

Reviewer comments on Shah & Jaegle 2017, "Surface deposition of oxidized mercury dominated by production in the upper and middle troposphere"

12 March 2017

General comments

The authors present a modeling study on important regions of Hg(II) production in the troposphere and how those regions contribute to surface deposition. The paper is logically organized and well written. The authors have clearly put a lot of time and thought into the analysis and writing the paper. This will make a valuable contribution to the literature. I recommend minor revisions before publication.

A few general comments to consider:

Consider placing less emphasis on findings about the middle and upper troposphere being important regions for Hg(II) production and deposition, and putting more emphasis on the importance of subtropical anticyclones. It's been established for a while that the free trop is a key region for Hg (II) (flight obs: Franz Slemr, Dan Jaffe, Seth Lyman, Murphy et al. 2006, Brooks et al. 2014; models: Selin & Jacob 2008, Holmes et al. 2010, Bieser et al. 2014, Shah et al., 2016, Horowitz et al., 2016). I would go as far as to consider changing the title of the manuscript to something about subtropical anticyclones -- that's the new, exciting piece and would draw in more readers.

The model spin-up (6 years) is less than half that of other GEOS-Chem Hg model studies (15 years; Holmes et al. 2010 and Horowitz et al. 2016). The rationale for the 15-yr spin-up provided by Holmes and Horowitz is that that's how long it takes to equilibrate the stratosphere. What's the justification for a 6-yr spin up? What are the implications if your model stratosphere hasn't reached equilibrium with the upper troposphere?

Section 3 could be improved by adding more insight and narrative. It presently feels a bit like a core dump of numbers. Having a lot of numbers can be useful, but perhaps might be better served in a table.

Section 6 *Implications* could be merged with Section 7 *Conclusions*. Combining the two sections would help trim some of the redundancy.

Line-by-line comments

Page 1

Line 18: How is "surface" defined? Is that the first level of the model? Or is it used synonymously with lower troposphere in this context?

Line 25: What accounts for the other 45%? That's surprising precip + Hg(II) production only account for 55%.

Lines 27-28: Statement is unclear. Is there a word missing? "Our simulation points to a large role of Hg(II) present in the dry subtropical subsidence regions..." Confused about the role of Hg(II).

Line 31: "Contribution of these dry regions..." Unclear what the dry regions are contributing to. Hg(II) concentrations? Hg(II) mass in the free troposphere?

Lines 32-34: "Our results highlight the importance of the upper and middle troposphere as key regions for Hg(II) production and of the subtropical anticyclones as the primary conduits for the production and export of Hg(II) to the global atmosphere." I might delete or reword the underlined part. The subtropical anticyclone part is new. I'd play that up in the abstract.

Page 2

Line 4: Recommend amending the sentence to say "most aquatic ecosystems".

Line 9: "Global dry deposition fluxes of gaseous elemental mercury (Hg(0)) and oxidized mercury in the gas and particle phases (Hg(II)) are comparable." Needs a citation. Jeroen Sonke's group published work in 2015 or 2016 looking at dry dep in peat. How does your statement line up with the Sonke lab's peat findings?

Line 16: Sproveiri et al. 2010 is a relevant citation.

Line 30: Please quantify "clean" and "dry".

Page 5

Lines 3-4: "We assume that stack emissions (emission height > 50m) of Hg consist of 90% Hg(0) and 10% Hg(II)." Needs some justification. Even better if you can include a citation.

Page 6

Line 27: Are the assumptions about Hg wet scavenging on lines 15-20 relevant? "Below clouds, gas-phase Hg(II) is washed out by dissolving in falling raindrops ($T > 268K$), but not in falling snow and ice (Amos et al., 2012). Particle-phase Hg(II) is washed out in collisions in falling rain, snow and ice with different efficiencies (Wang et al., 2011)."

Page 7

Lines 10-11: "We adjust the reduction rate to best match aircraft- and ground-based observations of Hg(0) over the mid-latitudes." What rate did you come up with? How does that compare to previous GEOS-Chem modeling studies?

Line 28-29: "...model spin-up period of six years." Is 6 years long enough to spin up the stratosphere? Holmes et al. (2010) and Horowitz et al. (2016) had to initialize their GEOS-Chem simulations with a 15-yr spin-up to equilibrate the stratosphere.

Page 8

Line 3: How does the subtropical subsidence in 2013 compare to other years? Was this a dry year with lots of subsidence? Or an average year? A sense of the interannual variability would be helpful.

Page 11

Line 28: "...while the contribution from E-Hg(II) is noticeable mainly in East Asia." Please quantify "noticeable".

Page 13

Line 13: Please quantify "strong influence".

Line 18: Please quantify "small".

Lines 20-23: How much confidence can be placed in the statement, "Surface Hg(II) in areas poleward of 40° is from anthropogenic emissions (Europe), is produced locally (polar regions)..." give that you have a step function in Br-concentrations at 45 N (Figure 4)?