26 August 2014

Revisions for “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, Y. Y. Cui, J. J. Schauer, M. Janssen, J. R. Turner, C. Wiedinmyer, Atmospheric Chemistry and Physics Discussion, 2014.

Thank you for the reviews of our manuscript. Please find below a point by point reply to the comments along with a list of the changes made to the text.
The complete original reviewer comments are in black below, and the responses and modifications made to the manuscript are listed in blue.

Sincerely,
B. de Foy, Y. Y. Cui, J. J. Schauer, M. Janssen, J. R. Turner, C. Wiedinmyer

Anonymous Referee #3, Received and published: 6 June 2014
This manuscript presents an inversion for emissions of elemental and organic carbon using data from the St. Louis – Midwest Supersite. While the topic is important, I am concerned that the physical models are not appropriate for assessing emissions of EC and OC. With respect to both gases, it would seem vital to explicitly model their chemistry (formation, sinks). With respect to EC, which is not formed in the plume, has the sink been included (deposition)? With regard to OC, both formation in the plume and the sink would need to be modeled. In its current form, the manuscript does not appear to include these important processes. Therefore, the derived emissions will surely be biased. The study needs to be revised to model these processes.

General comments:
1. As mentioned above, the chemistry and deposition needs to be included to accurately derive model sensitivities. For EC, this might be simpler if there is no chemical formation/destruction in the plume and only deposition would have to be modeled in CAMx/FLEXPART. In FLEXPART, it is important to include sink processes for shortlived species and it was not mentioned in the manuscript whether this was done. OC will require a full chemistry model and deposition, without which, emissions or impacts at the site cannot be assessed. If this is not possible, then OC should be removed from the analysis. The current manuscript assumes that the OC measurement at the site is indicative of emissions/impacts from the source. The plume will have a different distribution from the inventory distribution so it is not clear how that can be disentangled without having a chemistry model.

We apologize for failing to mention that the CAMx simulations used both wet and dry deposition. The following text was added:
"Dry deposition was calculated using the Zhang et al., 2003 scheme, and wet deposition using the standard scheme in CAMx."

CAMx simulations are used for estimates of known emissions, whereas FLEXPART is used to estimate concentration impacts from unknown sources. For FLEXPART we therefore stick to Residence Time Analysis grids which do not have deposition included. The role of the two models was clarified as follows:
"The purpose of combining Eulerian with Lagrangian simulations is therefore to estimate adjustments

to known emission inventories with the Eulerian simulations, and to estimate impacts from unknown area sources in an overlapping domain with the Lagrangian simulations."

Because EC is not formed in the atmosphere, our model includes the main processes involved in EC transport, and so the emissions estimates can be interpreted directly. For OC, you are right that chemistry is an important source. However, we feel that there is still valuable information in the OC analysis and that this merits inclusion in the present paper. Aerosols are a very complex subject, and we believe that having different results from different angles does not detract from alternative methodologies but rather contributes to the field as a whole.

The following text at the end of the introduction clarifies this situation:
"Our model is focused on transport and consequently the results for EC can be straightforwardly compared to emission inventories. For OC however, the model does not distinguish between primary OC that is emitted by a source and secondary OC that is created in the plume of that same source. The results are therefore best interpreted in terms of impacts at the measurement site rather than emissions at the source location."

2. I do not understand the purpose of using the CAMx model when FLEXPART could be used for the entire inversion (provided that the chemistry can be included) or vice versa. If the only sources were ones that exist within the domain (i.e. boundary conditions are negligible), FLEXPART would contain all of the necessary information for the inversion. What is the benefit of using the second model?
This was clarified in response to the comment above and the comments from reviewer 2. We use CAMx to simulate transport from the well-established LADCO inventory. We then use FLEXPART to estimate impacts from sources that may have been totally missed in the inventory. Clearly there are alternative choices that are also valuable.

3. If the source distributions are incorrect, then this would affect both the inventory scalings that are derived as well as the estimation of 'missing' sources. How well are the spatial distributions known for each source?
We believe that the LADCO inventory is the state-of-the-art for our region. However anyone working in emissions knows what a hugely complex task it is. We think that the results of our analysis provide a partial answer to your question: the Point Sources, "Other," MAR and Non-Road emissions seem to be adequately represented. The category needing the most work according to our inverse model is the On-Road category, bearing in mind that part of the problem could be related to inaccuracies in winter-time WRF winds. Please refer to Sec. 3.3 for a discussion of these issues.
Additionally, the uncertainties in the spatial distributions of the sources is the main reason why we believe that it is valuable to combine CAMx for the known sources and FLEXPART for the unknown sources.

4. Please provide a more in-depth description of the inverse method and the assumptions that go into this method and what they imply (diagonal errors, trust-region iterative algorithm). As one example, assuming independent hourly observations (though there is a mention that previous studies have diagnosed a correlation timescale of 12 hours) could lead to an over-weighting of the data. References are given for various aspects of the method but the method should be justified in the context of this work.
We have added a figure of the WRF density functions and auto-correlation coefficients in Sec 3.1. This shows more clearly what we are referring to, and also addresses Specific Comment #11.
Note that the measurement errors can be assumed to be uncorrelated in time, as is done by all the

studies we know of. For block-bootstrapping, we want to select separate episodes. The issue is not one of correlation of errors, but of length of meteorological events. By using 24 hours as our block-bootstrapping interval, we select independent weather events. The discussion of the uncertainties in the paper has been expanded, please see comment #6 below.

5. Why was 1 ug/m3 uncertainty on the measurements chosen? Has a model representation error been included?
This value was selected by expert judgment as a realistic estimate. Note that is only used in order to interpret the values of the regularization parameter. The model representation errors are included in the regularization parameters. Note that what matters in the inversion is the ratio of the model uncertainties to the measurement uncertainties. In our work, we determine these objectively in order to minimize the total error as explained in the text.

6. It would also be nice to see an outline of the inverse procedure for clarity (for example, is the vector of regularization parameter optimized in the same iterative routine as the emissions)?
The following was added at the end of Sec. 2.4:
"In outline, we first perform the optimization of the regularization parameters without bootstrapping for each set in turn: for the RTA grids, for the LADCO emissions, for the open burning emissions and for the biogenics. This is repeated to make sure the values are stable. We then use the set of regularization parameters to obtain inverse results with the full data set, and 100 realizations with block-bootstrapping."

How are uncertainties and correlations derived in the inversion accounted for? Have the authors analyzed the correlations in the bootstrapping results (e.g. from the realizations of ‘x’ that are derived)? Are uncertainties in the observations and uncertainties due to the prior (from the regularization parameter) propagated into emissions and associated uncertainties? Some of this material could go into the Supplemental section.
Thank you for bringing up this important point.
In addition to block-bootstrapping, we have performed a Monte Carlo error propagation to further understand the uncertainties and the cross-correlations in the model. A new figure was added to show the uncertainty in the results and the cross-correlations for EC using bootstrapping. The corresponding figures for the Monte Carlo error propagation in the EC inversion and for OC are included in a supplemental section. This shows that the results are not unduly correlated with each other, and that we have made a reasonable attempt at characterizing the errors.

New text at the end of Sec. 2.4:
"We estimate uncertainties in the inverse model by two different methods. The first is to use expert judgment to determine an uncertainty on the measurements (y) and on the model sensitivities H) and to use Monte Carlo error propagation. We perform 100 realizations of the inversion with randomized scaling of the entries in y and H in order to estimate the uncertainties in x. In practice, we assume that entries in y vary by plus or minus 20% and those in H by plus or minus 50%.

An alternative method is to assume that by randomly sampling the data included in the inversion we are randomly sampling both the measurement errors and the simulation errors at the same time. This can be done with the bootstrap algorithm. Although measurement errors are assumed to be uncorrelated in time, meteorological events vary on the order of hours to days. In order to obtain samples that have different meteorological conditions, we perform block-bootstrapping with a block length of 24 h. We therefore perform 100 inversions with random selection with replacement of the days included in the analysis. In this way, the bootstrapping yields an estimate of the combined uncertainty due to measurement errors and due to transport modeling errors."

New text added at the end of Sec. 3.2:
"We used both Monte Carlo error propagation and bootstrapping to estimate the uncertainties in the emissions estimates. Fig. 9 shows the histogram of total emissions for each of the main categories in the inversion, along with correlation scattergrams of the results for the bootstrapped simulations for EC. For EC, the standard deviation of the contributions is between 3% and 5% of the mean contribution for all emission categories except for open burning where it is 20%. There is little correlation in the emissions estimates from the different source groups. The highest r2 is 0.22 for realizations of the On-Road and Other emissions. Overall this suggests that our results are not excessively impacted by cross-correlation terms.

The results of the Monte Carlo error propagation are included in the supplementary material. The uncertainties vary between 1.5% and 3% except for open burning where they are 6%. These are noticeably lower than the bootstrapping estimates as well as what we expect from knowing about emission inventories and from the values of the regularization parameters that were determined from the inversion themselves. These suggest that using block-bootstrapping provides a better estimate of the uncertainties.

The results for OC are included in the supplementary material. The bootstrapped standard deviations are between 5% and 10% of the mean contributions for all emission categories except for open burning where they are 18%. This suggests that the emissions estimates are robust with respect to uncertainties in the model inputs."

Specific comments:
1. Abstract – This sentence (The inverse model combines forward Eulerian simulations with backward Lagrangian simulations to yield estimates of emissions from sources in current inventories as well as from area emissions that might be missing in the inventories.) is confusing if you haven't first read the paper. Perhaps reword 'area' emissions to something like emissions unaccounted for in the inventories.
The word "area" was removed, which leaves the following text which is similar to the one you suggest: "from emissions that might be missing in the inventories"

2. Page 12029 Paragraph 1 – it would helpful to have a short description of what Concentration Field Analysis is and what it shows (as was done for the Residence Time Analysis)
New text added:
"Concentration Field Analysis is based on scaling the Residence Time Analysis at each time step with the concentration at the measurement site. The sum over the entire measurement period is then normalized with the Residence Time Analysis. This highlights air flow patterns that are associated with high receptor concentrations."

3. Page 12029 Line 24 – An explanation for why the two models are used together would be helpful. At present, it is unclear what the need is for using both (i.e. couldn't FLEXPART be used alone?).
Please see the clarifications added to the text, outlined under General Comments #1 and #3.

4. Section 2.4 Least Squares Inverse Model – Please provide a short description of

the lifetimes of these species and whether it is assumed that the boundary conditions to the Lagrangian domain are negligible.
Text added: "In our case, the background levels of EC and OC are very low (see Fig. 4), and we expect minimal impacts from sources outside the study area."

5. Section 2.4 Least Squares Inverse Model - How are CAMx model sensitivities calculated (emissions from the inventory of that particular source/time period are perturbed)?
We have clarified the explanation (see also comments from reviewer #2):
"Hourly Eulerian simulations with CAMx were performed for the five different source groups in the LADCO inventory: On-Road, Non-Road, MAR, Other and Point Sources. Because we are interested in evaluating the temporal profiles of the sources, we carry out separate simulations for emissions during different times of the day and different days of the week. The time slots were selected based on the diurnal profile used in the emissions inventory: 11:00 p.m. to 05:00 a.m., 05:00 a.m. to 08:00 a.m., 08:00 a.m. to 02:00 p.m., 02:00 p.m. to 06:00 p.m., and 06:00 p.m. to 11:00 p.m. Days of the week were split into a weekday group and a group containing Saturdays, Sundays and Holidays. As an example, an hourly time series of concentrations was obtained from a CAMx simulation with On-Road emissions only between 05:00 a.m. to 08:00 a.m. on weekdays."

And further down:
"For the CAMx time series, the entries in x are scaling factors on the LADCO emissions that went into the CAMx simulations."

6. Section 2.4 Least Squares Inverse Model – There could be significant temporal correlation. There is a brief mention of 12 hours being the correlation timescale from previous studies, but hourly observations are used and are treated as independent. This could lead to over-weighting of observations in the inversion. Can the least squares method be reformulated to deal with a full covariance matrix? Otherwise, using daily averaged observations may be better.
Yes, we could use a full covariance matrix with this method. In practice, most inverse models of emissions in the atmospheric sciences use diagonal matrices and so we are following common approaches to this question. The brief mention of 12 hours is to do with the correlation time of meteorological events. We use this to justify the selection of block-bootstrapping on chunks of 24 hours in order to increase the variability of the meteorological conditions in our bootstrapped sample. We moved the mention of the auto-correlation to Sec 3.1 as it is misleading in this context. Please refer to the new text described for Comment #6 above.

7. Section 2.4 Least Squares Inverse Model – Are there assumptions that go into converting equation 1 to equation 2? Can you describe what an augmented H”, x” and y” are (what are the dimensions)? Describing the inverse methodology in more detail is needed and can go in the Supplement.
There are no assumptions, please refer to Aster et al., 2012 for more details along with our previous papers on the method.
Text added: "H’’ has dimensions of (7091 + 3486) by (3486), and y’’ has dimensions of (7091 + 3486)."

8. Page 12030 Line 29 – ‘Area sources’ is confusing. Sources unaccounted for in the inventories is more clear.

The paragraph was rewritten as follows (see also comments by reviewer #2):
"The inverse model derives a posterior estimate of emissions based on the Eulerian simulations that used the emissions inventory as a prior. In addition, the inverse model uses the Lagrangian simulations to derive an estimate of sources that may be missing from the inventory. This is done by using the polar grids of Residence Time Analysis that represent the impact that an emission in a given grid cell would have at the measurement site. As all the known sources were already included in the CAMx simulations with the emissions inventory, we use a field of zero prior emissions for the polar grids from the Lagrangian simulations."

9. Page 12031 Line 25 - What are the 606 emissions elements? Are they scaling factors of the prior distribution for that source/time? Please provide some text to clarify this.
This was clarified as follows:
"For the CAMx time series, the entries in x are scaling factors on the LADCO emissions that went into the CAMx simulations."

10. Page 12033 First Paragraph – Why are the results of the inversion for the regularization parameter described here rather in the Results section? Also why are single values given? Isn't 's' a vector of values? It would also be good to discuss these results more, for example, about which components of the inventories are most uncertain. The derived regularization parameter should give an indication of the relative uncertainties of various parts of the prior.
We appreciate the suggestion of moving some of this section to the results section which would be a logical place to find it. However, we felt that when we did this it broke up the flow of discussing the emissions, and it separated into two parts something that is best understood when it is kept in a single part. We would therefore prefer to keep this section as it is.
In principle, one can have as many values of s as there are entries in x. In practice this is neither feasible nor desirable. We have therefore elected to use common values by emission groups. The logic is that the estimate of the uncertainties for example of the 6 open burning parameters are similar to each other, but different than the ones for the LADCO emissions.

The text was modified as follows:
"While in principle we can ascribe different values for each entry in the sensitivity matrix, we decided to use common values by source groups. The values of s were therefore determined separately for the emissions inventory sources, for the open burning sources and for the emissions based on back-trajectories."

11. Page 12033 Line 18 – The claim that there are no systematic errors in the model is likely overstated.
We have included the figure for KCPS in Sec. 3.1 and added the following text:
"Fig. 6 shows the probability density function for both the measurements and the simulations at KCPS. The distributions are very similar, and all variables passed the Kolmogorov-Smirnov test to much lower than the 1% significance level, showing that the model does not suffer from significant systematic biases."

The original sentence was entirely removed as part of an improved discussion of model uncertainties, see comments #6 above.

12. Page 12038 Line 19 – 'explains why' should be 'low posterior emission causes the

total emissions to decrease'
We have replaced "explains" with "is": "which is why ..."

13. Page 12040 Line 19 – why is the inversion not able to simulate winter concentrations? Are there 'missing' sources at this time that are compensating for the lack of agreement with the inventories (if posterior is showing scaling from inventories are showing near 0 emissions)?
This is an area of future research - any statement we would make would be speculative at this point.

14. Page 12041 Line 1 – Are these swings statistically significant based on the derived uncertainties? The phrase 'This suggests that there are large uncertainties in these estimates' should be rephrased using the evidence from the uncertainties that are presented.
The following text was added: "These swings are mostly contained within the 90% confidence range displayed in the figure which suggests that they are not statistically significant."

Also, the conclusion that 'more data could stabilize the emissions is too narrow. There are other areas that could contribute such as in the spatial distribution of the inventories and lack of chemistry being modeled that are hard-wired into the system.
We have added the following text: "or an improved model that considered in-plume chemistry."