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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 #2

Received and published: 10 June 2014

General comments

This study examines one year of hourly measurements of elemental carbon (EC) and organic carbon (OC) from the St. Louis Midwest Supersite. Using a least squares inverse model and atmospheric transport modelling, the authors estimate the emissions from different source types. In addition, the difference between weekday versus weekend emissions and the diurnal cycle are resolved. The authors find reasonably good agreement of the emissions with the prior estimate, but that open burning emissions

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are likely significantly underestimated in their prior. I recommend this manuscript for publication after the following comments have been addressed.

Section 2.4, which describes the inverse modelling method, is difficult to follow. In particular, there appear to be a number of inconsistencies in the definitions of the variables in Eq. 1 and the physical units of these. Details are given in the specific comments below. I suggest that the authors review this section carefully to make it clear to the reader exactly what was done.

Specific comments

P12021, L22: Specify whether this is emissions of EC, OC or both.

P12023, L27: What is meant by “smoking vehicles”. Is this synonymous with “vehicle emissions”?

P12024, L10: This sentence appears to be incomplete, for instance, which “Potential Source contribution function” or were there more than one. Please also add a reference.

P12024, L15: Do the authors mean at the St. Louis Midwest Supersite, if so, this should be specified.

P12024, L18-20: Please rephrase this sentence to make it clearer that EC is a passive tracer whereas OC is produced also in the atmosphere. The way it is written it is not clear where OC is “created” and it implies that EC is not emitted, which of course is not the case.

P12026, L7: Fig. 2 is referred to before Fig. 1, suggest either referring to Fig. 1 beforehand or reversing the order of the figures.

P12026, L9: Table 1 has not yet been referred to, suggest reversing the order of Table 1 and 2.

P12026, L20: Suggest that the authors start this paragraph by mentioning that an

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alternative emission dataset was prepared, to compare with the LADCO one, from the NEI data. Otherwise it is difficult to follow the text.

P12028, L20: Is dry/wet deposition accounted for in the FLEXPART simulations? If yes, please indicate the scheme for this. If no, please comment on how this may impact your residence time analysis.

P12029, L12: What is the impact of not using the aerosol module in CAMx, for instance, does this mean that dry/wet deposition of aerosols is not accounted for. If so, how will this impact your simulations?

P12029, L25: Please specify the "two-step" method. In Rigby et al. and Rödenbeck et al., an Eulerian model is used to take into account the influence on the air masses which is not accounted for in the time frame of the back trajectories (in this study 4 days). It is not clear in this study, however, how the background influence is accounted for or how the Eulerian model simulations would provide the background influence information. Also, please note that Rigby et al. 2011 actually use a 1-step method.

P12030, L6-8: If I have understood correctly, the simulations are not made using actual meteorology of each hour/day. Are these then average hours and days for the given year (i.e. 2002) or other? Please specify. Also, please specify if this was using CAMx or FLEXPART.

P12030, L19-22: Please specify that these were the averaged timeseries since they are the averages of different the weekdays/weekends and 4 timeslots throughout 2002.

P12031, L1-2: This sentence does not follow from the preceding one. What does "these" refer to in this sentence, it is not clear.

P12031, L10: the elements of x' are not "factors" as there is no multiplication involved. Please use rather "vector of emission corrections" or other.

P12031, L11: From the preceding paragraph I understood that you use the emissions estimates themselves and that these are not parameterized. Please clarify, are the

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elements of x the prior emissions or prior parameters?

P12031, L11 and L20: These sentences are inconsistent. In L11 the authors state that H is the operator to convert emissions to concentrations, while in L20 the authors state that the columns of H contain the timeseries' from the CAMx and FLEXPART simulations (in units of concentration for CAMx and emission sensitivity for FLEXPART).

P12031, L20-25: Having read this section, I understand that the authors have made 606 + 2880 simulations hourly for all of 2002. Is this correct? I suggest that they re-write the section P12030, L17-27 to make this clearer as otherwise, the reader will think that these are averaged timeseries, in which case, the description of H does not follow.

P12031, L27: There is an inconsistency here with L10-11 of the same page. In L10-11the authors state that x0 are the prior emissions and x are emissions. However, in L27 the authors state that x are scaling factors such that posterior emissions are equal to x*x0. Furthermore, given the definition of H, Hx (for the 2880 emission sensitivities) cannot be equivalent to y if they are scaling factors. Lastly, again given the definition of H, the elements of x cannot all have the same units. Please clarify.

P12031, L25-26: the vector x includes entries for the "gridded area sources" as well as the "group sources". Please be clearer about which sources are gridded and which are grouped as to be sure that the source types are not being double counted.

P12037, L12-13: I do not understand this sentence. What is meant by "yielded the most consistent estimate of impacts in the inversion"?

P12037, L19: Is the 19% for the "Other" category refer to the posterior emissions. This should be stated and possibly also mentioned at L4-5.

P12037, L23-29: Bootstrapping will provide an estimate of the uncertainty that comes from sub-sampling the data. However, there is also data selection in that outliers of more than 3 SD are removed. Have the authors investigated the sensitivity of the

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results to outliers and the selection criteria?

Section 3.3: Suggest adding subheadings to this section to make it clear what type of emission is being discussed e.g. On-road emissions, Non-road emissions, etc.

P12044, L12: What is meant by “annualized” does this simply mean the emission for each period given as the emission per year? Please clarify.

Technical comments

P12021, L2: “a year-long” (since it is only one year)

P12022, L20: replace “under-prediction” with “underestimation” as it is something can only be predicted or not and not “under” or “over” predicted.

P12025, L6: “mixed-use neighbourhood”

P12025, L17-18: Please use SI units, i.e. metric units throughout.

P12029, L20: Please correct: Rödenbeck et al. 2009

P12040, L2: “during daylight hours” (remove “the”)

P12044, L16: missing full-stop after “December”.

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