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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Identification of mercury emissions from forest fires, lakes, regional and local sources using measurements in Milwaukee and an inverse method" *by* B. de Foy et al.

Anonymous Referee #2

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A inversion study is presented in which emissions of gaseous elemental mercury emissions are derived using measurements from a site downtown Milwaukee. A 'hybrid' regional model approach is implemented to determine emissions from lakes, forest fires and other sources. GEM emissions are thought to be relatively poorly constrained from bottom-up inventories and the use of inverse methods to constrain sources is therefore important and timely. The paper is well written and the techniques are thoroughly explained. However, clarification of some key points should be addressed before publication.

1 Major comments

- Suitability of the site is a major concern. Urban sites are rarely used for regional-scale inversions, due to the difficulty of simulating near-by sources in a regional model, and concerns about how representative the measurements are of the regional scale. Further details should be given about the sampling to address this. For example, is the instrument sited on a tower (where local source influence may be smaller), or in a street canyon (where local sources could be dominant)? Can we really believe that the variations seen are due to the interception of different large-scale air-masses, or the transport of local sources around the urban area? In reality, I suspect that few alternative sites exist for this work? However, if this is the case, I would suggest that more work should go into identifying times when the measurements can be considered representative of regional-scale air. For example, Manning et al., (2011) try to identify potential local contamination (even at background sites) by identifying days with low boundary layer ventilation. The authors note that the local emissions may be causing significant discrepancy between the observations and model. However, the apparent assumption that the local sources can simply contribute to a 'background' offset added to the measurements seems unlikely to be the whole story (P12952 L23): local sources usually cause high variability and large pollution events under certain conditions, and concentrations that are close to background levels are possible even in urban areas.

- Clarification of the inverse method is required to ensure that the results are robust. In particular it is very difficult to discern what constraints are actually being used in the inversion as it is presented:

    1. As I understand the procedure, forward simulations of an Eulerian model (CAMx) are run for certain sources (fires and lakes), and a scaling factor is sought to multiply these reference concentrations. In addition to this,

ACPD
12, C4852–C4858, 2012

emissions from each grid cell in the domain are also derived using WRF-FLEXPART residence time analysis (RTA). The results then discuss the scaling factors as a measure of the increase or decrease to be applied to the lake or fire emissions. However, I am confused as to whether the RTA also covers the lake and fire emissions regions? If so, the multiplying factors will be correlated with the emissions derived from the 'RTA part' of the inversion, and so the real lake/fire emissions would have to be increased or decreased accordingly.

2. A Bayesian inverse framework is derived in which a cost function containing measurement and prior emissions constraints is derived (Equation 4). Much of the next page or so discusses how the constraints on the observations or emissions are combined. However, on P12951 L8, the author states that a priori information is not used. I do not understand how the framework outlined in Section 2.6 can be used without a priori constraints? If a 'prior-free' inversion were desired, why not use a least-squares approach? In fact, the solution described on P12947 looks more like a tapered least-squares approach to me than a Bayesian method, in which case all of the discussion of measurement and model covariances etc. discussed in the previous page has been discarded. In which case, why mention it? Apologies if I'm confused by this section. A much clearer description of the inversion approach is required with clear justification for the choices made and how they are statistically robust.

3. If a 'Bayesian' approach was actually followed, then I have a number of concerns. Firstly, an emission covariance appears to be applied to the prior (Rb). This itself provides a weighting between the prior and the observations (which are weighted by Ra). To derive the cost function as written in Equation 4, the assumption is that the prior estimates and the measurements are uncorrelated. This is fine if Ra and x are not determined using the observations. However, on P12947 L17 onwards, it is stated that a scaling factor is

ACPD

12, C4852–C4858, 2012

iteratively applied that weights the Jobs and Jemis depending on the model-measurement mismatch. This means that independence between the two parts of the cost function is lost and in fact there is little point specifying Rb a priori anyway. Similarly, the procedure of iteratively decreasing the a priori uncertainty on grid cells where negative emissions are obtained also removes the independence between the prior and the observations.

4. The fit between the optimized model and the observations seems poor (Figure 10). In particular, the baseline seems to be higher than the observed background for much of the period (e.g. see Stohl et al., 2009 where the background is almost always below the smallest measurements). If this is true, does this not suggest that the derived emissions could be underestimated?

2 Specific comments

Introduction: Examples of previous regional inversions, specifically using Lagrangian Particle Dispersion models should be given.

The use of the term 'grid models' throughout is confusing. Does the author mean Eulerian chemical transport models in this case? All models use grids of some sort or other!

P12942, L10: Are 1000 particles/hour enough to calculate robust footprints? Typically an order of magnitude more particles are released to reduce noise (e.g. Stohl et al., 2009 use 40,000 every 3 hours).

Section 2.6: Un-accounted for sources of uncertainty in the inversion should be discussed. In particular, model-measurement mismatch errors do not seem to be accounted for. Furthermore, aggregation errors are likely to be significant in the time

ACPD
12, C4852–C4858, 2012

domain, as (I think) annual-mean emissions factors are derived, rather than shorter time periods (e.g. Thompson et al., 2011). Systematic model errors are also likely to be dominant for these types of inversion (e.g. Gurney et al., 2002).

P12946 L1: Is it simply measurement repeatability that goes into Ra? Usually some estimate of model-measurement mismatch are also included (e.g. Chen and Prinn, 2006).

P12946 L1: Ra is the uncertainty covariance corresponding to the measurement (and model prediction of the measurements), not the sensitivity matrix.

P12946 L26: Is D the identity matrix in this formulation? Further, I think that the transpose of the two matrices is required on this line (e.g. y"=(y, $x_{zero})^T$.

P12946 L28: I think that off-diagonal zeros are required for the combined R matrix.

P12947 L25: I think units are required for this scaling factor.

P12942, L16-19: Is this residence time analysis carried out for the surface grid cells? This needs to be clarified in this section (although it is mentioned later in the manuscript).

P12948, L11: It is stated that a 1000m cell height is used for the RTA so that sufficient particle counts are obtained. Usually a lower height is used to ensure that the particles can realistically be expected to interact with the surface. Could this situation not be improved by again using more particles when running the model?

P12948 L21: What is the basis for the 75

P12951 L5: Are 100 bootstraps sufficient to calculate robust statistics here?

P12952 L29: Does the seasonal cycle in the fire emissions not stem from the emissions dataset used, rather than the inversion, since a single factor multiplies the CAMx entire time series?

ACPD

12, C4852–C4858, 2012

Figure 4: There appears to be a step-change in the time series after a break in the measurements towards the end of March 2005. Can the authors provide some meteorological explanation for this apparent increase in the baseline, or could this indicate instrumental issues?

3 References

Chen, Yu-Han, and Ronald G. Prinn, 'Estimation of Atmospheric Methane Emissions Between 1996 and 2001 Using a Three-dimensional Global Chemical Transport Model', Journal of Geophysical Research, 111 (2006), D10307 <doi:10.1029/2005JD006058>

Gurney, Kevin Robert, Rachel M Law, A Scott Denning, Peter J Rayner, David Baker, Philippe Bousquet, and others, 'Towards Robust Regional Estimates of CO2 Sources and Sinks Using Atmospheric Transport Models', Nature, 415 (2002), 626 - 630.

Manning, A. J., S. O'Doherty, A. R. Jones, P. G. Simmonds, and R. G. Derwent, 'Estimating UK Methane and Nitrous Oxide Emissions from 1990 to 2007 Using an Inversion Modeling Approach', Journal of Geophysical Research, 116 (2011), D02305 <doi:10.1029/2010JD014763>.

Stohl, A., P. Seibert, J. Arduini, S. Eckhardt, P. Fraser, B. R. Greally, and others, 'An Analytical Inversion Method for Determining Regional and Global Emissions of Greenhouse GasesâĂŕ: Sensitivity Studies and Application to Halocarbons', Atmospheric Chemistry and Physics, 9 (2009), 1597-1620 <doi:10.5194/acp-9-1597-2009>.

Thompson, R. L., C. Gerbig, and C. Rödenbeck, 'A Bayesian Inversion Estimate of N2O Emissions for Western and Central Europe and the Assessment of Aggregation Errors', Atmospheric Chemistry and Physics, 11 (2011), 3443-3458 <doi:10.5194/acp-11-3443-2011>

ACPD

12, C4852–C4858, 2012

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ACPD

12, C4852–C4858, 2012

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