Response to referee comments

We thank the referee for their thorough review and helpful suggestions. The reviewer's comments (in black) and our responses follow.

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.

Excellent suggestion. We have revised the title to: "**Subtropical subsidence and surface deposition of oxidized mercury produced in the free troposphere.**" We have also extended our "dry-Hg(II)" simulation from one year to four (2013-2016), to get a sense of the interannual variability. We have also modified the abstract and the text to place more emphasis on the subtropical anticyclones.

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?

In the revised manuscript, we have conducted a 15-year spin up period and have updated the figures and tables accordingly. The revised version of Table 1 is below. We see only a small (5% or lower) change in the tropospheric budgets of the STRAT, UT, and MT tracers.

Table 1 Tropospheric budgets of Hg(II) and individual tagged Hg(II) tracers.

	Total Hg(II)	Tagged Hg(II) tracers[a]				
		UT	MT	LT	STRAT	E-Hg(II)
Tropospheric mass of Hg(II)[b] [Mg]	618	517	48	4	48	1
Mass located in UT [Mg]	480	432	3	0	45	0
Mass located in MT [Mg]	118	79	36	0	3	0
Mass located in LT [Mg]	20	7	8	4	0	1
Hg(II) production[b] [Mg a^{-1}]	15,790	8,560	4,190	2,460	410	170
Hg(II) reduction [Mg a^{-1}]	9,740	5,750	2,390	1,260	290	50
Hg(II) wet deposition [Mg a^{-1}]	3,740	2,250	1,150	230	80	30
Hg(II) dry deposition [Mg a^{-1}]	2,310	570	640	970	40	90
Hg(II) tropospheric lifetime [days]	14	22	4.1	0.6	43	2.2

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.
Good suggestion. We have deleted some of the redundant numbers from Section 3.1 that were already listed in Table 1, and added more explanation of our results. We have also added another table (see below) to summarize the results of the sensitivity simulations and reduce the numbers written in Sect. 3.3.

Table 2 Contribution of tagged Hg(II) tracers to the tropospheric mass and total deposition of Hg(II) for the base case and the sensitivity simulations.

	Simulation	Tagged Hg(II) tracer contribution [%]				
		UT	MT	LT	STRAT	E-Hg(II)
Contribution to Hg(II) tropospheric mass [%]	Base	84	8	<1	8	<1
	Lower UT+MT Br[a]	71	7	1	21	<1
	O_3/OH oxidation[b]	61	18	4	17	<1
	Higher Hg(II) emissions[c]	84	8	<1	8	<1
Contribution to Hg(II) deposition [%]	Base	47	30	19	2	2
	Lower UT+MT Br[a]	43	21	27	6	3
	O_3/OH oxidation[b]	20	38	38	2	2
	Higher Hg(II) emissions[c]	49	28	17	2	4

(a) Simulation using the original GEOS-Chem Br concentrations instead of the 3 times Br concentrations in the base simulation,
(b) Simulation using O_3 and OH as the Hg(0) oxidants instead of Br as in the base simulation,
(c) Simulation using the default UNEP/AMAP Hg(0):Hg(II) emission speciation of 55%:45% instead of the 90%:10% speciation as in the base simulation.

Section 6 *Implications* could be merged with Section 7 *Conclusions*. Combining the two sections would help trim some of the redundancy.
Following your suggestion, we have trimmed Sect. 6 of the most of the redundancy, but have decided to keep the *Implications* separate from the *Conclusions*.

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 is this context?
The surface is defined here as the first level in the model.

Line 25: What accounts for the other 45%? That's surprising precip + Hg(II) production only account for 55%.
We don't know. It could be because of variations in the amount of Hg(II) in the precipitating column, caused by spatial variation in production and loss rates. Hg(II) column amounts in the model are only moderately correlated to the *contribution* of the UT+MT tracers. They are almost perfectly correlated, of course, with the total amounts of UT+MT tracers.

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).
We have modified the text as follows: "Our simulation points to a large role of ~~Hg(II) present in~~ the dry subtropical subsidence regions. Hg(II) present in these regions ~~which~~ accounts for…"

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?
Modified to: "…the contribution of Hg(II) from the ~~these~~ dry subtropical regions was found…"

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.
We have deleted the underlined phrase to emphasize the anticyclone part. The modified sentence is "Our results highlight the importance the subtropical anticyclones as the primary conduits for the production and export of Hg(II) to the global atmosphere."

Page 2
Line 4: Recommend amending the sentence to say "most aquatic ecosystems".
We have modified the text accordingly.

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?
Our statement is based on model estimates, and we have added relevant citations.

Line 16: Sproveiri et al. 2010 is a relevant citation.
Agreed. We have added the citation.

Line 30: Please quantify "clean" and "dry".
Clean and dry is defined as RH below 35% and CO below 75 ppbv. We have specified this in the text now.

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.
We have some discussion about this assumption in Sect 2.2.1, including citations. We have added a reference to Sect. 2.2.1 to the sentence in question here.

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)."
Since Hg(II) wet deposition is an important part of our work, we wanted to state all relevant model assumptions in the manuscript. These assumptions affect the simulated wet deposition flux and the vertical distribution of Hg(II).

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?
The reduction rate is scaled to the photolysis rate of NO_2. We use a scaling coefficient of 0.1, which is a 16 times higher than the reduction rate used by Zhang et al. (2012).

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.
We have now extended the spin-up period to 15 years.

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.
To provide a sense of the interannual variability, we have extended our "dry-Hg(II)" simulation from one year to four, and have shown the variation (as anomaly) in Fig. 7. The revised Fig. 7 is below:

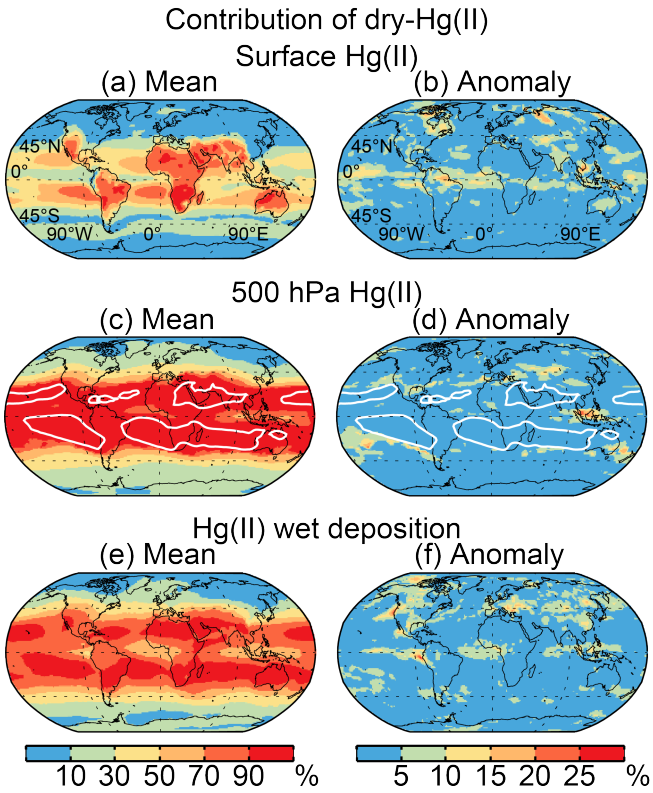

Figure 7: Mean and anomaly (maximum deviation from the mean) of the contributions of dry-Hg(II) to (a,b) surface Hg(II) concentrations, (c,d) 500 hPa Hg(II) concentrations, and (e,f) Hg(II) wet deposition flux for 2013-2016. The white contours in (c,d) show the boundaries at 500 hPa for areas with 2013-2016 RH less than 20% for a minimum of four months of the year.

Page 11
Line 28: "...while the contribution from E-Hg(II) is noticeable mainly in East Asia." Please quantify "noticeable".
We've changed the sentence as follows: "is greater than 10% mainly in over East Asia"

Page 13
Line 13: Please quantify "strong influence". Line 18: Please quantify "small".
The contribution of Hg(II) to surface deposition is 45 ± 25%.
We have added the following text to Line 13 quantifying the strong influence. "We see from Fig. 7a that dry-Hg(II) exerts a disproportionate influence on surface Hg(II) concentrations between 40°S and 40°N, where its contribution is 45 ± 25%."
And, on Line 18, by small we mean <20%. This has been clarified in the revised manuscript.

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)?
This higher Br in the subtropics should increase, if anything, the contribution of dry-Hg(II) poleward of 40°. That we don't see much of a contribution of dry-Hg(II), indicates other processes are involved. This statement relies on both Figures 5 and 7, and to clarify this we are citing both figures in the revised manuscript.

References:

Zhang, Y., Jaeglé, L., van Donkelaar, A., Martin, R. V., Holmes, C. D., Amos, H. M., Wang, Q., Talbot, R., Artz, R., Brooks, S., Luke, W., Holsen, T. M., Felton, D., Miller, E. K., Perry, K. D., Schmeltz, D., Steffen, A., Tordon, R., Weiss-Penzias, P. and Zsolway, R.: Nested-grid simulation of mercury over North America, Atmospheric Chem. Phys., 12(14), 6095–6111, doi:10.5194/acp-12-6095-2012, 2012.