acp-2017-628: Response to Anonymous Referee #1

We would like to thank the referee for their comments and suggestions. Below, the original comment provided by the referee is shown in gray. Our response, with revisions noted, are in bold font.

General comments
This study reports the molecular composition of cloud water collected under urban, biogenic, and biomass burning influences revealing key differences among dissolved organic compounds in each condition. The results are clearly presented, concise, and contain sufficient detail. I have only minor suggestions for strengthening this work. Upon revision, I would recommend that the editor accept this manuscript.

Specific comments

Abstract: The abstract would benefit from a concluding sentence in the abstract that draws the reader's attention to the most significant findings of this work, or to the larger takeaway from the measurements.

> **The following sentence was added to the end of the abstract: "Overall, cloud water molecular composition depended on air mass source influence and reflected aqueous-phase reactions involving biogenic, urban, and biomass burning precursors."**

Pg 4 Line 4: What were TOC concentrations before the concentration step? Since there was a concentration step, it's not clear what giving these values tells the reader, except that the concentrated samples were probably above LOD for most techniques. Even an estimate of the increase (doubled, 10-fold) would be useful here.

> **The cloud water TOC concentrations prior to the concentration step are shown in Table 1 of the main text, as now noted in the methods section, as requested.**

Line 11: Why negative ion mode? I'd think that you would miss any ammonium-based oligomers
including imines. Negative ion mode captures acids well, but not reduced nitrogen compounds. Could the authors at least comment on this point here?

> **Negative ion mode was used to target CHO compounds, which are more easily ionized in negative ion mode and are the primary component of cloud water. We now note on P4 L15 that we are targeting oxidized organic compounds by creating negative ions. While examining both positive and negative ion mode would have been useful, limited access to instrumentation required only one mode to be chosen for these samples.**

Pg 8 Lines 16-17: I'm not sure the data support the claim that acidity (as opposed to BB-specific VOCs, their concentration, or pyrolysis itself) is responsible for the increase in oligomeric material. It's certainly possible, but it's also possible that the BB cloud water simply had lower pH AND more oligomeric compounds. Higher concentrations of the same compounds could facilitate increased oligomerization too, right? Please revise to avoid overstating the connection.

This is an excellent point. We have revised this sentence (moved to P8 L 30-34) as follows: "Notably, there appear to be a greater diversity of oligomeric compounds present in the wildfire samples (Fig. 2, Fig. S2), which is likely a combination of the identities of the specific organic compounds present at high concentrations in the smoke, as well as the potential role of acidity (as observed through cloud water acidity), resulting in the production of the observed oligomeric species."

Technical corrections Pg 1 Line 26: "with a focus" Line 31: "was positively correlated"

The suggested changes were made.

Pg 2 Line 4: "water soluble organic gases" so as to distinguish SO2 from the organic compounds alluded to here Line 6: "depending on their solubility" for concision

The suggested changes were made.

Pg 4 Line 5: "including some organosulfate" right? Not all? I believe organosulfates are reported here.

This is correct. The suggested change was made.

Pg 5 Line 6: "compose" not comprise

The suggested change was made.

Pg 9 Line 12: "succinic acid based"

The suggested change was made.