

Interactive comment on “Atmospheric wet and litterfall mercury deposition in typical rural and urban areas in China” by Xuewu Fu et al.

Xuewu Fu et al.

fengxinbin@vip.skleg.cn

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RC- Reviewer's Comments; AC – Authors' Comments

RC: This study measured the Hg concentrations and deposition fluxes in precipitation and litterfall at six rural sites and an urban site across a broad geographic area in China. This is an important study and the results help us to better understand the spatial distributions of Hg deposition and mass balance of atmospheric Hg in China. The manuscript is generally well written. However, there are several issues to be clarified before the paper can be published at ACP. Some specific comments are given below.

AC: We would like to acknowledge the anonymous reviewer for dictating the time to read our original manuscript and provide valuable suggestions and editorial remarks. These suggestions are very helpful and constructive. We have made careful revision

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in the revised manuscript based on the reviewer's recommendations.

RC: 1. Line 145-146: Please give details of the sampling method, such as the type of wet-only collector used, sampling method, and key parameters of the collector.

AC: the sampling method was specified in line 145-168 on page 6 in the revised manuscript.

RC: 2. Line 149-150: The authors said “Before each of the new sampling cycle, the borosilicate glass sampler was rigorously rinsed by Milli-Q water.” Is the rinsing of Milli-Q water ENOUGH to clean the sampler?

AC: we agree with the reviewer that it will be better using diluted HCl solution rather than Milli-Q water to clean the collector. In this study, the collector was a borosilicate glass bucket. It is characterized by simple structure and was clean during field sampling (was baked for one hour in a muffle furnace at a temperature of 500 °C before field sampling and kept sealed during dry period). We therefore suggest it may have a minor adsorption of Hg and small cross contamination between samples. The filed blank was not conducted in this study, but a previous study using the similar collector observed a low filed blank (≈ 0.36 ng L⁻¹) (Burke et al., 1995). We therefore speculate that the sampling method used in this study would have minor artifacts.

RC: 3. Line 178-180: Please give details of the litterfall collectors, such as the design of the collector. The authors said the litterfall collectors are 0.25 to 1.0 m². Why do not use same collectors? Why did you use four, four, three, and eight collectors instead of same number of collectors at the MCB, MDM, MLG, and MAL sites?

AC: the design of the litterfall collectors were shown in line 195-198 on page 7 and 8 in the revised manuscript. There is no standard litterfall collectors in the studies of litterfall deposition. For example, the sample box of litterfall collectors used in the North America ranged from 0.06 to 0.25 m² (Graydon et al., 2008; Fisher and Wolfe, 2012; Risch et al., 2012). That indicates the size may not lead to significant artifacts

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during sampling of litterfall. The four forests in this study have different vegetation structures and species, that is why we use different number collectors at the sampling sites.

RC: 4. Line 224-226: This sentence says “VWM Hg concentrations in precipitation at all sites showed a clear season trend with higher concentrations in summer wet season and lower concentrations in winter dry season”. However, the following discussions are totally opposite (higher Hg concentration in winter). Please clarify.

AC: yes, this paragraph in line 224-226 in the original manuscript had the error. We changed it in line 254 on page 10 in the revised manuscript

RC: 5. Line 227-231: 1) Does snow contain higher Hg? It will be interesting to see the Hg concentration data in snow only compared with that in rainfall. 2) The higher Hg concentrations in winter might be affected by the higher Hg emissions from coal combustion for heating in north China. Is it possible to analyze the impacts of heating from coal?

AC: We appreciate the reviewer’s curiosity regarding difference of Hg concentrations between snow and rainfall. In this study, we did observe difference of Hg concentrations between snow and rainfall, however, there is no consistent difference of Hg concentrations at all the sampling sites. This indicates the difference of Hg concentrations were random can not be related to specific atmospheric mechanisms. We therefore did not discuss the difference in the revised manuscript. In the revised manuscript (line 256-258 on page 10) , we discussed the effect of atmospheric PBM concentrations in seasonal variations of VWM precipitation Hg concentrations. The PBM concentrations were generally elevated in winter in China (related to house heating cold burning) and have been discussed in many previous studies. We therefore did not dig further analysis for this.

RC: 6. Line 231-235: The lower Hg concentrations in precipitation during summer might also be associated with the lower ambient Hg concentrations during summer

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owing to the higher wind speed and higher mixing height. I suggest the author to add some comparisons and discussions on this.

AC: Hg in precipitation is mainly derived from scavenging of GOM and PBM in cloud and below cloud. In line 256-258 on page 10 in the revised manuscript, we have tried to establish the link between the seasonal trend of precipitation Hg concentrations and atmospheric PBM concentrations.

RC: 7. Line 253-263: It would be good to add a map showing the Hg concentrations in precipitation with both the data of this study and previous observations in China.

AC: this is a good suggestions. We added the Figure 4 in the revised manuscript to show the results of this and pervious studies.

RC: 8. Line 333: “which is 4.4 times lower than the mean ($24.8 \pm 17.8 \mu\text{g m}^{-2} \text{ yr}^{-1}$) at urban sites of China” shall be changed to “which is only 22.5% of the mean ($24.8 \pm 17.8 \mu\text{g m}^{-2} \text{ yr}^{-1}$) at urban sites of China” AC: the sentence in the manuscript and proposed by the reviewer is both correct.

RC: 9. Line 454: The authors state that “indicating that litterfall deposition contributes significantly to the Hg deposition budget in China”. Considering that this only applies to the forest area, it is suggested to change this sentence to “indicating that litterfall deposition contributes significantly to the Hg deposition budget in China forests”.

AC: yes, we changed the wording to ‘in forest ecosystems in China’ in line 490-491 on page 18 in the revised manuscript.

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