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Comment

Interactive comment on “Synthesis and coherent vibrational laser spectroscopy of putative molecular constituents in isoprene-derived secondary organic aerosol particles” by C. J. Ebb et al.

Anonymous Referee #1

Received and published: 6 January 2014

General comments:

This manuscript presents a novel method of investigating the presence and phase state of isoprene-derived secondary organic aerosol (SOA) constituents. After synthesizing a number of isoprene epoxydiols (IEPOX) and tetraols, the authors characterize the compounds by polarization-resolved sum frequency generation (SFG), and compare these spectra to those of isoprene-derived synthetic and authentic SOA particles. Their results indicate that the synthetic SOA is well represented only by trans-B-IEPOX,

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while ambient aerosol from the Amazon Basin does not match any of the epoxides or tetraols synthesized, suggesting that if such species are present, they may be found primarily on the interior of the particles rather than at the surface. While the experimental techniques described here may provide a new method for probing the phase state of SOA constituents, the results offered in this manuscript allow only for limited conclusions, as it appears a compound's existence in an SOA sample can neither be confirmed, discounted, nor quantified by SFG spectroscopy. However, the unique ability to probe the phase states of SOA constituents, particularly when used in conjunction with additional instrumentation to confirm the presence of compounds in SOA material, may open valuable new doors for investigating chemistry at the aerosol-gas interface. I would suggest the authors focus more attention on this novel aspect of their experiments, and on the possible atmospheric implications of a molecule's specific orientation at the gas-particle interface.

Specific comments:

The references need some revision. A large number of cited papers appear to address topics irrelevant to the manuscript (e.g., references #2, 6, 11, 12, 14, 17-20, 26, 27, 41, 42, 49, 51, and 55 discuss CdSe solar cells, nanocrystals, and other unrelated subjects), and journal abbreviations (e.g. ACP and ES&T) are inconsistent.

p. 29813 line 14: The compounds studied in this manuscript result not from reactions of BVOC oxidation products “with each other,” but from their reaction with atmospheric oxidants. This section would benefit from a clearer explanation of how IEPOX and the tetraols form, and therefore in what locations and phase states they may be expected to arise.

p. 29814 lines 1, 12: The oxidation pathways leading to SOA are not entirely “unknown,” and the epoxides and tetraols are not merely “proposed” but observed constituents of isoprene-derived SOA. Both the introduction and the discussion of results would benefit from greater description of how this manuscript fits with previous obser-

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vations of IEPOX and tetraol particle uptake and/or gas-phase chemistry.

p. 29816, section 2.2: It is important to note that IEPOX and the tetraols are presumed to be products of isoprene oxidation under low-NO conditions. Were the synthetic samples prepared in such conditions? Are NO levels known for the time and place where the AMAZE08 samples were collected?

p. 29817, line 5-6: Does the 8 cm⁻¹ variability refer to variations among spectra of the same compound, or between the various epoxides and tetraols?

p. 29821, lines 3-14: Are there no previous IR studies of epoxidic methylene groups to which IEPOX can be compared?

p. 29824, line 7: Trans- and cis-B-IEPOX switch between compounds #1 and #2 in figures 1 & 2; they appear to be consistent in the text, but should be double-checked.

p. 29826, line 16: "Title" should be "tilt."

p. 29826, lines 21-25: It seems unsurprising that the synthetic isoprene-derived SOA (itself in the condensed phase) is better matched by condensed-phase epoxides than gas-phase epoxides. Should the gas-phase epoxides even be compared to condensed-phase SOA samples? What is gained by this comparison?

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