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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The formation, properties and impact of secondary organic aerosol: current and emerging issues" *by* M. Hallquist et al.

Anonymous Referee #4

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Review of Hallquist et al. The Formation, Properties....

The manuscript is an extensive review of the current state of understanding of secondary organic aerosol (SOA). The authors of this review are among the most highly regarded researchers in SOA chemistry for both field and laboratory work. In fact, many are the foremost world's authorities in their areas of specialty. As a result, from my reading of the manuscript, the areas under consideration are technically sound and generally complete. My comments, in general, represent personal preferences in how certain subjects are treated or how arguments are structured. Thus, in this type of manuscript, I would defer to the author(s) in the final treatment and emphasis of the topics. This manuscript should be published following a consideration of the minor comments that follow.

One objective in a review article is to provide a comprehensive list of references for the subject area. I believe the authors did an admirable effort in providing the vast majority of the important references over the past 15 years including some legacy references from the 1980s. In some specific cases I have suggested some additional references that I believe are also valuable.

Criticisms of measurement methods, instrumental techniques, and laboratory and field measurements have been made in good taste. Recommendations found throughout the text tend to be reasonable. The only case where I believe some repetition is valuable is in assembling all recommendations found throughout the text into Section 4, Future Research Directions.

While this is meant to be a comprehensive review, some of the sections are particularly long and the section would be more readable by editing to shorten the presentation. (E.g., the discussion of the upper and lower limits for the global budget on pp. 3561-3565; Sections 2.2.1 and 2.2.2 on atmospheric degradation mechanisms on pp. 3570-3576; the discussion of particle formation from emissions of boreal forests on 3652-3653.)

By contrast, some subjects are treated insufficiently such as, FTIR analysis of aerosol (page 3586 lines 12-16); the use of PILS (page 2590 line 9-15).

I find the organization to be ad hoc; The heart of the manuscript is in Sections 2 and 3. I find the titles 'Overview' and 'Current and Emerging Issues in Secondary Organic Aerosol Research'; to be non-descriptive and arbitrary. The review gives little rationale for decisions made for which topics appear in which sections. Perhaps better titles could be adopted. Alternatively, the end of the Introduction could have a paragraph giving some guidance in the placement of topics in sections 2 and 3.

In many place throughout the review there is unneeded duplication, e.g., in the definition of the term oligomer in page 3591 line 29 and page 3627 line 11. While this is unavoidable with multiple authors, the lead author should make an effort to eliminate

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obvious duplications.

Page 3560, line 5-10, I would include a broader range of review articles, for example, – Daniel J. Jacob Heterogeneous chemistry and tropospheric ozone Atmospheric Environment 34 (2000) 2131-2159

Page 3560 line 25, I would recommend using more formal terminology than 'bottom-up' and 'top-down' (e.g., inductive/deductive approach?). The determination of the upper and lower bounds of globally produced SOA appears to be an original calculation and perhaps somewhat outside the scope of this article. While I believe the authors' will ultimately decide to retain this calculation in the revised version, it could be edited to be more concise. For example, the comments included in line 24 (p.3563) to line 4 (p.3564) could easily be removed without detracting from the argument.

Page 3565 line 24, the sentence starting on this line should have a reference.

Page 3568 line 15, what time scale is being referred to. If a VOC process produces SOA which is then removed by wet or dry deposition, can the carbon lost be converted to CO2? If one were to take seriously the VOC conversion to 910 TgC y-1 SOA, very little of the degraded carbon would be left in the atmosphere to be converted to CO2.

Page 3570 line 6, any precursor compounds that can generate high yields of dicarbonyl compounds, particularly, glyoxal are potential SOA producers regardless of how large or small the precursor is. This is basically the same reason that laboratory experiments show the formation of SOA from benzene.

In the top paragraph on p. 3567, I would suggest including the reference of – M. J. Molina, A. V. Ivanov, S. Trakhtenberg, and L. T. Molina et al. Atmospheric evolution of organic aerosol GEOPHYSICAL RESEARCH LETTERS, VOL. 31, L22104, doi:10.1029/2004GL020910, 2004

In Section 2.2.1, many of the sentences are poorly constructed (e.g., Page 3570 line 17). The whole paragraph needs considerable editing. The last sentence in the section

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is incomprehensible. The use of the word, respectively on page 3573 line 12, is unclear.

Page 3570 line 27, use significantly.

Page 3574 line 27-29, the sentence needs a reference.

Page 3575, the paragraph starting on line 9 lacks a level of precision needed to understand what the author is actually trying to say (e.g., 'Such methods invariably identify exotic reaction pathways which are not predicted by extrapolation methods....').

In my opinion, the issues addressed starting on page 3579 have been very well stated. I would suggest that they should also be better incorporated into Section 4. It may already be there and I simply do not see it.

On p. 3579 under point 3, the reference of – Hideto Takekawa, Hiroaki Minoura, Satoshi Yamazaki Temperature dependence of secondary organic aerosol formation by photo-oxidation of hydrocarbons Atmospheric Environment 37 (2003) 3413-3424, should be included.

On page 3582 line 3, the author might discuss in a bit more detail the meaning and implications of Figure 4. It is not self evident.

Page 3595 line 5, An alternative approach to yield determination given in 2001 should be included in the references – Michael D. Hurley , Oleg Sokolov, Timothy J . Wallington, Hideto Takekawa, Masa Yoshikarasawa, Bjorn Klotz, Ian Barnes, Karl H . Becker Organic Aerosol Formation during the Atmospheric Degradation of Toluene Environ. Sci. Technol. 2001, 35, 1358-1366

Page 3601 line 9-11, include recommendations in Section 4.

Page 3602 line 2-6, there is no need for a section outline since it is not used elsewhere. As noted above, an overall of the entire paper could be valuable if included in the Introduction.

Page 3611 line 14-29, the following references should be incorporated – Manish K.

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Shrivastava, Timothy E. Lane, Neil M. Donahue, Spyros N. Pandis, Allen L. Robinson. Effects of gas particle partitioning and aging of primary emissions on urban and regional organic aerosol concentrations, JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 113, D18301, doi:10.1029/2007JD009735, 2008

Timothy E. Lane, Neil M. Donahue, Spyros N. Pandis Simulating secondary organic aerosol formation using the volatility basis-set approach in a chemical transport model Atmospheric Environment 42 (2008) 7439-7451

Page 3613-3614, much of this is repetitive from Section 2.3, and should be reduced to cover only the essential ideas required for modeling.

Page 3627 line 20-22, The sentence needs a reference.

Page 3629 line 20, It is unclear how writers are using the term reference compounds; while oligomers do not have reference compounds neither do a wide variety of individual multifunctional organic compounds. Where standards are considered, it should also be emphasized that many of the calibration standards are frequently synthesized in-house at low, uncertain synthetic yields thus mitigating their value as reference compounds. Verification of the reference compounds by NMR and assays for their concentrations are also essential. I recognize that the use of laboratory generated standards may be sufficient for the types of studies being considered in this review but not in the classical sense of reference compounds such as those generated by NIST.

Page 3643 line 1-5, It is not clear as to what exactly needs further investigation. Perhaps the paragraph should be expanded somewhat.

Page 3630 line 24, It would be of great benefit if additional comments can be made to critically evaluate the quantification of oligomers in laboratory experiments.

Page 3650 lines 20-29, one or more references are needed for the final 5 sentences in the paragraph.

Page 3652 lines 5-28, The detail for these measurements are far in excess of other

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sections in the review. I would suggest reducing this paragraph.

Page 3676 line 9, The introduction should also point out that a section on abbreviations is included immediately before the references.

In the reference list, manually reorder Pöschl to precede Poulain.

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