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Interactive Comment

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Interactive comment on "A numerical modelling study on regional mercury budget for eastern North America" by X. Lin and Y. Tao

X. Lin and Y. Tao

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Reply to Referee #2

We would like to thank the referee for his/her interest, time and comments. Our detailed response is as follows.

(1) We agree with the referee that the uncertainties exist in the Hg content in soil and water and boundary conditions. As we can see from Table 5 of the discussion paper, the re-emissions of Hg0 only account for a very small part (~2%) of wet deposition over the entire domain due to the physio-chemical nature of Hg0. On the other hand, the Hg background levels are relatively well established from both measurements and global modelling studies. An arbitrary 'tune' on the background would lead to unrealistic deviations from the averaged ambient measurements since the background dominance in the Hg modelling practice (also see (4) below). During the process of comparing

model results against the wet deposition data available, these considerations with others prompted us to consider the suggestions raised by Bergan and Rodhe (2001), Travnikov and Ryaboshapko (2002), and Ryaboshapko et al. (2002). As a matter of fact, the 3-fold reduction of the rate constant of Hg0(g) oxidation by hydroxyl radical OH preserves a \sim one-year lifetime for Hg0, which is presently accepted. On the contrary, as discussed in Section 5.3, the run with the original rate constant leads to a 'net sink' of more than 6 tons of Hg. Such a large sink is not consistent with the human and industrial activities within the region in a global context.

(2) We agree with the referee that the 'net source' conclusion needs to be presented in a proper way. The 'net' came from the small difference between two large terms. The sign could change because of not only the uncertainty mentioned above, but also the uncertainty in other inputs such as anthropogenic emissions etc. We will reword the 'net source' conclusion in the Abstract, Sections 5.2 and 6 according to the referee's suggestion and will elaborate it more in Sections 5.2 and 6 of the revised manuscript. Besides, we will delete the decimals of all numbers in units of Kg to avoid a false perception associated with the ignorance of uncertainties involved in the study.

(3) As recommended by the referee, we will include a data analysis on the Hg cycling between air, water and land in Section 5.2 of the revised manuscript.

(4) The study indicates >80% of the global background contribution to Hg regional deposition. The background dominance reflects the 'long-living' nature of Hg. Consequently, it implies that a proper Hg modelling needs to be performed on a large spatial scale such as a continental scale unless boundary conditions can be defined reasonably well. We will elaborate this aspect in Section 6 of the revised manuscript following the referee's suggestion.

(5) We agree with the referee on the low correlation between modelled wet deposition and MDN measurements. Although the 3-fold reduction of the rate constant of Hg0(g) oxidation by hydroxyl radical OH, reported by Sommar et al. (2001), brings the

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modelled wet deposition significantly closer to the limited MDN data (with an improved correlation), the correlation coefficient is still low. This is mainly linked to the meteorological input data generated from MM5. We will add a discussion of the meteorological issue in Section 5.1 of the revised manuscript.

(6) As for the Hg0 content in surface soil water, we related it to the current emission sources as an approximation at the first order in the discussion paper. In our view, the proportional relation of the Hg0 content to the product of the source strength and the squared reciprocal of the distance between the source and receptor does not necessarily imply an equilibrium status between the sources and the distribution of Hg0 content. Based on the proportional relation, the Hg0 content was determined through a calibration approach based on measurements. We agree, there is an uncertainty associated with the Hg0 determination. Since the re-emissions of Hg0 only account for a very small part (~2%) of wet deposition over the entire domain, we do not expect a substantial impact of the uncertainty on the deposition simulations in the study. In the revised manuscript, we will exclusively address the issue in Section 4.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 983, 2003.

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