We appreciate the referee's additional comments and valuable suggestions on our revised manuscript. We have carefully revised the manuscript again by fully taking account of the comments to improve it. Our responses to the specific comments are given below.

Responses to the comments of Referee#2:

Comment 1: TSP sampling: I understand now that the authors sampled with TSP due to previously seeing some BSOA tracers in the coarse mode. The authors use the word "some" to quantify this. How much have you seen previously in the coarse mode? I suspect this is very small fraction of the total as most of the biogenic SOA (like from isoprene-derived oxidation products) should reside in the fine aerosol mode based on the fact that sulfate is also predominant there and provides the necessary sites for heterogeneous chemical reactions.

It is good to see the authors noted the potential of positive and negative artifacts. I want to stress, I'm not a big fan of TSP sampling for SOA tracers, especially if the filter isn't covered, etc. It has been my experience in the past that TSP samplers don't do a lot to ensure debris, dust from work area, etc. deposit onto the filter. Also, it has been my experience the aerosol is not well protected. I don't mean to nit-pick on this issue, but just pointing out that I think PM10 or PM2.5 samplers in my experience do a better job in protecting the integrity of the aerosol sample. I'm certainly not saying that the PM10 and PM2.5 samplers are the best, as they have their problems due to the positive and negative artifacts that the authors have now discussed and added to the manuscript.

***Reply 1*:**

Our previous measurements at the forest site showed that on average ~56% of the mass of pinonic acid resided in the coarse mode in summer. For 2-MTLs, ~33% of the mass was found in the coarse mode. Taking account of the comment, we have added these quantitative information on the mass size distribution of the SOA tracers as follows: "*Our previous study showed that BSOA mass resided in the coarse mode observed at the forest site in summer (e.g., pinonic acid: ~56% and 2-methyltetrols (2-MTLs): ~33%) (Miyazaki et al., 2014).*" (Page 6, Lines 4-6)

Comment 2: Compounds used for quantification of BSOA tracers: The description is much improved now as it is clear how this was done. I still think the authors need to add a cautionary note in the discussion that the absolute concentrations are still tentative due to the use of surrogate standards.

***Reply 2*:**

Taking account of the comment, we have added the following sentence to the text: "*It is noted that the quantification of these compounds includes some uncertainty due to the use of surrogate standards.*" (Page 7, Lines 2-3)

Comment 3: Determination of aerosol pH from water extraction of quartz filters: Although the authors clearly had good intentions to address my original suggestion to estimate aerosol pH, extracting filters in water to estimate aerosol pH is not appropriate as the pH of the extract solution doesn't represent the "true" aerosol pH. The aerosol pH is only representative of the aqueous layer on the inorganic/organic mixed aerosol. The best way to estimate the "true" aerosol pH is by using an inorganic thermodynamic model, such as ISORROPIA-II (Fountoukis and Nenes, 2007) or E-AIM (see papers by Wexler and Clegg). The groups at GA Tech have had some recent papers discussing this important issue in ACP (Guo et al., 2015; Hennigan et al., 2015). Don't the authors have aerosol sulfate, nitrate, and ammonium data at least from their measurements? In the ideal case, they might also have gaseous ammonia data to better constrain the model estimate. If they don't, then it might make sense as to why they extracted the filters to address this previous comment I raised in my last review. The authors should note that work from the southeastern U.S. has shown at two different sites that aerosol pH is acidic (pH <2) but doesn't vary much during summertime conditions, and thus, doesn't seem to be the controlling variable due to the lack of variation in this parameter (Xu et al. 2015, PNAS; Budisulistiorini et al., 2015, ACP). I wonder if you would find the same thing if the aerosol pH is estimated using one of these thermodynamic models?

***Reply 3*:**

We have recognized that the pH of the extract solution doesn't represent the true aerosol pH, but is used here as an "indicator" of the aerosol pH. In fact, we have used ISORROPIA-II to estimate pH with the input data of the measured inorganic ions, ambient temperature, and relative humidity. However, the estimated pH values are not realistic or seem to contain large uncertainty. Possible reasons for the result are: (1) TSP samples may contain debris or dust, as the referee pointed out, which may partly make the thermodynamic equilibrium constrain difficult, (2) the lack of measurements of total organic mass and its hygroscopicity parameter, (3) the lack of gaseous ammonia data, etc. Thus, we have decided not to present the estimated pH using ISORROPIA-II in this manuscript. Instead, we have modified the statement on

the measurement of the pH in the extract samples as follows:
"*In order to further examine the effect of aerosol acidity on the formation of 2-MTLs, we measured pH in the water-extracted samples (section 2.3). The pH in the extracted samples may not represent the true aerosol pH, but is used here as an indicator of the aerosol pH, which offers insights into the aerosol acidity.*" (Page 14, Lines 17-20)

Comment 4: Other GC/MS tracers for isoprene SOA:
Did the authors not observe the known C5-alkene triols from isoprene oxidation (or really from reactive uptake of IEPOX) (Wang et al., 2005, Rapid Communications in Mass Spectrometry; Lin et al., 2012, ES&T)? These compounds should be seen by GC/MS with prior trimethylsillylation (as they use here) and are typically as abundant as the 2-methyltetrols. You can easily check the data for the C_5-alkene triols by using the extracted ion chromatogram (EIC) of m/z 231. I'm assuming you saw no dimers of IEPOX previously seen by Surratt et al. (2006, J. Phys. Chem. A) or Lin et al. (2012, ES&T) using the EIC of m/z 335?
I should point out if you don't see the C_5-alkene triols that is kind of interesting, as a recent paper by Riva et al. (2015, Atmos. Environ.) showed that isoprene ozonolysis in the presence of acidic sulfate aerosol only produced the 2-methyltetrols but no C_5-alkene triols were produced, suggesting an alternative mechanism from IEPOX uptake chemistry that leads to 2-methyltetrol formation.

***Reply 4*:**
As the referee pointed out, we observed C_5-alkene triols in all the samples, whereas dimers of IEPOX from the EIC of m/z=335 were not obvious in our analysis. The average concentration of C_5-alkene triols was comparable to (~74% of) that of 2-MTLs. The presence of C_5-alkene triols with its significant positive correlation with 2-methyltetrols ($r^2 = 0.67$) supports the IEPOX uptake chemistry previously suggested that leads to 2-methyltetrol formation. We have now added the following statement in response to the referee's comment.
"*Indeed, we also observed C_5-alkene triols, which is a particle-phase product from reactive uptake of IEPOX as well. The average concentration of C_5-alkene triols was 7.8±6.7 ng m^{-3}, which is comparable to that of 2-MTLs. The presence of C_5-alkene triols with its significant positive correlation with 2-MTLs ($r^2 = 0.67$) supports the IEPOX uptake chemistry that leads to 2-MTLs formation suggested by previous studies (e.g., Surratt et al., 2010).*" (Page 14, Lines 11-16)

Comment 5: Filter sampling size: Since the authors state clearly "that the goal of this study is to investigate controlling factors of BSOA formation from BVOC in a forest environment located near anthropogenic sources," I must state that I'm still cautious by the limited filter sampling used in this study. The paper kind of reads that this research activity was an afterthought to the flux measurements. Since the filter data is used for the PMF analyses, it seems the PMF results can only be interpreted with extreme caution. It would have been better of course if the authors had considered conducting longer sampling of filters, especially if you wanted to capture more variability in biogenic only vs. biogenic influenced by anthropogenic pollutants. Although the authors now argue this is the right time of year for biogenic emissions based on previous measurements, how do you conclude 5 days of sampling is enough for measuring the trends of BSOA tracers versus other co-located data? I realize the authors cannot go back and make more measurements, but I think the authors must acknowledge that extreme caution must be used with how to interpret the PMF results. I noted the other reviewer shared some of my concerns with the PMF.

***Reply 5*:**
Taking account of the referee's concern, we have added the following sentence to the text: "*Although the the data were obtained under typical meteorological conditions at this study site in summer, the PMF results can be interpreted with caution due the limited number of the data.*" (Page 13, Lines 7-9)