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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimating sources of elemental and organic carbon and their temporal emission patterns using a Least Squares Inverse model and hourly measurements from the St. Louis-Midwest Supersite" *by* B. de Foy et al.

Anonymous Referee #1

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1 Overview

The manuscript by de Foy et al. uses least-absolute value regression to constrain emissions of EC and OC that contribute to year-long hourly measurements in St. Louis. There modeling setup allows them to specifically investigate temporal emissions patterns in some detail. Overall, the manuscript is very well written and easy to follow. The introduction and abstract might be enhanced a bit in terms of framing the value of their

work in examining an already much studied dataset. I believe the biggest scientific issue I see is the discussion of sinks, which are mentioned by the other reviewers. Mostly I have comments and clarifications about the inversion methods. This manuscript will be suitable for publication after revision to address the comments and corrections noted below.

2 Comments

- Title (and throughout): I feel like using the term "least squares inverse" as the name of the method in the form of a proper noun is a bit odd. The least squares method is ubiquitous, and by definition it is an inverse molding approach. So it doesn't seem to warrant capitalization in this form.
- 12032: Regarding the IRLS scheme, this is in general a method to perform least-absolute value regression, i.e., L_1 regression. The textbook by Aster shows this equivalence. It is thus further confusing that the authors would refer to their method as "Least Squares Inversion" when in fact it is actually a least-absolute value regression.
- It might be useful if an introductory sentence was added to the beginning of the abstract to help emphasize the value of this study.
- 12021.13: A subtle point on methodology: it is not necessary for error covariances to be diagonal in order for a Bayesian inversion to be cast as a standard least squarest problem. See for example the textbook by Aster, wherein augmented matrices involving the square roots of the error covariances are used to turn the standard Bayesian cost function into a standard least squares regression (Chap 11 perhaps? Sorry, I don't have it with me.). Maybe it is just then not clear what the authors mean by "single" in this context.

- 12021.25: The text refers to "the inventory" as if we knew specifically of one being discussed (e.g., NEI, or LADCO), but we don't yet at this point.
- Could the authors comment a bit more on the disconnect between the time periods covered by the different emissions inventories, and the observations? There have been significant trends (mostly reductions) in BC concentrations in the U.S. in the past decade. To what extent are inventories for years several after 2002 possibly impacted by these trends? Would this explain some of the deficiencies notes e.g., on lines 12038.23?
- 12022.7: An additional (better?) citation for BC-specific health impacts is Janssen et al., Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM10 and PM2.5. Environmental Health Perspective, 119(12):1691-1699, 2012.
- 12024: At this point in the manuscript, it seems that many previous works have used this dataset to look at source attribution questions. It might be good to state here what the angle of the present work is in terms of questions that remained to be answered or additional analysis that will be brought to bear.
- 12025: Given that later parts of the article emphasize the importance of micrometeorology, to what extent to the authors expect that the meteorological data from 15 miles away from the measurement site are relevant?
- 12026: Could it be clarified how these were updated?
- 12029: I'm not sure if CFA is a widely used technique. Can the authors explain, in a sentence or two, what this does?
- 12031.17: Another minor point about the methods: this statement is true only if the error covariance matrices can be reduced to αI, where α is a constant and I is the identity matrix. This is a more restrictive condition than just being diagonal.

- 12041.5: An alternative explanation is that estimates could be stabilized with more prior constraints, i.e., the current setup is under-smoother or ill-conditioned.
- 12042.2: I'm concerned about the large relative increases in emissions, factors of 20 and 30. This again seems like the system is under constrained (either to lack of data or lack of prior constraints). At the very least, these posterior estimates are vary inconsistent with the a priori uniform error assumption of 100% (12033.8).
- 12033.8: It seems odd that all emissions would be ascribed equal a priori uncertainty. Wouldn't we expect some sectors to be constrained much more or less than others?
- 12041.22: Alternatively, generating and using different meteorological fields from WRF using different physics schemes could provide some diversity to test the impact of the dynamos on the results.
- 12034.24: Could the authors clarify which features of the inventory that they know about are being referred to here?

3 Corrections

- 12035: Low Level Jet → low-level jet
- 12040.6: has a more → has more
- 12043: The phrase "LADCO inventory is slightly larger than the NEI" is written twice in this paragraph.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 12019, 2014.