Response to Reviewer #1

General comments:

In this paper the authors report further on a framework they have been developing to extend the utility of functional group analysis of organic aerosol to obtain more accurate information on possible missing chemical components, organic mass/organic carbon ratio, elemental composition (O/C ratio), and carbon oxidation state. A thorough discussion of the conceptual and analytical aspects of the methods are presented and discussed, and software is made available that can be downloaded by others wishing to use these methods. The methods were applied to a data set of SOA composition predicted for a-pinene photooxidation using the MCM coupled with gas-particle partitioning, and for which FTIR functional group data were also available. A variety of useful results are extracted from the analysis. The paper is technically very dense and beyond my ability to fully understand without a much greater investment of time and effort, and so I am unable to evaluate many of the details that are presented. Nonetheless, the approach seems reasonable to me.

Overall, I think this is likely to be a useful paper as more people begin to adapt these methods for analysis of functional group data. Not many group conduct functional group analysis, but there are reasons to think this could increase in the future because of its unique value compared with molecular and AMS analysis. Im very pleased to see that the authors have made the software needed to conduct the analysis freely available. I recommend the paper be published after these very minor comments are addressed.

We thank the reviewer for this encouraging assessment.

Specific comments:

1. Page 5, line 7: I have never heard it said that FTIR measures the total abundance of bonds. The spectrum depends on vibrations and bending of molecules, but I dont see how this translates directly into bond abundance. This should be clarified.

   The absorbances associated with stretching and bending modes of molecular bonds can be calibrated (according to Beer-Lambert law) to quantify their abundances, but it may have been confusing to include the word, "total", which was meant to indicate the independence from extraction or ionization efficiencies often used in other types of chemical analysis. Also, describing the method of quantification is not the main purpose of this Section 2.2. To remedy these issues, we have first simplified the expression to in Section 2.2:

   "The molar abundance of molecules $\boldsymbol{n}_{\mathrm{molec}} = [n_i]$ in a mixture (consisting of a set of molecules denoted by $\mathcal{M}$) can be related to FG abundance $\boldsymbol{n}_{\mathrm{group}} = [n_j]$ (for each FG in $\mathcal{J}$) obtained by FT-IR — or other means — by invoking a group composition matrix $\mathbf{X} = [x_{ij}]$, which describes the FG makeup of each molecule."

   We have also included the appropriate references for obtaining bond or FG abundances from FT-IR spectra and other methods in Section 1:

   "However, studies on this topic have thus far been very limited on account of challenges in quantitative characterization of FGs, which requires either advanced algorithms (e.g.,

Takahama et al., 2013; Ruthenburg et al., 2014; Takahama and Dillner, 2015) for spectral interpretation or derivitization steps (e.g., Dron et al., 2010; Aimanant and Ziemann, 2013) for chemical analysis."

Furthermore, we have edited Section 1 to make note of the fact that we are not considering less than "total recovery" of bonds as we do not chemically extract our samples:

"The benefit of developing a systematic approach is that we can precisely understand the achievable mass recovery, and biases incurred on the calculated O/C and OM/OC for a given set of molecules and FGs analyzed (when chemical extraction is not required, OM mass recovery is primarily dependent on the completeness of FG calibration models constructed)."

2. Page 13, lines 4–5: Baltensperger and co-workers (AMT) measured a peroxide lifetime of a few hours in chamber SOA generated from a-pinene + ozone.

We thank the reviewer for pointing out this important reference (in *Cell*) — the text has been modified to include this citation:

"The rate of transformation of these FGs remains uncertain — for instance, reported lifetimes of hydroperoxides range from less than an hour to many days (Epstein et al., 2014; Krapf et al., 2016); resolving their reaction pathways may play a critical role in understanding model-measurement discrepancies (McVay et al., 2016)."

Technical comments:

1. Page 1, line 17: Insert "a" before "framework".

2. Page 3, line 1: Should "metric" be inserted after "carbon-centric"?

3. Page 4, line 8: Delete "a" after "specific".

4. Page 5, line 10 or 11. Delete "weighted" on one line or the other. Page 6, line 1: Can probably delete "expressed".

5. Page 6, line 2: Should be "below" not "blow".

6. Page 7, line 4: Should "atoms" be inserted after "carbon"?

7. Page 7, line 11: Should a comma be added after "eCH"?

We thank the reviewer for these technical corrections — the changes have been made in the manuscript.