This manuscript describes the characterization of air parcels observed at the Pico Mountain Observatory. Transport of air particles to the observatory are determined using FLEXPART analysis along with observations of gas and particle phase species. Monitored gas phase species include non-methane hydrocarbons and ozone. Particle phase species were determined using continuous measurements of black carbon, aerosol light scattering, and number concentration. Molecular characterization was conducted for 24 hr filter collected aerosol samples using an OC/EC analyzer, ion chromatography, SEM, and ultrahigh-resolution electrospray ionization fourier transform ion cyclotron resonance mass spectrometry (ESI-FT-ICR-MS). Based on the suite of measurements, FLEXPART back trajectories, and satellite data from the time periods, two plume sources were determined and studied in detail, one representing aged biomass burning aerosol from 12 days traversing the Atlantic and the other representing 15 day old marine aerosol. The detailed molecular composition analysis of the two air parcels and aerosol particle constituents represents a unique contribution to the literature. The manuscript is well written, however, is rather lengthy with no clear discussion of the results and implications. I support publication in ACP after the following comments are addressed.

Major Comments:

1) Page 24755 line 19-22: Why would increased fragmentation reduce the O/C? Most studies suggest aging and fragmentation actually increases O/C? See Comment #16.

2) Page 24762 line 4-7: Why is the Cl- in the blank such a large fraction of the aerosol samples? Is this due to very low Cl- concentrations in the aerosol? It seems the errors in the estimation of Cl- in the ambient aerosol (Table 2) would be higher with such a large background subtraction.

b) Along the same lines. For marine aerosol of 9/25, one would think that the concentration of Cl- would increase.

3) Data processing section is very wordy. Most of this information has been published already. I would suggest substantial shortening of text. Most of section 2.3.3 could reside in the supplemental information.

4) The average amounts listed in the text should also be listed in Table 1. Perhaps the last row could contain information of the averaged ambient mass concentrations.

5) Page 24768 line 1-11: This is a long winded way of saying this. It would be easier to just state that you measure 1.7 for the OM/OC ratio. The authors already state they are using the suggested literature value of 1.8 from Pitchford et al. (2007). Include a similar statement: the measured value (1.7) is the lower limit of OM/OC due to low retention of highly oxygenated low molecular weight species (Hallar et al. 2013).

6) Through-out the results section, small sentences of discussion are inserted into the text and feel out of place. The flow of the paper would be better, if these comments were deleted or moved to one section. The authors conclusions would be stronger if these comments were grouped together. I believe the

authors prove the source of the air masses from sections 3.1-3.2. I would say you should rephrase the discussion to state that your data is representitative of the these air masses and therefore the chemical composition is obtained. I do not believe you have enough evidence to say that any one feature from the mass spectra proves the aerosol source. Rather, the authors should say because they know the source of the aerosol, the obtained mass spectra represent the chemical composition of these aerosols. I will list the sentences I think should either be removed or moved to a consolidated section.

Page 24768 line 21-23: I would adjust this statement to: This indicates your 24 hr filter samples capture the major trends observed by the continuous measuremensts.

Page 24774 line 4-5: How is this statement supported? It would be better to say that the higher molecular weight ranges could be an indicator of aging processes associated in long range transport? However, as mentioned in #11 it is a long stretch to compare ESI-MS spectra and make conclusions based on intensity.

Page 24774 line 26 – Page 24775 line 1: Why is this an indication of highly processed aerosol? Other than Suwannee fulvic acid, because although it has similar optical properties to HULIS (definition of HULIS), it does not mean it has similar molecular composition. It also does not confirm the aged nature of the aerosol (Page 24775 line 3-4). Please include references to the use of suwanne river fulvic acid as a marker of the age of an aerosol if the authors wish to use this as an indicator.

Page 24775 line 19-21: Please move this discussion to the end of all the results and discuss with all the results, rather than one at a time.

Page 24776 Line3-5: Include in discussion section.

Page 24777 Line 26-29: Include in discussion section. Also explain how a high DBE value suggests more SOA formation?

Page 24779 Lines 1-6: Include in discussion section. I think it would be better worded as; biomass burning aerosol processed by clouds have the characteristics (…) seen in the mass spec. Your obervations are not confirming the source, but rather informing the community what the chemical composition of these aerosol types are.

Page 24779 line 16-18: Why would similar average values indicate the same emission source? Should not the greater O/C value go to the more aged sample 09/25?

Page 24779 line 23-25: Include in discussion.

Page 24780 line 24-29: Include in discussion, How would molecular fragmentation, mentioned in the other sections as a sign of aging, fit in with high DBE values?

Page 24784 line 20-21: Please include in the discussion section why increased fragmentation of molecular species would produce an aerosol with a lower O/C ratio and a reference to the literature articles supporting this?

7) There is no description of the gas phase measurements mentioned in the abstract in the experimental section. Page 24770 page 25-27: At least list the name of the methods and a small description of the measurements methods. This should be included in the experimental.

8) Page 24771 lines 4-18: This is a confusing paragraph with no explanation of why this is important. Are the authors suggesting ethane and propane are gas-phase indicators of aged biomass burning plumes? Or is this already established in the literature? The authors need to explain why ethane and propane matter and what they suggest about the age of aerosol in an air parcel, including references.

b. Page 24771 lines 6-8: I do not understand the notation in this sentence. What does "ethane (propane) increased from 0.78 (0.09)" mean? Do you mean "ethane and propane" increased?

c. Page 24771 line 13: What is the significance of "ln(propane/ethane)" ? Is this a quantity often reported in the literature?

9) Page 24772 lines 4-22: I am also confused by this section. What is the purpose of mentioning the other studies in the results sections? Are you confirming your results with those from other literature? That does not make sense because you are studying aerosol aged for 12 days and comparing with fresh and very near fresh (1-2h) aerosols in the literature. Please revise this section for clarity.

10) Page 24772 lines 25-27: The authors mention the low O/C measured in this study and here mention that there is likely a large portion of material that is not included. This portion of material not extracted would likely have a very low O/C and would reduce the measured O/C even further. This caveat should be mentioned again in a discussion section.

11) Page 247723 Lines 23 – 24774 line 3: Can this be attributed to tuning of the MS electronics? Can one compare the ESI intensity between different instruments and compare the concentrations in each sample? ESI intensity is not necessarily correlated to solution concentration, due to charging and matrix effects among others factors.

12) Page 24781 line 1-13: This paragraph can be deleted the topic was already discussed on page 24780 lines 10-15.

13) Section on CHOS group. This seems a lot of text for such a small subset of compounds. I think the authors should stress in the beginning that the statistics are not great. The authors should not bother trying to report in the text all the subtleties of the dataset. Table 2 and Figure 8 are enough. In addition, there was recent study of organosulfates in the atmosphere from CalNex and China.[1] It could be interesting to note the absence of organosulfates in the Pico Mountain Observatory compared with urban aerosols.

14) Page 24784 line 19-20: I thought the air masses were aged 12 and 15 days? I am confused by the 9.5 and 7 days statement.

15) Page 24785 line 22-23: It would be nice to explicitly mention what the critical insights this work provided.

16) Page 24786 lines 9-10: I am not sure of the authors conclusions that aged SOA have low O/C and that the low O/C can be explained by fragmentation. The authors have certainly not proven this in this manuscript. In fact papers have found an increase in O/C from again processes.[2-5] The authors need to discuss these and/or similar papers and support their conclusion that low O/C SOA can be an indicator of aging through a fragmentation pathway.

Minor Comments:

Figure 1: The caption of Figure 1 mentions that the filter samples in (a) do not align with the continuous measurements in (b) (c) and (d) because of being shown for the entire overlapping period. Why not line them up? There are already breaks in the axis of (b) (c) and (d). A small break in (a) around 09/24 would seem like it would line them up.

Figure 2: It is difficult to read the white numbers. Perhaps they can be outlined?

Page 24755 line 2: spell out acronym, m.a.s.l.

Page 24755 line 8-10: What about the other 5%?

Page 24755 line 11: delete “with an”

Page 24758 line 27-29: Insert reference for the article described

Page 24762 line 24: Include location of woods hole oceanographic institute.

Page 24767 line 17: delete “during 2012”

Page 24769 line 11-17: This seems rather long winded.

Page 24772 line 6-7: Include a brief decription of where 12 days comes from, i.e. FLEXPART analysis.

Page 24774 line 11- 12: The statement about CHO has already been made in this manuscript.

Page 24775 line 11-13: Delete sentence: This statement is unnecessary at this point in the manuscript.

Page 24776 line 17: Define isoabundance. What is the difference from abundance?

Page 24784 line 29: This is a confusing sentence. The unique formulas are fewer than what?

Page 24785 line 19: Include the uncertainty of the average mass concentration.

Page 24786 line 3-5: Was the pollution events both days? I thought one day was clean and one day was polluted.

References

1. Tao, S.; Lu, X.; Levac, N.; Bateman, A. P.; Nguyen, T. B.; Bones, D. L.; Nizkorodov, S. A.; Laskin, J.; Laskin, A.; Yang, X., Molecular Characterization of Organosulfates in Organic Aerosols from Shanghai and Los Angeles Urban Areas by Nanospray-Desorption Electrospray Ionization High-Resolution Mass Spectrometry. *Environ. Sci. Technol.* **2014,** *48*, 10993-11001.
2. Bateman, A. P.; Nizkorodov, S. A.; Laskin, J.; Laskin, A., Photolytic processing of secondary organic aerosols dissolved in cloud droplets. *Phys. Chem. Chem. Phys.* **2011,** *13*, 12199-12212.
3. Cubison, M. J.; Ortega, A. M.; Hayes, P. L.; Farmer, D. K.; Day, D.; Lechner, M. J.; Brune, W. H.; Apel, E.; Diskin, G. S.; Fisher, J. A.; Fuelberg, H. E.; Hecobian, A.; Knapp, D. J.; Mikoviny, T.; Riemer, D.; Sachse, G. W.; Sessions, W.; Weber, R. J.; Weinheimer, A. J.; Wisthaler, A.; Jimenez, J. L., Effects of aging on organic aerosol from open biomass burning smoke in aircraft and laboratory studies. *Atmos. Chem. Phys.* **2011,** *11*, 12049-12064.
4. Hawkins, L. N.; Russell, L. M., Oxidation of ketone groups in transported biomass burning aerosol from the 2008 Northern California Lightning Series fires. *Atmos. Environ.* **2010,** *44*, 4142-4154.
5. Kroll, J. H.; Donahue, N. M.; Jimenez, J. L.; Kessler, S. H.; Canagaratna, M. R.; Wilson, K. R.; Altieri, K. E.; Mazzoleni, L. R.; Wozniak, A. S.; Bluhm, H.; Mysak, E. R.; Smith, J. D.; Kolb, C. E.; Worsnop, D. R., Carbon oxidation state as a metric for describing the chemistry of atmospheric organic aerosol. *Nature Chemistry* **2011,** *3*, 133-139.