Author responses to comments from RC2 are given below in blue font. The original referee comments are provided in *black italicized font.*

This manuscript presents molecular-level analyses of fresh versus aged fog and samples influenced by biomass burning. The authors aim to explore the potential importance of aqueous phase processing on alteration of organic matter chemical compositions. The authors reported that aged aerosols and fresh fog samples show similarity in composition, indicating the possibility of aged aerosols that served as fog nuclei. One of my major concerns for this manuscript is that the authors attributed the CHON and CHOS compounds exclusively to organonitrates and organosulfates based on FT–ICR MS analyses, but this is not supported by NMR spectra! This seems to be a major finding but it was not discussed in great detail. It looks to me that other types of organic nitrogen and organosulfur compounds may contribute to formation of detected CHON and CHOS that need further investigations. In addition, nitro groups (R-NO2) and nitrooxy groups (R-ONO2) are different. They have distinct formation processes and physiochemical properties as well (e.g., lifetime against hydrolysis). The authors need to be clear when discussing their findings in context of literature.

For the results and discussion, the current form of manuscript is a bit lengthy and repetitive when reporting the FT-ICR MS data. A more concise presentation will greatly improve the readers' reading experience. Also, reactions in aerosol liquid water content and in fog should be discussed separately. Based on the results presented (with only 1 sample in each category), the aqueous processing does alter the chemical compositions of organic matter, but the pathways are rather inconclusive.

Overall, this is still a nice case study that provides useful information. Below I provide a few more specific comments for the authors' consideration and clarification.

We thank the referee for their helpful comments. As suggested, we made several edits to improve the readability and reduce redundancies in the manuscript. We also clarified the comparisons of fresh fog to aged fog compositions and fresh aerosol to aged aerosol compositions (Sections 3.3.3 and 3.3.4 respectively). Another comparison between the two types of samples was made because of the interesting observation of similar compositions between the aged aerosol and the fresh fog. It is plausible that some reactions in aerosol liquid water may also occur during fog activation as droplets begin to grow, helping to explain the similar compositions between these two samples of different types.

The results of the ^{1}H-NMR and FT-ICR MS measurements are only apparently in contradiction. While the ^{1}H-NMR analyses were performed on bulk aqueous samples, the electrospray ionization FT-ICR MS analysis requires the removal of inorganic ions present in the bulk aqueous extracts. Reversed phase SPE cartridges were used to isolate the water-soluble organic aerosol components. However, some losses of low molecular weight and ionic water-soluble organic compounds are expected. This may have included the low-molecular weight alkyl amines observed by ^{1}H-NMR analysis. Furthermore, the negative ion electrospray favors the detection of acidic compounds and thus is not ideal for the detection of reduced nitrogen or sulfur compounds. A statement was added to the end of section 2.4 (p. 6 line 19-22) to clarify this: "The resulting data set represents the SPE-recovered higher molecular weight water soluble organic aerosol and is expected to predominantly contain acidic compounds due to the negative ion ESI analytical bias. The observed molecular compositions represent the oxidized fraction of the atmospheric samples thus, useful insights can be made with these limitations in mind."

The concern as to whether the ^{1}H-NMR data actually support the hypothesis of the occurrence of organic nitrates and organic sulfates in these samples can be reassured by the clear signals in the spectral regions where aliphatic hydrogen atoms in alpha position to such functional groups are expected to occur (specifically between 4 and 5 ppm chemical shift, as described by Hsieh et al. (2014)). However, it is not as clear as to whether the ^{1}H-NMR analysis provides the same information on the relative abundance of organic nitrates and organic sulfates between samples as derived from the FT-ICR MS datasets, as the same spectral region can host many other possible functionalities (e.g., esters, peroxides, hydroxy-carboxylic acids). In summary, ^{1}H-NMR spectroscopy was not specific enough to trace the abundance of organic nitrates and sulfates in these samples in a useful manner for comparison with the FT-ICR MS data. The N and S containing molecular formulas observed with FT-ICR were attributed to organonitrates, organosulfates and nitrooxy-organosulfates, based on a previous study using negative mode electrospray ionization and MS/MS analysis for functional group determination of water-soluble atmospheric organic matter (LeClair et al., 2012), and the observed O:N and O:S ratios, which we clarified in the text.

Specific Comments:

1) Page 4, lines 7-9: The aerosol filter extracts were filtered with 0.45 um PTFE membrane, while the fog water was filtered through 47 mm quartz fiber filters. What is the pore size of 47 mm quartz fiber filters? Why did the authors use two different filtering methods here? Since the FT–ICR MS analysis is very sensitive, potential artifacts (even trace amounts) during sample preparation should be avoided.

These method differences result from the laboratory methods for the different analysis techniques. Aerosol filter extractions were performed at Michigan Tech (MTU) for FT-ICR analysis (Section 2.4) and at the Institute of Atmospheric Sciences and Climate (CNR-ISAC) for total carbon, ^{1}H-NMR analysis and HR-ToF-AMS analysis for fog samples (Section 2.1). Fog and aerosol samples prepared at MTU for FT-ICR analysis were prepared consistently with 25 mm quartz fiber filters, as described in section 2.4. We are not aware of a uniform pore size for quartz fiber filters, due to the nature of the material. Sample blanks were used to correct for artifacts that may have been introduced by the quartz fiber filter due to the sensitivity of FT-ICR MS. Additional statements were added to section 2.1 (p. 4 line 18-25): "The aerosol filters were extracted with deionized ultra-pure water (Milli-Q) in an ultrasonic bath for 1 h. The water extract was filtered with a 0.45 μm PTFE membrane in order to remove suspended particles. Fog water was filtered through 47 mm quartz fiber filters within a few hours of collection and conductivity and pH measurements were taken ... Aliquots of both aerosol water extracts and fog water prepared in this way were used to determine the total organic carbon content … and water soluble organic carbon (WSOC) concentration, (Rinaldi et al., 2007) as well as for ^{1}H-NMR analysis and HR-ToF-AMS analysis of fog samples described below (HR-ToF-AMS data for aerosol samples was collected in real time)." and 2.4 (p. 5 line 22-24): "Fog samples were later re-filtered using a 25 mm quartz filter before SPE. A portion of the aerosol filter samples were extracted with ultrapure water using sonication and the extracts were then filtered using a 25 mm quartz filter to remove insoluble materials…" to clarify these differences in methods.

2) Page 7, line 6: Does "SOA-like" mean oxygenated/or functionalized/or fragmented? It is not clear here.

We have revised the text to more accurately convey that the fog compositions were more oxidized, and thus more similar to SOA than the aerosols, according to the simplified source attribution scheme of Fig. 1. The text was changed to reflect this in the abstract (p.1 line 25-27): "Fog compositions were more

oxidized and "SOA-like" than aerosols as indicated by their NMR measured acyl vs alkoxyl ratios and the observed molecular formula similarity between the aged aerosol and fresh fog, implying that fog nuclei must be somewhat aged." In the ^{1}H-NMR discussion (p. 8 line 3-5): "So, according to the simple source-attribution scheme based on the major ^{1}H-NMR functionalities presented here, the fog compositions were more oxidized and "SOA-like" than aerosols." And in the conclusions (p. 14 line 18-20): "Overall, the fog composition was generally more oxidized and "SOA-like" than the aerosol, where the fresh fog composition was similar to the aged aerosol composition in both the ^{1}H-NMR analysis and the molecular formula trends."

3) Page 10, lines 25-35: Since aged aerosols could act as fog nuclei, scavenging of organosulfates resided in aged aerosols into fog might have contributed to the observed organosulfates in fresh fog water. Based on the data presented, I don't really see direct evidence here showing that aqueous processing leads to CHOS production. Similarly, on Page 13 lines 12-17: the authors concluded that the current data provide strong evidence of aqueous processing that dominates the production of S-containing organic matter. I would tone down this statement.

We have revised this paragraph to include statements on nucleation scavenging. The revised paragraph (p. 12 line 3-13) now reads: "The unique molecular formulas found in the fresh fog (SPC0106F) were mostly of the $O_{5-13}S$ and $NO_{7-12}S$ subclasses. Organosulfates are known products of aqueous secondary processes, (Darer et al., 2011; Ervens et al., 2011; McNeill, 2015; Schindelka et al., 2013) and nucleation scavenging from the preceding fog nuclei composition likely plays a significant role as well (Darer et al., 2011; Gilardoni et al., 2014; Herckes et al., 2007; Hu et al., 2011). The aromatic organosulfates and nitooxy-organosulfates observed in fresh biomass burning aerosol (Staudt et al., 2014) were not observed here. In general, organosulfates are the products of aqueous-phase SOA reactions which are expected to be enhanced at acidic pH (Ervens et al., 2011; McNeill et al., 2012; Noziere et al., 2010). Because the pH of SPC0106F was only slightly acidic at 5.81, we propose that the formation of these organosulfates may have been promoted by low LWC, and thus relatively high solute concentrations, during the activation of the fog droplets or possibly in the fully formed fog droplets. Organosulfates may also efficiently nucleate droplets, leading to their eventual presence in the fog samples."

The conclusion statement (now p. 14 line 21-23) was revised to the following: "CHOS and CHNOS formulas were detected with high frequencies in samples with high water content during collection (all samples except BO0204N). This supports an enhanced production of S-containing SOA species via reactions in the aqueous phase."

4) Page 12, line 32: "hygroscopic" is a better term to describe aged/oxygenated organics that contribute to droplet formation.

This sentence (now p. 14 line 4-5) now reads: "Hygroscopic species are expected to enhance droplet formation, indicating that organics acting as fog nuclei must be somewhat aged."

5) Page 13, lines 8-9: it is confusing when the authors stated "some evidence of dimerization" here. This was not presented in "results and discussion" but suddenly mentioned in summary.

We have revised the text to remove all references to dimerization.

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