First of all, we'd like to thank anonymous referee #2 for the detailed comments, which help improve the manuscript. All the reviewer's points have been carefully considered and our responses are given below after each review comment (given in italics).

In this manuscript, the authors use a global model framework to explore various SOA formation mechanisms in the aqueous phase. Detailed multiphase reaction schemes are compared to simpler surface-limited uptake processes. In addition, the effect of dissolved iron chemistry on aqSOA formation is investigated. Finally, the model results are compared to observations.

There are quite a few comparisons that the authors conduct in this study, but it is unclear what are the conclusions or the take-away messages. Which of their case studies would be closest to reality and should be incorporated in future models? I am not sure whether the comparisons that they make to the observations necessarily validate their model, they might be a little far-reaching. This paper presents good ideas of the different mechanisms of aqSOA formation that can be used in global models and I think it would be most valuable if they could provide recommendations on why and which of these would be most "ideal" for future studies. For purposes of global modeling what parameters is aqSOA formation most sensitive to? Also, they should do a thorough read through again and try to make the language succinct and more clear.

Reply: We will revise the abstract and add one paragraph in the end of Section 5 conclusion (see below) to clarify the take-away messages and to provide recommendations for future work. We have done the sensitivity studies associated with the uncertainties resulting from different aqSOA schemes, different cloud field, and with or without dissolved iron chemistry in cloud and found the aqSOA formation rate is sensitive to the aqSOA scheme used in the model, cloud water content, and iron chemistry. Other sensitivity studies (e.g., based on altering cloud properties other than cloud water content, aerosol size distributions, and the parameters within the chemical mechanisms) are beyond the scope of this paper. Many of these sensitivity tests have been done in other published studies and we will point to them in the revised manuscript (see our replies below as well as replies to Referee #1). Meanwhile, we have done an additional sensitivity test by increasing the effective Henry's law constants of glyoxal and methylglyoxal in aerosol water content due to the ion strength effect (for details, also see our responses to Referee #1). Last but not least, we will check through the full paper again and do as much as we can to clarify the paper.

The revised abstract will be as follows:
"There is growing interest in the formation of secondary organic aerosol (SOA) through condensed aqueous phase reactions. In this study, we use a global model (IMPACT) to investigate the potential formation of SOA in the aqueous phase. We compare results from several multiphase process schemes with detailed aqueous phase reactions to schemes that use a first order gas-to-particle formation rate based on uptake coefficients. The net global SOA production rate in cloud water ranges from 13.9 Tg/yr to 46.8 Tg/yr while that in aerosol water ranges from -1.5 Tg/yr to 12.6 Tg/yr. The global burden of SOA formed in the aqueous phase ranges from 0.09 Tg to 0.51 Tg. A sensitivity test to

investigate two representations of cloud water content from two global models shows that increasing cloud water by an average factor of 2.7 can increase the net SOA production rate in cloud by a factor of 4 at low altitudes (below approximately 900 hPa). We also investigated the importance of including dissolved iron chemistry in cloud water aqueous reactions. Adding these reactions increases the formation rate of aqueous phase OH by a factor of 2.2 and decreases the amount of global aqueous SOA formed by 44%. None of the mechanisms discussed here is able to provide a best fit for all observations. Rather, the use of an uptake coefficient method for aerosol water and a multi-phase scheme for cloud water provides the best fit in the Northern Hemisphere and the use of multiphase process scheme for aerosol water and cloud water provides the best fit in the tropics. The model with iron chemistry under predicts oxalate measurements in almost all regions. Finally, the comparison of O/C ratios estimated in the model with those estimated from measurements shows that the modeled SOA has a slightly higher O/C ratio than the observed SOA for all cases. ”

One more paragraph will be added in the end of Section 5 Conclusions as follows:
“Currently, there is no mechanism that is able to capture all observations well. The cases that use an uptake coefficient method for aerosol water perform better than the cases using the multiphase reaction scheme in comparison to the Northern Hemisphere AMS measurements. However, in the tropics, the use of a multiphase process scheme for both aerosol water and cloud water perform best, while the case using the uptake coefficient method in cloud water clearly overestimates the observed OA by as high as more than a factor 3. The multiphase reaction scheme including iron chemistry under predicts the observed oxalate concentrations at all sites. Future work is needed to close the gap between simulations and observations. In particular, more lab and model studies are needed to improve the representation of chemical reactions within the aqueous phase and at the gas/aerosol interface, since there is still a large inconsistency between existing gas/aerosol transport representation and reaction mechanism derived from lab studies and field measurements Also, more work is needed to improve our understanding of oxalate sources and sinks because the model tended to underestimate observed oxalate concentrations for all cases studied. In addition, the high sensitivity of aqSOA to cloud water content shows the importance of improving the representation of cloud water content in general circulation models in order to improve aqSOA formation.”

Specific comments:
p.29632, l.2: thereby instead of there

Reply: We will change “there” to “thereby”.

p.29632, l.20: observed to “be” part of

Reply: We will add “be” in the front of “part of”.

p.29635, l.9: Did the authors mean formation “of” SOA instead of on?

Reply: Yes. We will change “on” to “of”.

p.29636, l.9: Can you include a source for using the value of 0.0029 as the uptake coefficient?

Reply: We will change the sentence to "In addition, we also accounted for SOA formation due to heterogeneous reactions of epoxides on the surface of wet sulfate aerosol by assuming an uptake coefficient of 0.0029, the same value as that for uptake of glyoxal derived by Liggio et al. (2005)."

p.29636, l.10: Please elaborate on why this is the most realistic mechanism.

Reply: We will revise this sentence to "We used the Simulation C in Table 1 in Lin et al. (2012), since this mechanism was shown to best capture the observations of OH and the first generation products of isoprene-OH reactions. ".

p.29636, l.17-21: Include a reference

Reply: We will add two references as follows: "In the presence of cloud droplets or aqueous particles, water-soluble gases (e.g., glyoxal, methylglyoxal, and glycolaldehyde) will dissolve in the aqueous phase and be further oxidized by OH and NO_3 radicals to form products with lower volatility (e.g., dicarboxylic acids and oligomers) (Ervens et al., 2011; Lim et al., 2010)."

p.29637, l.3-4: Should it be "are" instead of is, and "equations" instead of equation?

Reply: Yes. Thanks. We will revise this sentence as follows: "The change of aqueous and gas phase species due to photochemical reactions and the exchange between the gas and aqueous phase are expressed by the following equation, ".

p.29637, l.22: What is the rationale behind using the value for gas diffusivity for all species?

Reply: Because not all species' gas diffusivities are known and the values of available gas diffusivities don't differ much from the value we used here (Schwartz (1986); Lelieveld and Crutzen (1991)). So we will clarify this by changing this sentence to: "A gas diffusivity of 0.1 $cm^2 s^{-1}$ is assumed for all gaseous species, because not all species' gas diffusivities are known and the values of available gas diffusivities do not differ by much from the value we used here (Schwartz, 1986; Lelieveld and Crutzen, 1991).".

p.29638, l.3-5: Can you elaborate on how the effective radius of the particles is calculated?
Were any sensitivity calculations conducted for the cloud droplet radius? And what, if any, affect do the sizes used for the study have on the uptake rates?

Reply: For the particles we use the ratio of third to second moment of the wet sulfate aerosol size distributions. As stated on line 4-5, page 29638, it is humidity dependent. We will clarify this by revising the sentence on line 3-5, page 29638, to "In addition, an effective cloud droplet radius of 10 μm is assumed for all clouds, while the effective radius for aqueous sulfate particles (the ratio of third to second moment of the sulfate aerosol size distributions) is calculated explicitly according to their relative humidity dependent size distributions".

Liu et al. (2012) have studied the sensitivities of predicted aqSOA formed in cloud to the cloud droplet size by using a global model. They found that the predicted aqSOA depended on the cloud droplet size weakly. We will discuss the effect of cloud droplet size on the uptake rates by citing Liu et al. (2012) paper.

p.29642, l. 2: add "the" before supplement

Reply: We will do it.

p.29642, l.3-7: Can you briefly describe the differences between the two models used for the cloud field output? And, why the AM3 is picked instead of others?

Reply: Cloud water content is prognosed in GFDL AM3 while the cloud water content is diagnosed in CCM2. AM3 was picked primarily because it was easily assessable to us. Scientifically, any other GCM cloud field output can be used to study the effect of cloud water on aqSOA formation. Jiang et al (2012) compared the liquid water path (LWP) output from 19 climate models submitted to CMIP5 to NASA A-train satellite observations. Root mean square (RMS) of the difference between the modeled and observed LWP is shown to range from 0.7 to 2.0 g/m^2, among which GFDL AM3 has a RMS of 0.9 g/m^2.

p.29642, l.17: Consider changing it to Results & Discussion

Reply: We will change it to "Results & Discussion".

p.29642, l.21: missing word between shown and the

Reply: We will revise this sentence to "We note that the predicted oligomers in the model can consist of different numbers of monomers (e.g., dimers, trimers and tetramers), so that the total oligomers shown in different cases in Table 2 do not necessarily consist of identical species.".

p.29643, l.13-19: For comparison purposes, stay consistent with using exact values or percentages or both for all compounds discussed.

Reply: We will revise these sentences as follows: "However, the contribution of aerosol water to the formation of organic acids formations is very small. The net production rate of glyoxylic acid in the aerosol water accounts for only 0.025 Tg yr^{-1} of the total 4.5 Tg yr^{-1} net production rate; the net production rate for pyruvic acid formed in aerosol water is only 3.68×10^{-4} Tg yr^{-1} of the total 1.84×10^{-1} Tg yr^{-1}; and the net production rate for oxalic acid in the aerosol water (after consumption by reaction with OH) is −0.7 Tg yr^{-1}, compared to the total net production rate of 15.7 Tgyr^{-1}."

p.29644, l.3: "contributor" instead of contribution

Reply: We will change it to "contributor".

p.29645, l.21: : : :and "for" oligomer concentrations instead of in

Reply: We will correct it.

p.29647, l.7-8: Be consistent with using or not using the parenthesis for the references

Reply: We will revise it to "The reason for the discrepancy between measurement studies (i.e. Waxman et al. (2013) and Liggio et al. (2005))".

p.29649, l.2: Could you compare the rates for oxalate destruction due to addition of iron chemistry and increase in SOA due to OH.

Reply: Fe-complex photolysis accounts for 26.5 Tg/yr of oxalate destruction, while the destruction rate through OH and NO_3 reactions is 1.30 Tg/yr. We will add this comparison in this paragraph.

p.29650, l.4: Elaborate on the effect of iron chemistry on SOA formation in clouds vs aerosol water.

Reply: We have discussed the effect of iron chemistry on SOA formation in cloud vs. aerosol water in Section 3.5. We will clarify this by changing this sentence to "In case 5 with the iron chemistry in cloud, the SOA production rate in cloud explains 63.3 % of total aqSOA production rate (see the Sect. 3.5. for details).

p.29650, l.7: Since there are a few different comparisons to observations in this section, a short introductory paragraph about what is being discussed and the relevance of these comparisons (simple clarifications like why compare oxalate, O/A ratios etc.) would help the flow of the manuscript. Somewhere in this section a brief discussion comparing the oxalate results to those from Myriokefalitakis et al. (2011) could be included.

Reply: We will add an introductory paragraph as follows: "In this section, we will compare model results to measured oxalate, AMS measurements of SOA, and SOA O/C ratios. Oxalate is a major component of aqSOA formed in cloud, thus the comparison of oxalate gives us a direct evaluation for the modeled aqSOA, while comparison of SOA measured by AMS data and O/C ratios will indirectly constrain the modeled aqSOA.". As suggested by Reviewer #1, we will also divide this section into three subsections as 4.1. Oxalate; 4.2. AMS measurements; 4.3. O/C ratios. We will also add a paragraph on line 12, page 29652, to briefly discuss the comparison with Myriokefalitakis et al. (2011) as follows: "The global oxalate burden in the simulation that Myriokefalitakis et al.

(2011) predicted in their case S1 is 5 times larger than the burden in Case 1 in our predictions. This is partly because Myriokefalitakis et al. (2011) increased the solubility of glyoxal and methylglyoxal in cloud water by 2 orders of magnitude, which is at the high end of measured values, causing a larger oxalate source. Also, Myriokefalitakis et al. (2011) included a set of additional reactions performed by Carlton et al. (2007) under higher glyoxal concentration conditions than those present in real cloud water, which results in more oxalate formation. In addition, a longer oxalate lifetime with respect to deposition was predicted by these authors. The larger oxalate burden in Myriokefalitakis et al. (2011) leads to a closer agreement with measurements over rural sites. However, the simulations in Myriokefalitakis et al. (2011) did not include iron chemistry in cloud which can decrease oxalate concentrations significantly (compare Case 1 and Case 5).".

P.29652, l.5: something missing in Florida Sydney?

Reply: We will clarify this by changing the sentence to: "The three blue circles in Fig. 6 show the comparisons for the sites in US, and indicate that the model does reasonably well, except for the site in Sydney, Florida, at which the model still under-predicts the oxalate source or/and overestimates its sink.".

p.29655, l.3-26: This discussion could be rephrased and rearranged as its easy to get lost in all the information. The ideas of organic hydroperoxides and organic nitrate are introduced without relaying their significance.

Reply: The reason we mention organic hydroperoxides and organic nitrate alone in the paper is that in our model the aerosol phase reactions of organic hydroperoxides and organic nitrate dominate the formation of SOA formed from aerosol phase reactions of condensed SVOCs (Lin et al., 2012). To make this 'comparison of O/C ratios' part to be easy to follow, we will rearrange this long paragraph as below: divide it to several short paragraphs, list the method of calculating the O/C ratio separately for each SOA component, and point the importance of organic hydroperoxides and organic nitrate.

"Aiken et al. (2008) derived a significant correlation between the O/C ratios of OA and their f44 signal (the ratio of m/z 44 to the total signal in the mass spectrum). By using this correlation, Ng et al. (2010) estimated the O/C ratio of oxygenated OA (OOA) obtained from a factor analysis of the Northern Hemisphere AMS dataset. Here, we compare the O/C ratios estimated in the model with those reported by Ng et al. (2010) (Fig.8).

In the model, we have four different SOA components: SOA from the gas-particle partitioning of semi-volatile organic compounds (SVOCs), SOA from aerosol phase reactions of these condensed SVOCs, SOA from the uptake of epoxide on sulfate aerosol, and SOA formed in the aqueous phase as described above (aqSOA). We used the following methods to estimate the predicted O/C ratio of these SOA components:

(1). For the SOA from the gas-particle partitioning of SVOCS, there are 26 explicit SVOCs that contribute to the SOA formed from gas-particle partitioning. We calculated the O/C ratio for each species based on their chemical formulas (See Table S1 in Lin et al. 2012).

(2). For the SOA from aerosol phase reactions of condensed SVOCs, however, it is not straightforward to calculate O/C ratio. First, the aerosol phase reactions of condensed SVOCs were simply treated in the model as first-order reactions to form oligomers with an assumed time constant (nominally 1 day), without any information on the products from these aerosol phase reactions. However, Chen et al. (2011) and Liu et al. (2012) suggested specific aerosol phase reactions for organic hydroperoxides and organic nitrate, respectively, both of which are major components of SVOCs in the model (Lin et al., 2012). Chen et al. (2011) proposed a new aerosol phase reactions for organic hydroperoxides: decomposition, followed by radical-radical oligomerization to explain the smaller measured O/C ratios than those predicted in their model. This aerosol phase reaction was shown to remove 1~2 O atoms from organic hydroperoxides. Organic nitrate is thought to undergo hydrolysis in the particle phase (Liu et al., 2012), which removes two O atoms from organic nitrate. Despite the simple treatment for oligomer formation in the model, we assume that the oligomers from organic hydroperoxides and organic nitrate have 1.5 O atoms and two O atoms less than condensed organic hydroperoxides and organic nitrate have, respectively, based on Chen et al. (2011) and Liu et al. (2012). For the oligomers formed from other condensed SVOCs, we assume they have the same O/C ratio as their corresponding SVOCs. An additional complexity is that in the model the 26 SOA species formed from the oligomerization of condensed SVOCs are lumped together as one species when they are transported in the atmosphere. Therefore, here we used their global-averaged SOA formation rates (listed in Table S1 in Lin et al. 2012) rather than their mass concentrations to weight their relative contributions to the O/C ratio. Based on these assumptions, we estimate the average O/C ratio for the SOA from aerosol phase reactions of condensed SVOCS to be around 0.687, with values for some of the SVOCs with smaller burdens ranging from 3.0 to 0.3.

(3). For SOA from the uptake of epoxide, the O/C ratio is estimated to be 0.6 since the O/C ratio of epoxide from isoprene oxidation is 0.6 in the model, and its oligomerization would not be expected to change the O/C ratio (Surratt et al., 2010).

(4). The O/C ratio for oxalic acid is 2, for glyoxylic acid is 1.5, and for pyruvic acid is 1.0. The O/C ratio for oligomers from glyoxal and methylglyoxal is assumed to be 1.5, which is consistent with the measurements by Lim et al. (2010).

We then averaged the O/C ratios of these four SOA components by weighting their mass concentrations to obtain the O/C ratio for total SOA. As shown in Fig. 8, the modeled O/C ratios are higher than those reported by Ng et al. (2010) and have a normalized mean bias of 38%. The higher O/C ratios estimated in the model might suggest an overestimation of the contribution of multiple generation oxidation products to SOA formation or a missing aerosol phase reaction of condensed SVOCs that would lead to products with lower O/C ratios. Alternatively, the AMS instrument might underestimate O/C ratios."

References not already shown in the original manuscript or the reviewer comments:

Jiang, J. H., Su, H., Zhai, C., Perun, V. S., Del Genio, A., Nazarenko, L. S., Donner, L. J., Horowitz, L., Seman, C., Cole, J., Gettelman, A., Ringer, M. A., Rotstayn, L., Jeffrey, S., Wu, T., Brient, F., Dufresne, J.-L., Kawai, H., Koshiro, T., Watanabe, M., LÉcuyer, T. S., Volodin, E. M., Iversen, T., Drange, H., Mesquita, M. D. S., Read, W. G., Waters, J. W., Tian, B., Teixeira, J. and Stephens, G. L.: Evaluation of cloud and water vapor simulations in CMIP5 climate models using NASA "A-Train" satellite observations, J. Geophys. Res., 117(D14), D14105, doi:10.1029/2011JD017237, 2012.