

## ***Interactive comment on “Mercury dynamics and mass balance in a subtropical forest, southwestern China” by M. Ma et al.***

**Anonymous Referee #2**

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This manuscript presents measurements of litterfall, throughfall, output stream water, forest floor and soil mercury concentrations, as well as air-soil exchange and other fluxes in a one year study in China. Those measurements were used to estimate mass balance of the forest under investigation. The topic is relevant to ACP, however, some results could be erratic. Furthermore, the scientific contribution is hindered by a lack of in-depth discussion considering results of similar studies conducted in the same region had been published. Moreover, the presentation has much room for improvement. My comments are listed below. Major concerns:

1. The title reads “Mercury dynamics and mass balance in a subtropical forest, southwestern China”. However, the reviewer could not find any analysis of the dynamics. If data were indeed collected in different seasons, a seasonal analysis would provide the

C11730

changes of Hg in different compartments of the forest system throughout the year.

2. It is uncommon that the forest field (defined by land cover) and watershed (defined by hydrological features) under investigation had the same size, “The study area is typical of the region with hills of 1394m and watersheds of about 100.1 km<sup>2</sup>.” (pg~60, L12) “The subtropical forest field in the study area is 100.1km<sup>2</sup>” (pg~68, L18). This could be one of the reasons an annual water discharge rate (1.86X 10<sup>+8</sup> m<sup>3</sup>, from hydrological departments of Jiangjin district, pg~68, L16) greater than the water input rate (annual precipitation of 1508 mm in Table 1 times 100.1km<sup>2</sup> = 1.52X 10<sup>+8</sup> m<sup>3</sup>) was reported and used in the Hg flux calculation. This implies a negative evapotranspiration flux from a subtropical forest (assume the net water flow to/from the groundwater reservoirs is negligible), which is almost absent from the literature. Therefore, the representativeness of the Hg mass balance as presented could be challenged due to a large discrepancy in the water balance. In that case, the Hg mass balance may need to be reconsidered.

3. Volume-weighted average Hg concentrations of precipitation, throughfall, and output stream water should be used in flux calculation. Please include those in Table 1 and the main body, in addition to (arithmetic) means as the table caption and standard deviation imply. If the values presented are indeed volume-weighted average Hg concentrations, please provide the method of standard deviation calculation.

4. The comparison with other studies was at times hard to follow due to the use of general statements of higher/lower, e.g. “The annual mean gaseous elemental Hg (GEM) concentration in the middle of Chongqing city (more than 200 km away from the study site) tripled comparing with global background level”; “The MeHg flux was 0.45 μgm<sup>-2</sup> yr<sup>-1</sup>, which was higher than those measured in other areas.”; “While MeHg/THg in the throughfall samples was 1.3 %, which was a relatively high value compared with other studies.”; “And it is also considerably higher than litterfall fluxes reported from other regions.”; “But the emission of GEM elevated in comparison with those reported from other places.”; “THg and MeHg concentrations in stream water draining

C11731

the upland in our research were slightly higher than those reported in literature.”; “The ratio between output and input of THg was 0.34 at the subtropical forest field of Mt. Simian, which was significantly higher than others.” The authors may want to provide data support of others’ results in tables as in Ma et al. (2013) and Ma et al. (2015) or provide the values or ranges in the main body.

5. Nearly identical methodology was employed in this study and that reported by Fu et al. (2010a: Elevated atmospheric deposition and dynamics of mercury in a remote upland forest of southwestern China), and both are in southwest China. The authors may want to compare the soil profiles, mass balance, and seasonal variations of the two studies, instead of just a comparison of the output to input ratio as presented. In addition to the study by Feng et al. (2009a: Mercury mass balance study in Wujiangdu and Dongfeng Reservoirs, Guizhou, China), another recent Hg mass balance study in south/southwest China is by Wang et al. (2009, Mercury fluxes and pools in three subtropical forested catchments, southwest China. *Environmental Pollution* 157, 801–808), see Table 4 in Fu et al. (2010a). Inclusion of these studies in your mass balance discussion would highlight anything new that we have learned from this study.

6. Some statements are not supported by data presented in this manuscript, e.g. “The annual precipitation of the sampling site is slightly lower than the annual discharge.”; “The robustness of the approach for THg and MeHg was 5 and 9% respectively.”; “There may be very few errors of estimates of Hg output from the soil pool.”

7. Please reference your sources when presenting data/results from others work, e.g. pg ~60, L6-16; pg ~67, L27; pg ~68, L1-3.

#### Clarification issues

1. Please provide the division of the 12 months into the four seasons, and the dry and rainy seasons.

2. Section 2.2.4 could be condensed by referencing Ma et al., 2013, because the

C11732

method employed is identical.

3. Pg ~62, L6-14, suggest removing this passage because it is not related to sampling method of litterfall and soil.

4. Pg ~63, L12, “No blank value was needed to be subtracted from the flux results due to no significant difference found.” Please clarify.

5. Pg ~64, L4, “Canopy density did have an effect on THg and MeHg concentrations (the forest cover is more than 90% and the canopy density is 0.9).” The reviewer could not find any density levels or any ranges of canopy density to support the association or a lack of association between canopy density and Hg concentrations.

6. Pg ~65, L23, “GEM concentration in the study area is as high as 3.8\_1.5 ng m<sup>-3</sup> (Ma et al. (2015).” In Ma et al. (2015), this value was reported as mean concentration in another study. Furthermore, please comment on the distance between the monitoring site in this study and that in Ma et al. (2015).

7. Pg ~67, L16, “So we assumed that there still existed Hg<sub>0</sub> emission in December and February in winter.” Please provide in Fig 2 the month when each seasonal sampling (pg ~63, L4) was conducted. Similarly, in the main body please provide the month when each of the “eight intensive field campaigns” (pg ~66, L16) was conducted.

8. Pg ~71, L7-9, the total input as presented (57.1% plus 40%) is less than 100%.

9. The second part (pg ~71, L14-21) of the Conclusions, those general statements could be better placed in the Introduction or Results and discussion section.

10. Table 1, please define DHg, PHg, DMgHg, PMeHg and provide methods of analytical analyses in the main body.

11. Table 2, please provide a column header for the last two values (5148.7, 20 192.6).

12. The meaning of the three Os in Table 2 and in the main body should be defined.

C11733

13. Fig 2 caption, "Volume-weighted mean concentrations. . ." Did you mean "Sample concentrations"? If not, please provide method to calculate volume-weighted mean concentrations for each sample.

14. The redundancy in the presentation should be eliminated, e.g. pg ~69, L5-8, L8-11, L18-22.

Editorial comments/suggestions:

The use of English language is overall satisfactory. However, there is much room for improvement. There are quite a few awkward word choices, phrases and sentences. The authors may want to enlist the help of a native English speaker to improve the readability. Some of my suggestions are listed below.

1. Examples of awkward word choices: obvious, obviously, formerly, fishes, whole (one whole year, whole sampling period, need to be considered as a whole, the whole forest ecosystem).

2. Examples of awkward phrases: Hg as a gas phase can travel; higher data; bound up; One of the possible reasons perhaps was; Normally it was supposed; mean average.

3. Examples of awkward sentences, "Hg transformation processes in the forest is considered as a vital part of global Hg cycling and possible climate changes."; "The study area has . . . , which means that this area has . . ."; "The stream/runoff was carried out at the edge of the forest catchment. "; "The measured data of . . . were collected by the local hydrological departments in the outlets"; "It is, however, still in September and October that a higher throughfall flux is observed."; "thus only several data were observed with Hg deposition in the night"; "Numerous studies showed that the remote forest already considered the forested catchments as filters between. . ."; "The ultimate fate of Hg in the terrestrial ecosystem may depend upon the means of delivery and incorporation of Hg into the forest floor. And the average Hg fluxes were also estimated."; "An amount of the atmospherically deposited THg was released through Hg<sup>0</sup> emission

C11734

at a rate of 18.6  $\mu\text{gm}^{-2} \text{yr}^{-1}$ ."

4. Example of contradicting statements, "The deposition fluxes of THg through throughfall in Mt. Simian were lower than those investigated in the southwestern cities of China, . . . approximately 2–10 times higher than those reported in remote areas of North America and Europe. . . It was obviously that the THg fluxes at Mt. Simian were higher than other sites at home and abroad." (pg ~65, L2-10)

5. Example of incorrect statements, "Unlike some other studies, in which average fluxes of Hg in spring (12.25.1  $\text{ngm}^{-2} \text{h}^{-1}$ ) were slightly lower than that in summer (14.24.7  $\text{ngm}^{-2} \text{h}^{-1}$ ); the seasonal fluxes reported here seem to be yours.

6. The word "and" may not be used to start a sentence.

7. Pg ~61, L3, "through" could be replaced by "throughfall".

8. Pg ~66, L4, "are shown in Table 1".

9. Pg ~68, L19, "export mass of THg through stream water was 7.23  $\mu\text{gm}^{-2} \text{yr}^{-1}$ ". Either change to "export flux" or report total mass.

10. Pg ~70, L19, "Compared the ratios of output flux with other places", it should read "the ratios of output to input flux", or "the output flux".

11. The significant figures seem to be a bit excessive at times, e.g. "982.2 times", "337.6 times", suggest using integers in those two incidents.

12. The readability of sections 3.3 and 3.4 could be improved by avoiding long paragraphs and eliminating redundancy with sections 3.1 and 3.2, e.g. comparison of individual concentration/flux with other studies.

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 35857, 2015.

C11735