

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

**Xun Wang et al.**

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The paper proposes a new parametrization of surface fluxes of elemental mercury based on different pathways of reduction of reactive mercury. The model is applied to China, taking into account landuse patterns to estimate emissions of elemental mercury.

Response: We thank the reviewer for recognizing the new contribution of our work, and appreciate the reviewer's constructive comments.

The main problem with the study is the minimal evaluation of the model and the lack of details concerning the sensitivity studies. The organization of the results section was puzzling: Sec 3.4 has the evaluation of the model, which I would have expected at the beginning of Section 3. I would have liked to see more details about the evaluation.

Response: We thank reviewer for the suggestion and have provided additional data in Figures S8-S9 to address the reviewer's concern regarding model evaluation. We respectfully disagree regarding the organization of the section describing the results. Compared to earlier air-surface models (Bash, 2010; Wang et al., 2014), we built a new scheme for air-soil flux exchange. We think the evaluation for this scheme is necessary before running the air-surface exchange model and choose to present the evaluation results of individual model components in section 3.1, and show the verification results after multiple model components are integrated in section 3.4.

Figure 8 shows only measurements and was difficult to figure out, it needs a better legend (eg. Units) and the information should be presented in such a way as to help evaluate the model results. Figure 9 contains the information for evaluating the model, but it is difficult to get a clear sense of model performance from this.

Response: We have provided additional data in Figures S8-S9 to address such concerns. Figure S8 and S9 display the comparison for the air-foilage flux exchange by the model and field data of Hg deposition through litterfall. We have revised this section in Line 441-452 as: "Figures S8, S9 and 8.1 compare the model estimates to the mean and variability level predicted by Monte Carlo simulation using field data. The annual Hg uptake simulated by the bidirectional exchange model is not significantly different from the field observations ( $p > 0.05$ , t-test) and the spatial patterns are similar (Figure S8) in coniferous forest ecosystems, demonstrating the model capability for simulating the air-foilage flux. However, the bidirectional exchange model did not capture the spatial distribution of air-foilage flux in broadleaf forest ecosystems (particularly in evergreen broadleaf forest, Figure S9). One possible explanation is that the resistance terms obtained from temperate/boreal forests (Zhang et al., 2012b) may not appropriately represent the value in evergreen broadleaf forests. Filed measurements suggests that the leaf stomatal conductance of broadleaf is usually higher than the value of needleleaf (Wang et al., 2015; Ishida et al., 2006; Sobrado, 1991; Eamus, 1999), leading to a larger air-foilage Hg<sub>0</sub> exchange (Graydon et al., 2006). Further studies on the

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Hg transport and chemical reactions at the air-foilage interface in evergreen broadleaf forests will help constrain the model. ”

The sensitivity study seemed very limited in scope, with only a low and a high level. It seems there could be a more thorough way of doing this.

Response: We thank the reviewer for pointing this out and want to clarify here. The 2-level factorial design of experiments is meant to gauge the extreme variation of flux caused by the possible range of all parameters. This method is statistically robust, and therefore the synergistic and antagonistic interactions among model parameters can be estimated with indications of statistical significance. In reality, since the actual variation of the parameters is much smaller than the possible range, the flux change will also be much milder. To illustrate this, we run the model using the center value (i.e., showing the model results by running the model at half of the experimental level). By using the center values of soil Hg content, LAI, soil bulk density, solar radiation and soil temperature in Table 2 (close to the environmental parameters in a typical forest ecosystem), the air-soil flux is  $4.5 \text{ ng m}^{-2} \text{ hr}^{-1}$ . Such flux is close to the measures fluxes ( $0.5\text{-}9.3 \text{ ng m}^{-2} \text{ hr}^{-1}$ ) in forest ecosystems of China (Fu et al., 2012;Fu et al., 2008). We have supplement the above discussion in the Line 267-287.

Figure 2 was difficult to see – cross-sections would probably be preferable.

Response: We have added the cross-sections for a better presentation.

For Figure 3, I was surprised at the magnitude of the changes (around  $100 \text{ ng m}^{-2} \text{ hr}^{-1}$ ) when the fluxes listed in Table 3 are 1 to 2 orders of magnitude smaller.

Response: We carefully checked the process of the sensitivity analysis, and found that we omitted the parameter of ratio of UV radiation over total radiation (Table 1). In revised Figure 3, the flux change is about from  $20\text{-}30 \text{ ng m}^{-2} \text{ h}^{-1}$ . Filed measurements suggest the combined effects of soil Hg content (from  $60$  to  $590 \text{ ng g}^{-1}$ ) and soil temperature (from  $5$  to  $30 \text{ }^{\circ}\text{C}$ ) enhance the flux by  $\sim 40 \text{ ng m}^{-2} \text{ hr}^{-1}$  (Fu et al., 2012;Fu

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et al., 2008). (Line 274-276).

Because the model is very specific in inputs, it seems to me that the model development part requires a very specific evaluation which is distinct from the application of the model on the national scale. The paper therefore seems to be a curious combination of 2 papers: one paper on model development and one on application of the model to a national scale. However, I think the paper would be acceptable with an expanded description of the model evaluation and an improved sensitivity analysis.

Response: We appreciate the reviewer's suggestion and want to clarify here. We agree that a specific evaluation for the model development is necessary and this have been thoroughly discussed in section 3.1 (Line 238-296). It is our view that a comprehensive paper describing the model development and application serve readers better.

Specific comments:

Sec 2.1.1: It would be good to explain how the model differs from prior work in more detail.

Response: We agree with the reviewer on the comment and have added a section for this purpose in Line 111-115: "Compared to the earlier mechanistic schemes (Wang et al., 2014; Bash, 2010; Scholtz et al., 2003; Zhang et al., 2012), this model (1) builds a new scheme for the air-soil flux based on the reduction pathways of reactive Hg in soil identified in the literature, (2) develops a scheme for the Hg flux exchange in rice paddy, which is an important landuse feature in China, and (3) updates the scheme for the air-snow interface and chemical parameters for air-foilage flux (Table 1)."

Sec 3.1: It is preferable to talk about "evaluation" rather than "verification." Model evaluation seems to be in Sec 3.4. Sec 3.1 seems to be a comparison with other studies – a graphical representation may help some of the discussion.

Response: We thank the reviewer for the constructive suggestion. We have used the evaluation of the air-soil flux scheme in section 3.1 of revised manuscript.

Line 222: Putting uncertainty on the bounds of the ranges seemed like an odd thing to do. Isn't it enough to state the range? Response: Thanks for constructive suggestions. This sentence has been revised as "The soil Hg content in 0-20 cm surface soil varies with landuse types, containing mean concentrations of 119~211, 61~197, 80~82, 80~82 and 31~162 of Hg for forest ecosystems".

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