Reviewer 1

We would like to thank for the valuable comments from the reviewer.

This manuscript reports the results from a field study conducted in a forested mountain region at Whistler, BC, in summer 2010. A main focus of this study was to investigate biogenic SOA formation in a coniferous forest and the discussions are primarily based on the measurement data acquired with an HR-ToF-AMS and a PTR-ToF-MS. The manuscript provides new information on SOA formation pathways in biogenic emissions and the discussions are thoughtful. Overall, this work fits well within the scope of ACP and I recommend it to be accepted for publication after the authors address the following comments.

PTR-ToF-MS can measure a large number of VOC species but only 8 are reported in Table S1. It would be useful to provide information about additional VOC species, e.g., BTEX for anthropogenic emissions and acetonitrile for biomass burning emissions. For example, showing the time series of BTEX in Fig. 1 could provide additional support for the classification of periods into anthropogenic, biogenic, or mixed.

Response: Table S1 has been updated to report other VOCs measured in this study, including benzene, toluene and acetonitrile. The NO_x (average ± standard deviation = 0.7 ± 0.9 ppbv), benzene (0.02 ± 0.01 ppbv), toluene (0.11± 0.16 ppbv) and acetonitrile (0.08 ± 0.04 ppbv) mixing ratios were generally low throughout the whole study, indicating limited influence by local anthropogenic emissions and biomass burning. However, a relatively high level of secondary sulfate originated from regional background was observed in the later period of this study (July 13-19, Period 2) compared to the biogenic period. Therefore, we re-define the later period as "the period with both biogenic and regional background influence" and is referred as Period 2. The biogenic period is referred as Period 1. The above information has been added to the first paragraph of Section 3 in the revised manuscript.

Given that most HR-ToF-AMS studies so far reported O/C and H/C values using the Aiken's method, in order to understand how the values reported here compare to those from other locations, it would be useful that the authors provide some information about how the elemental ratios of OA determined from the Aiken and the Canagaratna methods compare for this study. Maybe providing in the supplemental material scatter plots of the O/C and H/C values from the two methods.

Response: Thanks for the reviewer's suggestion. We agree that it would be useful to compare the results between Aiken's and improved-methods. Comparison of the two methods is shown in Figure S5 of the revised supplementary information.

There was very little information on how PMF was performed and how the solution was selected and validated. Given that this study relies heavily on the mass spectra and the time trends of the PMF OA factors to infer BSOA properties and formation pathways, it is necessary that more technical details are provided about the PMF work in this study.

Response: Additional information and Figures S9-12 have been added to the revised manuscript and supplementary information, respectively, to better describe the PMF analysis performed in this study. The following paragraph has been added to Section 2.2.

"Positive matrix factorization (PMF) was performed to analyze the high resolution AMS organic measurement for identification of the forms of organic aerosol in this study. The bilinear model was solved using the PMF2 algorithm in robust mode (Paatero and Tapper, 1994) and the final solution was selected using the PMF Evaluation Tool (PET) version 2.06 (Ulbrich et al., 2009). A three-factor solution was selected as the optimum solution based on examination of the PMF quality of fit parameter (Q/Qexpected) as a function of the number of PMF factors up to seven factors (Figure S9). Further increasing the number of factors from three to four factors only splits a factor representing background organic aerosol, and thus more than three factors was not considered in this study. Time series and mass spectra of two-, three- and four-factor PMF solutions are shown in Figure S10, S11 and S12 respectively."

Line 8, page 28012, the chemical formula for hexafluoropropene is wrong

Response: Thanks for pointing out the error. The formula has been corrected to C_3F_6.

Line 1, page 28014, methanol has both primary and secondary sources. What is the relative importance of the two sources for methanol during this study?

Response: We cannot differentiate the relative contribution of primary and secondary sources of methanol in this study. However, we believe that the observed methanol was dominated by terrestrial vegetation in forest areas with limited influence of anthropogenic emissions. The wordings have been changed when we describe the correlations between VOCs and organic aerosol mass in Section 3.1.

Line 28 2013, Fig. 4 shows the correlations of OA with methanol, acetone, and acetic acid. However, the term "SOA" is used at here. I think it is necessary that the authors justify first the assumption that all OA mass was secondary in this study.

Response: We agree that it is inappropriate to use the term SOA here. "Total OA mass" is used in the revised version.

Line 24, 28014, change "values" to "value".

Response: Done

Same page, the volatility of an OA does not necessarily correlate with its O/C. Unless volatility measurements (e.g., using a thermodenuder) were available during this study, it is not appropriate to claim the volatility order for the 3 OA factors.

Response: Thanks for pointing out this error. We have removed the argument regarding the volatility order for the 3 OA factors.

Line 6, page 28017, define LT and its relationship to UTC.

Response: LT = local time, UTC -8 hours. This information has been added to the manuscript.

Line 15, page 28017, what are the f82 values in different OA factors? Give the ion formula at m/z 82 for isoprene SOA.

Response: According to previous studies, the ion formula at m/z 82 for isoprene SOA is $C_5H_6O^+$ (Robinson et al., 2011, Hu et al., 2015). The f82 values of background OA, BSOA-1 and BSOA-2 are 0.003, 0.007 and 0.006, respectively, which match previous AMS measurements from studies strongly influenced by monoterpene emissions (Hu et al., 2015). The discussion has been moved to Section 3.2 of the revised manuscript.

References:

Hu, W. W., Campuzano-Jost, P., Palm, B. B., Day, D. A., Ortega, A. M., Hayes, P. L., Krechmer, J. E., Chen, Q., Kuwata, M., Liu, Y. J., de Sá, S. S., McKinney, K., Martin, S. T., Hu, M., Budisulistiorini, S. H., Riva, M., Surratt, J. D., St. Clair, J. M., Isaacman-Van Wertz, G., Yee, L. D., Goldstein, A. H., Carbone, S., Brito, J., Artaxo, P., de Gouw, J. A., Koss, A., Wisthaler, A., Mikoviny, T., Karl, T., Kaser, L., Jud, W., Hansel, A., Docherty, K. S., Alexander, M. L., Robinson, N. H., Coe, H., Allan, J. D., Canagaratna, M. R., Paulot, F., and Jimenez, J. L.: Characterization of a real-time tracer for isoprene epoxydiols-derived secondary organic aerosol (IEPOX-SOA) from aerosol mass spectrometer measurements, Atmos. Chem. Phys., 15, 11807-11833, 2015.

Robinson, N. H., Hamilton, J. F., Allan, J. D., Langford, B., Oram, D. E., Chen, Q., Docherty, K., Farmer, D. K., Jimenez, J. L., Ward, M. W., Hewitt, C. N., Barley, M. H., Jenkin, M. E., Rickard, A. R., Martin, S. T., McFiggans, G., and Coe, H.: Evidence for a significant proportion of Secondary Organic Aerosol from isoprene above a maritime tropical forest, Atmos. Chem. Phys., 11, 1039–1050, 2011.

Section 3.5, I would like to ask the authors to elaborate a bit more on f91 and its utility as an indicator of BSOA. It could be useful to show f91 values in the mass spectra of the other OA types, such as urban/anthropogenic SOA, BBOA, HOA, and COA. What compound(s) or classes of compounds in BSOA that could produce C7H7+ in the AMS?

Response: Thanks for the reviewer's suggestions. As discussed in the original manuscript, f91 is unlikely a unique tracer for BSOA but it can be potentially used to evaluate formation pathways of BSOA in this unpolluted, forest environment (e.g., evaluate relative importance of BSOA formation pathways and precursors). Therefore, to avoid potential confusion, we decide not to make an in-depth comparison of f91 values in the mass spectra of different organic aerosols. The following paragraph has been added to Section 3.5 for discussing potential sources of $C_7H_7^+$ fragment.

"Although major contributors of $C_7H_7^+$ observed in this study are not well characterized, it is well known that formation of tropylium ion ($C_7H_7^+$) by electron impact ionization of benzyl compounds can attribute to this organic fragment (McLafferty and Turecek, 1993). While combustion processes are major sources of aromatic compounds, Gratien et al. (2011) observed formation of p-cymene via oxidation of a-pinene in laboratory experiments, suggesting possible biogenic origins of aromatic compounds. Other terpenes perhaps undergo similar chemistry but their atmospheric significance remains unclear."

References:

McLafferty, L. W. and Turecek, F.: Interpretation of Mass Spectra; University Science Books: Mill Valley, CA, 1993

Gratien, A., Johnson, S. N., Ezell, M. J., Dawson, M. L., Bennett, R. and Finlayson-Pitts, B. J.: Surprising Formation of p-Cymene in the Oxidation of alpha-Pinene in Air by the Atmospheric Oxidants OH, O-3, and NO3, Environ. Sci. Technol., 45, 2755-2760, 2011.

Showing the ion-speciated mass spectra of different OA factors in Fig. 3 is more informative. For example, it is interesting that the mass spectra of BSOA-1 and BSOA-2 look quite different but their O/C ratios are very similar. Do the m/z 43 peaks in the three factors have the same composition or do they have different ratios of C3H7+ to C2H3O+? I would like to see the relative contributions of CxHy ions and oxygen-containing ions at each m/z.

Response: We have modified Figure 3 a-c to show the ion-speciated mass spectra of different PMF factors. It is clear that m/z 43 peaks in the three factors are primarily due to presence of $C_2H_3O^+$. This observation may explain the similar O:C ratio observed in BSOA-1 and BSOA-2. This information has been added to Section 3.2 of the revised manuscript.

It may be helpful to scale the y axis of Fig. 3d to allow the H/C differences among data points more clearly seen.

Response: The scale of y-axis of Figure 3d has been changed.

Fig. S5 was not discussed. It was only mentioned in the figure caption of Fig. 3 and the purpose of showing it is not clear. In fact, how were the "biogenic" data points selected?

Response: The purpose of this figure is to illustrate the distributions of O:C and H:C ratios observed in the biogenic period and the whole sampling period. Since this information can be obtained from the Van-Krevelen diagram, Figure S5 has been removed in the revised manuscript.