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General comments:

This is a very comprehensive and fine study on the formation of isoprene-derived SOA during an intensive field campaign carried out at Look Rock, a site in the Ozark Mountains in the Eastern USA, where isoprene emissions during summer are known to be high. The authors have combined on-line measurements using the Aerodyne Aerosol Chemical Speciation Monitor (ACSM) with off-line chemical measurements of a suite of isoprene-related SOA tracers. The isoprene-derived SOA tracers contributed with ~9% (up to 26%) to the organic aerosol (OA) mass, and almost exclusively were made up by IEPOX-related tracers. An interesting result is that using PMF analysis of the ACSM data an IEPOX-OA factor could be derived that correlates well with off-line measured concentrations of 2-methyltetrols, C5-alkene triols, and IEPOX-derived organosulfates. There is, however, a substantial gap between the IEPOX-OA factor mass (~25%, up to 47%) accounting for 32% of the total OA, whereas that estimated by off-line isoprene SOA tracer measurements is substantially lower, i.e. ~9%, which is at present not understood but suggesting that in addition to the IEPOX-related SOA tracers there are other isoprene-related SOA tracers that are not covered by the off-line measurements. As expected, good correlations were found between particle-sulfate with both IEPOXand MAE-derived SOA tracers and the IEPOX-OA factor, supporting the important role of sulfate in isoprene SOA formation. The authors have also successfully modeled the IEPOX-derived SOA tracers 2-methyltetrols and corresponding sulfates.

We thank Professor Claeys for her very careful review of our manuscript. Her comments have improved the clarity of our manuscript.

Specific comments:

Page 7369/7370 – lines 27 – 5: The only pathway considered for the formation of the sulfate ester of 2-methylglyceric acid is the methacrylic acid epoxide (MAE) pathway (Line et al., 2013b). There is sufficient evidence in my opinion that the pathway involving reactive uptake of methacrolein onto highly acidic aqueous aerosol and reaction with the sulfate radical anion should also be taken into account and mentioned. See, for example, Schindelka et al. (Faraday Discussions, 165, 237-259, 2013). The latter pathway allows to explain the formation of other C2-C5 isoprene-related organosulfates, for example, the sulfate ester of glycolic acid (MW 156), which is difficult to explain otherwise.

We have added this pathway and reference to the main text as follows:

"Under both high- and low-NO conditions, acid-catalyzed reactive uptake and multiphase chemistry of isoprene-derived epoxides (IEPOX and MAE) as well as aqueous reactions of MACR and methyl vinyl ketone (MVK) with sulfate radical anion are now known to enhance SOA formation from isoprene (Surratt et al., 2007b, Surratt et al., 2010, Lin et al., 2013b, Schindelka et al., 2013).

Page 7388 – line 1 (and Figure 5): The elemental composition of terpenylic acid (MW 172) should be C8H12O4, instead of C8H12NO4.

This has been corrected as suggested.

Page 7389 – lines 19-28: Here, oligomeric IEPOX-derived HULIS is mentioned and the suggestion is made that quantification of these compounds could help to close the IEPOX-OA mass budget. Is there already any evidence for the presence of these compounds in ambient fine aerosol from an isoprene-rich site? Other compounds that may help to close the isoprene SOA mass budget are the C2-C5 isoprene-related organosulfates, formed through aqueous-phase reactions of methacrolein or methyl vinyl ketone, first-generation gas-phase oxidation products of isoprene, with the sulfate radical anion (mentioned above).

Lin et al. (2014) reported IEPOX-derived HULIS observation at Look Rock and Centerville sites during 2013 SOAS campaign. These sites are characterized by high isoprene emissions, particularly at the Look Rock site.

To clarify this we have revised the text as:

"An interesting and potentially important observation is that oligomeric IEPOX-derived humic-like substances (HULIS) have been reported in both reactive uptake experiments onto acidified sulfate seed aerosol and in ambient fine aerosol collected from LRK and Centerville sites during the 2013 SOAS campaign (Lin et al. 2014)."

It is noted that we didn't include the C2-C5 organosulfates from aqueous reactions of methyl vinyl ketone or methacrolein in this discussion here since we were referring to IEPOX-derived SOA mass closure only.

Page 7390 – line 5: The statement "However, it should be noted that in all previous studies 2-methyltetrols and C5-alkene triols were quantified by surrogate standards structurally unrelated to the targeted analytes." is too general. Authentic standards are a better choice but a surrogate standard used in previous studies is the C4-tetrol erythritol, which is structurally related (homologous) to the 2-methyltetrols (e.g. Kourtchev et al., Plant Biology 10, 138-149, 2008; Claeys et al., ACP 10, 9319-9331, 2010). The situation is different for previous measurements of the sulfate esters of the 2- methyltetrols (MW 216), where indeed a surrogate standard that is unrelated to the analytes (i.e. n-propylsulfate) has been used.

We revised the sentence as follows:

"It should be noted that in all previous studies sulfate esters of 2-methyltetrol were quantified by surrogate standard structurally unrelated to the target analytes (i.e., sodium propyl sulfate). While in current study, a mixture of authentic 2-methyltetrol sulfate esters was used as a standard for quantifying IEPOX-derived organosulfates."

Technical corrections:

Page 7373 – line 13: . . .. a Nafion dryer . . ...

This has been changed as suggested.

Page 7377 – line 11: . . .. by using a T-piece in . . .. . .

This has been changed as suggested.

Page 7377 – line 27: the abbreviation "sLPM"should be defined.

We changed "sLpm" to "standard L min^{-1}".

Page 7384 – line 28: . . ... higher than those . . ...

This has been changed as suggested.

Page 7388 – line 27: . . .. the abundance of . . ..

This has been changed as suggested.

Page 7393 – line 3: . . .. in the predicted IEPOX SOA . . ...

This has been changed as suggested.

Page 7409 – line 8: 2-methylglyceric acid

This has been changed as suggested.

Supplement – page 3 – Table S2: . . ... and reference mass spectra [Note: the abbreviation "MS"stands for "mass spectrometry" and not for "mass spectra"].

We changed "MS" into "mass spectra".