Comments to the author:
Dear Authors,

Thank you for addressing the 3 reviewer comments. Upon reviewing your replies and your new version of the manuscript, I will now accept with minor revisions noted. Please reply point-by-point to these remaining comments and outline how you changed the manuscript as a result.

Thanks so much, Jason Surratt

Response: Thank you so much for your kindly comments and good suggestions. We have addressed each comment in the following point by point and revised the manuscript accordingly. In addition, we check our manuscript again, according to the ACP guideline.

1.) What are the uncertainties of using GC/MS and the SOC tracer method to estimate total SOC? This is the largest issue to me that isn't addressed in this manuscript. For example, recent studies have raised serious questions about how thermal methods like GC/MS might cause for misinterpretation of isoprene SOA (e.g., Cui et al., 2018, ESPI; Lopez-Hilfiker et al., 2016, ES&T). Specifically, these prior studies show that isoprene SOA has a very low volatile nature, which is inconsistent with chemical constituents like C5-alkene triols and even to some degree 2-methyltetrols (e.g. Lopez-Hilfiker et al., 2016, ES&T, Hu et al., 2016, ACP). I think the authors need to acknowledge at least for isoprene SOA (but this is likely true for other SOA types) that low-volatility oligomers (including some organosulfates) may break down into monomers like C5-alkene triols and 2-methyltetrols. The SOC estimates made in this study likely have a large degree of uncertainty due to the thermal breakdown of the "actual" SOA constituents and also due to the lack of more complete anthropogenic SOA tracers. I think the authors need to acknowledge these uncertainties for their study before publication can be fully considered. Lastly, the foc values described in Kleindienst et al. that are used in this study to estimate the SOA amounts has a lot of uncertainty, since these were determined for these tracers of ONE representative experimental scenario. I worry that this SOA tracer method gets overused and not properly acknowledged for its high degree of uncertainty.

Response: Thank you very much for your good comments and suggestions. You raised a very good point. Indeed, due to their inherent low volatility of isoprene SOA tracers, and more importantly a lack of knowledge about their identity and thus available authentic standards, quantifying the abundance of such accretion products has remained an analytical challenge. Nowadays, in our observations sites, online

chemical characterization of SOA was performed using a chemical ionization mass spectrometer (CIMS) equipped with a filter inlet for gases and aerosols (FIGAERO) and aerosol mass spectrometer (AMS-Aerodyne Research Inc.).

Some details of the uncertainties of using GC/MS has been added in the revised manuscript, as follows:

However, inherent low volatility of isoprene SOA tracers could cause the uncertainties of using the GC/MS method, and low-volatility oligomers might break down into monomers, such as C5-alkene triols and 2-methyltetrols (Lopez-Hilfiker et al., 2016; Hu et al., 2016). Therefore, quantifying the abundance of certain SOA tracers remained a lot of uncertainties.

Hu, W., Palm, B. B., Day, D. A., Campuzano-Jost, P., Krechmer, J. E., Peng, Z., de Sa, S. S., Martin, S. T., Alexander, M. L., Baumann, K., Hacker, L., Kiendler-Scharr, A., Koss, A. R., de Gouw, J. A., Goldstein, A. H., Seco, R., Sjostedt, S. J., Park, J.-H., Guenther, A. B., Kim, S., Canonaco, F., Prevot, A. S. H., Brune, W. H., and Jimenez, J. L.: Volatility and lifetime against OH heterogeneous reaction of ambient isoprene-epoxydiols-derived secondary organic aerosol (IEPOX-SOA), Atmospheric Chemistry and Physics, 16, 11563-11580, 10.5194/acp-16-11563-2016, 2016.

Lopez-Hilfiker, F. D., Mohr, C., D'Ambro, E. L., Lutz, A., Riedel, T. P., Gaston, C. J., Iyer, S., Zhang, Z., Gold, A., Surratt, J. D., Lee, B. H., Kurten, T., Hu, W. W., Jimenez, J., Hallquist, M., and Thornton, J. A.: Molecular Composition and Volatility of Organic Aerosol in the Southeastern US: Implications for IEPDX Derived SOA, Environmental Science & Technology, 50, 2200-2209, 10.1021/acs.est.5b04769, 2016.

In addition, the foc values in this study cited from Kleindienst et al. based on simulation experiments, also has a lot of uncertainty. Therefore, the results of SOC tracer method used to estimate total SOC have been simplified and moved to the supporting information section. The trends and percentages of different types of SOC were only demonstrated in this study.

2.) Line 44: Change "Compare" to "Compared"
Response: Corrected.

3.) Lines 44-46: I would insert the result of your regression analyses in the abstract for the BSOA tracers versus Ox, HONO, UV, and T. Readers will want to assess from themselves how well correlated are these parameters.

Response: Thank you for your suggestions. The sentence has been rewritten, as follows:

Compared to those in winter, the majority of BSOA tracers in summer showed significant positive correlations with Ox (O_3+NO_2) (r = 0.443～0.808), HONO (r = 0.299～0.601), ultraviolet (UV) (r = 0.382～0.588) and temperature (T) (r = 0.529～0.852), indicating the influence of photochemical oxidation under relatively clean conditions.

4.) Lines 47-49: I would insert the result of your regression analyses in the abstract for the BSOA tracers versus PM2.5, NO3-, SO42-, and NH3. Readers will want to assess from themselves how well correlated are these parameters.
Response: Thank you for your suggestions. The sentence has been rewritten, as follows:

However, in winter, BSOA tracers were significantly correlated with $PM_{2.5}$ (r = 0.407～0.867), NO_3^- (r = 0.416～0.884), SO_4^{2-} (r = 0.419～0.813), and NH_3 (r = 0.440～0.757), attributed to the contributions of anthropogenic emissions.

5.) Lines 49-50: I would insert the result of your regression analyses in the abstract for the BSOA tracers versus aerosol acidity, LWC, and SO42-. Readers will want to assess from themselves how well correlated are these parameters.
Response: Thank you for your suggestions. The sentence has been rewritten, as follows:

Major BSOA tracers in both seasons was linearly correlated with aerosol acidity (pH) (r = 0.421～0.752), liquid water content (LWC) (r = 0.403～0.876) and SO_4^{2-} (r = 0.419～0.813).

6.) Lines 53-54: I would insert the result of your regression analyses in the abstract for the SOA tracers versus HCl and Cl- ions in PM2.5. Readers will want to assess from themselves how correlated are these parameters.
Response: Thank you for your suggestions. The sentence has been rewritten, as follows:

We also found that concentrations of the total SOA tracers was correlated with HCl (R^2 = 0.545) and chlorine ions (r = 0.280～0.639) in $PM_{2.5}$, reflecting the contribution of Cl-initiated VOC oxidations to the formation of SOA.

7.) Lines 64-65: change "researchers" to "research"

Response: Corrected.

8.) Line 73: Change "was" to "is"

Response: Corrected.

9.) Line 78: Delete "observation" and change "model" to "modeling'

Response: Corrected.

10.) Line 88: Change "nitrates" to "nitrate formation"

Response: Corrected.

11.) Lines 114-115: Please rewrite the sentence "We also demonstrated the indications of SOA tracers for air pollution process." This sentence is not well-written and is unclear what it adds here to the introduction.

Response: Thank you for your suggestions. The sentence has been rewritten, as follows:

Atmospheric process identified by SOA tracers in different seasons were further analyzed.

12.) Looking at your figure comparing H+ insitu derived from ISORROPIA and EIAM IV it is hard to see they agreed perfectly. How did the authors conclude this? Did they from a linear correlation, and if so, what was the result of that correlation? I raise this issue as there are several time periods where EAIM calculates higher H+ insitu. Seeing this difference at many time periods raising the question for me as to which model is the best to use?

Response: Thank you for your good suggestions and comments. At first, both ISORROPIA and EIAM IV were tried to calculate the aerosol acidity in this study. But, different data were input into these two models. ISORROPIA II calculated the equilibrium H_{air}^{+} and aerosol liquid water content of inorganic material ( ) by inputting the concentrations of the total SO_4^{2-} (TH_2SO_4, replaced by observed SO_4^{2-}), total NO_3^-(TNO_3, gas HNO_3 plus particle NO_3^-), total ammonia (NHx, gas NH_3 plus particle NH_4^+), total Cl^- (TCl, replaced by observed Cl^- due to the low concentration and measurement uncertainties of HCl)(Rumsey et al., 2014). However, the related air pollutants (including gas HNO_3 and NH_3) were not used in the EIAM IV.

Indeed, as you mentioned, there are several time periods where EAIM calculates higher H+ insitu. I think it has a lot of uncertainty, due to the 10-20 day observation period and different input data set. I regret not to answer the issue in this article. Anyway, the editor raised a very good point. In other study, we are comparing the difference of these two models based on multi-year monitoring data. We hope to find the answer in the future. Thank you for your good advice.

The sentence has been rewritten in the revised manuscript, as follows:

ISORROPIA II can calculate liquid water content (LWC), based on total SO_4^{2-}, NO_3^- (gas HNO_3 plus particle NO_3^-), Cl^-, ammonia (gas NH_3 plus particle NH_4^+), non-volatile cations (Na^+, K^+, Ca^{2+}, Mg^{2+}), and meteorological factors (RH and T).