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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Surface deposition of oxidized mercury dominated by production in the upper and middle troposphere" *by* Viral Shah and Lyatt Jaeglé

Anonymous Referee #1

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General comments

This paper investigates – using the GEOS-Chem global chemical transport model - how surface deposition of divalent mercury species (Hg(II)) is influenced by Hg(II) production at different atmospheric heights. The authors show that surface deposition is dominated by production in the upper and middle troposphere and highlight the large role of subtropical anticyclones as a global reservoir of Hg(II). This study also shows that regional decreases in anthropogenic mercury emissions will not lead to a proportional regional decrease in wet deposition. The paper is organized clearly, easy to follow, well written, and will make a valuable contribution to the literature. However, I find the evaluation of the model with observations insufficient and not up to date. This

paper will be suitable for publication after the authors address the following issues.

Major comments: Comparison with observations

The two-year simulation (2012-2014) is evaluated with ground-based observations of Hg(II) concentrations and wet deposition. Section 2.2.3 concludes that the simulation reproduces quite well the spatial distribution and seasonal cycle of Hg(II) and wet deposition over the US but displays a 46% underestimate of wet deposition observed at EMEP sites. So what? How might this uncertainty affect the distribution of the tagged Hg(II) and ultimately their contributions to wet/dry distribution fluxes in different regions of the world? Additionally, the model is evaluated over the US and Europe only, using ground-based observations. The authors should consider using recent data from ground-based sites, aircraft campaigns and high-altitude sites to evaluate the model in different regions of the world and at different heights. To me, evaluating a model used to investigate the global distribution of Hg(II) at different heights a) over the US only, and b) at ground level only is not convincing enough.

1. Ground-based observations

1.1 Hg(II) concentrations The authors use the 2009-2012 AMNet observations to evaluate the model over the US. I understand that the authors use data that are publicly available. However, evaluating 2013-2014 model outputs with 2009-2012 observations is not satisfying unless inter-annual variability is discussed at some point. In Europe, the authors highlight a discrepancy between modeled/observed wet deposition and suggest that this could "indicate an underestimate in the modeled Hg(II) concentrations over the region". The authors could easily check that since Hg(II) data are available for 2013-2014 (Sprovieri et al., 2016) at Iskrba (Slovenia), Longobucco (Italy), and Rao (Sweden – see also Wängberg et al., 2016). Additionally, how well can the model reproduce Hg(II) concentrations elsewhere? Still according to Sprovieri et al. (2016), Hg(II) data are available around the world for years 2013-2014 at Amsterdam Island (see also Angot et al., 2014), Bariloche (Argentina), Cape Hedo (Japan), Manaus (Brazil), and

Minamata (Japan).

1.2 Wet deposition Same as above, why don't the authors use recent wet deposition data collected around the world to evaluate the model in different regions of the world? A recent paper (Sprovieri et al., 2017) present seasonal and annual variations of Hg wet deposition and concentration collected at 17 ground-based sites in the Northern and Southern Hemispheres as part of the GMOS project. Additionally, page 9, lines 2-4: "Over the southeast US, the modeled VWM concentrations are higher than observations during winter and spring, suggesting a model overestimate in atmospheric Hg(II) concentrations in that region or an overestimate in the amount of Hg(II) scavenged by precipitation". If the model overestimates the amount of Hg(II) scavenged by precipitation, what is the possible influence on results presented in section 3.2, i.e. on the modeled contribution of MT and UT? I would like to see a discussion on how results presented in section 2.2.3 (comparison of modeled and measured Hg(II)) affect results presented thereafter.

2. Vertical profiles

The authors should consider using recent data from aircraft campaigns and high-elevation sites to evaluate the model in different regions of the world. How well can the model reproduce these observations (see for instance Bieser et al., 2016).

2.1 Aircraft campaigns An evaluation of the model is done, over the US, in a previous paper (Shah et al., 2016) during the NOMADSS campaign. The authors could refer to this paper here. Within the GMOS project, vertical profiles were taken on board research aircraft in August 2013 in background air over different locations in Slovenia and Germany (Weigelt et al., 2016). Additionally, Hg(0), Hg(II), and Hg(p) profiles were collected on 28 flights between August 2012 and July 2013 (1000 to 6000 m, Brooks et al., 2014). Finally, the authors could use data from the intercontinental flights between Germany and North/South America under the umbrella of the CARABIC project (Slemr et al., 2014, 2016).

2.2 High-elevation ground sites The authors could use data collected at various high-elevation sites such as Mt. Walinguan (China), Mt. Ailao (China), Kodaicanal (India), Everest/K2 (Nepal) and Col Margherita (Italy) (Sprovieri et al., 2016) to evaluate Hg(0) and/or Hg(II) concentrations. Note that mercury data discussed in this paper are available upon request at: http://sdi.iia.cnr.it/geoint/publicpage/GMOS/gmos.historical.zul.

Other comments: Model sensitivity to oxidation chemistry and emission speciation

The authors perform an additional one-year sensitivity simulation using the original GEOS-Chem Br concentrations instead of the 3 times Br concentrations in the base simulation. Given that updates by Schmidt et al. (2016) have resulted in an improved agreement with satellite and in situ observation of BrO, I wonder why the authors did not perform an additional simulation using these updated fields. Page 9, line 17: "suggesting that the modeled oxidation rate is too slow over this region". Using Br fields from Schmidt et al. (2016), i.e., a factor 2.3 increase in free tropospheric Br concentrations north of 45N might lead to a better agreement between modeled/observed data over Europe.

Page 12, lines 24-33. How do these results compare to the results by Bieser et al. (2016)? According to the latter, "high RM concentrations in the UT could be reproduced by oxidation by Br while elevated concentrations in the LT were better reproduced by OH and ozone". Does it sound feasible and adequate to implement two different mechanisms in GEOS-Chem depending on the altitude?

Line by line comments

Section 2.2: Which version of GEOS-Chem do you use?

Page 8, lines 18-20: "The model reproduces the observed seasonal variations in the central and northeast regions, but underestimates the summer deposition fluxes in the southeast because of a factor of 2 underestimate in summertime precipitation by the GEOS-FP meteorological fields". Is that also the case for other (GEOS-5, MERRA)

meteorological fields? If not, why don't the authors use them? MERRA meteorological data are available for 2013-2014.

Page 9, line 2: there is a typo "Over the southeast US, tmodeled (. . .)".

Page 9, lines 10-12: "(. . .) likely because the upward scaling of the Br concentrations in our simulation did not extend north of 45N and covered only parts of Southern Europe". Could you please add the latitude on the various figures?

Figure 4e: I am just curious; how can you explain the elevated contribution of MT tracer over the Antarctic continent?

Figure 10b: Why is NY95 excluded from the regression calculation? I agree that it is an outlier here, but the question is why? According to info found on AMNet website (and not in the paper. . .), the collection of Hg(II) concentrations stopped in November 2009 at this site. This suggests that the authors only have a few months of data at this site, and not data for the entire 2009-2012 period. That kind of information would be useful (in supplementary?) in order to get a better insight on which observation data are used to evaluate the model.

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