Anonymous referee #3

This manuscript describes results obtained at the ground site of Look Rock, TN, during the 2013 Southern Oxidant and Aerosol Study (SOAS). A large set of instruments was deployed to measure the particle chemical composition (with on-line and off-line techniques) and gas-phase compounds. Results reported in this manuscript concern mainly non-refractory submicron particles (NR-PM1) with an Aerodyne aerosol chemical speciation monitor (ACSM), isoprene-derived secondary organic aerosol (SOA) tracers from filter samplings, and gaseous compounds with a high-resolution time-of-flight chemical ionization mass spectrometer (HR-ToF-CIMS) and a proton transfer reaction time-of-flight mass spectrometer (PTR-ToF-MS).

The authors showed that isoprene-derived SOA contributed significantly to the total organic mass, and that almost all the tracers quantified with off-line techniques were isoprene epoxydiol (IEPOX)- derived compounds. Results obtained suggest that IEPOX-derived SOA was not formed locally but rather during long-range transport, during which anthropogenic and biogenic emissions mix and interact.

This manuscript is well written, fits the scope of the journal, and provides interesting information on the complex mechanisms leading to the formation of isoprene-derived SOA. I recommend its publication in Atmospheric Chemistry and Physics after minor revisions.

We thank the reviewer for their careful review of our manuscript and the suggestions made below that help with improving clarity of our manuscript.

Specific comments:

1) Section 2.1: A better description of the sampling site is needed to fully understand the rest of the manuscript. If the authors include just one figure with a map of the region and a wind rose plot for the entire campaign, it will help a lot to better understand the different air masses, where the anthropogenic influences come from, etc. Without this information, even the back-trajectories given in the supplementary material (Figures S12 and S13) are impossible to understand, because we have no idea on the locations of biogenic or anthropogenic sources.

As suggested by the reviewer, we have added maps and description of study location in the SI.

2) Section 2.2: According to results shown later (section 3.4.1, Figure 6a), particles seem rather acidic. In these conditions, the use of a constant collection efficiency (CE) of 0.5 for the ACSM is not appropriate. I suggest that the authors introduce a time-dependant CE using equation 4 in Middlebrook et al. (2012).

The CE of 0.5 was calculated using Equation 4 of Middlebrook et al. (2012). The calculated CE values were around 0.5 during the entire field campaign.

We added this information into the main text of the experimental section describing the ACSM as follows:

"A collection efficiency (CE) of 0.5 calculated using Eq. 4 of Middlebrook et al. (2012) was applied to the ACSM data in order to accommodate composition-dependent CE."

Reference:

Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, *Aerosol Sci. Technol.*, *46*, 258-271, 10.1080/02786826.2011.620041, 2012.

3) Section 3.2: Additional information is needed in the supplementary material to support the choice of the 3-factor solution. In particular, it would be important to show mass spectra of the PMF factors for the 2-, 4-, and eventually 5-factor solution, in order to see how the OOA split into different factors. In addition to that, it would be useful to show a few diagnostic plots, such as the correlation among the PMF factors based on time series and mass spectra (so the same graph as Figure S3, panel d) for the 2-, 4-, and eventually 5-factor solution.

Moreover, can the authors confirm that they do not resolve a hydrocarbon-like organic aerosol (HOA) factor, even if they go up to 10 factors? This is a bit surprising for a site which is supposed to have anthropogenic influences. This result, coupled to the low concentration of primary pollutants (BC, NOx, CO), suggests that anthropogenic influences were quite limited at the sampling site.

There was no primary anthropogenic emission at the site since it was located atop of a mountain located in the densely forested Great Smoky Mountain National Park. Some primary pollutants could be transported to the site from the valley, which is more populated, however it was still very low. Low concentration of primary OA did not show as distinct factor from PMF analysis. This could also be attributed to the 30-minute time resolution of the ACSM, preventing it from being sensitive enough to pick up low concentrations of primary OA plume.

We have added time series, mass spectra, and factor inter-correlation plots of 2-, 4-, and 5-factor solutions in the SI section. HOA factor was observed in the 4- and 5-factor solutions, however, its temporal variation could not be distinguished from other factors and it was not well correlated with primary pollutants (i.e., CO and NO_x). Therefore, we concluded that the ACSM could not resolve the HOA factor from the organic mass spectral data collected during this study.

4) Section 3.3: It seems there is a mistake in the percentages of isoprene-derived SOA tracers reported in this section. Thus, the contribution of IEPOX- (96.8%) and MAE- (8.8%) derived tracers to the total isoprene-derived SOA mass is higher than 100% (page 7389, line 5). Moreover, the sum of all the tracers given in Table 1 reaches 101.6%.

This has been corrected in Section 3.3 and in Table 1.

Technical corrections:

1) Page 7368, line 1: "methacrylic acid epoxide (MAE)". Actually, MAE appears for the first time 2 lines earlier (page 7367, line 27), so the abbreviation should be defined already there.

This has been changed as suggested.

2) Page 7384, line 28: "but higher than that those".

This has been corrected as: "but higher than those "

3) Page 7393, line 3: "decrease in the in predicted IEPOX SOA".

This has been corrected as: "decrease in the predicted IEPOX SOA"

4) Supplementary material, page 13, line 5: "organic aerosol mass (OM)"

This has been corrected as: "organic matter (OM)"

5) Supplementary material, page 15, line 2: "the 2014 2013 SOAS field study".

This has been corrected as: "the 2013 SOAS field study."

6) Supplementary material, page 16, line 1: "24-hr model during_the first".

This has been corrected as: "24-hr model during the first"