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Interactive comment on "Inverse modelling for mercury over Europe" *by* Y. Roustan and M. Bocquet

Anonymous Referee #2

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——————— General Comments ———————

The article is an excellent contribution and should definitely be published.

The authors may wish to consider additional efforts to make the article more accessible to those not intimately involved in the field of adjoint modeling. This could be accomplished by more plain-language explanations of the meaning and significance of the many formulas employed in the paper. In some cases the authors have already done this, but wherever practical, it could be done for every formula.

The article relies in many cases on Roustan et al 2005, and it is difficult to fully understand certain aspects of the article without reference to this source. However, it

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appears to be a "grey literature" document, apparently contained within the proceedings of a conference. Is this manuscript widely available? Could the URL of a web-based version be provided in the reference? Perhaps it could be made available on the author's institutional web page or linked via the SRef system.

The article correctly points out that deposition is a more important aspect of atmospheric mercury analysis. This analysis deals primarily with gas-phase concentrations of elemental mercury [Hg(0)], and this species does not contribute significantly to deposition. So, the analysis is only an initial, somewhat limited foray into the complicated issue of boundary conditions for regional models. The authors are to be commended for providing this context for the paper.

——————— Specific Comments ———————

As mentioned above, the article is an excellent contribution and should definitely be published. However, the following are issues that the authors may wish to consider.

1. Why weren't the southern boundary conditions and the "top" boundary conditions chosen as a variables? Perhaps this could be explained in the text.

2. The mass balance (e.g., Table 1) is very useful and informative. However, it is a bit surprising that wet deposition is negligible. Of course, the wet deposition of elemental mercury is probably negligible, but, elemental mercury can be oxidized and these oxidized forms are certainly subject to wet deposition. The chemical model used for this mass balance – with the Petersen chemical module – certainly includes this phenomenon. So, for a mass balance of elemental mercury, it would seem that one component would have to be chemical conversion and eventual wet deposition.

3. It is not always clear which year the various analytical inputs are based on. For example, was 2001 meteorological data and/or 2001 emissions used throughout, even though analyses were done for 2001 and 2002? If this is the case, it should be made more clear. And if this is the case, then it raises the question of the appropriateness

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of using 2001 meteorology and/or 2001 emissions data to analyze 2002 monitoring data. Travnikov and Ilyin found in a sensitivity analysis that while boundary conditions were the most important influence on TGM concentrations, inter-annual meteorological variability was also very significant. [Travnikov, O., Ilyin, I. (2005). Regional Model MSCE-HM of Heavy Metal Transboundary Air Pollution in Europe. EMEP/MSC-E Technical Report 6/2005. Meteorological Synthesizing Centre - East, Moscow. Available at http://www.msceast.org].

4. The analysis uses "climatic boundary conditions" in some cases, provided by MSC-East, for 2001 and 2002. It would be helpful if there was at least a brief description of the methodologies used to derive these boundary conditions, and what their values were.

5. The author's certainly realize this, but in actuality, the boundary conditions are varying temporally at much greater frequencies than yearly or monthly, and are varying spatially (both vertically and horizontally) at much shorter length scales than the whole-side boundaries (e.g., "West boundary"). In other words, the assumption of a "constant boundary condition" – even for a month – on a particular boundary seems very unrealistic. As the authors point out, there are few measurements to use. But, the measurments that do exist (e.g., at Mace Head) seem to suggest very signficant temporal variability at time scales shorter than one year. In a way, the authors are estimating the "average" boundary conditions – at a very coarse time and spatial resolution – that give the best results. Its a fine starting point, but the issue of spatial and temporal variability of boundary conditions should be discussed.

6. This question no doubt shows the reviewer's unfamiliarity with inverse modeling, but what is the physical meaning of the "assimilated parameters", e.g., in Table 3? This comment is related to the general comment above regarding accessibility to a broader scientific audience.

7. Is is possible that there might be other data available, besides the few stations

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used? Maybe not... but, are data available for the Neuglobsow site? Or for NO42? The Topolniky station is briefly mentioned – are there no TGM measurements there that could be used? The analysis presented is a great start, and the author's point out that more data are needed, and that the procedure can be extended to include more data, if and when it would become available.

8. The authors point out the need for RGM measurments in addition to TGM measurements. TPM measurements would also be useful. It is recognized that the technology does not yet exist, but measurements of actual chemical species (e.g., HgCl2, Hg(OH)2, etc.) would be helpful. Moreover, measurements at different heights in the atmosphere would be helpful, as well as measurements on the "boundaries" to characterize the spatial and temporal variability of boundary conditions.

———————- Technical Corrections ———————-

The article is certainly readable, but it could benefit from a little editing, to clean up the grammar, etc... as I'm sure could be said of this review as well :)...But it is not too bad, given that english is probably not the author's first language.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 795, 2006.