

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

Anonymous Referee #2

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General comments

The paper describes a set of improvements introduced to the mercury chemistry and transport model CMAQ and discusses an application of this new model version to computation of regional Hg budget for part of North American continent. The model improvements include: modified bi-directional flux in soil - air and air - water interfaces, studying of the chemical oxidation of elemental mercury with OH radicals, split of particulate mercury to soluble and insoluble fractions, etc. As it is stressed by the authors, the uncertainty of the oxidation rate of Hg by hydroxyl radicals is huge – a factor of times at least – so a special caution is necessary. It is too easy to “tune” this crucial coefficient to get nice agreement with observations, which are themselves are scarce and uncertain. Two other “easy” parameters are the amount and distribution of Hg in soil and water and boundary conditions (global Hg background). These difficulties are

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objective and just reflect level of our knowledge, but I was surprised when the authors put all attention to oxidation rate and neglected two other sources.

In this regard, the main result of the paper – the regional Hg budget – becomes an easy target for criticism. Indeed, the claimed net outflow is just ~8% of the total in- and out- Hg fluxes, which totally lies inside the uncertainties. For example, agreement with few available deposition measurements is worse than these 8%, not to speak about the spatial representativeness of the sites. And, of course, 461.2 kg looks very strange – one can try to speculate about 0.5 ton plus/minus, say, 200%, but nobody can defend 4-digit precision here. The same is true for many other numbers provided in the paper – they should come with uncertainty ranges and should not contain un-justified number of digits.

At least for me, the most interesting was not the budget figure, which probably means “do not know, insufficient accuracy”, but rather an attempt to simulate the Hg cycling between air, water and soil explicitly resolving the inter-compartmental interfaces. I would recommend stressing this very point, together with “do not know, probably net source” conclusion concerning the regional budget.

This can also be related to a more general question – may be, regional models can not be used for Hg simulations at all? There is no discussion of this matter in the paper, while >80% of the global background contribution to the regional deposition seems to answer negatively. Another argument in the same direction is a low correlation with observations. Again, the mean level means nothing – it reflects just the background value attributed voluntarily. But low correlation tells that some significant processes have been missed or misinterpreted, and we have few chances to study them due to fully dominating artificial factors.

Specific comments

I have some reservations about distribution of Hg in soil (p.5). Its direct link with the current emission sources implies that this is an equilibrium distribution, which, in turn,

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means that the emission has been quite stable for a relaxation time of the soil concentrations. The last assumption has to be justified \hat{U} we may be talking about Hg accumulation in soil during very long time, so the stationarity assumption may not work.

The simulations have covered just few weeks, so the influence of the meteorological conditions should be severe. Unfortunately, I have not found any analysis of meteo situation and its relation to climatological parameters.

Recomendation

Summarizing the above, I would suggest a major revision of the paper with special stress to correct representation of all involved uncertainties.

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