Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-314-AC2, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Emission-dominated gas exchange of elemental mercury vapor over natural surfaces in China" *by* Xun Wang et al.

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Received and published: 7 August 2016

This paper provides an updated model of estimating the exchange of gaseous elemental mercury between the atmosphere and underlying surfaces in China. This topic is relevant to the scope of ACP and of great importance as very high uncertainty still exists in its estimate and it is closely related to Hg input into the ecosystem in China and Hg export to downwind regions. In general, the manuscript is very well written. I suggest the acceptance of this paper after the authors addressing the following general and specific comments. The manuscript also needs a careful proofread to correct many grammatical errors.

Response: We thank the reviewer for recognizing the contribution of our work and for the constructive comments, which have been addressed in our response below and in

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the revised manuscript. We also appreciate the reviewer for taking the time to provide the editorial comment.

General Comments:

1. In this paper, the "natural" emission refers to the sum of the primary natural activities (i.e. geogenic) and the re-emission of legacy Hg stored in the terrestrial and water surfaces. However, in some other papers, the "natural" emission only refers to geogenic activities. This may cause some confusion. I suggest a clear definition of the "natural" emission be given in the introduction section.

Response: We totally agree with the reviewer on this comment and have clarified this in Line 41-44: The inventories of Hg emission include the emission from anthropogenic sources, and the so-called "natural" emission. In this study, the term "natural emission" refers to the sum of the primary natural release (i.e., from geogenic activities) and the re-emission of legacy Hg stored in the terrestrial and water surfaces, because the geogenic release and re-emission cannot be separated analytically.

2. There is a logical question. It is mentioned in the paper that "the feedback of the air-surface exchange to the air concentration does not significantly modify the atmospheric Hg0 concentration" (Lines 226-229), and thus I think that a different air-surface exchange mechanism may not significantly affect the outflow of Hg from China. But the authors do argue in the paper that the effect may be large.

Response: We understand the reviewer's point and wat to clarify here. In short, the total mass of release is significant but the concentration forced by the release is insignificant. Outflow is a mass budget and therefore will be affected by the quantity of Hg natural emission. Given the $\sim$ 1 km mixing layer for each grid cell, the feedback of the air-surface exchange to the air Hg concentration is 0.01-0.1 ng m-3. However, if we consider the area of our domain (9,600,000,000,000 m2), and the several days of Hg0 residence time in our domain, the Hg output during this period can be up to several tons (Mg).

3. The updated soil Hg concentration dataset is based on the NMPRGS survey. What is the depth of soil sampling, 0-10 cm? 0-20 cm? It seems that most of HgII reduction occurs within the very top soil layer. What is the vertical profile of Hg concentration?

Response: The surface soil depth is 0-20 cm. Though the mean Hg concentration in 0-20 cm soil profile could somewhat underestimates Hg concentration in the top soil layer, the dataset is the best available one describing the soil Hg concentration in China. We have made this clear in the revised manuscript (Line 223-226): Datasets of Hg concentration in the top soil layer (e.g., 0-5 cm depth) are recommended for simulations when they become available.

4. It may be beyond the scope of this specific paper, but I wonder is it possible to calculate an uncertainty range of natural Hg emission?

Response: That is a very good suggestion. Mathematically, it is possible to calculate the uncertainty for a mechanistic model. However, it is beyond the scope of this study because the uncertainty is controlled by the input parameters/datasets, and the uncertainty for certain dataset has not been systematically quantified, the uncertainty range for the Hg concentration in surface soil. With that said, on the basis of the mode verification results, the estimates in this study is generally in agreement with the field observations.

Specific Comments:

Lines 51-52: The trend of future anthropogenic Hg emission depends on many factors and the Minamata Convention does not necessarily lead to a decrease. Also, the reference De Simone et al. 2015 is not relevant here.

Response: We agree that future anthropogenic Hg emission depends on many factors. We have replaced this reference by Pacyna et al. (2016) and revised the sentence as “In light of the control of anthropogenic Hg emission by the legally binding Minamata Convention, a better quantification of natural Hg emission is critical in evaluating the

effectiveness of policy actions (Selin, 2009;Pirrone et al., 2010;Song et al., 2015)" in Line 51-54.

Lines 195-197: Is the value of the parameter of soil interfaces compensation point (3 ng/m3) only based on the measurements at air-snow interface? What about other types of underlying surfaces?

Response: Yes, the measured compensation point (3 ng/m3) is only for the surface covered by snow. The compensation point for other types of soil surface is calculated based on Equations 1-18. We have clarified in Line 194-195 : The χ_g for the air-snow interface is assumed to be 3 ng m-3 based on field measurements at air-snow interface.

Lines 224-226: Are the CMAQ modeled atmospheric Hg0 concentrations validated against real world observations in China?

Response: Yes, the CMAQ model results have been verified. More details of the model simulations are presented in Lin et al. (2010).

Lines 248-251: Do the measurements in the surface forest floor have similar environmental conditions with the modeling? In addition, the average measured concentration of 4.1 ng/m3 seems to be very high, if we consider vegetation serves as a large sink.

Response: Yes, the measurements in the surface forest floor have similar environmental conditions to the modeling, and we have clarified this in Line 822-823. We want to clarify here the case in which the mean measured concentration is 4.1 ng/m3. This is the Hg0 vapor concentration in soil porous media, not the concentration in ambient air.

Lines 257: What does the 1-2 ng/m3 refer to? Is it the difference between soil and air Hg concentrations? Response: It refers to the atmospheric Hg concentration at background site. To clarify it, we have revised the sentence as "Hg0 soil gas concentrations are typically lower than the 1-2 ng m-3 atmospheric Hg concentration in background forest ecosystems at night." (Line 259-260)

Technical Corrections: Line 64: “may be appropriate”; Response: The wording has been revised as suggested. Line 69: “estimate”; Response: The wording has been revised as suggested. Line 96: “at a resolution of”; Response: The wording has been revised as suggested. Line 98: “a recent database”; Response: The wording has been revised as suggested. Line 199: “at a 36-km”; Response: The wording has been revised as suggested. Line 286: “can improve”; Response: The wording has been revised as suggested. Line 402: “are more comprehensive”; Response: The wording has been revised as suggested. Line 481: “to earlier estimates”; Response: The wording has been revised as suggested. Line 487: “also enhances”; Response: The wording has been revised as suggested. Figure 9.4: missing legend; Response: The missing legend has been added in Figure 8.4.

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