

***Interactive comment on* “The continental source of glyoxal estimated by the synergistic use of spaceborne measurements and inverse modelling” by T. Stavrakou et al.**

Anonymous Referee #1

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Overall evaluation:

1. Does the paper address relevant scientific questions within the scope of ACP? Yes
2. Does the paper present novel concepts, ideas, tools, or data? Yes
3. Are substantial conclusions reached? Yes
4. Are the scientific methods and assumptions valid and clearly outlined? No
5. Are the results sufficient to support the interpretations and conclusions? No
6. Is the description of experiments and calculations sufficiently complete and precise

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to allow their reproduction by fellow scientists (traceability of results)? No

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution? Yes

8. Does the title clearly reflect the contents of the paper? Yes

9. Does the abstract provide a concise and complete summary? No, the abstract is too long and too detailed.

10. Is the overall presentation well structured and clear? Yes

11. Is the language fluent and precise? Yes

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? Yes

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? Figure fonts are too small

14. Are the number and quality of references appropriate? Yes

15. Is the amount and quality of supplementary material appropriate? N/A

Main comments:

This study presents a two-species inversion of global glyoxal and formaldehyde using the chemical transport model IMAGESv2 and satellite observations from SCIAMACHY. The purpose of the inversion is to constrain the continental sources of glyoxal. Glyoxal is produced by the oxidation of NMVOC from biogenic (mainly isoprene), anthropogenic, and pyrogenic sources; there is also a direct pyrogenic emission of glyoxal. Glyoxal is removed by photolysis, oxidation, and deposition. In addition, it is assumed that glyoxal is irreversibly absorbed by clouds and aqueous aerosols to form SOA mass. Model based on a priori emissions underestimates satellite-observed glyoxal, leading to the assumption that there is an unidentified VOC (UVOC) precursor of glyoxal. The

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authors conducted a couple of inversions, assuming either a direct glyoxal source (Inversion A) or a secondary production from a UVOC of 5-day lifetime (Inversion B). They found that inversion B showed better agreement with satellite observation and in situ surface concentration measurements (taken in mid-latitude forests).

The main weakness in this paper is that the scientific basis of the UVOC assumption is not well explained. Why is the missing precursor assumed to be biogenic (by scaling to LAI)? It seems also possible (from looking at Figure 2) that pyrogenic or anthropogenic emissions may be underestimated over some continents.

Assuming that UVOC is biogenic, what is the basis for the assumption of a 5-day lifetime? The authors must have reasons to suspect one (or some) particular candidate VOC species to make such an assumption. Is the resulting glyoxal production from UVOC from the inversion consistent with the emission of that (those) candidate UVOC(s)? At a lifetime of 5 days, a significant fraction of glyoxal production will be over the ocean instead of over land. Therefore, the inverted continental UVOC emission will be directly related to the assumed lifetime (as shown by the sensitivity studies). If the assumed lifetime is 3 days, for example, the inverted emission will undoubtedly be much lower, while still maintaining a reasonable agreement between modeled and measured surface concentrations.

Page 13596, lines 16~17: If the release of glyoxal from SOA is a potential source, is the magnitude of that glyoxal source consistent with any SOA production mechanism? In Section 2.4, SOA is assumed to be a glyoxal sink. If the glyoxal uptake is irreversible [e.g., Liggio et al., 2005], then how can SOA also be a glyoxal source? If the glyoxal uptake is reversible [e.g., Kroll et al., 2005], then the release from SOA cannot be counted as an additional source. If some other precursors is absorbed by aerosols, undergo chemical reactions, then released as glyoxal from SOA, then is the magnitude of that release consistent with the budget of the precursors?

Page 13599, lines 26~27: This is confusing; is methylbutenol included in the model or

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not?

Page 13600, lines 3~7: How is the irreversible uptake calculated for clouds and aqueous aerosols? How are the cloud droplet and aqueous aerosol surface areas calculated?

Page 13600, lines 11~13: In the present study, is uptake calculated only under light conditions?

Section 2.3, 2.4: How are dry/wet depositions of glyoxal and formaldehyde calculated? What about aerosol depositions?

Page 13604, lines 6~7: I am not convinced that simulated glyoxal is low over all continents. For example over North America, where isoprene is the dominant NMVOC and its emission well known, the model seems to be doing quite well. Over other continents, the comparisons between model and satellite data suggest that different NMVOC sources are may be at fault over different regions.

Page 13607, lines 6~9; Figure 6: There are considerable information on isoprene emissions over North America [e.g., Palmer et al., 2006], and some information over South America [e.g., Barkley et al., 2008], Africa [e.g., Shim et al., 2005], and Asia [e.g., Fu et al., 2007]. How do those compare to the isoprene inversions here? By looking at Figure 6, North American isoprene emission is greatly reduced in Inversion B, which contradicts earlier inversion studies. This may represent an over-emphasis of biogenic UVOC emission in the inversion.

Page 13607, lines 13~16: The authors should at least offer an order of magnitude estimates for the uncertainties. Otherwise it is difficult to gauge the relevance of inversion A and B.

Page 13610, lines 1~4: The better comparison between measurement and inversion B is not a robust way of rejecting a direct glyoxal source (inversion A). As the authors pointed out, all surface concentration measurements were taken in mid-latitude forests,

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where the a priori isoprene emission in the model is already high (which is why the a priori model already agrees well with the measurements). A secondary source (with a 5-day lifetime precursor) assumed in inversion B of course will not cause large increase over the source region.

Conclusion: The authors should briefly summarize the scientific implication of the present study in a broad context.

Table 3: What are these concentrations? Afternoon concentrations, 24-hour averages?

Minor comments: Page 13595, line 27: 'lanfd' should be 'land'. Page 13598, lines 12~14: Is there a reference for the C₂H₂ emission adjustment? Figures: Fonts in all figures are too small (e.g., colorbar legends, labels, axis labels)

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 13593, 2009.

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