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Interactive Comment

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

### Anonymous Referee #1

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#### **General Comments**

The authors present the application of an adjoint method to solve the inverse problem for the fate and transport of atmospheric mercury over Europe. They show that using the inverse modeling results to adjust the boundary conditions leads to improved model performance. They also show that the inverse modeling does not provide much improvement when adjusting the emissions because emissions affect mostly local receptors and only a few receptors were available for this study.

This work provides a valuable application of the adjoint method to air quality modeling. Because mercury is a global pollutant, boundary conditions affect mercury concentra-



tions and deposition significantly on regional/continental domains. Also, the speciation of emissions is particularly important.

The authors correctly demonstrate the importance of resolving the temporal variation of the boundary conditions for the inverse problem. Moreover, the northern boundary conditions is shown to be particularly influential during the period of the Arctic springtime, i.e., when mercury depletion events significantly affect mercury concentrations and speciation in northern latitudes.

The results presented by the authors are quite consistent with our current knowledge of the atmospheric cycle. Their work provides an important quantitative tool to analyze the results of atmospheric mercury models and improve their performance.

The methodology, assumptions, results and conclusions are clearly presented with appropriate numbers of tables, figures and references.

Therefore, I recommend publication of this manuscript in Atmospheric Chemistry and Physics.

#### **Specific Comments**

In Section 5.1, first sentence: The authors assumed that the fraction of oxidized mercury is low at the boundary and they set HgII to zero. This assumption may not be correct as Hg0 is continuously oxidized to HgII. Results of global simulations show that HgII concentrations are small but not negligible (on the order of 10 to 30 ng/m3 over the oceans in the northern hemisphere). Measurements of speciated Hg at Mace Head on the west coast of Ireland (station IE31) show commensurate values for HgII (Ebinghaus et al., Atmos. Environ., 33, 3063-3073, 1999). The author address this issue to some extent in Section 5.2 and one should note that this assumption does not affect the interesting implementation of the inverse modeling technique to mercury nor the valuable conclusions of the study. Nevertheless, the authors may want to further discuss the possible implications of this assumption. For example, if the rate of reACPD

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moval of HgII from the atmosphere is fairly short, then the HgII boundary condition will have little impact on Hg concentrations in the mainland; on the other hand, if the HgII lifetime is commensurate with the transport time to the measurement sites, then, one may expect the HgII boundary condition to be influential. The authors correctly point out that HgII boundary conditions could be considered in the analysis if one focused on Hg deposition data since those pertain mostly to HgII.

#### **Editorial Comment**

On p. 796, line 15: I suggest replacing ", and their location far from Europe centre" by ", and most of them are located at coastal and/or northern locations that may not be representative of the European mainland".

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