**Response to Anonymous Referee #1**

*This study discusses the sensitivity of cloud production of secondary organic aerosol (SOA) to several parameters including LWC, T, VOC/NOx, and oxidant concentrations. The analysis is conducted using a climate model to examine these sensitivities on a global scale. The key finding is that in-cloud SOA production is most sensitive to LWC, followed by total carbon chemical loss rate. The topic is of relevance to this journal and is an important one to understand since SOA formation mechanisms are not fully understood, especially in the aqueous phase. The presentation quality of the paper was at an average level.*

*My issue with this study is that the analysis is superficial and does not significantly advance the community's knowledge of factors influencing SOA production in clouds. The key finding is based on correlation coefficients (does not prove cause-and-effect) and is arguably a repeat of previous work (e.g. Ervens et al., 2008), including the importance of LWC. In contrast to this older 2008 study (just to name one), the current one under review is conducted at coarser resolution, which will not provide the level of detail needed at the scale of clouds to capture aqueous phase processes in cloud drops. Thus I was left struggling to find the critical mass of new findings needed that would warrant publication. I cannot support publication of this work. More comments can be found below.*

We thank the reviewer for thoughtful comments that improve this manuscript. Here we would like to clarify the primary differences between the previous work (i.e. Ervens et al., 2008, hereafter ER08), which informs and influences the work here. These manuscripts differ fundamentally in model scale and application, factor evaluation, and key conclusion. First, the former study (i.e., ER08) is a process level modeling investigation based on a cloud parcel model (including detailed gas-phase and liquid-phase chemistry) driven by a prescribed set of trajectories generated in 3D large eddy simulations. This study is based on a fully coupled chemistry-climate model which solves large-scale clouds at a level to reasonably represent historical global climate. The bulk prognostic microphysics cloud scheme for large-scale stratocumulus cloud as well as the shallow cumulus and deep convective cloud schemes implemented in AM3 have been evaluated against satellite observations for actual cloud fraction, cloud-drop radius, liquid water path and precipitation (see Donner et al., 2011, J Climate). This information is sufficient to drive a detailed multiphase chemistry scheme and resolve secondary aerosol formation from cloud processes, where cloud droplet size, cloud liquid water content, and cloud droplet lifetime are the key factors influencing gas-liquid transfer, aqueous-phase chemistry and secondary aerosol production. In a cloud parcel model, for droplet lifetime, the cloud parcel model tracks the time of passage of an air parcel through cloud (i.e., the cloud contact time in ER08), while GCM solves the production and sink of cloud liquid water in one grid cell which can be used to quantify cloud cycles in each model time step [Liu et al., 2012]. Therefore, in terms of SOA formation from cloud processes, both GCM and cloud parcel model provide suitable details to drive the cloud chemistry. The earlier results from cloud parcel model have suggested that "$SOA_{drop}$ can be modeled in bulk cloud water, similar to sulfate, avoiding the solution of differential equations for individual drop classes" [Ervens et al., 2008]. This suggests implementation of aqueous phase organic chemistry and subsequent SOA formation in a larger scale model, such as the GCM employed here, is an appropriate progression in the development of chemical schemes for atmospheric models. There are also a number of issues related to differences in mechanisms of gas-phase and aqueous phase chemistry (e.g., nighttime chemistry, contribution of acetic acid to $SOA_{cld}$, formation of LMC, SOA precursors (i.e., isoprene, terpene and aromatic species), photolysis rate, etc.) between ER08 and this work. Further, this study employs new lab results, not included in earlier work, such as new insights regarding photooxidation products of glycoaldedhye.

Approaches from process level modeling (ER08) to GCMs (work described here) are valid and needed to add to the community's understanding of multiphase chemistry. The method we used to determine model sensitivities to physical cloud parameters is inherently different from the process-level modeling of ER08. In ER08, factors determining production of $SOA_{cld}$ are constrained by setting up 20 sensitivity tests with different initial conditions in a closed parcel system. Then it counts the SOA accumulation from a 1-hour trajectory (totally 45 trajectories are derived from the large eddy simulations) which is repeated five times starting at 10am (solar time). Then the production of $SOA_{cld}$ from these trajectories is evaluated against the change from initial conditions. Therefore, the results derived from ER08 may depend on the experimental design and specific model configuration. However, this study is not limited to a set of prescribed cloud physics, rather there is large spatiotemporal variability in global scale clouds, enabling targeted understanding the key factors influencing the spatiotemporal variability of $SOA_{cld}$ production in a global scale.

Studied factors varied substantially in space and time when studied with a GCM, and may result in significant difference in SOA production rates, a research interest of this work. We archived $SOA_{cld}$ production every 3-hours during the 1-year simulation period for each grid box and derive the relationship between individual factors and $SOA_{cld}$ production based on spatiotemporal statistical inference. Within each 3 hour period, the production of $SOA_{cld}$ is based on the detailed physiochemical processes within the model. Therefore, the production of $SOA_{cld}$ is determined by these factors leading to $SOA_{cld}$ changes. Rather than using 20 sensitivity tests, this study includes millions of 3-hour tests covering all seasons and locations all over the world and evaluates the most influential factors. Our results implicitly reflect true globally averaged situations, and are not limited by prescribed configurations.

Finally, we believe our conclusions are inarguably distinct to ER08. In ER08, key findings include in-cloud SOA yield is mostly affected by VOC to $NO_x$ ratio and cloud contact time, but is less affected by LWC. However, based on global spatiotemporal statistical inference, this study found LWC is a dominant factor determining the global spatial and temporal distribution of $P_{SOAcld}$. VOC/$NO_x$ could be important in specific locations and season, but is insignificant, relative to LWC and VOC loss in the broader scale studied here. In addition, this study found a robust and stable non-linear relationship that suggests globally, the increase in $P_{SOAcld}$ is slower than the increase in VOC loss rate. These new findings could be important in global climate models to understand the feedbacks between VOCs, oxidants, SOA, and multiphase atmospheric chemistry.

We concur that ER08 is pioneering work which informs our experimental design. To address the reviewer's comments, we made substantial changes in the revised manuscript to clarify the unique features of this study to ER08. Please see our reply to each comment below.

Comments:

*Introduction: A weakness of this manuscript is that it completely ignores the body of literature describing field measurements related to SOA formation in clouds and more generally in the aqueous phase. Sufficient treatment is given to model and lab papers, but a better balance is needed with field results.*

Good comment. We added a paragraph in the introduction to address the importance of field measurements related to SOA formation in the aqueous phase. Now we add the following in section 1:

*"A number of field measurements have confirmed SOA formation in the aqueous phase. Sorooshian et al. (2006) found a strong correlation between measured oxalate and* $SO_4^{2-}$ *aloft. Sulfate and oxalate are well-known to form during aqueous phase photooxidation from common water-soluble atmospheric gases. The strong correlations despite large differences in precursors and their emission sources, provide evidence for a common production process (i.e., in-cloud production). Sorooshian et al. (2007) observed ubiquitous organic aerosol layers above clouds with enhanced organic acid levels and their data suggested that aqueous-phase reactions to produce organic acids are a source of this elevated organic aerosol level. Moreover, based on several airborne field missions, organic acids and a larger body of oxygenated species with a mass spectral peak in m/z 44 were found to contribute to the total organic aerosol fraction, which increased as a function of RH and aerosol hygroscopicity, further indicating the role of aqueous-phase chemistry played on the formation organic aerosols (Sorooshian et al., 2010)."*

*Pg 26932, line 26 and forward: it is not clear to me what is new about this paper as compared to the studies outlined in the previous category. Is there reason to believe a more accurate assessment of factors affecting in-cloud SOA production can be obtained with a global model rather than the parcel model in the Ervens et al. study? It seems that at coarser resolution, it would be much harder to get a solid understanding of factors affecting SOA production in clouds, which occur at a scale more relevant to what a cloud parcel model can provide.*

The key question here is not model resolution. Instead, it should be what kinds of cloud information that cloud chemistry requires to accurately simulate in-cloud production of secondary aerosols. Based on the development of aqueous-phase chemistry scheme in AM3, the key cloud information needed includes cloud liquid water content, cloud droplet size, cloud fraction, and cloud droplet lifetime. As mentioned above, this information can be reasonably resolved in AM3 by comparing to observations. In addition, both OA and sulfate (well-known in-cloud production pathway) simulations are significantly improved after the detailed cloud chemistry scheme (developed with an understanding of ER08 in mind) is being used in AM3. Even results from cloud parcel model have also shown: "$SOA_{drop}$ can be modeled in bulk cloud water, similar to sulfate, avoiding the solution of differential equations for individual drop classes". Therefore, factors influencing cloud parcel model should have a similar effect in GCM at some specific location and time. However, rather than focusing on prescribed set of small scale, process-level cloud features (e.g., ER08), this study targets on understanding the spatiotemporal features of $P_{SOAcld}$ over the full range of cloud and chemical conditions experienced globally. For example, Liu et al., 2012 has found that huge amount of $SOA_{cld}$ is produced over the western Amazon and southern Africa in DJF, and southern China and the boreal forests in Eurasia and North America in JJA. Based on spatiotemporal factor analysis, this study confirms that $P_{SOAcld}$ is largely controlled horizontally and vertically by the availability of cloud water and VOC oxidation, and their combined effects generally determine $P_{SOAcld}$ global distribution pattern. However, at specific locations or seasons we have also found unique features that require information beyond LWC and $TC_{loss}$, giving raise to the importance in understanding the spatially heterogeneous feature of $P_{SOAcld}$. This will be addressed in our follow-up studies based on process-based models. Therefore, our results might reflect the results of repeating ER08 experiments for a larger variety of clouds, for example, at all seasons and locations over the world.

To articulate the linkages between ER08, Liu et al 2012, and this study, now we include the following text in the introduction:

"*Ervens et al. (2008) used a cloud parcel model with explicit kinetic aqueous-phase chemistry to study the effects of initial VOC/$NO_x$, cloud PH, cloud contact time, and liquid water content (LWC) on $SOA_{cld}$ formation from isoprene. They found that $SOA_{cld}$ yield was a function of LWC and cloud contact time under different VOC/$NO_x$ conditions, and $SOA_{cld}$ could be modeled in bulk cloud water without solving* *individual drop classes. This suggests implementation of aqueous phase organic chemistry and subsequent SOA formation in a larger scale model is an appropriate progression in the development of chemical schemes for atmospheric models. Liu et al. (2012) implemented a detailed cloud chemistry scheme into a climate model AM3 and evaluated the effects of cloud properties on the global* $SOA_{cld}$ *production and distribution. They found that formation of* $SOA_{cld}$ *varied significantly by time and space (i.e., dense production occurs over southern Africa and western Amazon tropical forests in DJF, but over southern China and the boreal forests in JJA) and is tied to the distribution of cloud water up to 400hPa above the surface.*

*This study is framed on Liu et al. (2012), but aims firstly to quantitatively understand the potential factors contributing significantly to the global spatial and temporal diversity of in-cloud formation of SOA, and secondly to establish a relationship between* $SOA_{cld}$ *production and the key factors determining this process spatiotemporally. The approach is based on the global chemistry-climate model AM3 (Donner et al., 2011) with detailed multiphase chemistry developed in Liu et al. (2012, more description is given in Section 2.1).*"

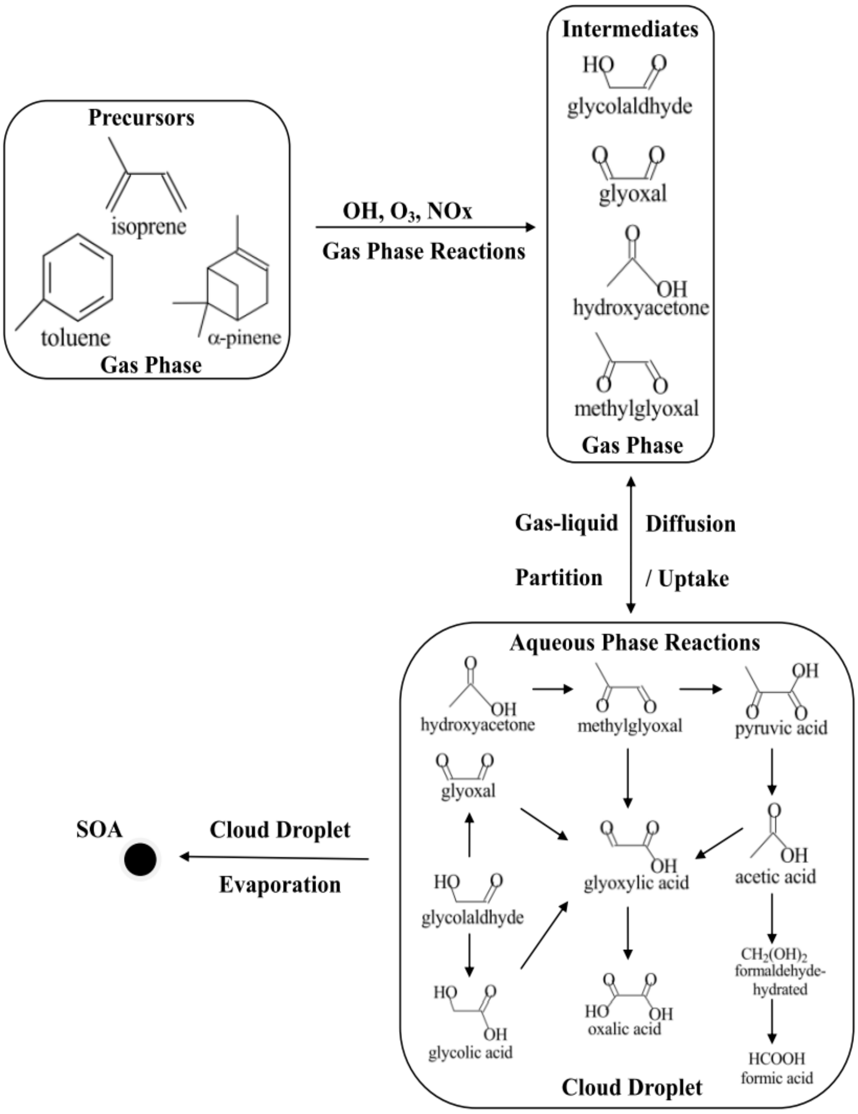
*Pg 26932, last line: describe the "framework developed in Liu et al." Do not expect the reader will know what this means when reading this line.*

The detailed aqueous-phase chemistry in the framework developed by Liu et al. (2012) is described in Section. 2.1. To make it clear, now we have:

*"This study aims firstly to quantitatively understand the potential factors contributing significantly to the global spatial and temporal variability of in-cloud formation of SOA, and secondly to establish a relationship between* $SOA_{cld}$ *production and the key factors determining this process spatiotemporally. The approach is based on the global chemistry-climate model AM3 (Donner et al., 2011) with detailed multiphase chemistry developed in Liu et al. (2012, more description is given in Section 2.1)"*

*Figure 1. Hard to see the species and text. Make it larger and provide for better visual quality.*

Thanks for the suggestion. Now we made Figure 1 larger, see below.



*Section 3: It would be helpful at the beginning of Section 3 to provide background as to why these factors are chosen and how they are expected to affect in-cloud SOA production. For example, what is the significance of the VOC/NOx ratio. I get the impression that the authors assume that the readers know exactly how these factors may affect cloud processing, when in fact, this is not the case always.*

Good comment. Some factors (i.e., LWC, VOC/NO$_x$ ratio, VOC loss rate, oxidants …) are chosen because they influence the chemical pathways to generate SOA$_{cld}$ (we admit some factors have already been evaluated in previous studies (e.g., ER08, Carlton et al., 2010)), others (e.g., temperature) are used because they influence chemical kinetics (Liu et al., 2012). We expand our previous discussion (i.e., Section 2.3) on factor selection in the revised manuscript. Now we have:

*"Significance of factors driving the variability of the rate of SOA$_{cld}$ production (P$_{SOAcld}$, expressed as the total SOA mass formed through cloud processes in one unit*

*time, unit: kg·m$^{-3}$·s$^{-1}$) is examined by spatiotemporal statistical inference. Based on results from previous studies (e.g., Ervens et al., 2008 and Carlton et al., 2009) as well as the physical and chemical mechanisms of in-cloud formation of SOA (see Section 2.1 or Liu et al., 2012), factors mainly influence SOA$_{cld}$ production include LWC, temperature, the chemical loss rate of total carbon mass of primary precursors (i.e., isoprene, α-pinene, and toluene), the concentration of OH and O$_3$, and the VOC to NO$_x$ ratio (VOC/NO$_x$).*

*Cloud water (i.e., LWC, unit: kg(water)/kg(air) ) serves as the media where SOA$_{cld}$ is formed. More cloud water is usually associated with more SOA$_{cld}$ production (Liu et al., 2012). Temperature governs the overall reaction rates and is shown in laboratory studies to be one of the key factors in SOA formation (Takekawa et al., 2003). VOC oxidation rate is an essential factor during the transformation among different organics. It indicates the availability of water-soluble gases (e.g., glyoxal and methylglyoxal) that are able to form SOA$_{cld}$. Here we use total carbon chemical loss rate (TC$_{loss}$) of isoprene, α-pinene, and toluene to represent the precursors' oxidation rate:*

$$TC_{loss}(kgC \cdot m^{-3} \cdot s^{-1}) = 0.012 \times (5 \times L_{isop} + 7 \times L_{tol} + 10 \times L_{pine}) \qquad (1)$$

*where L$_{isop}$, L$_{pine}$, and L$_{tol}$ are the loss rates (unit: mol·m$^{-3}$·s$^{-1}$) of isoprene, α-pinene, and toluene, respectively.*

*The concentrations of OH and O$_3$ (unit: mol·m$^{-3}$) reflect the oxidation power of the atmosphere and can affect the reaction rate and pathway of SOA formation in both gaseous and liquid phases (Lim and Ziemann, 2005). VOC to NO$_x$ ratio could also be another factor to influence oxidants and SOA$_{cld}$ yields, which is shown to be the most effective factor controlling SOA$_{cld}$ yields in the cloud parcel model (Ervens et al., 2008). We ignore the factor of cloud fraction since it has been partially included in LWC when grid-box averaged LWC is used. Other factors related to cloud properties are not considered in this study and will be evaluated in the follow-up studies. "*

*Section 3: Correlations are not evidence of a cause-and-effect relationship and thus much of the conclusions being reached need to be toned down. For example, line 22-24 of pg 26937 is one such place. Authors must seriously consider what their results can unambiguously prove as the analysis is not too detailed (only correlations) and the relationships at play may be highly non-linear.*

It is true that correlations are not causality. There are two steps in our analysis and each represents a cause-and-effect relationship from different angle. In the first step, we generate a cause-and-effect relationship between factors and P$_{SOAcld}$ for each grid box in every 3-hour period for a whole year. In such a process, P$_{SOAcld}$ is determined by the availability of cloud water, precursors' oxidation, temperature and overall oxidation level, which are physically built in AM3 (see Figure 1 in the manuscript) and are represented by all the linear/nonlinear processes related to the clouds and multi-phase chemistry resolved in AM3. In this step, our method is close to ER08 except that model is configured to a few specific conditions in ER08, whereas we archived all the possibilities globally during all seasons. In the second step, we use spatiotemporal statistical inference to understand, in a global scale, to what extent the spatiotemporal variability of these factors individually/combined determines the spatiotemporal distribution of $P_{SOAcld}$. Here, the cause is the spatiotemporal variability of each factor which is determined by many other linear/nonlinear processes and factors (e.g., human activity, ecosystem growth, incoming solar radiation, atmospheric transport and climate) except $P_{SOAcld}$. We admit that the relationship we derived (i.e., the linear relationship between $P_{SOAcld}$ and LWC, and the nonlinear relationship between $P_{SOAcld}$ and $TC_{loss}$) is not perfect ($R^2 \sim 0.5$) since only two factors are employed globally. In our parameterization, the variance of $P_{SOAcld}$ has been addressed internally by the nonlinear solving of LWC and $TC_{loss}$ in AM3, and externally by the non-linear dependence on oxidation of hydrocarbons (i.e., gamma). This differs substantially to ER08, in which $P_{SOAcld}$ depends linearly on hydrocarbons. Assuming gamma equal to 1 will reduce the explained variance ($R^2$) of $P_{SOAcld}$ to 0.35. The unexplained variance by our parameterization should be mainly imbedded in the non-linearly solving of gas-phase and aqueous-phase chemistry, which we believe could not be easily addressed by adding a few more predictors.

In sum, the overarching goal of this work is to explore the key factors determining the global spatiotemporal variability of $P_{SOAcld}$. In the follow-up studies, we will focus more on specific region and season, and testify the predictability of other factors.

To make this clear and to tone down the conclusions, we explicitly expressed our goals and method in the revised manuscript. Now we have:

Last paragraph of Section 1: *"This study aims firstly to quantitatively understand the potential factors contributing significantly to the global spatial and temporal variability of in-cloud formation of SOA, and secondly to establish a relationship between $SOA_{cld}$ production and the key factors determining this process spatiotemporally. The approach is based on the global chemistry-climate model AM3 (Donner et al., 2011) with detailed multiphase chemistry developed in Liu et al. (2012, more description is given in Section 2.1)"*

Section 2.2: *"In this study, the emission inventories used for both gas and aerosol species are obtained from the database developed for IPCC AR5 studies (Lamarque et al., 2010). Sea ice cover and sea surface temperature (SST) are prescribed using databases developed at the Hadley Center (Rayner et al., 2003). Cloud droplet lifetime is set to be 30 minutes. Cloud droplets sizes and cloud fraction are using the model default values as described in Liu et al. (2012). Entrainment between the cloudy and non-cloudy areas is neglected here. We conduct a one-year simulation, and factors potentially influencing formation of $SOA_{cld}$ (see Section 2.1 and Figure 1)* *are archived every 3 hours. Therefore, for each grid box, the modeled production of* $SOA_{cld}$ *in each timestep is physicochemically determined by the availability of cloud water, water-soluble gases, oxidants, etc."*

Last paragraph of Section 2.3: "*The spatiotemporal dependence of* $P_{SOAcld}$ *on a variety of individual meteorological and chemical factors and their combinations are examined based on correlation analysis and multiple regression models with/without nonlinear transformations (e.g., logarithm or categorizing our analysis by* $VOC/NO_x$*). We note that factors could be significantly involved in the non-linear production of* $SOA_{cld}$ *but show little direct spatiotemporal association to* $P_{SOAcld}$*. Such factors will not typically be given strong weight by spatiotemporal statistical inference, consistent with our goal of identifying the key factors (or best predictors) best explaining the spatial and temporal diversity of* $P_{SOAcld}$*. The final parameterization predicting* $P_{SOAcld}$ *is explored based on both statistical inference and sensitivity analysis."*

Beginning of Section 3.1: "*Spatiotemporal associations between daily* $P_{SOAcld}$ *and each of the six factors (i.e., LWC,* $TC_{loss}$*, concentrations of OH and* $O_3$*,* $VOC/NO_x$*, and temperature) are examined by a correlation analysis with a sample size of approximately 95 million. Here we apply a linear relationship to non-linear processes of* $P_{SOAcld}$*, but those non-linear processes are partially accounted for in the prognostic model simulation (though not perfect).*"

*Section 3.1/3.2: What factors co-vary with LWC and Total Carbon Loss Rate, which could potentially assist with SOA production? When putting too much weight into correlations, it is critical to provide perspective on what co-varies with the most important parameters.*

As we mentioned before, all the factors we chosen in our analysis can physically determining $P_{SOAcld}$ directly or indirectly. The cause-and-effect relationship is already built in our model. Correlation is used to determine the most critical factors to investigate spatiotemporal differences and examine relationships. In a fully coupled climate system, basically every two variables can co-vary either linearly or non-linearly to some extent. For example, LWC should co-vary with processes related to large-scale condensation. Total carbon loss rate is directly determined by VOCs, oxidants and factors influencing their reaction rates, and is indirectly determined by the VOCs emissions/transport and other factors controlling oxidants and temperature. Since LWC and $TC_{loss}$ could be solved in most global chemical transport models and new-generation climate models with online gas-phase chemistry, factors physically plausible confounding LWC or $TC_{loss}$ are investigated. For example, in section 3, we list the correlation matrix among the six factors in the supplementary material (see Table S1). We find that the correlation coefficients are low (<0.2) between LWC (or $TC_{loss}$) and other factors.

Table S1. Correlation matrix between the six factors

| | LWC | $TC_{loss}$ | Temp | OH | O3 | $VOC/NO_x$ |
|---|---|---|---|---|---|---|
| LWC | 1 | 0.07 | 0.18 | 0.05 | -0.1 | -0.04 |
| $TC_{loss}$ | | 1 | 0.17 | 0.08 | -0.03 | 0.07 |
| Temp | | | 1 | 0.66 | -0.22 | -0.13 |
| OH | | | | 1 | -0.08 | -0.1 |
| O3 | | | | | 1 | 0.08 |
| $VOC/NO_x$ | | | | | | 1 |

In ER08, high correlation between LWC and cloud contact time was observed, but both terms were included in their parameterization. However, in this study, if LWC is replaced by cloud volume fraction, the $R^2$ will decrease by half. Therefore, to screen out best factors we present the final factors with the highest $R^2$. To clarify this, we add the following in the revised manuscript:

Section 3: "*Above analysis indicates that LWC and $TC_{loss}$ have the highest correlation with $P_{SOAcld}$. To understand the inter-dependence between the six factors, Table S1 in the supplementary material lists their correlation matrix. In general, neither LWC nor $TC_{loss}$ is correlated well (r<0.2) with the remaining factors.*"

Section 4: "*The cross regional $R^2$ of Eq. (2) is approximately 0.5, indicating that the combination of LWC and $TC_{loss}$ alone can explain nearly half of the spatial and temporal variability of $P_{SOAcld}$ in a global scale. To investigate factors that potentially confounding with LWC and $TC_{loss}$, other physically plausible relationships were investigated. In Ervens et al., 2008, cloud contact time ($\propto$ cloud fraction) is significantly associated with cloud liquid water. In this study, if LWC is replaced by cloud fraction, $R^2$ will decrease by half. Therefore, we present the factors with the highest $R^2$.*"

*Section 3: Many of the results agree with those of Ervens et al. (2008), which again begs the question as to why this type of study is being repeated with a model that cannot examine the relevant processes at the scale of single clouds. Example: Line 10-14 on pg 26939.*

Actually, many of our results are inconsistent to those of ER08. For example, as we mentioned on line 11-14, Page 26939 in the original manuscript, $VOC/NO_x$ ratio is a trivial factor influencing $P_{SOAcld}$ in a global scale, though it is critical in the work of ER08. We also analyzed the data grouped by $VOC/NO_x$ (i.e., >100, 5-100, and <5) but found little improvement in model overall predictability. Therefore, this factor might be important at few locations where the conditions are similar to the configuration of the clouds used in ER08. However, in a global scale, much more possibilities exist where results based on the single clouds in ER08 may not fully capture. Here we summarized all these differences between this work and ER08 in

Table S2 in the supplementary material:

Table S2. Major difference between this work and ER08

| | **This work** | ***Ervens et al.,* 2008** |
|---|---|---|
| **Model** | GCM coupled with detailed gas and cloud chemistry | Cloud parcel model with detailed gas and cloud chemistry |
| **$SOA_{cld}$ precursors** | isoprene, toluene, α-pinene | isoprene |
| **Multiphase chemistry** | *Jacob,1986; Liang and Jacobson, 1999; Lim et al.*, 2005; *Tan et al.*, 2010; *Emmons et al*., 2010; *Liu et al.*, 2012 | *Ervens et al., 2004a*; *Lim et al.*, 2005; *Altieri et al.*, 2006; *Carlton et al.*, 2006,2007 |
| **Transport** | Coupled chemistry-climate GCM simulated continuously for one year | Offline 45 1-h trajectories describing passages of an air parcel through stratocumulus clouds, derived from three-dimensional Eulerian large-eddy-simulations. Repeat 5 times from 10 am. |
| **Methods** | Statistical analysis on the spatiotemporal association of different factors with respect to in-cloud SOA formation | Sensitivity of in-cloud SOA production against initial chemical composition in a closed system. |
| **Parameterization** | $P_{SOAcld} = \alpha \cdot LWC \cdot TC_{loss}^{\gamma} + \beta$ | $P_{SOAcld} = (a \cdot LWC + b \cdot \tau + c) \cdot TC_{initial}$ |
| **Major findings** | 1. LWC linearly determines the spatiotemporal variability of $P_{SOAcld}$.<br>2. The spatiotemporal association between $TC_{loss}$ and $P_{SOAcld}$ is non-linear.<br>3. $VOC/NO_x$ is not spatiotemporally correlated ($\|R\| < 0.1$) well with $P_{SOAcld}$. | 1. Initial $VOC/NO_x$ and τ are the most important factors.<br>2. In-cloud SOA yield is less affected by LWC. |

*Section 4 and Tables 2-3: In my opinion, this analysis really does nothing to advance our knowledge of in-cloud SOA production and its sensitivity to parameters such as LWC. It is just an extension of Table 1, which I still struggle to understand as to why* *it would be better than a modeling study at a more cloud-relevant resolution (and what is being shown that is significantly new as compared to older work).*

Table 1 shows the linear least squares correlation between $P_{SOAcld}$ and each individual factor. Data is paired in space (horizontal and vertical) and time (daily average). Tests in Table 2 show regression coefficients (i.e., the slope of each predictor) for multiple factor linear regressions for two scenarios, 1) if all six listed factors are simultaneously associated with $P_{SOAcld}$ and 2) when LWC and $TC_{loss}$ are the only considered factors. The meaning of each value in Table 1 is correlation (i.e., the r value), whereas values in Table 2 show the regression coefficients (i.e., the slope of each predictor). Therefore, their meanings are different between these two tables. The role of table 1 is to show the relative importance of factors influencing $P_{SOAcld}$ and help to screen out factors with significant association to the spatiotemporal variability of $P_{SOAcld}$. In table 2, we use multiple regression models and screen out predictors by standardized regression coefficients (i.e., the B values which indicate the relative contribution of each factor to the predicted variable $P_{SOAcld}$). The b values in table 2 suggest the exponents of LWC and $TC_{loss}$ used in Eq. (2). Table 3 is discussed in Section 6 in the original manuscript. These values in addition to Figure 7 in the original manuscript show spatial sensitivity of $P_{SOAcld}$ to each parameter $\alpha$, $\beta$, $\gamma$ in Eq. (2). This would be useful when tuning Eq (2) to fit either global mean values or observations.

Section 4 and Table 2 have summarized our new findings related to the spatiotemporal relationship between $P_{SOAcld}$ and these key factors in a global scale. We find that (1) LWC is the dominant factor controlling $P_{SOAcld}$ globally (Table 2), whereas ER08 indicated "in-cloud SOA yield is less affected by LWC". (2) $P_{SOAcld}$ varies linearly with LWC, but nonlinearly (concavely) with VOCs oxidation rate, whereas in ER08 in-cloud SOA production varies linearly with VOCs initial mass concentration. (3) VOC to $NO_x$ ratio is not a key factor consistently determining the spatiotemporal distribution of $P_{SOAcld}$ (and no significant improvement is observed by reanalyzing the data grouped by $VOC/NO_x$), whereas in ER08 $P_{SOAcld}$ is mostly determined by $VOC/NO_x$ and cloud contact time. Cloud contact time is actually difficult to employ in a GCM (the physical meaning of cloud contact time is different to cloud volume fraction). Finally, for model resolutions (i.e., bulk cloud *vs* small scale cloud), it was stated clearly in ER08 that "$SOA_{cld}$ can be modeled in bulk cloud water similar to sulfate". Therefore, section 4 (in the original manuscript) and table 2 have shown how the key factors influence spatiotemporal variability of $P_{SOAcld}$ in a global scale, which to our knowledge is new to our community.

In the revised manuscript, we have edited many places to clarify the meanings of values in Tables and include a table (i.e., Table S2 in the supplementary material) to compare the major difference between this work and ER08.

*Section 5: This analysis also does not do much beyond what already was shown in Table 1 in terms of trying to advance understanding of what factors influence SOA formation. Pg 26940, Line 24-25: I don't understand what is meant by ". . .than the estimation based on detailed cloud chemistry".*

Section 5 evaluates how well AM3 would perform using the new parameterization (i.e., Eq 2, developed in section 4) rather than using online detailed cloud chemistry as developed in Liu et al., 2012. Then we compare the parameterized result (based on Eq 2 alone) to what GCM with detailed cloud chemistry predicts. We find that our new parameterization can capture well the features generated by GCM with detailed cloud chemistry. This also indicates the difference in $P_{SOAcld}$ parameterization between this work and ER08. Features in ER08 parameterization are suitable for application in cloud parcel model or global models which track air parcels' cloud contact time, but may not adequately capture the actual processes at play in most GCM. To avoid misunderstanding, we combined sections 4 and 5 into one section "Section 4 Parameterization of $SOA_{cld}$ production and its evaluation". We also made revisions to Pg 26940, Line 24-25 in the original manuscript:

"*After implementing Eq. (2) into AM3 to replace the detailed cloud chemistry scheme, the parameterized global $SOA_{cld}$ production is 23.0 Tg•yr$^{-1}$, close to what detailed cloud chemistry shows (23.4 Tg•yr$^{-1}$), as well as the results based on uptake coefficients (~22 Tg•yr$^{-1}$, Fu et al., 2008, assuming OM:OC=2. We did not compare our results to Ervens et al., 2008 since implementation of the parameterization as suggested by Ervens et al., 2008 would require additional information (i.e., cloud contact time, not equivalent to cloud volume fraction) beyond the processes at play in AM3).*"

*Figures 3-7: I find it troubling to invest this many figures and discussion on such a simplified parameterization (Eqn 2), which cannot be expected to capture all of the aspects influencing SOA production in clouds. Section 6.2 thankfully tries to address some of these issues, but a more important question is what is being learned in this analysis (Figs 3-7) that can significantly advance our knowledge of factors influencing SOA formation.*

Figures 3-6 are used to show that although our parameterization (Eq 2) is simple, it works (shown in figures 3-6) to capture the global spatiotemporal distribution of $SOA_{cld}$ production. Most GCMs do not have detailed cloud chemistry scheme and cannot afford the computational expense. Therefore the $SOA_{cld}$ source is totally neglected in their models. Even though we implement the detailed multiphase chemistry in AM3 (i.e., Liu et al., 2012), it is expensive to use it for long-term climate simulation (computer hours increase nearly a factor of 2 when the detailed cloud chemistry is simulated online). Therefore, in this study we explored and screened for the most influential factors (i.e., LWC and $TC_{loss}$) and derived the empirical parameterization which represents the essential of the role of detailed multiphase chemistry on global production of SOA from cloud processes. This is the exact approach employed in ER08, but our findings suggest different factors regulate $SOA_{cld}$ production and we develop an alternate parameterization. Similar to ER08, Carlton et al., 2008 as well as Fu et al., 2008, this study delivers additional insights regarding global spatiotemporal variability of $SOA_{cld}$ formation and provides an additional parameterization which can capture well in-cloud SOA production in a global scale.

To make the paper concise, and describe this better we put Figure 4 into supplementary material and combined Figures 5 and 6 into one figure. Figure 7 shows the sensitivity of $P_{SOAcld}$ to each tunable parameter in Eq 2. It provides the guidelines on how to implement our parameterization into other global models with different time resolution.