

Interactive comment on “Modeling the sensitivity of soil mercury storage to climate-induced changes in soil carbon pools” by O. Hararuk et al.

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We would like to thank reviewer 2 for the thorough feedback to the manuscript. Below find the detailed responses to all comments and suggestions (our response is in italic font).

"General comments: 1-Even though Hg has been almost only been addressed as a global contaminant in the past century of research and this climate link is novel, the former should not be out-casted completely. The authors should also consider that surface soil has (surely) received anthropogenic Hg, which might result in elevated Hg/C ratios, e.g. archives of atmospheric deposition, such as ombrothrophic peat bogs, are commonly used to trace this."

C4509

We absolutely agree with this statement. We already mention in the introduction that ‘atmospheric Hg loads are estimated to have increased three-to-five orders of magnitude’, and provide references for this, including from sediment, peat, and ice core archives (Biester et al., 2003; Fitzgerald et al., 1998; Schuster et al., 2002; Streets et al., 2011). We further clarify in the introduction that ‘large pools of past Hg pollution – or “legacy” pollution – reside in surface litter and soil horizons (Grigal, 2003; Obrist et al., 2009)’.

2-"A major flaw is that their major findings are backed up by a single reference (Natali et al., 2008) and this is not a benchmark piece (3 citations). Better arguments are needed."

There is indeed a dearth of experimental studies that has addressed effects of elevated carbon dioxide concentrations on mercury cycling. We performed a careful literature review and to our knowledge there are only these two cited studies (Natali et al., 2008, Millhollen et al 2006 which is co-authored by co-author Obrist) that have experimentally addressed effects of CO₂. This is a reason we strongly suggest in this paragraph that ‘Further studies are needed to assess additional effects that changes in CO₂ concentrations may have on Hg cycling to accurately assess effects of global change’.

3-"The manuscript diverges a bit from the figures. The model is thoroughly explained, but the spatial heterogeneity of the results is not addressed in details."

We improved the discussion of spatial heterogeneity in the text. We now clarify spatial heterogeneity in regards to the following results in the following section: (i) present-day C distribution (page 14, line 15 to page 15, line 14); (ii) present-day Hg distribution (page 16, line 9-19); (iii) response of C to temperature (page 18, line 5 to page 19, line 4); (iv) response of Hg to temperature (page 19, line 11-17); (v) response of C to precipitation (page 20, line 19 to page 21, line 23); (vi) response of Hg to precipitation (page 22, line 1, to page 22, line 16); and (vii) response of C and Hg to changes in CO₂ concentrations (Page 23, line 8 to page 24, line 10).

C4510

"References are still in Endnote style (I guess) and need to be checked."

We edited references so they are all consistent, cited correctly in the text, and correspond to the required format.

"Abstract P1119: GEOS-Chem model uses the top 15cm, why the difference?"

There are two reasons for us to use the top 40 cm of the soils. First, the empirical equations used for the prediction soil Hg is derived for the top 40 cm of soil; second, we observed significant concentrations of Hg in soils below depth of 15 cm, and we therefore preferred to include soils to a depth of 40 cm.

"P2113: Hg is not a pollutant because of its atmospheric residence time"

We rephrased this.

"P2119: put in order of magnitude"

Done as suggested.

"P2120: add/check Streets et al., 2012"

Citations added for Streets et al., 2011

"P2123: top predators and humans"

Corrected as suggested.

"P3114: bound to"

Done as suggested.

"P3120: positive correlation"

Corrected as suggested.

"P412: remove stores"

Done as suggested.

C4511

"P4112: remove highly"

Done as suggested.

"P5111: remove across the contiguous U.S."

We clarified that simulations are performed for the contiguous USA; this is necessary as CLM is often used for global simulations.

"P1019-14: needs to be shortened"

We shortened this sentence.

"P14115: duplicate"

Corrected.

"P1515: replace continent by U.S."

Done as suggested.

"P1615: address anthropogenic Hg here"

It is important to note that our study cannot distinguish between natural or anthropogenic sources, so we have to be careful to single out anthropogenic effects. We clarified that many factors (biogeochemical processes, spatial/temporal scale of investigation, regional pollution, etc.) could explain the discrepancies between observations and the model.

"P1617-21: better shown in a table"

We tried several iterations of visualizing the general agreements.

"P1711-9: too long and complicated to read, shorten"

The sentences were slightly modified.

"P17113: end phrase with simulations."

C4512

Done as suggested.

"P17117: end phrase with precipitation."

Done as suggested.

"P17122: end paragraph here."

We removed the last sentence of this paragraph.

"P1814-9: this is a figure caption not a text"

Paragraph was deleted.

"P2011-16: address oxidation-reduction briefly"

We added that the terrestrial cycling of Hg is very complex and provide a few examples of processes (including redox processes) and references (Page 20, line 13-14). We also re-iterate that our statistical approach in this study does not allow simulating individual biogeochemical processes.

"P2119: results of..."

Sentence rephrased.

"P23123: CO₂, CO₂..."

Sentence rephrased.

"P26122: go beyond and describe in detail the implication for the studies sub-areas."

We added a final paragraph where we discuss the implications for changes in soil Hg for regional changes (both for increased accumulation and losses); we also discussed what changes in terrestrial storage might have for atmospheric cycling and global re-distribution processes.

"Fig 2: poor linear correlation, explain"

C4513

We explain reasons for the fits of the linear regression on page on page 15 (lines 3-14), and page 16 (lines 1-19). Please note that using concentrations (instead of areal mass) would have increased the linear correlations, but this approach would have added other areas of uncertainties as detailed in the manuscript.

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C4514