

Interactive comment on “Global Observations and Modeling of Atmosphere-Surface Exchange of Elemental Mercury – A Critical Review” by W. Zhu et al.

Anonymous Referee #1

Received and published: 2 February 2016

Overall comments: Overall this is a very well written and fully researched review paper. In addition to simply reviewing existing studies, this paper performs original analysis of the compiled datasets in order to make large scale observations. This paper provides a comprehensive assessment of the current understanding of the atmospheric-surface exchange of Hg and recommend it for publication in Atmospheric Chemistry and Physics.

The only complicating factor with the publication of this paper is that a separate group published a fairly similar paper very recently: Agnan et al., New Constraints on Terrestrial Surface-Atmosphere Fluxes of Gaseous Elemental Mercury Using a Global Database. ES&T 2016 Abstract Despite 30 years of study, gaseous elemental mercury

C1

(Hg(0)) exchange magnitude and controls between terrestrial surfaces and the atmosphere still remain uncertain. We compiled data from 132 studies, including 1290 reported fluxes from more than 200 000 individual measurements, into a database to statistically examine flux magnitudes and controls. We found that fluxes were unevenly distributed, both spatially and temporally, with strong biases toward Hg-enriched sites, daytime and summertime measurements. Fluxes at Hg-enriched sites were positively correlated with substrate concentrations, but this was absent at background sites. Median fluxes over litter- and snow-covered soils were lower than over bare soils, and chamber measurements showed higher emission compared to micrometeorological measurements. Due to low spatial extent, estimated emissions from Hg-enriched areas (217 Mg Åa(-1)) were lower than previous estimates. Globally, areas with enhanced atmospheric Hg(0) levels (particularly East Asia) showed an emerging importance of Hg(0) emissions accounting for half of the total global emissions estimated at 607 Mg Åa(-1), although with a large uncertainty range (-513 to 1353 Mg Åa(-1) [range of 37.5th and 62.5th percentiles]). The largest uncertainties in Hg(0) fluxes stem from forests (-513 to 1353 Mg Åa(-1) [range of 37.5th and 62.5th percentiles]), largely driven by a shortage of whole-ecosystem fluxes and uncertain contributions of leaf-atmosphere exchanges, questioning to what degree ecosystems are net sinks or sources of atmospheric Hg(0).

Given the similarities in objective and scope, I think this paper by Zhu et al, needs to: 1) acknowledge this separate paper in results and discussion and 2) specifically identify how their paper is unique from Agnan et al., 2016, and 3) discuss similarities and differences in the two papers findings.

Specific comments: Line 261: Very interesting result. Glad to see this analysis. A little more information is needed. The samples sizes are 229 and 39, but it is not clear if these numbers represent daily average values, hourly values, etc. A little more discussion about what constitutes a measurement would be helpful. Also, within the <0.3 ug/g cutoff, were there significant differences in the Hg concentrations between

C2

DFC and MM areas? If not, this would help build the case for the analysis.

Line 262: Why was 0.3 ug/g used as a cut-off point.

Line 288: change matters to matter.

Line 285: Two factors that have been shown to affect soil-air Hg fluxes are grain size and soil disturbance. Only a couple of studies have shown this, but may want to consider including these two factors in the discussion if the goal is to be comprehensive as possible.

Line 339. There is a paper by Mazur et al. 2014 in Science of the Total Environment that has a similar focus: the impact of forestry operations on surface-air Hg fluxes.

Line 346: Suggest changing to "more recent" instead of just "recent". This idea has been around for more than a decade now.

Line 352: remove excess Hg₀.

Line 422: Need more information to support this statement. Earlier the text focuses on photo-pathways and this is a big jump without sufficient explanation.

Line 472: remove "got flux calculation".

Line 476: in "the" literature

Line 558: This paragraph should also discuss the work of Kuiken et al, 2008 part 1, which shows the opposite trend...lower emission in summer due to drier conditions and lower light from more leaf cover. In the scaling paper, Hartman et al, 2009 comes to the same conclusion.

Line 601. Double check that Gustin et al, 2003 used a multivariate approach using soil Hg, flux and solar radiation. Or did that paper look at these variables separately.

Line 700. Remove "in"

Line 713. This is a great summary of knowledge gaps, glad to see this in the paper.

C3

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2015-1064, 2016.

C4