Anonymous Referee #1

General comments:

This paper reports a series of measurements of the composition of cloud water and interstitial aerosols collected from Whistler mountains in Canada. In addition to comparing the aerosol vs. aerosolized/dried cloud water composition using aerosol mass spectrometry (AMS), the authors performed aqueous OH oxidation of the samples and characterized the degree of chemical change induced by the aqueous photochemical processing. The experiments described in this paper are novel, and the information is highly relevant to the atmospheric chemistry community. The main message of the paper is that aqueous oxidation of relatively volatile organics by OH produces oxidized organics with sufficiently low volatility to remain in residual particles after the cloud dissipation. The paper is well-written and the figures are appropriate. The following comments and suggestions are aimed at further improving the readability and the logical flow of the paper.

Abstract:

o According to the experiments described in the text, the enhancement of organics was observed only during the initial stages of the aqueous OH oxidation; this was followed by a decline with the amount of organics in aerosol extracts decreasing. The statement made in the abstract does not accurately reflect this point, and may give the false impression that organic mass always increases as a result of photooxidation.

Response:
Yes, we agree with the reviewer's comment and this information has been added to the revised manuscript.

Page 1, line 21-23: "By contrast, enhancement of AMS-measurable organic mass by up to 30% was observed during the initial stage of oxidation of cloud water organics, which was followed by a decline at the later stages of oxidation."

Introduction:

o Page 6021, line 5: it would be helpful to insert a citation to support the "usually hygroscopic" nature of atmospheric aerosols. The authors' group have a conducted a number of comprehensive studies on this topic in the past, so they should be in a good position to point the readers to appropriate references.

Response:
We appreciate the reviewer's suggestion. Because there can be many different types of hydrophobic aerosol particles in the atmosphere, it is more appropriate to just say atmospheric aerosol can absorb water under humid environments and activate to form cloud droplets under supersaturation. We decided to revise this sentence in the Introduction as follows.

Page 2, line 15-16: "Atmospheric aerosols can absorb water to become aqueous particles under humid conditions and activate to form cloud droplets upon water supersaturation."

o Page 6022, line 18: the authors assert that "cloud water" in this work actually refers to fog water and that they are expected to be similar for a given elevation. This is not entirely accurate. There have been numerous measurements of cloud vs. fog water composition for a given site, and the conclusion is that the composition is not necessarily similar. Fog waters tend to contain more dissolved material. For example, concentration of dissolved inorganic compounds in fog water can be an order of magnitude greater (Fisak et al. 2002), possibly due to smaller drop size. Fog water has a wider range of pH values compared to cloud water (Collett et al. 2002). Finally, the OH concentration in clouds is expected to be higher than that in fogs (Ervens et al. 2011), especially in the interstitial areas of clouds where OH may be enhanced (Maudlin et al. 1997). The authors should elaborate on the justification of their assumption.

Response:
We agree with the reviewer that differences between fog and cloud composition can occur at a specific site. Those differences are largely routed in the meteorological conditions and the resulting type of cloud/fog forming with differences in LWC, drop sizes and pollution levels in the respective air mass (local stable air for fog vs. transport for cloud). However, in our study, the sampled water droplets likely originated from cloud in the valley that approached our mountain site. In this case, we assume that the cloud droplet composition would roughly remain unchanged. We have revised the statement as follows:

Page 3, line 30-32: "The term "cloud water" used in this paper refers to the cloud droplets approaching our forest mountain sites and we assume it has similar composition to the cloud, at the same elevations, that did not impact the mountain."

o For the completeness of the introduction, a few other studies should be mentioned. Bateman et al. (2011) focused on direct photolysis of dissolved organics, but they probably could not have avoided oxidation by OH. They observed fragmentation of dissolved organics accompanied by an increase in the O/C ratio – this is similar to the observations of the authors for the later stages of the photooxidation. Chang and Thompson (2011) found that OH oxidation of phenolic compounds in water produced complex molecules that absorbed light. There might be other relevant papers this reviewer is not aware of, I would check a recent comprehensive review on this topic by Ervens et al. (2011). References to these and other papers are supplied at the end of this review.

Response:
We thank the reviewer for his/her suggestion. Some additional references have been added and briefly discussed in the introduction.

Page 2, line 27 to Page 3, line 1: "Bateman et al. (2011) found that photolysis of aqueous extracts of SOA derived from dark ozonolysis of d-limonene might generate OH radicals and the combined results of photo-dissociation and OH oxidation led to fragmentation of dissolved SOA materials with increasing O/C ratio. Chang and Thompson (2011) observed formation of visible light absorbing materials with similar IR spectral features to atmospheric humic-like substances (HULIS) via aqueous OH oxidation of phenolic compounds."

Page 2, line 20-21: "Furthermore, Kaul et al. (2011) recently reported that aqueous-phase processing likely enhanced the production of SOA in foggy periods in Kanpur, India."

Experimental:

o The aqueous OH oxidation experiments are performed with irradiation of H2O2. However, this irradiation happens in the presence of sulfate, nitrate, and ammonium in the cloud water and aerosol extracts. Can the authors estimate the degree of possible interference from these ions? Sulfate UV photolysis can produce radicals that generate organosulfates in the presence of organics (Nozierre et al. 2010). Nitrate is also a photolytic source of OH and other reactive species in water. Ammonium ion can be oxidized by OH to form hydroxylamine which is quite reactive. One way to assess the contribution from these ions is kinetics modeling; another way is conducting control studies with inorganic ions intentionally added to (or removed from) the solution. The reviewer's guess is that the contribution of other OH precursors is negligible because of the high concentration of H2O2.

Response:
We didn't conduct such control experiments to determine the role of inorganic ions in producing OH radicals and other reactive species. As per reviewer's comment, we assume that the contribution of other OH precursors is negligible because of the high initial concentration of H_2O_2. Based on the control experiments done in our previous works (Lee et al. 2011b), we did not observe glyoxal decay during photolysis in the presence of ammonium sulfate without addition of H_2O_2. The information below has been added to the revised manuscript to clarify this concern.

Page 6, line 6-8: "Based on the control experiments performed in our laboratory, other potential photolytic sources of OH radicals were assumed to be negligible due to the high initial concentration of H_2O_2."

o Please state the actual mass resolving power of the HR-AMS achieved in these measurements.

Response:
This information has been added to the revised manuscript.

Page 4, line 27-28: "The mass resolving power of the HR-AMS in our measurement was about 4000."

o The authors state that only one sample is analyzed from the peak site, so the discussion is focused on RN site. But data from the peak site are included in figures (e.g. Fig 1, 2, 3…) and the discussion includes this site. Also it is not clear how many samples from the RN site are included in the analysis.

Response:
That sentence has been deleted and the numbers of cloud water samples from RN and the peak site have been added to the manuscript.

Page 5, line 3-4: “Six cloud water samples were analyzed from RN and only one was from the peak site”

o The experimental section could benefit from clarifying sentences to introduce sampling techniques and explain why they are used. For example, it would help to briefly explain the benefit of sampling techniques like CVI.

Response:
We appreciate the reviewer’s suggestion. A few sentences have been added to briefly introduce the reasons behind our sampling approach.

Page 4, line 17-20: “To better understand aqueous oxidative processing of WSOC in SOA and cloud organics, aerosol filter samples and cloud water were collected at Whistler, British Columbia, during summer 2010 for chemical characterization and performing aqueous oxidation experiments (see details in Section 2.2).”

Page 5, line 6-8: “The CVI allows real-time measurement of cloud residuals using different online aerosol particle instruments by switching normal ambient inlets to the CVI output during cloud periods.”

o The high 70 mM concentration of H2O2 may pose a problem because hydrogen peroxide has oxidative potential on its own. This concentration will be further increased during evaporation, and result in chemistry that the authors attribute to OH chemistry. Have the authors performed a control measurement wherein the solution with added H2O2 was aerosolized without prior photolysis?

Response:
We agree that it is ideal to atomize the samples without photolysis for evaluating the oxidative potential of H_2O_2. However, we didn't performed this control experiment due to the limited volume of aerosol and cloud samples. Instead, we monitored the composition of atomized aerosol filter extracts and cloud water after the photoxidation for about 1 hour (without UV). We expected that the oxidized solution should still contain high concentration of H_2O_2. We observed that the AMS organic composition remained roughly constant (data not shown). We now add a sentence to the paper expressing this assumption.

Page 6, line 8-11: “We also assumed that the oxidative potential of H_2O_2 did not significantly affect the organic composition because the organic spectra of the oxidized solution remained roughly constant in a dark condition for about 1 h.”

o An estimation of the steady state OH radical concentration during irradiation, and an estimation of the total organic concentration achieved from extraction in 30 mL of water would be useful.

Response:
Yes, we agree with the reviewer's comment. The information has been added in the revised manuscript. The total organic concentrations were estimated using the organic mass loadings measured by the HR-AMS. Also, we estimate the photolysis rate constant of H_2O_2 in our experimental setup and use a simple model of hydrogen peroxide photolysis to determine the steady state OH concentration (see supplementary materials).

Page 5, line 20-23: "The estimated total organic concentration of aerosol filter extracts was on the order of 10^{-5} M based on the organic aerosol mass loading measured by the HR-AMS, assuming their average molecular weight of about 200 g/mole and that all the material was water soluble."

Page 6, line 4-6: "We estimated the steady state OH radical concentration to be about 10^{-11} M using a simple chemical model with a calculated H_2O_2 photolysis rate constant of 6.9×10^{-4} s^{-1} (see details in supplementary material)."

o Acronym "CToF" should be spelled out the first time it is used.

Response:
The term "CToF-AMS" has been defined in the revised manuscript. The AMS community conventionally uses CToF to represent the Compact Time of flight-AMS with unit mass resolution.

Page 5, line 12-14: "The sampled droplets were dried in the CVI inlet and the non-refractory composition of cloud residuals was measured by a compact time of flight-AMS with unit mass resolution (CToF-AMS)."

o As samples with low organic loadings were excluded, it would be a good idea to mention explicitly how many samples of each kind were used for analyses.

Response:
That's a good idea. This information has been added to the revised manuscript.

Page 6, line 22-23: "Eleven aerosol filter extracts with organic loadings at least 5 times higher than the average of the background organic level were included in the analysis."

Results/Discussion:

o Page 6026, line 14: volatile aldehydes account for a significant portion of the WSOC. However, upon droplet evaporation, they need not volatilize. De Haan et al. (2009) showed that methylglyoxal when evaporated in the presence of ammonium (which is present in ambient samples) results in low volatility nitrogen compounds. Nguyen et al. (2012) observed similar behavior during evaporation of solutions of more complicated aldehydes. To what extent such reactions occurring during atomization of solutions may contribute to the authors' observations? Perhaps they can be ruled out with appropriate control measurements (e.g., atomization without any irradiation and without H_2O_2 addition)?

Response:
We entirely agree that not all of these volatile species would evaporate to gas phase during droplet evaporation due to the reactions described by the reviewer. However, we believe that the observed organic mass loading accounts for the effects from such reactions throughout the oxidation process.

Page 8, line 12-17: "Although formation of less volatile and stable organic salts (e.g., dicarboxylates) and oligomers from these volatile species during droplet evaporation were possible (see ammonium ion as an example in the next paragraph), the observed organic mass accounts for the potential effects from such reactions throughout the oxidation process. It is believed that a portion of volatile species may not be retained in the evaporated droplets and hence cannot be fully detected by the AMS."

o The authors discuss nitrate and sulfate extensively throughout the paper and use quantities like nitrate to sulfate ratios to characterize filter samples and cloud water. However, they do not discuss ammonium ion. Ammonium ion concentration can be quite high in cloud and fog water, as also demonstrated by Figure 1 of this work. Ammonium ion has been shown to be catalytic in the formation of oligomers from glyoxal (Noziere et al. 2009) and participate in complex chemistry during droplet evaporation (De Haan et al. 2011, Nguyen et al. 2012), so it may be interesting to expand upon this discussion.

Response:
We thank for the reviewer's comment. Some discussion on ammonium ion and related chemistry has been added in the revised version as below.

Page 8, line 29 to Page 9, line 4: "Ammonium ion contributed about 13% and 8% of aerosol filter extracts and cloud water constituents as shown in Figure 1c. Previous laboratory studies have shown that ammonium ion can catalyze the formation of oligomers from glyoxal (Noziere et al. 2009) and participate in complex chemistry to produce organic-nitrogen compounds in the particle phase after droplet evaporation (De Haan et al. 2011; Nguyen et al. 2012). A large fraction of ammonium ion may thus reduce the overall volatility of organics in the evaporated droplets. Unfortunately, our CToF-AMS measurement with unit mass resolution cannot discriminate between the fragments of organic carbon and organic nitrogen that form as a result."

o The reviewer is not an expert in the analysis of AMS data, and as a result, has some reservations about the ability of AMS to achieve the degree of chemical specificity implied by the discussion in paper. For example, it is not obvious that information on functional groups can be unequivocally derived from generic fragments. The fragment f29 was linked to carbonyls on page 6028, line 3 and fragment f44 was linked to carboxylic acids. These assumptions might work well for relatively simple mixtures (e.g., SOA prepared from a single precursor), but a number of complicated interferences are expected for ambient SOA. For example, m/z 29 can be either CHO+ or C2H5+, m/z 44 can be CO2+ or C2H4O+, and m/z 43 can be also be C3H7+. These interferences must have been discussed extensively in the AMS literature. It would help to cite appropriate examples from the prior work to help the readers better understand the level of confidence in the attribution of peaks to different functional groups.

Response:
We agree that m/z 29, 43, and 44 can arise from many different fragments as described by the reviewer. In addition to providing a few related citations to support our fragment assignment, we also analyzed the HR-AMS organic spectra of organic components in Whistler. The HR-AMS confirms that m/z 29, 43, and 44 are mainly due to the presence of CHO^+, $C_2H_3O^+$, and CO_2^+, respectively. The sentences below have been revised/added.

Page 9, line 10-14: "While the organic fragment at m/z 44 (CO_2^+) is primarily due to the fragmentation of organic acids (e.g., Duplissy et al. 2011), m/z 43 of oxygenated organic aerosol mostly originates from $C_2H_3O^+$ (e.g., Ng et al. 2010), indicating the presence of less oxygenated functional characteristics. The above organic fragment assignment is also supported by our online HR-AMS measurement."

Page 9, line 26-28: "Our HR-AMS data suggests that CHO^+ fragment was the main contributor to organic fragments at m/z 29, likely due to the increased concentration of carbonyl functional groups."

o Page 6027, line 13 (and also abstract). Because the mass spectra are similar you conclude that there must be a common biogenic source of particles. Please insert a reference to previous AMS studies confirming that biogenic origin can be deduced unequivocally from the AMS spectra.

Response:
We agree that this is not an appropriate statement and has been deleted in the revised manuscript.

o Page 6020, line 4-5. It would be useful to compare the O/C ratios in this work with other lab and ambient SOA studies.

Response:
This information has been added to the revised version as shown below.

Page 10, line 25-32: "In general, the O/C ratios of our aerosol and cloud water samples are higher than those of chamber SOA produced by gas-phase oxidation chemistry as summarized in some previous studies (e.g., Aiken et al. 2008; Chhabra et al. 2010; Ng et al. 2011) but much lower than those obtained from aqueous SOA oxidation and glyoxal uptake experiments (Lee et al. 2011a; Lambe et al. 2011; Chhabra et al. 2010). Note that the O/C ratios of aerosol filter samples collected within the biogenic period are comparable to those measured from the Amazon Basin (0.42-0.49) (Chen et al. 2009), which is also strongly influenced by biogenic sources."

o Page 6029, line 27-28: The authors state that the differences from OH oxidation in filter and cloud samples are almost identical but when the spectra are normalized with respect to sulfate there is a only "minor production of m/z 44" or even a small reduction in the filter sample and not the cloud samples. This reviewer thinks that this point is confusing and can be better explained.

Response:
The spectral difference obtained by normalizing the AMS spectra to total AMS organic mass only represents the relative fractional changes in organic composition. In contrast, assuming sulfate mass is roughly constant during oxidation, the organic mass spectra normalized by sulfate mass can be used to determine the absolute changes in individual organic fragments. Below is a revised version of the explanation.

Page 11, line 20-22: "Note that the spectra used for this subtraction were normalized to their corresponding total AMS organic mass so that the spectral difference only represents the fractional changes in organic composition."

Page 11, line 31 to Page 12, line 2: "It is important to note that the spectral differences obtained by comparing spectra normalized to total AMS organics do not reflect the absolute changes (i.e. net mass production or reduction) in individual organic fragments, which should be independent of the total amount of organic mass remaining in the evaporated droplets."

o Page 6030, line 2-3: Please explain what f18 and f28 are. They must correspond to H2O+ and CO+, respectively. H2O+ and CO+ are not specific fragments, so this reviewer has reservations about the statement that they are "indicating organic acids are predominant condensed-phase products…".

Response:
We agree that H_2O^+ and CO^+ are the major organic fragments at m/z 18 and m/z 28, respectively and they can be due to the presence of other species such as water for m/z 18 and CO and N_2 for m/z 28. However, organic acids can also give H_2O^+ and CO^+ fragments in the AMS organic spectrum and we cannot differentiate the organic fragments of m/z 18 and m/z 28 from other species. So, the AMS community has developed a fragmentation table to estimate the organic intensities at m/z 18 and m/z 28 from the organic intensity of m/z 44 (CO_2^+), which is predominately due to the presence of organic acids. Therefore, the qualitative changes of m/z 18, 28 and 44 are the same in our AMS data. In the original manuscript, we have mentioned this information as follows:

Page 11, line 27-30: "Note that the organic intensities at m/z 28 (CO^+) and m/z 18 (H_2O^+) are directly estimated from m/z 44 signals using the standard fragmentation table in Squirrel so that their changes cannot provide extra information for identifying the functional characteristics of oxidation products."

o Page 6030, line 20-21: The authors state that the organic composition of cloud organics is different than that of WSOC in aerosols, which contradicts previous points that "in general, the aerosol and cloud organic spectra were very similar, indicating that the cloud water organics likely originated from secondary organic aerosol." Please explain.

Response:
The second argument is not appropriate and has been deleted in the revised manuscript.

o Two assumptions are used in this paper: (1) high O/C compounds have in general lower volatility (e.g. page 6029, line 11); (2) high O/C is indicative of significant fragmentation, which leads to mass loss through the production of high volatility products (e.g. page 6032, line 19 and page 6036, line 14). These two seemingly contradictory ideas need to be reconciled. Strictly speaking, O/C is not well correlated with volatility. Many other factors such as molecular size and degree of chemical interaction with other organics contribute to volatility.

Response:
We agree with the reviewer that the O/C ratio is not necessarily correlated with volatility. On page 6029, we just wanted to point out that the f44 (or O/C) of oxidized filter extracts and cloud water fall in the LV-OOA region of f44-f43 space. In the later part of the manuscript, we believe that fragmentation and functionalization should occur continuously throughout the oxidation. Therefore, oxidation can increase the O/C ratio of the particle-phase organics and induce particle mass loss at the same time. We read through the whole manuscript and we now try to avoid misunderstanding related to these issues.

o Page 6033, line 26: This statement can be made in a clearer way. The authors are probably trying to say that when one atomizes pinonic acid solution some of the compound will volatilize. But when one oxidizes it in solution, and then atomizes the solution, more mass will be in the particle phase due to the less volatile products and therefore the particles will appear to have accumulated mass.

Response:
Yes, this was our intention. We revised the related sentences to make it clear as shown below.

Page 11, line 3-10: "Since the organic mass increased by a factor of 2.2 at its maximum, this cannot be realistically achieved by functionalization reactions of the AMS measured *cis*-pinonic acid alone (e.g., this would require addition of about 10 OH groups, without fragmentation occurring). However, due to the fact that a large fraction of atomized *cis*-pinonic acid escapes to the gas phase after droplet evaporation and aqueous OH oxidation of *cis*-pinonic acid can produce less volatile first- and/or second-generation oxidation products (Jimenez et al. 2009), it is expected that more organic mass can accumulate in the particle phase upon evaporation."

o It is interesting that the authors found a decrease in organic mass from aqueous OH oxidation of glyoxal and methylglyoxal. This seems to be in contrast to other reports, for example from Barbara Turpin group, where oligomer formation was observed for this same system. What can the origin of the discrepancy?

Response:
Concerning mass loss, it is hard to compare our results to those of Turpin and co-workers given that the latter work is performed in the bulk whereas ours includes a volatilization step to form aerosol. The origin of the potential discrepancy with regards to oligomer formation has been discussed in our previous publications (Lee et al., 2011b; Zhao et al., 2012) and thus we decide to not go through all the details in this paper. Also, to avoid confusion and to make the point more clearly, we have removed the results and discussion pertaining to glyoxal and methylglyoxal from the paper, focussing instead on the more easily interpreted laboratory results for cis-pinonic acid and the ambient measurements instead.

Figures:

o Figure 2: Is there a reason the cloud-influenced sample collected on July 12 was not included in Figure 2? The authors should insert a brief explanation.

Response:
A brief reason has been added to the figure caption of Figure 2 as follow:

"The cloud water collected on July 12 at RN has much higher intensity at m/z 29 compared to all other cloud water samples and thus it is not included in the average organic spectrum of cloud water (d)."

o Figure 4: The difference spectra look very similar for filter and cloud samples when they are normalized with respect to the organic mass. However, when normalization is done with respect to sulfate, the difference spectra are no longer similar, for example large peaks at m/z 28 and m/z 44 essentially disappear from panel c but not from panel f. This approach is a little confusing; it would be a good idea to explain the normalization procedure in the text and expand on why it is beneficial.

Response:
This comment is similar to one of the previous comments in the section of "Results and discussion". The revised sentence is shown below.

Page 11, line 20-22: "Note that the spectra used for this subtraction were normalized to their corresponding total AMS organic mass so that the spectral difference only represents the fractional changes in organic composition."

Page 11, line 31 to Page 12, line 2: "It is important to note that the spectral differences obtained by comparing spectra normalized to total AMS organics do not reflect the absolute changes (i.e.

net mass production or reduction) in individual organic fragments, which should be independent of the total amount of organic mass remaining in the evaporated droplets."

Technical corrections:

• In general, the use of "WSOC compounds" is redundant. Please use just "WSOC," e.g., on page 6026, line 3 you can write "WSOC rapidly increased…"

Response:
All "WSOC compounds" have been changed to "WSOC".

• The term "peak site" that is used in figure captions and further in the text needs to be defined first in Section 2.1.

Response:
The "peak site" has been defined in the text as follow:

Page 4, line 28-31: "The cloud water samples were collected by automated versions of the Caltech Active Stand Cloudwater Collector (CASCC) (Hutchings et al. 2009; Macdonald et al. 2012) at RN and also at another site at the Whistler mountain peak (peak site), located at an elevation of 2182 m-asl."

• Page 6024, line 10: change "ml" to "mL"

Response:
Done

• Page 6024, line 10: change "18 MΩ water" to "purified water (18 MΩ cm)"

Response:
Done

• Page 6024, line 17: change "colume" to "volume"

Response:
Done

• Page 6029, line 9: delete "as described"

Response:
Done

- Page 6034, line 4 and Page 6036, line 7: correct the spelling of "pinonaldehyde"

Response:
Done

- Figure 2: the labeling scheme is confusing as the letters a-h are not arranged in order by column nor by row. Please fix.

Response:
Thanks for the suggestion. However, we would like to keep this arrangement so that the reader can make a side-by-side spectral comparison easily. The top left and right columns are the organic spectra of aerosol filter extracts and cloud water, respectively. The bottom spectra are the online AMS mass spectra of aerosol sampled from the normal inlet and cloud residual sampled from the CVI.

References:

Bateman et al., Photolytic processing of secondary organic aerosols dissolved in cloud droplets. *Phys. Chem. Chem. Phys.* **2011,** *13* (26), 12199-12212.

Chang, J. L.; Thompson, J. E., Characterization of colored products formed during irradiation of aqueous solutions containing H2O2 and phenolic compounds. Atm. Environ. 2010, 44 (4), 541-551.

Collett et al., The chemical composition of fogs and intercepted clouds in the United States. *Atmos. Res.* **2002,** *64* (1), 29-40.

De Haan et al., Secondary organic aerosol formation by self-reactions of methylglyoxal and glyoxal in evaporating droplets. *Environ. Sci. Technol.* **2009,** *43* (21), 8184-8190.

Ervens et al., Secondary organic aerosol formation in cloud droplets and aqueous particles (aqSOA): a review of laboratory, field and model studies. *Atm. Chem. Phys.* **2011,** *11* (21), 11069-11102.

Fisak et al., Pollutant concentrations in fog and low cloud water at selected sites of the Czech Republic. *Atmos. Res.* **2002,** *64* (1-4), 75-87.

Mauldin et al., New insights on OH: Measurements around and in clouds GRL. **1997,** *24,* 3033–3036.

Nguyen et al., Formation of nitrogen- and sulfur-containing light-absorbing compounds accelerated by evaporation of water from secondary organic aerosols. *J. Geophys. Res. D* **2012,** *117*, D01207, doi:10.1029/2011JD016944.

Noziere et al., Radical-initiated formation of organosulfates and surfactants in atmospheric aerosols. *Geophys. Res. Lett.* **2010,** *37* (5), L05806, doi:10.1029/2009GL041683.

Noziere et al., Products and kinetics of the liquid-phase reaction of glyoxal catalyzed by ammonium ions (NH4+). *J. Phys. Chem. A* **2009,** *113* (1), 231-237.