>dependent. Our analysis</u> shows a significant increase of  $E_q$  from the stratocumulus (Sc) to 49 cumulus (Cu) regions. Furthermore, the enhancement factor  $E_N$  due to the subgrid variation of 50 CDNC is derived from satellite observation for the first time, and results reveal several regions 51 downwind of biomass burning aerosols (e.g., Gulf of Guinea, East Coast of South Africa), air 52 pollution (i.e., Eastern China Sea), and active volcanos (e.g., Kilauea Hawaii and Ambae Vanuatu), 53 where the  $E_N$  is comparable, or even larger than  $E_q$ , suggesting an important role of aerosol in 54 influencing the  $E_N$ . MODIS observations suggest that the subgrid variations of cloud liquid water 55 path (LWP) and CDNC are generally positively correlated. As a result, the combined enhancement 56 factor, including the effect of LWP and CDNC correlation, is significantly smaller than the simple 57 product of  $E_q \cdot E_N$ . Given the importance of warm rain processes in understanding the Earth 58 system dynamics and water cycle, we conclude that more observational studies are needed to 59 provide a better constraint on the warm rain processes in GCMs.

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### 1. Introduction

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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 GCMs is 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 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

$$\frac{\partial q_r}{\partial t} = C(q_c)^{\beta q} (N_c)^{\beta N_L} \tag{1}$$

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.

and  $N_c$  of cm<sup>-3</sup>. The three parameters C=1350,  $\beta_q=2.47$  and  $\beta_N=-1.79$  are derived

114 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-

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mean autoconversion efficiency requires the knowledge of subgrid distributions of LWC and 127 128 CDNC, but in the GCMs only grid-mean quantities  $(q_c)$  and  $(N_c)$  are known and available for use 129 in the computation of autoconversion rate. As pointed out by Pincus and Klein (2000), for a 130 process f(x) such as autoconversion that is nonlinearly dependent on subgrid variables, x, the 131 grid-mean value (f(x)) is not equal to the value estimated based on the grid-mean (x), i.e.,  $\langle f(x) \rangle \neq f(\langle x \rangle)$ . Mathematically, if f(x) is convex, then  $f(\langle x \rangle) < \langle f(x) \rangle$  (Larson and Griffin, 132 133 2013; Larson et al., 2001). To take this effect into account, a parameter E is often introduced in 134 the GCM as part of the parameterization such that  $f(x) = E \cdot f(x)$ . It is referred to as the "enhancement factor" in many studies and this study too because E > 1 for a convex function. 135 136 Such a nonlinear effect is not just limited to the autoconversion process. Some other examples 137 are the plane-parallel albedo bias (Barker, 1996; Cahalan et al., 1994; Oreopoulos and Davies, 138 1998a), subgrid cloud droplet activation (Morales and Nenes, 2010) and accretion (Boutle et al., 139 2014; Lebsock et al., 2013).

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

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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 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 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 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 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 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

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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  $q_c$  have been overlooked so far. Given the same amount of  $q_c$ , a cloud with a smaller  $N_c$  would have larger droplets and therefore larger precipitation efficiency than another cloud with a larger  $N_c$ . 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 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

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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 for the first time the subgrid CDNC variation and investigate its

enhancement factor for the autoconversion modeling in GCM, the picture is far from clear due to the small sampling rate of CloudSat observations.

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 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 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):

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

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

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

400 and variance given by

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

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

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

- where  $\eta = \frac{var(x)}{\langle x \rangle^2}$  is the relative variance. If x follows the Gamma distribution, for a physical
- 403 process M(x) that is a power function of x,

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

404 then the expected value (M(x)) is given by

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

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

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

- The PDF of a Lognormal distribution is given as follows (Larson and Griffin, 2013;
- 408 <u>Lebsock et al., 2013):</u>

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

- where  $\mu = (\ln x)$  and  $\sigma^2 = Var(\ln x)$ , correspond to the mean and variance of  $\ln x$ , respectively.
- 410 The mean value of the Lognormal distribution is given by

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

411 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)

- 412 It follows from Eq. (10) and (11) that the inverse relative variance can be derived from the
- 413 following equation

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

423 If x follows the Lognormal distribution, the expected value of (M(x)) is

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

Evidently, the corresponding enhancement factor is given by

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$$E(P_L, \nu, \beta) = \frac{\langle Kx^\beta \rangle}{K(x)^\beta} = \left(1 + \frac{1}{\nu}\right)^{\frac{\beta^2 - \beta}{2}}.$$

Note that Eq. (7) and (8) are only valid when  $\beta > -v$  because Gamma function  $\Gamma(v + \beta)$  can

run into singular values when  $v + \beta < 0$ . In contrast, Eq. (13) and (14) are valid for any real value

 $\beta$ . This is one advantage of the Lognormal distribution over the Gamma distribution.

An example of the Gamma and Lognormal distributions for  $q_c$  is shown in Figure 1a. In this example, both distributions have the same mean  $\langle q_c \rangle = 0.5g/kg$  and also the same inverse relative variance  $v_q = 3$ . Although the general shapes of the two PDFs are similar, they differ significantly at the two ends: the Gamma PDF is larger than Lognormal PDF over the small values of  $q_{cz}$  and the opposite is true over the large values of  $q_{cz}$ . The Gamma and Lognormal distributions can also be used to describe the spatial variation of  $v_c$  (Gultepe and Isaac, 2004). An example is given in Figure 1c, in which  $v_c$  is a constant of  $v_c$  (Gultepe and  $v_c$ ) = 50 cm<sup>-3</sup>, and  $v_c$  = 5.0.

Figure 1 b shows the autoconversion rate based on the KK2000 parameterization scheme for the Gamma  $P_G(q_c)$  and Lognormal  $P_L(q_c)$  that are shown in Figure 1a. Interestingly, although the cumulative autoconversion rates based on the two types of PDFs are almost identical, the contribution to the total autoconversion rate from the different LWC bins are quite different. As 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 large  $q_{C_L}$  (e.g.,  $q_c > 2.0g/kg$ ) is much higher in the Lognormal than in Gamma PDF. This difference is further amplified in the autoconversion rate computation in Figure 1b because the autoconversion rate is proportional to  $q_c^{2.47}$ .

The enhancement factors based on the Gamma (i.e.,  $E(P_G, \beta)$  in Eq. (8)) and Lognormal (i.e.,  $E(P_L, \beta)$  in Eq. (14)) PDF for  $\beta_q = 2.47$  are plotted as a function of the inverse relative variance v in Figure 2. When subgrid clouds are more homogenous i.e., v > 1, the enhancement

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Both Gamma and Lognormal distributions are mathematically convenient. For example, if any physical process M(x) is a power function of x,  $\dots$  [3]

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$$(M(x))_L = K \int_0^\infty x^\beta P_L(x) dx = \left(e^{\sigma^2}\right)^{\frac{\beta^2 - \beta}{2}} K(x)^\beta. \qquad \dots [4]$$

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factor based on the two PDFs are similar. However, for more inhomogeneous grids with i.e., v < 1, the  $E(P_L, \beta)$  is significantly larger than that  $E(P_G, \beta)$ , which is probably because of the longer tail of  $P_L(q_c)$  as shown in Figure 1 a and b.

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## 2.2. Impacts of subgrid cloud variations on warm rain <u>parameterization</u> in <u>GCM</u>

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).

Ideally, if the subgrid variations of  $q_{c}$  and  $N_{c}$  are known, then the grid-mean in-cloud autoconversion rate should be derived from the following integral

$$\sqrt{\frac{\partial q_r}{\partial t}} = \int_0^\infty \int_0^\infty C(q_c)^{\beta q} (N_c)^{\beta N} P(q_c, N_c) dq_c dN_{c_{\frac{1}{q_c}}}$$
(15)

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

$$\sqrt[4]{\frac{\partial q_r}{\partial t}} = E \cdot C(\langle q_c \rangle)^{\beta_q} (\langle N_c \rangle)^{\beta_{N_{\frac{1}{2}}}}$$
(16)

where *E* is the enhancement factor defined as:

$$E = \frac{\int_0^\infty \int_0^\infty (q_c)^{\beta_q} (N_c)^{\beta_N P} (q_c, N_c) dq_c dN_c}{((q_c))^{\beta_q} ((N_c))^{\beta_N}} \dot{z}$$
(17)

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

$$\langle \frac{\partial q_r}{\partial t} \rangle = C \int_0^\infty (q_c)^{\beta_q} P(q_c) dq_c \int_0^\infty (N_c)^{\beta_N} P(N_c) dN_{c}$$
(18)

Moved up [11]: This is one advantage of the Lognormal distribution over the Gamma distribution.

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**Deleted:** where  $\frac{\partial q_r}{\partial t}$  is the rain water tendency due to the auto-conversion process,  $q_c$  is the cloud water mixing ratio in the unit of kg/kg,  $N_c$  is the CDNC in the unit

Moved up [2]: of cm<sup>-3</sup>. The three parameters

**Moved up [3]:** The three parameters C=1350,  $\beta_q=2.47$  and  $\beta_N=-1.79$  are derived through a

**Moved up [18]:** Ideally, if the subgrid variations of  $q_c$ 

**Deleted:** least-square fitting of the rain rate results from a large-eddy simulation. The KK2000 scheme has been adopted in the popular two-moment cloud microphysics. [5]

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**Moved up [20]:** where  $P(q_c, N_c)$  is the joint PDF of  $q_c$ 

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

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

$$E_{q} = \frac{\int_{0}^{\infty} (q_{c})^{\beta q} P(q_{c}) dq_{c}}{((q_{c}))^{\beta q}}$$
(20)

and the  $\mathcal{E}_N$  is the enhancement factor due to the subgrid variation of <u>CDNC</u> which has the form,

$$E_{N} = \frac{\int_{0}^{\infty} (N_{c})^{\beta_{N}} P(N_{c}) dN_{c}}{((N_{c}))^{\beta_{N}}}.$$
(21)

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

If  $q_c$  and  $N_c$  both have significant subgrid variations and they are not independent, the enhancement factor should ideally be diagnosed from Eq. (17). However, the joint PDF  $P(q_c, N_c)$  may not be known and the integration can be time-consuming. Some previous studies proposed 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)

where  $\rho$  is the correlation coefficient between  $q_c$  and  $N_c$  (Larson and Griffin, 2013; Lebsock et

al., 2013). As such, both  $q_c$  and  $N_c$  follow a marginal lognormal distribution in Eq. (9). Substituting

Eq. (22) into Eq. (17), we obtain the enhancement factor for the bivariate lognormal distribution

that consists of three terms

$$E = E_q(P_L, \nu_q, \beta_q) \cdot E_N(P_L, \nu_N, \beta_N) \cdot E_{COV}(\rho, \beta_q, \beta_N \nu_q, \nu_N)_{\perp}$$
(23)

where 
$$E_q(P_L, v_q, \beta_q) = \left(1 + \frac{1}{v_q}\right)^{\frac{\beta_q^2 - \beta_q}{2}}$$
 and  $E_N(P_L, v_N, \beta_N) = \left(1 + \frac{1}{v_N}\right)^{\frac{\beta_N^2 - \beta_N}{2}}$  correspond to the

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

$$E_{COV}(\rho, \beta_q, \beta_N, \nu_q, \nu_N) = \exp(\rho \beta_q \beta_N \sigma_q \sigma_N)_{L}$$
 [24]

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

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

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

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Because most current GCMs do not have the capability to simulate the subgrid cloud property variations, models usually use pre-defined subgrid cloud variations in the computation of grid-mean auto-conversion rate instead of using prognostic values. For example, in the MG scheme for the CAMS.3, the subgrid LWC

**Deleted:** (1). Furthermore, it is assumed that the subgrid variation of CDNC is small and therefore the enhancement factor due to CDNC variation is negligible (i.e., close to unity). Substituting the Gamma distribution in Eq. (1) into the definition equation of enhancement factor in Eq. (18), and with help from Eq. (10), one can derive that  $\P$ 

**Moved up [12]:** As show in Figure 1a, the  $P_L(q_c)$  has a longer tail than the  $P_G(q_c)$ , i.e., the occurrence probability

**Moved up [13]:** (e.g.,  $q_c > 2.0 g/kg$  ) is much higher in the Lognormal than in Gamma

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**Moved up [14]:** —The enhancement factors based on the Gamma (i.e.,  $E(P_G, \beta)$  in Eq.

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**Moved up [16]:** ) PDF for  $\beta_q=2.47$  are plotted as a function of the inverse relative variance v in Figure 2. When

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

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

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

### 3. Data and Methodology

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To derive the above-mentioned enhancement factors , we will use 10 years (2007 ~ 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 bispectral solar reflectance method (Nakajima and King, 1990), All MODIS level-2 atmosphere products, including the cloud, aerosol and water vapor products, are aggregated to 1°×1° 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 level-3 product provides both their "in-cloud" grid-mean values ((x)) and subgrid standard <u>deviations</u>  $(\sigma_x)$ . 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 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., 2012; 2016), the 3.7 μm band CER retrieval is more resilient to the 3-D effects and retrieval failure than the 2.1 µm band retrievals. For these reasons, it is used as the observational reference in this study.

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

**Deleted:** Finally, it has to be noted that when both  $q_c$  and  $N_c$  have significant subgrid variations, their covariation also becomes important. As explained in Griffin and Larson (2013), if the  $q_c$  and  $N_c$  are negatively correlated, ¶

**Moved up [24]:** clouds with larger  $q_c$  would tend to have smaller  $N_c$ . The autoconversion rate in such a case

**Moved up [25]:** The autoconversion rate in such a case would be larger than that in the case where  $q_c$  and  $N_c$  are positively correlated (i.e.,

**Deleted:** larger  $q_c$  would tend to have larger  $N_c$ ). As explained in Eq. (17), only when they are uncorrelated can the total enhancement factor be decomposed into the product of two independent factors  $E = E_q \cdot E_N$ . Otherwise additional terms are necessary to take into account the effect of  $q_c$  and  $N_c$  correlation. Although potentially important, the correlation of  $q_c$  and  $N_c$  from satellite remote sensing data is difficult to derive from the satellite remote sensing observations due to the retrieval uncertainties. We will return to this point later in Section 5.3.

#### Moved (insertion) [28]

**Deleted:** Of particular interest to this study are the gridmean value and subgrid variation of several key properties of liquid-phase clouds, namely, COT, CER, LWP and CDNC, in the tropical regions. For this purpose, we use the latest collection 6 (C6) *daily mean* level-3 cloud retrieval product from the Aqua-MODIS instrument (product name "MYDO8\_D3"). The MODIS level-3 (i.e., grid-level) product contains statistics computed from a set of level-2 (

### Moved up [27]: i.e.,

**Deleted:** pixel-level) MODIS granules. 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, CER and LWP retrievals based on the bi-spectral solar reflectance method (Nakajima and King, 1990)

**Moved up [28]:** All MODIS level-2 atmosphere products, including the cloud, aerosol and water vapor products, are aggregated to  $1^{\circ}$ X1° 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 level-3 product provides both their "in-cloud" grid-mean values ((x)) and subgrid standard deviations ( $\sigma_x$ ).

**Deleted:** The inverse relative variance v can then be derived from Eq. ...

**Deleted:** , i.e.,  $v=\langle x\rangle^2/\sigma_x^2$ . Note that the operational MODIS product provides two CER retrievals, one based on 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 [13]

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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 it is a common factor in the calculation of v. The choice of the coefficient has no impact on our study, because we are interested in the relative inverse variance  $v = (x)^2/\sigma_x^2$ . We note here that it is the LWC  $q_{CL}$  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. <u>Airborne in situ measurement faces similar</u> challenge. For example, Boutle et al. (2014) use the LWC observation along "horizontal flight 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. <u>Following</u> previous studies (Bennartz, 2007; Bennartz and Rausch, 2017; Grosvenor and Wood, 2014; McCoy et al., 2017a), we estimate  $N_c$  of liquid-phase clouds from the MODIS retrieved COT ( $\tau$ ) and CER ( $r_e$ ) based on the classic adiabatic cloud model

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$$N_{c}(\tau, r_{e}) = \frac{\sqrt{5}}{2\pi k} \frac{\sqrt{f_{aa}\Gamma_{w}}}{\sqrt{\rho_{w}Q_{e}}} \tau^{\frac{1}{2}} r_{e}^{-\frac{5}{2}} = \frac{\sqrt{15}}{2\pi k} \frac{\sqrt{f_{aa}\Gamma_{w}}}{\rho_{w}\sqrt{2Q_{e}}} LWP^{\frac{1}{2}} r_{e}^{-3}, \tag{27}$$

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

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  $1^{\circ}\times1^{\circ}$  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 boundaries, ranging from 0 to 150, and 10 CER bin boundaries, ranging from 4  $\mu$ m to 30  $\mu$ m. With 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

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

$$Var(x) = \int \left[ (x(\tau, r_e) - \langle N_c \rangle)^2 P(\tau, r_e) d\tau dr_e, \right]$$
(28)

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

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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

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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 annual mean total cloud fraction  $(f_{tot})$ , liquid-phase cloud fraction  $(f_{lia})$ , in-cloud COT, CER from the 3.7 µm band, LWP and estimated CDNC over the tropical oceans based on 10 years Aqua-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 introduction, simulating these features in the GCMs proves to be an extremely challenging task, 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).

**Deleted:** The efficiency of using the level-3 product is accompanied by two important limitations. First, 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., CMIPS (Taylor et al., 2012), future GCMs may have significantly finer resolution. Second, it is difficult to sub-sample the pixels with the best retrieval quality. As reviewed in Grosvenor et al. (2018), the main source of uncertainty in the CDNC retrieval is the MODIS retrieval uncertainties, particularly in CER because of

 $N_c \sim r_e^{-\frac{5}{2}}$  dependence. In the pixel-by-pixel method, the pixel-level retrieval uncertainties, as well as some other metrics such as the sub-pixel inhomogeneity index, provided in the level-2 product can be used to select the pixels with the best retrieval quality. Here, because we use the static COT-CER joint histogram provided in the operational level-3 product, we do not have the flexibility to sub-sample the data using retrieval quality. Alternatively, we can subsample the data using the COT. It is well known that the bispectral retrieval method has a large uncertainty for thin clouds. Indeed, the clouds with COT thinner than about 4 have often been screened out in previous studies (Quaas et al., 2008). Such screening can be easily done with the joint PDF of COT and CER, but it would obviously lead to sampling bias in LWP. The impact on CDNC is dependent on whether the CDNC is correlated with the COT, i.e., whether thin clouds have the similar CDNC as the thick clouds. We will revisit this point later. It should be noted that because thin clouds in MODIS retrieval tend to have large uncertainty, any type of data quality-based data screening would inevitably lead to the sampling bias. ¶

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Switching the focus now from grid-mean values to subgrid variability, we will show the grid-level inverse relative variances  $v = \langle x \rangle^2 / Var(x)$ ) for several key cloud properties. Here, we first derive the daily mean values. Therefore, for each grid box we have 120 samples (i.e., 10 years x 12 months) of monthly mean v for analysis and visualization. Because the value of v can be ill-behaved when Var(x) approaches zero, instead of the mean value, we plot the median value of v based on 120 months of MODIS observations in Figure 5. There are several interesting and important features in Figure 5. First of all, the v of all four sets of cloud properties (i.e., COT, CER, LWP and CDNC) all exhibits a clear and similar Sc-to-Cu transition, with larger values in the Sc region and smaller value in the broken Cu regions. This indicates that cloud properties, including both optical and microphysical properties, are more homogenous, in terms of spatial distribution within the grid, in the Sc region than in the Cu region. Secondly, the value of v of CER (i.e.,  $10^{\sim}100$  in Figure 5b) is larger than that of the other properties (i.e., 1~10) by almost an order of magnitude, indicating that the subgrid variability of CER is very small. On the other hand, however, it is important to note that the v of CDNC (Figure 5d) is comparable with that of COT (Figure 5a) and LWP (Figure 5c). The reason is probably in part because the highly nonlinear relationship between CDNC and CER (i.e.,  $N_c \sim r_e^{-\frac{5}{2}}$  ) leads to a stronger variability of CDNC than CER, and also in part because the variability of CDNC is also contributed by the subgrid variation of COT. In some regions, the Gulf of Guinea, East and South China Sea, and Bay of Bengal for example, the v of CDNC is close to unity, indicating the subgrid standard deviation of CDNC is comparable to the grid-mean values in these regions. As discussed in the next section, the significant subgrid variability of CDNC in these regions should be taken into account when modeling the nonlinear processes, such as the autoconversion, in GCM to avoid systematic biases due to the nonlinearity effect.

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

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COT on a global scale from the level-3 Terra-MODIS retrievals. Although using a different metric (i.e., their inhomogeneity parameter is defined as  $\chi = \exp(\ln(\tau))/(\tau)$ ), they also found 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 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).

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As explained in section 2, the correlation between cloud water and CDNC can also influence the computation of enhancement factor and thereby the grid-mean autoconversion rate. Figure 5e shows the median value of the LWP and CDNC correlation coefficient p. Similar to the derivation of median v, we first compute the monthly mean  $\rho$  from daily MODIS observations and then derive the median value of  $\rho_{-}$  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 **5**b and c, the subgrid variability of  $r_e$  is order of magnitude smaller than that of LWP. Since CDNC 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 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.

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

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

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

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

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As explained in the Theoretical Background, in GCMs the influences of subgrid cloud water variability on the simulation of highly nonlinear autoconversion process are accounted for using the enhancement factors defined...

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

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

Figure 7a shows the variation of inverse relative variance v as a function of the grid-mean liquid-phase cloud fraction  $f_{liq}$ . In general, the value of v increases with the increasing  $f_{liq}$ , which is expected from the Sc-to-Cu increase of  $f_{liq}$  in Figure 4b and the Sc-to-Cu decrease of v in Figure 5c. The  $v(f_{liq})$  pattern in Figure 7a is also consistent with the results reported in Wood and Hartmann (2006) and Lebsock et al. (2013). In the hope of obtaining a simple parameterization scheme for  $v(f_{liq})$  that can be used in GCMs, we fit the median value of v as a simple v0 order polynomial of v1 as follows:

$$v(f_{liq}) = 2.38 - 4.95 f_{liq} + 8.74 f_{liq}^2 - 0.49 f_{liq}^3, f_{liq} \in [0,1].$$
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To test the performance of this simple parameterization, we first substitute the  $f_{liq}$  from MODIS daily mean level-3 product into the above equation and then use the resultant v to compute the enhancement factor  $E_q$ . Unfortunately, the enhancement factor  $E_q$  computed based on the parameterized  $v(f_{liq})$  as shown in Figure 8a substantially underestimate the observation-based results in Figure 6, especially over the Cu regions. The deviation is probably because the relationship between  $E_q$  and v is highly nonlinear (e.g., Eq. (8) and (14)) and therefore the above parameterization scheme that only fits the value of v is not able to capture the variability of  $E_q$ . Based on this consideration, we tried an alternative approach. Instead of parameterization of v, we directly parameterize the enhancement factor  $E_q$  as a function of  $f_{liq}$ . Figure 7b shows the variation of  $E_q$  as a function of  $E_q$  as a function of  $E_q$  as a function of  $E_q$  is fitted with the following  $e^{ir}$ 0 order polynomial of  $e^{ir}$ 1 is fitted with the following  $e^{ir}$ 2 order polynomial of  $e^{ir}$ 3 is fitted with the following  $e^{ir}$ 3 order polynomial of  $e^{ir}$ 4.

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

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

## 5.2. Influence of subgrid variance of CDNC

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

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

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

**Commented [V18]:** Importantly (and confusingly), E\_N is small in the main Sc regions off the coasts of California, Peru, and Namibia. Why aren't there variations in Nc in those regions off the coasts? Autoconversion will be not be enhanced in those regions by variations in Nc (or qc).

To me, it looks like E\_N is controlled by variations in the (remote) source of aerosol, whereas E\_Q is controlled by variability in cumulus clouds that is locally induced by turbulence. But neither is large in Sc.

Is it possible that the high values of E\_N are an artifact of time variability in the aerosol as a plume of pollution from, e.g., a fire, meanders across the ocean? Is there such large variability in instantaneous snapshots? Can the MODIS observations work on instantaneous data instead of time averages?

Commented [zz19R18]: These are very good questions. I don't have clear answers at the moment. Some small scale CDNC variation is due to retrieval artifacts. But as shown later, even we screen out the COT<5 data, the results are still similar.

I'm in favor of your hypothesis about the spatial variation of E\_N. In this paper, I just want to hold on the "observation" and leave the in-depth study of the causes to future work.

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**Commented [V21]:** It might be reassuring to compare with aircraft observations. Do any aircraft observations show CER varying from 4 to 30 microns?

**Commented** [zz22R21]: Yes, in situ measurement would be highly useful. But it will be left for future work.

**Deleted:** . On one hand, the pattern of  $E_N$  in Figure 9a seems to suggest that there are some underlying physical mechanisms controlling the subgrid variation of CDNC, in which aerosols seem to play an important role. On the other hand, the magnitude of  $\boldsymbol{E}_{N}$  is surprisingly large. As explained in section 3, the CDNC is estimated based on Eq. (23) from the MODIS retrieval of COT and CER. Could retrieval uncertainty contribute to the large subgrid variation of CDNC and therefore  $E_N$ ? In order to better understand the large value of  $E_N$ , we selected a case during the biomass burning season in the Gulf of Guinea, which is shown in Figure 10. During the boreal winter, the grassland and savanna fires in the southern West Africa generate a thick layer of smoke aerosols that are clearly visible in the satellite image (Andreae and Merlet, 2001). On this day, the Gulf of Guinea is quite cloudy, filled with broken cumulus clouds in the northern coastal region and stratiform clouds in the south. We arbitrarily selected a smaller region, marked with the red box, for detailed analysis. Although the cloud fraction in this region is about 60%, the clouds are [19] 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. The subgrid variation of updraft leads to subgrid variation CDNC and thereby large  $E_N$ .

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

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

Finally, in this section we examine the combined effect of subgrid variations of cloud water and CDNC, as well as their correlation, on the autoconversion rate simulation. The annual mean combined enhancement factor E derived based on Eq. (17) from 10 years of MODIS COT and CER observation is shown in Figure 11a. Comparing to the  $E_q$  in Figure 6 and  $E_N$  in Figure 9, the combined enhancement factor is generally larger, It is easy to see that the in some regions (e.g., Gulf of Guinea, East Coast of South Africa and Eastern China Sea) the combined enhancement factor E resembles the  $E_N$  while in other regions (i.e., trade wind cumulus regions over open ocean) it resembles more of  $E_q$ . Interestingly, because both  $E_q$  and  $E_N$  are small over the Sc decks, those regions have the smallest combined enhancement factor E. As discussed in Section 2.2, only when the subgrid variation of cloud water is uncorrelated with the subgrid variation of CDNC can the combined enhancement factor E be decomposed into the simple product of  $E_q$  and  $E_N$  (i.e., Eq. (19)). Figure 11b shows the annual mean value of the simple product  $E_q \cdot E_N$ , without considering the correlation between cloud water and CDNC. Evidently, the simple product substantially overestimates the combined enhancement factor derived from

**Deleted:** The results from the above case study raises some concerns. It seems that the large variations of CER and therefore CDNC are usually associated with thin clouds. While there could be a physical explanation (e.g., CCN activation), it seems more likely to be caused, or at least contributed, by retrieval uncertainty. It is well known that the bispectral method has large uncertainties for thin clouds, especially when they are broken. Several previous studies have shown that the sub-pixel level surface contamination, subpixel inhomogeneity, and threedimensional radiative transfer effects, tend to cause overestimated CER retrieval on top of large uncertainties (Zhang and Platnick, 2011; Zhang et al., 2012; 2016). Therefore, for such a challenging case in Figure 10, it is not surprising that the large CDNC variation and  $\boldsymbol{E}_{N}$  are partly caused by retrieval uncertainty. Based on this consideration, we did a sensitivity test, in which we screen out the thin clouds with COT < 4 in the computation and analysis of CDNC and  $E_N$ . The result from this test is shown in Figure 9c. Indeed, the removal of thin clouds substantially reduces the value of  $\,E_{N}.\,$  For example, in the Gulf of Guinea, the median value of  $E_N$  reduces by a factor of 4 from about 10 to only about 2.5. Nevertheless, the global pattern of  $E_N$ still remains, i.e., nonnegligible values of  $\boldsymbol{E}_{N}$  are found in the downwind regions of biomass burning, air pollution and volcano emission. ¶

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**Deleted:** , which in our opinion should focus on the regions with large  $E_N$ , e.g., Gulf of Guinea, East Coast of South Africa and Eastern China Sea. Last, but not least, the example in Figure 10 clearly exposes the limitation of the current satellite remote sensing method. There are alternative methods for retrieving the CDNC from satellite 0

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**Deleted:** can be derived from joint PDF  $P(q, N_c)$  based on Eq. (15). Because both q and  $N_c$  are a function of the retrieved COT and CER, we can easily derive the

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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.

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 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 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 <u>parameterization</u> 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:

 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. Moved down [30]: (i.e.,

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**Deleted:** optically thin clouds with less cloud water tend to have larger CER and smaller CDNC). This correlation mainly exists among optically thin clouds as a result of retrieval bias and uncertainty and it tends to counteract the effect of  $E_q$  and  $E_N$  making the combined enhancement factor  $E_q$  substantially smaller than the simple product of  $E_q \cdot E_N$  (i.e., assuming no correlation).

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2. The wide spatial coverage of the Level-3 MODIS product enables us to depict a detailed quantitative picture of the enhancement factor  $E_{q_\perp}$  which shows a clear cloud regime dependence, i.e., a Sc-to-Cu increase. The constant  $E_q=3.2$  used in the current CAM5.3 model overestimates and estimates the observed  $E_q$  in the Sc and Cu regions, respectively.

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- 3. The  $E_q$  based on the Lognormal PDF assumption performs <u>significantly</u> better than that based on the Gamma PDF assumption. A simple parameterization scheme is provided to relate  $E_q$  to the grid-mean liquid cloud fraction, which can be readily used in GCMs.
- 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 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 than the simple product of  $E_q \cdot E_{N_\bullet}$  (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

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### Acknowledgement:

Z. Zhang acknowledges the financial support from the Regional and Global Climate Modeling Program (Grant DE-SC0014641) funded by the Office of Biological and Environmental Research in the US DOE Office of Science.

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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# **Acknowledgement:**

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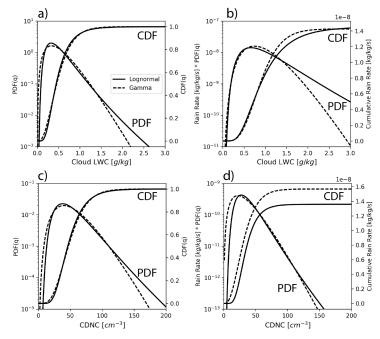


Figure 1 a) The probability density function (PDF) and cumulative distribution function (CDF) of cloud LWC  $(q_c)$  that follow the Gamma (dashed) and Lognormal (solid) distribution. For the both distributions,  $\langle q_c \rangle = 0.5g/kg$  and  $v_q = 3.0$ . b) The PDF and CDF of autoconversion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

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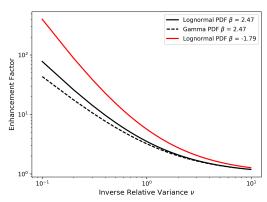


Figure 2 Enhancement factors based on Lognormal  $E(P_L,\beta)$  and Gamma  $E(P_G,\beta)$  subgrid PDF for different  $\beta$  as a function of the inverse relative variance v.



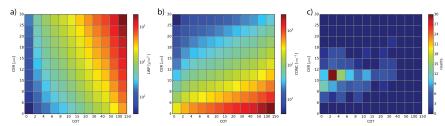


Figure 3 The (a) LWP and (b) CDNC as a function of COT and CER. (c) An exmaple of the COT-CER joint histogram observed by Aqua-MODIS on Jan.  $09^{th}$ , 2007 at 1°S and 1°W.

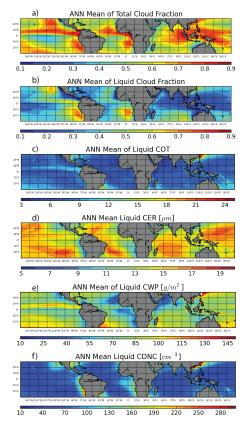


Figure 4 10-year (2007~2016) averaged annual mean a) total cloud fraction, b) liquid cloud fraction, c) cloud optical thickness, d) cloud effective radius retrieved from the 3.7 µm band, e) cloud wather path and f) cloud droplet concentration retrievals from Aqua-MODIS over the tropical (30° S-30° N) oceans. All quantaties are "in-cloud" mean that are averaged over the cloudy-part of the grid only.

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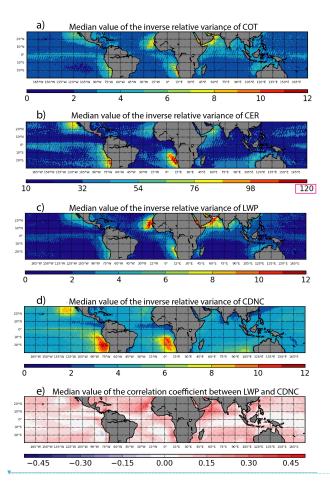
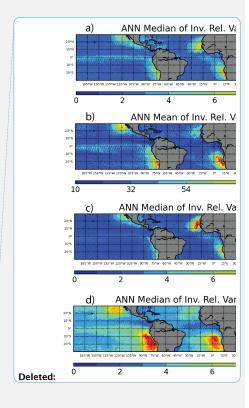


Figure 5 Median value of the inverse relative variance (i.e.,  $v = (x)^2/Var(x))$  for a) COT, b) CER, c) LWP and d) CDNC, and e) median value of the correlation coefficient between LWP and CDNC derived from 10 years of MODIS observations. Note that the color scale of CER is different from others'.



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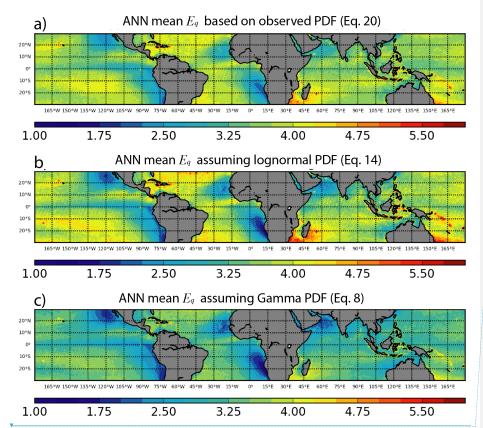
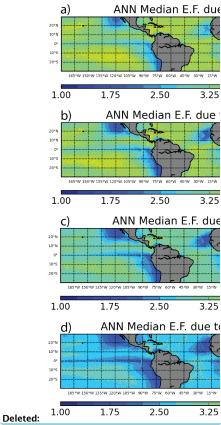


Figure 6 The annual mean factor for the KK2000 scheme due to subgrid variation of LWP computed a) directly from observation, i.e.,  $E_q$  in Eq.(20), b) from relative variance assuming Lognormal PDF of LWP, i.e.,  $E_q$  in Eq.(14) and c) from relative variance assuming the Gamma PDF of LWP, i.e.,  $E_q$  in Eq.(8).



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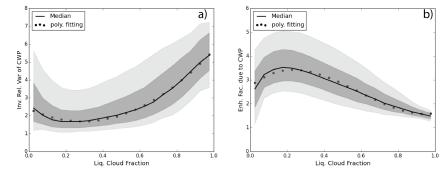


Figure 7 a) The inverse relative variance v and b) autoconversion enhancement factor due to LWP subgrid variability assuming Log-normal PDF as a function of grid-mean liquid cloud fraction, where the solid line, dark shaded area, and light shaded area correspond to the median value, 25%~75% percentiles, and 10~90% percentiles, respectively. The dotted lines correspond to simple 3-rd order polynomial fitting.

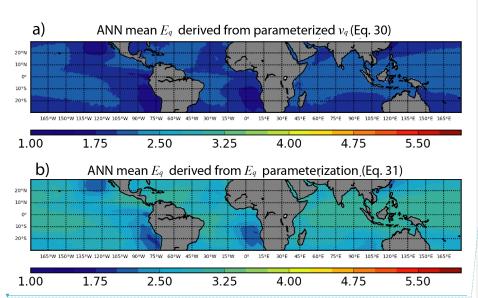
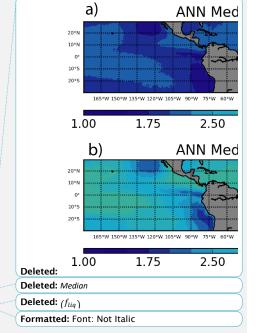


Figure 8 <u>Annual mean</u> value of the enhancement factor  $E_N$  computed based on the a)  $v(f_{liq}) = 2.38 - 4.95 f_{liq} + 8.74 f_{liq}^2 - 0.49 f_{liq}^3$  parameterization scheme in Eq. (30) and b)  $E_{q_q}(f_{liq}) = 2.72 + 7.33 f_{liq} - 19.17 f_{liq}^2 + 10.69 f_{liq}^3$  parameterization scheme in Eq. (31).





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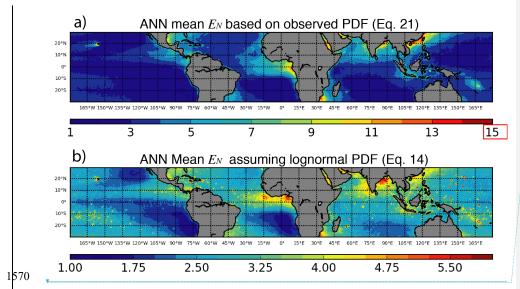
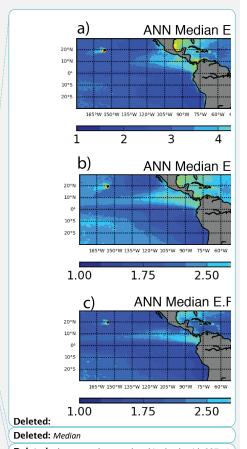


Figure 9 <u>Annual mean</u> value of the enhancement factor  $E_N$  derived from a) observation based on Eq. (21) and b) from Eq. (14) assuming Lognormal subgrid CDNC distribution.



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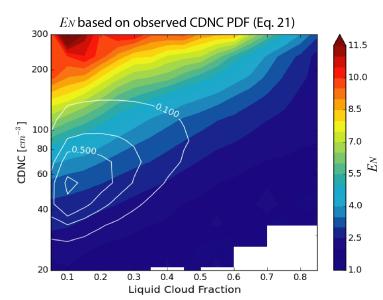
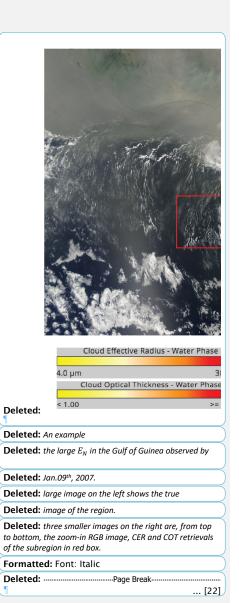


Figure 10 <u>Dependence</u> of  $E_N$  on  $f_{liq}$  and  $N_d$ . The color <u>map corresponds to the mean value of  $E_N$  for a given  $N_d$  and  $f_{liq}$  bin. The white contour lines correspond to the relative sampling frequency of  $N_d$  and  $f_{liq}$  bins (i.e., the most frequently observed combination is  $N_d \sim 50 cm^{-3}$  and  $f_{liq} \sim 0.1$ ).</u>



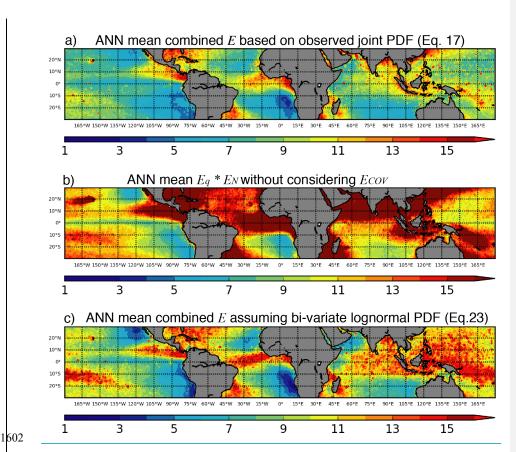


Figure 11 <u>The</u> combined enhancement factor <u>derived a)</u> based on Eq. (17) <u>from the observed joint PDF of LWP and CDNC</u>, b) <u>assuming</u> that subgrid variations of LWP and CDNC are uncorrelated, i.e.,  $E_q \cdot E_{N_q}$  only and c) based on Eq. (23) <u>assuming that the subgrid LWP and CDNC</u> following the bi-variate lognormal distribution,

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