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Atmospheric Chemistry and Physics Discussions

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

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Dear referee, Thanks you for your kindly comment concerning our manuscript entitled "Mercury dynamics and mass balance in a subtropical forest, southwestern China" (acp-2015-664). Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. P35858, L11: We appreciate very much for this advice of the referee about the digits in L11. These are exact results of real-time investigation by our research. More significant digits, more accurate. P35860, L12: The study site is the most representative one because Mt. Simian is one of the most complete forest located in the place between Chongqing and Guizhou. And the study area is almost all subtropical forest and until recent decades was one of Asia's least populated and most

inaccessible areas. P35860, L21: We will delete "self-made" according to the referee's comments. P35860, L23: We are sure the precipitation samples were collected at an open-air site near a meteorological station, which belongs to the Simian Mountain Alpine Ecosystem station of the Southwest University. Precipitation was taken from the nearest meteorological station (1318 m from the sample site) at an catchment of Mt. Simian with an elevation of 1427 m, which can be fully representative of the study area. Precipitation samples were collected by automatic APS-3A rain collectors (Changsha Xianglan Scientific Instruments Co., Hunan, China) installed in an unshadowed field at the sampling sites. We calculated the annual ratio (0.91) between precipitation amount and water discharge from the lake catchment. P35861,L6: Steam outflow of THg was estimated by multiplying the mean THg concentration in stream water (3.89 $\pm$2.03 ng L–1) and water discharge rate in the forest field of Mt. Simian (annual water discharge: 1.86$\times$108 m3, from hydrological departments of Jiangjin district). The export mass flux of THg via runoff/stream was 0.73 kg yr-1, and subtropical forest field in the study area is 100.1 km-2. So the export mass of THg via stream water was 7.23 μg m$-$2 yr$-$1, which was 3 times higher than those reported in the catchments of Sweden. Soil erosion and runoff were additional sources to stream water. THg concentrations in streamwater were slightly higher compared with precipitation, which was probably derived from soil erosion and runoff during stream transport. P35861, L11: Yes, the collected rain samples were transported in rigorously acidcleaned Teflon bottles (250 ml) and preserved by adding trace metal grade HCl. All Teflon bottles were cleaned with detergent, thoroughly rinsed with tap water, boiled in a 30% HNO3 solution (v/v) for 1.5 h and rinsed and filled with Milli-Q water. P35861, L13: As far as the catchment are concerned, this is by far the most ideal place. Choosing smaller catchment or using soil water Hg leaching instead may be better, we will pay attention to this in future research. P35861, L19: We appreciate very much for this question of the referee. Precision was determined by relative standard deviations. For duplicate samples, which were 5% for THg analysis in water samples, 9% for MeHg analysis inwater samples, 8% for THg analysis in soil samples, and 4.1% for THg

analysis in leaf tissues. P30862, L7-9: We appreciate very much for this advice of the referee. We added the references at L7-9 in page 35862(Rea et al., 2001; St. Louis et al., 2001), this will be modified in the coming manuscript. In P35862, L25, Cout and Cin are the TGM concentrations of the outlet and inlet air stream in ng m-3; TGM concentrations were measured twice in the ambient air entering the inlet of the chamber and twice in the air exiting through outlet of the chamber. P30863, L12: Blanks of the flux sampling system were routinely measured by placing the chamber on a Quartz glass surface. Here we thought the blanks were small based on the background values of TGM of China. And we did not need to make blank correction for measured flux. P30865, L9: Yes, "at home" will be changed to "in China" in the coming manuscript. In P30866, L5: Thanks for the reviewer's suggestion. Mercury that is delivered to the forest floor with litterfall is likely retained in the soil profile, whereas mercury delivered to the forest floor in throughfall is either incorporated into decomposing leaf litter or re-volatilized. At the same time, Hg accumulation in soil is a long process that the pool in soil is not comparable with Hg deposition fluxes, and delivered Hg might not be considered a new output on the basis of origin in forest soil, it would still be considered a more biologically available form of Hg. Thus, new and recycled mercury are difficult to differentiate in the soil pool. The next step should consider this problem. There may be very little errors of estimate of Hg output from the soil pool. We appreciate very much for this advice of the referee. We plan to add the following text to the coming manuscript: "Unlike some other studies, in which average litter fluxes of Hg in spring was slightly higher than that in autumn, it appeared that warm temperature with low canopy density in spring at mid subtropical forest were more likely to release Hg through litterfall. Although the branches and leaves were not so luxuriant, they received more deposition Hg in winter, which increased the Hg deposition in litterfall." In P30868, L21: Yes, Research in recent years showed that if there is no other obvious mercury pollution sources, mercury levels in its runoff/stream water can reflect the characteristics of mercury inputs and risks of a remote forest. Studies in remote areas of forest already emphasized the role of forested catchments

as filters between atmosphere and hydrosphere. THg concentration in stream/runoff was lower than that in contaminated sites under the same geological background, indicating that the study area did not suffer from severe anthropogenic Hg pollution. As for MeHg, we found that the methylation rate was not so high, the mechanisms of this may need to do further work in the future. P30868, L24: The referee is totally right. In the coming manuscript, we'd like to change the sentence "Here the elevated Hg fluxes in stream water were probably attributed to the great atmospheric Hg depositions" to: "As we mentioned above that Mt. Simian was one of the most complete forest and until recent decades was one of Asia's least populated and most inaccessible areas. Average Hg concentration in soil detected in previous research was 0.28 mg kg-1(Ma et al., 2013), which indicated that there was not an obvious geological Hg hotspot. Therefore, the elevated Hg fluxes in stream water were probably attributed to the great atmospheric Hg depositions." In P30869, L5: Soil samples were collected from soil profiles; 3 layers (nearly 65 cm) were collected from each profile according to diagnostic horizons (Oi, Oe, and Oa horizons). Horizon thickness was measured and soil cores were taken to measure soil bulk density. P30869, L20: The steam outflow of THg was estimated by multiplying the mean THg concentration in stream water and the water discharge rate in the forest field of Mt. Simian. The export mass flux of THg via runoff/stream was 0.73 kg yr-1, and subtropical forest field in the study area is 100.1 km-2. So the export mass of THg via stream water was 7.23 μg m$-$2 yr$-$1. The referee is right that choosing smaller catchment or using soil water Hg leaching instead may be better, so we will pay attention to it in future research. But now, this area is still the ideal area considering on all aspects. P30870, L20: The referee is totally right. In the coming manuscript, we'd like to add the following sentences before "Therefore" in line 21: "As we mentioned above that Mt. Simian was one of the most typical subtropical forest systems and the least accessed area, average Hg concentrations in all soil surfaces of this area were below 0.30 mg kg-1(Fu et al., 2010a; Ma et al., 2013)." Figure 2: We've already showed some GEM data and discussions as references in 3.1: "The area, especially the Chongqing city, has

a large demand of energy, about 70% of which is derived from coal combustion. The annual mean GEM concentration of Hg in the main districts of Chongqing (more than 200 km away from the study site) is three-fold higher than the global background level (Yang et al., 2009), corresponding to the high annual deposition flux of Hg in the study area. It is also reported that the GEM concentration in the study area is as high as 3.8 $\pm$1.5 ng m-3(Ma et al., 2015), even it located in a natural subtropical forest reserve."

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C12929/2016/acpd-15-C12929-2016-supplement.pdf

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