Comments from referee #2:

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.

We have provided responses to each specific comment below (in blue). Our changes to the original text are shown **in bold** in the quotation.

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:

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

In the revised manuscript, we have expanded the section of inversion method in order to include the additional information mentioned in the above comments. See below for detailed responses to each point.

a. The GEOS-Chem model is used to calculate the sensitivity matrix which describes how monthly Hg^0 concentrations at different observational sites respond to changes in the emissions/parameters. To make this more clear, we have added in Sect. 2.4 that "**The GEOS-Chem model acts as a mathematical operator relating emissions/parameters to Hg^0 concentrations**". The responses to point (b) give more details of the way in which we calculated the sensitivities using the GEOS-Chem model.

b. For the calculation of the sentivitity matrix, we have added in Sect. 2.4 that "**For the emission inversion, sensitivities for the seasonal and aseasonal sources are generated by two different**

types of simulations. The aseasonal Asian anthropogenic emission is perturbed above the reference level by 50%, and we run the GEOS-Chem CTM until steady state is reached. For the seasonal sources (e.g. the NH ocean emission from March), a one-month pulse of Hg^0 is emitted, and we track modeled Hg^0 concentrations by GEOS-Chem for the next three years. After this, we assume that the perturbed concentrations at all observational sites will exponentially decrease".

c. The dimensions and structures of the error matrices (**P**, **R**, and **Q**) and the vectors ($\boldsymbol{x}$, $\boldsymbol{y}^{obs}$, and $\boldsymbol{y}^{ref}$) are explicitly given in the revised manuscript, for the emission inversion and parameter inversion. See Sects. 2.4 and 2.5: "In the emission inversion, ..., **the vector $\boldsymbol{x}$ contains 37 elements. P is a 37x37 diagonal matrix with each diagonal element equal to the square of one-sigma *a priori* error of the corresponding element in $\boldsymbol{x}$ ... $\boldsymbol{y}^{obs}$ and $\boldsymbol{y}^{ref}$ both have 12 (number of months per year) × 27 (number of observational sites) = 324 elements ... the matrix R, a diagonal 324×324 matrix, represents ... The size of Q is the same as the matrix P. Each diagonal element in Q is the square of one-sigma a posteriori error of the corresponding element in $\boldsymbol{x}$. ... In the parameter inversion, the state vector $\boldsymbol{x}$ contains 4 elements (corresponding to the 4 parameters), and P and Q are 4x4 matrices**".

d. The overall optimization procedure includes the preparations of several vectors and matrices, and the calculations of the *a posteriori* state and its error matrix based on the equations given in Sect. 2.4. We have added in Sect. 2.4 that "**As shown in Eqs. (6-7), several vectors and matrices need to be calculated during the optimization procedure, including the observational vector $\boldsymbol{y}^{obs}$ and its error matrix R, the error matrix P of the *a priori* state, the sensitivity matrix H, and the vector $\boldsymbol{y}^{ref}$ which is obtained from the reference simulation of the GEOS-Chem CTM**".

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.

The two types of inversion are performed separately. For the emission inversion, as shown in Table 2, we optimize annual Asian anthropogenic emission and monthly emissions from the ocean (further divided into two hemispheres) and soil. For the parameter inversion (see Sect. 2.5), we retain two parameters related to the soil and Asian anthropogenic emissions (ER_{Soil} and ER_{Asia}), and also include two additional parameters affecting ocean evasions. In summary, the emission inversion and parameter inversion are conducted indepedently but have some connections. We also find that they lead to similar changes in the soil and Asian anthropogenic emissions (see Sect. 3.3). To make this more clear, we have added a sentence in Sect. 2.5: "**It is noted that the emission inversion and the parameter inversion are carried out separately**."

3. Page 19, lines 9-10. "The parameter inversion decreases soil emission but increases Asian anthropogenic emission..." How optimization of the parameters of Hg transformation in seawater can affect anthropogenic emissions? This statement needs additional explanation.

As we mentioned in the response #2, in the parameter inversion, we include two parameters related to the soil and Asian anthropogenic emissions (ER_{Soil} and ER_{Asia}). In the revised manuscirpt, we have made this statement more clearly (see Sect. 3.3): "**As for the other two parameters (ER_{soil} and ER_{Asia})**, the parameter inversion ..., consistent with the emission inversion **(see Table 4)**".

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.

We have mentioned that it is the difference between GEM and TGM concentrations in this sentence (see Sect. 2.1): " The concentration difference **between measured GEM and TGM concentrations** in remote near-surface air is usually < 1%".

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.

Fisher et al. (2012) found that circumpolar rivers could deliver large quantities of mercury to the Arctic Ocean during summer, and the subsequent evasion of this riverine mercury to the atmosphere can explain the summertime peak in atmospheric mercury levels observed in Arctic. In the revised manuscript, we have edited this sentence to make it more clear (see Sect. 2.1): "Volatilization of **the** deposited Hg and **the large quantities of imported mercury from circumpolar rivers to the Arctic Ocean are hypothesized to** contribute to the observed summer Hg^0 peak **in the Arctic region**".

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).

As shown in Sect. 2.1 and Table 1, we use TGM concentration data at several observational sites. The oxidized mercury emissions may contribute a small amount to these observed TGM concentrations. In the revised manuscript, we have clarified that we are mentioning TGM concentrations (see Sect. 2.3.1): "We do not optimize oxidized mercury emissions ... because this form ... may not significantly contribute to observed **TGM concentrations**".

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

We have revised this sentence to (see Sect. 2.5) :"For simplicity they are expressed in **decimal logarithms** (-log K_{OX2} and log K_D)".

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.

In the revised manuscript, we have added a more clear definition for this term and discussed its sources (see Sect. 2.6.2): " **Here an intercomparison error (σ_{IC}) is used to represent the comparability of Hg^0 concentrations measured by different research groups using the Tekran. In principle, it includes several inaccuracies during the measurement process (e.g. the instrument's flow control and the permeation source rate for the automated calibration) and also arises from the different data management and quality control protocols taken by different research groups**".

Cited references:

Fisher, J. A., Jacob, D. J., Soerensen, A. L., Amos, H. M., Steffen, A., and Sunderland, E. M.: Riverine source of Arctic Ocean mercury inferred from atmospheric observations, Nat. Geosci., 5, 499-504, doi:10.1038/ngeo1478, 2012.