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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #1

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This manuscript presents wet deposition and litterfall mercury (Hg) measurements from multiple sites across China. Samples were collected over varying time frames, ranging from one to four years for wet deposition and from one month to one year for litterfall. The objective of this study is to quantify Hg inputs to the ecosystem through these two processes, and inform the Hg budget for China.

This is an important contribution to the scientific literature given the relatively limited understanding of Hg biogeochemical cycling, and given the relative contribution that emission sources in China apparently make to the global Hg pool. However, there are several major issues with the sampling methods as described here and also with the data interpretation that must be addressed.

Section 2.2: The wet-only precipitation sampling and collection method needs signifi-

cant clarification. This is probably the biggest issue I find with the manuscript. Specifically:

What type of wet-only collector was used? The name of a common commercial collector is not provided, so was the collector self-constructed? In this case more details should be provided if this particular collector has not already been described in the published literature.

Why were the samples collected into such large (15-L bottles) when most wet-only collectors employ 1-L or 2-L bottles? Why was the sample transferred to a 250 mL bottle? Was the sample acidified (i.e. with BrCl as a preservative) prior to this transfer? If not, then are the authors certain that no Hg was lost to the walls of the 15-mL bottle? Was a 250-mL bottle always sufficient to hold the entire sample that was collected into the 15-L bottle?

The text says that before each new sample the borosilicate glass sampler was rinsed with Milli-Q water. Does this mean the sample funnel and the 15-L bottle were not replaced between samples? Milli-Q would not be sufficient to clean the funnel or other parts of the sampling train appropriately between samples. New sampling materials should be installed or the materials should be cleaned with a dilute acid. This needs significant clarification. The authors should discuss any potential uncertainties or carryover introduced with this method and/or discuss their collection of field blanks to determine a lack of carryover.

Section 2.3: Are the litterfall collectors described here similar to or modeled after others reported in the literature? It would be helpful to know if this is a common method, or a new method developed by the authors.

In several places it is stated that samples were handled or analyzed in a "clean environment". What does this mean? Is this referring to a certified clean room or some other trace metal clean environment? Please elaborate.

Section 3.1 The paragraph on lines 224-234 is confusing. The paragraph starts by saying that higher concentrations are found in the summer wet season and lower concentrations are found in the winter dry season. But the rest of the paragraph says the opposite. Perhaps this was just a typographical area but it should be addressed.

The findings on lines 241-245 that wetter zones had more Hg wet deposition seem a little obvious. If there is more rain then, all else being equal, would we not expect more Hg deposition? Please provide more information on what these findings imply.

Section 3.2: If TGM was not significantly correlated with Hg concentrations in litterfall at any site, this implies that of the possible factors controlling Hg concentrations in litterfall the ambient Hg concentration was not the most important here. In this case, what do the authors propose to be the driving factor controlling Hg concentrations in litterfall?

Section 3.3 The statements on lines 308-310 contradict the findings discussed in Section 3.2 (referenced above). Here it is stated that Hg in litterfall is mostly due to uptake of atmospheric GEM, but previously you stated no correlation between litterfall Hg and ambient Hg. How do you resolve this?

Then, on lines 326-327 it is deduced that Hg deposition through litterfall played a predominant role in total Hg deposition in forested ecosystems in China. Is the "total" assumed to simply be wet deposition + litterfall? What about throughfall? Or dry deposition? Even if those two things were not measured they should be considered when drawing conclusions about total Hg input.

Conclusions: The manuscript ends with a statement that litterfall deposition contributes significantly to Hg deposition in China. Is this true for all of China? Or only for forested areas such as those sampled here? How much of China is forested compared to the total land area? Some clarification to these implications should be given.

Other comments: At no point in the text is the range of dates given for the sampling campaigns. It seems this can only be obtained from consulting Tables 1 and 2. Even

though the sampling spanned different date ranges at each site, there should be at least some statement in the text describing the overall time frame. Also, any limitations of comparing sites to one another where the samples were not collected concurrently or even during overlapping years should be discussed.

The manuscript title is a little misleading ("typical rural and urban areas") when in fact only one urban area was sampled. And what do the authors mean by "typical"?

Figure 2: The data points for rain depth should not be connected to one another, especially if the record is not continuous. Lines connecting data points that are weeks or months apart imply that the values along that line are known, which they are not (or maybe it simply did not rain then). Rain depth measurements should be represented as bars rather than points on a line just as the THg concentrations are presented.

Figure 3: This graph is difficult to interpret in black and white. It is also a little misleading since samples were not collected at all sites in the same year(s). What is it that the authors want the reader to take away from this graph, and is there a way it could be presented more clearly?

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