On behalf of all of the co-authors, I would like to thank all of the reviewers for their extremely thoughtful and comprehensive reviews. Unless specifically indicated, table and figure numbers refer to the ACPD verison. The reviewers' comments are reproduced in black, our responses are detailed below in red:

Interactive comment on “Direct photolysis of carbonyl compounds dissolved in cloud and fog droplets” by S. A. Epstein et al.
D. Vione (Referee)
davide.vione@unito.it
Received and published: 18 June 2013
This work is very interesting and gives insight into the carbonyl compounds that could undergo direct photochemical transformation in the atmospheric aqueous phase. The work is well done and the results relevant to atmospheric chemistry, thus acceptance can be recommended after MINOR TO MODERATE REVISION. Recommendations for improvement are given below.

GENERAL AND SPECIFIC REMARKS

1) I perfectly agree with the authors’ strategy to exclude all the compounds the direct photolysis of which cannot possibly be important, to finally come up with a shortlist of the interesting ones. However, when a few compounds (2 to 3) are finally highlighted, I wonder if it really makes sense to make a simulation of how their properties could be.
To my opinion, it would be much better to make experiments, if possible, to have definite answers (including the measurement of actual absorption spectra). The authors should address this observation in the manuscript, and state why did they decide to make calculations instead of experiments at the very last step (which sounds as a less effective strategy).
At the start of this project we did not know which compounds would fall in the "both highly-soluble and quickly-photolyzable" category (Z<1 and Q<1). We were actually quite surprised to discover that there were only a few compounds (two out 92 investigated) that could have significant aqueous photolysis rates. The paper was partially written to detail these findings. Carbonyls encompass a very large and important group of atmospheric organic compounds, much larger than the 92 relatively simple carbonyls included in this study. Therefore, the other main goal of the paper is to present this framework and its resulting chemical intuition with hopes that it can be used for other compounds besides the 92 carbonyl compounds investigated in this paper. We hope that people will carry out such a calculation for any given compound they want to investigate before starting experiments to avoid investigating molecules that do not photolyze in atmospheric waters with appreciable rates.

2) The authors compared direct photolysis with OH reaction as potential transformation processes in the atmospheric aqueous phase. This approach is made reasonable by the fact that chromophoric dissolved organic matter in atmospheric waters is poorly photoactive (see Albinet

et al., Science of the total Environment 2010, 408, 3367-3373), differently for instance from surface waters where other photoinduced processes can be important. A brief comment over this issue in the manuscript would be useful.
We added the following sentence in the text to address this issue: "Photosynthesized production of OH from dissolved organics is a possible source of aqueous OH, but it appears to be a minor contributor in cloud droplets (Albinet et al., 2010a)."

3) Page 10908, top. Note that among the processes that should be taken into account to carry out a proper modelling of the photochemistry in droplets, there are the photoreactions occurring at the air-water interface. Although the interface is thin (but not so thin in smaller droplets), it is a very effective photoreactor and the weight of the interface processes may not be negligible. See for instance Nissenson et al., Atmos. Environ. 2010, 44, 4859-4866. Some hint about interface reactivity should be added in the manuscript.
We added the following statement to the text addressing interfacial photolysis when discussing the glyceraldehyde photolysis yield results: "The average quantum yield may be slightly elevated in cloud droplets as a higher-fraction of molecules are at the air/water interface relative to our bulk measurements. Photolysis occurring at the air/water interface can be more efficient than in bulk due to an incomplete solvent molecule cage (Nissenson et al., 2010)."

4) Page 10915, 1st half. To understand what a Nosé-Hoover thermostat is I had to make a net search. It is true that most article readers will have easy Internet access while reading it, but it is strongly advisable to add a brief explanation.
Use of a Nosé-Hoover thermostat is a widely implemented method in computational chemistry circles. We decided it would be best to not use space to define it, but we did add a brief explanation about how it works and its accuracy. We also added an additional general reference to the text: " A Nosé-Hoover thermostat (Nose, 1984;Hoover, 1985) with a target temperature of 300 K and a characteristic time of 120 fs was to generate a NVT/Boltzmann ensemble of structures (constant number of particles, volume, and temperature). This was achieved by coupling nuclear motion to a fictitious heat bath. For sufficiently long trajectories, this thermostat has been shown to accurately reproduce macroscopic thermodynamic properties (Frenkel and Smit, 2001)."

5) Page 10915, bottom. The FTIR instrument was evidently used, but it was not described in the experimental section. Please add the instrument description where relevant (section 2.1?).
Experiments with the FTIR were only used to identify CO as one of the photolysis products. The instrument description of the FTIR is in the supplementary material.

6) Page 1096. Is there a reason for the choice (at least, I presume it was a choice by the authors) to have the different groups occupy the terminal positions in the chain?
Please add a brief rationale.

We added the following sentence addressing this: “We choose these series of molecules to systematically investigate how different functional groups and molecular chain lengths affect the relevant chemical properties.”

7) Figure 5 is not very clear. Is there something missing? It would be better to explain how were the k_{OH} curves obtained and/or give their equations, and (if possible) to put some reference points related to different compounds. In the present form, the figure looks rather puzzling.
Figure 5 has been modified to make it more understandable. We replaced the letters on the plot with markers and a legend. Arrows pointing to the corresponding k_{OH} for each compound were added to the plot. We also added information to the figure caption to make the plot clearer.

8) Page 10919, lines 25 to the end. The sentence "The lower left … by OH" is rather awkward, please check and rephrase.
This sentence was rephrased to read: “Pyruvic acid [2-oxopropanoic acid] and acetoacetic acid [3-oxobutanoic acid] lie in the lower-left quadrant; for these compounds, aqueous photolysis may be faster than both gaseous photolysis and aqueous oxidation by OH.”

9) Page 10923, top. Please spell out SZA (solar zenith angle, I presume, but the acronym was not defined before).
We added “(SZA)” to the first instance of “solar zenith angle” in the text.

10) Page 10923, line 6. "in Ref. in". Awkward phrase, please check.
Changed.

MINOR ISSUES

a) Page 10907, line 23. Delete "radical" (repetition).
Changed.

b) Page 10914. Please check "an absorption spectra". Is it rather "an absorption spectrum"?
Changed.

c) Page 10918, line 23. "slow" should read "low".
Changed.