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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Top-down constraints on atmospheric mercury emissions and implications for global biogeochemical cycling" *by* S. Song et al.

Anonymous Referee #2

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General comments

The manuscript presents description and application of a Bayesian inversion method for the top-down estimates of Hg anthropogenic and legacy emissions on a global scale. Available inventories of Hg anthropogenic emissions, which are mostly based on the bottom-up approach, contain significant uncertainties (within a factor of 2). In its turn, this hampers correct evaluation of Hg dispersion in the environment, current and future levels of Hg exposure. Application of the inverse modelling, which is based on direct Hg measurements allows re-evaluation of Hg emissions estimates and refining the key model parameters responsible for Hg cycling between the atmosphere and the

ocean. The authors also discuss possible implications of their findings for the global Hg boigeochemical cycle and formulate priority research directions needed for further improvement of the top-down approach for Hg.

The subject of the manuscript is relevant to the scope of the journal and the work makes up a new and original contribution. The data collection and interpretation techniques are sound and the drawn conclusions are convincing and justified. The manuscript will be suitable for publication after addressing the specific comments mentioned below.

Specific comments

1. The weakest part of the paper is description of the applied inversion method. Appropriate section of the manuscript is very short and contains just very general formulas of the Bayesian inversion. There is no explanation how it was implemented for the particular task. This section should be extended with some additional information of the method application and, probably, more detailed description should be given in the Supplement. Below there are some particular issues, which require some explanation:

- How the GEOS-Chem model was used in the inversion?
- How the sensitivity matrix was calculated in practice?
- What are the dimensions and structure of the errors matrices P and R?
- What was the overall optimization procedure?

2. As it follows from the text the overall inversion procedure was divided into the 'emission inversion' and the 'parameter inversion'. The former relates to anthropogenic emissions and emission from terrestrial areas, whereas the latter optimizes parameters governing evasion from the ocean. It is not clear whether these two types of inversion were performed independently or in combination.

3. Page 19, lines 9-10. *"The parameter inversion decreases soil emission but increases Asian anthropogenic emission..."*

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How optimization of the parameters of Hg transformation in seawater can affect anthropogenic emissions? This statement needs additional explanation.

4. Page 4, line 2. *"...The concentration difference ... is usually < 1%..."*
It is not evident that the difference between GEM and TGM is mentioned here. This sentence requires some editing.

5. Page 4, lines 18-19. *"...river input may contribute to the observed summer Hg0 peak..."*
It seems some intermediate chain is missed in this statement. How the river input can contribute to air concentration? The sentence requires rewording.

6. Page 6, lines 20-23. *"We do not optimize oxidized mercury emissions ... because this form has a short atmospheric lifetime (days to weeks) and may not significantly contribute to observed Hg0"*
It is not clear how oxidized mercury can contribute to Hg0 concentration taking into account that atmospheric reduction of oxidized Hg is not included in the simulations (page 6, line 7).

7. Page 10, line 20 and hereafter. *"For simplicity they are expressed in logarithmic forms ($-logK_{OX2}$ and $logK_D$)."*
I would suggest to note explicitly that the decimal logarithm is implied here to avoid any confusion.

8. Page 12, line 4. The term 'intercomparison error' is used throughout the paper This error presents the largest part of the total observation error and is discussed as a priority aim for further research. Probably, this term requires more clear definition and discussion of its possible sources.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 5269, 2015.