We thank the reviewers for careful reading and helpful comments that improve the quality of our manuscript. Reviewer comments have been copied followed by our responses in bold.

Anonymous Referee #4)

In recent years water-soluble di-carbonyls draw more attention due to their great potential in forming highly oxidized products in the atmospheric waters. The present work integrates recently developed methylglyoxal aqueous-phase chemistry (spans from dilute to concentrated conditions) with previously published glyoxal chemistry. This manuscript provides insights for the detailed chemistry and leads to a better understandings. The manuscript is well-written and I recommend publication after the author addresses some minor concerns outlined below.

General comments:

R4C1) Page 1, line 23-25, "At cloud relevant concentrations (10ˆ-6 ~ 10ˆ-3 M; Munger et al., 1995) of glyoxal and methylglyoxal, the major photooxidation products are oxalic acid and pyruvic acid, and simulated SOA yields (by mass) are ~120% for glyoxal and ~80% for methylglyoxal." Apparently according to Fig 9 the overall mass yield depends on the initial glyoxal/methylglyoxal level, e.g. at 10ˆ-3 M glyoxal the mass yield drops to ~80% (i.e. net mass loss?). This is interesting and consistent with Lee et al (ACPD, 2012) who indicated that under CERTAIN conditions (3mM glyoxal + 13.3mM H2O2 + light) fragmentation could dominate the whole glyoxal/methylglyoxal processing and eventually lead to mass loss. However, it seems that in Lee et al (ACPD, 2012) the mass reduction is due to the presence of H2O2 (the major product would be HCOOH rather than oxalic acid), i.e. Lee et al created cloud-relevant OH level (by means of the H2O2 photolysis) but probably a much higher H2O2 level than cloud condition. What is the major reason for the mass loss at 1e-3 - 1e-1 M glyoxal, as shown in Figure 9? What is the gas-/aqueous-phase H2O2 level in the atmospheric simulations?

Response: The comment pertains to modeling under atmospheric conditions. The findings by Lee et al are consistent with ours. The major reason for the mass loss is due to H_2O_2 reactions with glyoxylic acid forming formic acid and this was actually already discussed in our previous paper (Lim et al., 2010). The large mass loss occurred when the initial glyoxal concentration was 1e-2 M. In our atmospheric simulations, the initial H_2O_2 concentration is set to be zero, but as OH oxidation of glyoxal proceeds, H_2O_2 concentration builds up to 4.6 mM. This makes the lifetime of glyoxylic acid due to H_2O_2 as short as 12 minutes. The lifetime of glyoxylic acid due to OH radicals (1e-12 M) forming oxalic acid is about 45 minutes. So, at this condition, glyoxylic acid mainly reacts with H_2O_2 forming formic acid. Normally, at cloud conditions (1e-5 – 1e-4 M glyoxal) H_2O_2 concentration builds up to only 6 – 80 μM and the corresponding lifetime of glyoxylic acid due to H_2O_2 is ~11 hrs to ~6 days, which are too slow to compete with glyoxylic acid reactions with OH radicals. Note that in our model we assumed there is no evaporation of H_2O_2 formed in the aqueous phase (and there is no gas-phase H_2O_2 uptake, either) because H_2O_2 is very water soluble (H = 1e5 M/atm).

To address this, we added the following at the end of the Section 4.2A.

Note that at 1e-2 M of initial glyoxal in Fig. 9A, SOA yield is at a minimum (56%) for the following reason. For this initial glyoxal concentration substantial (up to 4.6 mM) H_2O_2 forms by HO_2 + HO_2 and HO_2 + O_2^- reactions, and the reaction of glyoxylic acid + H_2O_2 forming formic acid competes with the reaction of glyoxylic acid + OH radicals to form oxalate and organic radical-radical reactions to form oligomers (Lim et al., 2010). This

finding from atmospheric simulation is consistent with previous experimental results at similar concentrations suggesting a role for H_2O_2 (Lee et al., 2012).

R4C2) Page 9, line 1: I myself don't think the SOA yield here is well defined. Glyoxal in the atmospheric waters can undergo various pathways and subsequently contribute to SOA, such as reversible reactions (depends on many factors such as sulfate level), irreversible reactions (glyoxal+OH, glyoxal+NH4+) or surface-limited processes. In the present work the SOA yield only covers the glyoxal+OH pathways, which doesn't always play a dominant role in the aqueous-phase chemistry of glyoxal.
In addition, to my knowledge, currently many 3D chemical transport models only include basic aqueous-phase chemistry for cloud, not for wet aerosols, simply because they cannot afford big calculations. In order to use these yields, phase-transfer needs to be included for glyoxal/methylglyoxal as well as OH. Given that glyoxal might only contribute to limited mass to the total SOA (California: Washenfelder et al., 2011; Mexico City: Waxman et al., 2013), these parameters (yields derived in this work) might not be entirely feasible for 3D modelers at this moment.

Response: We accept that organic-inorganic reactions can also be important in wet aerosols and evaporating droplets. We have made that more clear by adding following at the end of Section 2.5.

It should be recognized the chemistry in wet aerosols is highly complex and poorly understood. It undoubtedly includes organic-inorganic interactions as well as oxidation reactions; the yields reported here are not intended to represent all processes. This paper represents a modest step forward in understanding organic photooxidation in wet aerosols. We expect OH oxidation to be a dominant daytime aqSOA formation pathway in clouds and an important contributor to wet aerosol chemistry (Lim et al., 2010).

Also, to be more clear we changed the section title to "SOA yields from atmospheric photochemical simulations"

By the way, our model for atmospheric conditions does include phase transfer for glyoxal/methylglyoxal and OH radicals and we clearly mentioned this in the text. In Section 2.5, "In atmospheric simulations, the OH radical concentration in the aqueous phase was set to be constant at 2.44×10^{-12} M, a value maintained by Henry's law equilibrium with the gas-phase OH radical concentration of 2×10^{6} molecule cm^{-3}." And "…glyoxal and methylglyoxal uptake by atmospheric waters (i.e., Henry's law equilibrium between gas- and aqueous-phase glyoxal and methylglyoxal)."

While adding this chemistry or incorporating these yields may not be feasible for all models, the insights provided by the modeling are, in our opinion, worthy of sharing with the scientific community. Incorporation of glyoxal, methylglyoxal, acetic acid and glycolaldehyde cloud chemistry (validated by our past experiments) in the GFDL global model has proved informative (Liu et al., 2012; He et al., 2013*)

R4C3) Page 10, line 6-7, "Oligomers were calculated as the sum of products with higher carbon number than the precursor (Lim et al., 2010)." Please provide the detailed characteristics of the oligomers since they dominate the SOA mass under concentrated conditions, e.g. what products are actually summed into this oligomer, also please show the product distributions of oligomers formed from glyoxal and methylglyoxal, respectively.

Response: We now include a plot (pie chart) of carboxylic acid oligomers differentiating by carbon number (C_nD, n = carbon number), formed from 1 M glyoxal/methylglyoxal + OH in Supplementary Material (Fig S3). We added the following to the Section 4.2A:

(Note that detailed oligomer distributions for 1 M glyoxal and 1 M methylglyoxal are provided in Fig. S3)

R4C4) In addition, it will be great if the authors could discuss in the context of ambient measurements available since some of the model scenarios in this work are ambient relevant. I am particularly interested in looking for evidence or tracers for the wet aerosol processes and apparently more insights of the oligomers would help a lot. According to Lim et al (2010), compounds such as tartaric acid and malonic acid are major products of the radical-radical chemistry of glyoxal. It seems to me that malonic acid has been reported in aerosol samples in many locations, but tartaric acid, on the other hand, is rarely seen at remarkable level.

Response: In this work, we use glyoxal and methylglyoxal as surrogates to study condensed phase organic chemistry, recognizing that there are many water soluble compounds that will exist in atmospheric waters and will form radicals via reaction with OH. These radicals can react with each other. Thus, we expect that a variety of oligomeric products will form in wet aerosols, not just tartaric acid.
To address this, we added the following after the second paragraph of the conclusion:

It should be pointed out that in this work, we use glyoxal and methylglyoxal as surrogates to study condensed phase organic chemistry, recognizing that there are many water soluble compounds that will exist in atmospheric waters and will form radicals via reaction with OH. In wet aerosols we expect that these radicals will all react with each other. Thus, we expect that a complex array of oligomeric products will form in wet aerosols, not just tartaric and malonic acids.

R4C5) The present manuscript is an important extension of Lim et al (2010). Based on Lim et al (2010) and Table S1 of this work, I guess tartaric acid is probably not speciated in this work (i.e. lumped into C4D which is included in the oligomers formed from glyoxal under concentrated condition).

Response: Yes, in this paper, we do not speciate tartaric acid, which is lumped in C4D.

Supplementary Material:
R4C6) Table S1: Reaction 207 and 208 show the OH reaction of MA and TA, but these two don't come in the abbreviation list. I go back to Lim et al (2010) and found that MA and TA refer to malonic and tartaric acid, respectively. Are they lumped in those dimers (e.g. C3D or C4D)? Anyway please double check the reaction list.

Response: We now deleted Reaction 207 and 208. In the modeling we did, all products are characterized merely as C3D or C4D, etc. These reactions were not used.

***Reference**

He, C., Liu, J., Carlton, A. G., Fan, S., Horowitz, L. W., Levy II, H., and Tao, S., Evaluation of factors controlling global secondary organic aerosol production from cloud processes, Atmos. Chem. Phys., 13, 1913-1926, 2013.