

Dear editors and referee,

Thanks for the reviewers and editors' comments concerning our manuscript entitled "**Mercury dynamics and mass balance/transport based on forest field in an old-growth subtropical forest, southwestern China**" (acp-2015-664). The comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our research. We have studied the comments carefully and made corrections which we hope meet with approval.

Here are our reply to the referee #2:

A1: We did collect continuous data of Hg fluxes through throughfall, litterfall and runoff (stream) in different seasons (Fig.1). We collected the air-surface exchange of gaseous elemental Hg from the subtropical forest field during eight intensive field campaigns in 2012 and 2013 (Fig.2). We introduced the changes in throughfall deposition flux in the whole year in the cold and warm seasons; we also analyzed the Hg exchange fluxes between forest soil and air in the four seasons during the whole year, respectively.

"The study station has obscure seasons and clear rainy and dry seasons, throughfall deposition fluxes were also seasonal, with higher monthly THg deposition appearing during rainy months (June to August) (Fig. 1). THg throughfall deposition in summer at the study sites represents over 40% of total annual Hg deposition. It is, however, still a higher throughfall flux observed in September and October. This may be because that the rainfall in this month was influenced by Indian Monsoon resulting in a high rainfall (Fu et al., 2008a). The lowest monthly values of THg deposition appeared in the cold season." in 3.1.

"Unlike some other studies, in which average fluxes of Hg in spring ($12.24 \pm 5.1 \text{ ng m}^{-2} \text{ h}^{-1}$) was slightly lower than that in summer ($14.24 \pm 4.7 \text{ ng m}^{-2} \text{ h}^{-1}$), it appeared that warm temperature with low canopy density in spring at mid subtropical forest were more likely to release GEM. Because the branches and leaves were not so luxuriant but well spaced in spring and received more sunlight, which increased the rate of reduction of Hg^{2+} by photochemical and thermal processes and biological activities." in 3.2.

A2: We appreciate very much for this question of the referee. 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 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 $7.23 \mu\text{g m}^{-2} \text{ yr}^{-1}$. 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 all aspects.

A3: Yes, you are right.

Volume-weighted mean concentration (VWM) is calculated with the formula introduced in Acid Deposition Monitoring Network in East Asia, 2012:

$$\text{VWM} = (X_1 \times V_1 + X_2 \times V_2 + \dots + X_t \times V_t) / (V_1 + V_2 + \dots + V_t) = \sum (X_t \times V_t) / \sum V_t$$

where, X_t means the ion concentration in each precipitation event (ng L^{-1}), V_t means the volume of each rainfall (mm).

Hg flux was determined by multiplying Hg concentrations by the volume of precipitation collected. Wet deposition fluxes of THg and MeHg were calculated according to the following equation:

$$F_w = \frac{1}{1000} \sum_{i=1}^{i=n} (C_R^i p^i)$$

Where, F_w is the annual THg or MeHg wet deposition flux ($\text{mg m}^{-2} \text{ yr}^{-1}$), C_i is the volume-weighted concentration (ng L^{-1}) of each rain sample, and P_i (mm) is the precipitation or throughfall amount.

We averaged concentrations of MeHg and THg in throughfall first for all sites. Throughfall deposition of MeHg and THg was estimated by multiplying an overall volumeweighted mean concentration of MeHg and THg calculated from all throughfall event by the mean annual throughfall volume.

We will add the two formulas in the coming version.

A4: We appreciate very much for this advice of the referee. The suggestions will benefit the improvement of the paper. We will re-write the ambiguous sentence in the coming manuscript (provide data support).

A5: Thanks for the referee's question. We know that atmospheric Hg emissions in China are predicted to increase due to fast economic development. The huge emissions have resulted in elevated atmospheric Hg depositions in industrial and urban areas, and have the potential to cause Hg pollution in surrounding areas and even remote areas of China via long-range atmospheric transport. Some Studies already showed extremely high Hg deposition fluxes. These results suggested that many urbanized areas of China were exposed to atmospheric Hg contaminations due to regional anthropogenic emissions. However, there are still limitations to fully describe temporal and spatial distributions of Hg in China. Hence, there is a great need to conduct long-term continuous measurements of Hg and deposition fluxes in remote areas of China. At the sometime, 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. As for Hg accumulated in the forest soils, one part of them transfers through food webs, threatening the balance of forest ecosystems; one part of them releases into the atmosphere again; the other parts of them probably transfer with the runoff/stream, becoming one of the Hg sources of downstream aquatic ecosystem. Therefore, to a certain extent, the role of forested catchments as Hg filters can be characterized by Hg output (runoff/stream) from forest field. Therefore, even the higher Hg deposition fluxes in throughfall, it can be concluded that "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." On one hand, this means that forested field has the filtering effect of Hg in precipitation and throughfall, even in the elevated atmospheric Hg area.

A6: According to the advice of the referee, we made the following changes:

"The annual precipitation of the sampling site is slightly lower than the annual discharge" changed to:

"The subtropical forest field in the study area is 100.1 km^2 . Annual water discharge of the study site is $1.86 \times 10^8 \text{ m}^3$ (hydrological departments of Jiangjin district). The annual precipitation (Table 1) 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.” was cancelled and add the following sentences in 2.3 “Quality control”:

“The precision was determined by relative standard deviations. For duplicate samples, which were 5% for THg analysis in water samples, 9% for MeHg analysis in water samples, 8% for THg analysis in soil samples, and 4.1% for THg analysis in leaf tissues.”

“There may be very few errors of estimates of Hg output from the soil pool.”

No change.

A7: According to the advice of the referee, we made the following changes:

P35860, L6-16, add the reference: (Lv et al., 2014)

Add the following reference in the reference list:

Lv, W., Zhang, H., Wang, W., Du, S., Wu, Y., He, P., Xiao, L. Characteristics of Soil Aggregates in Different Forestlands in Simian Mountains, Chongqing. *J. Soil Water Conserv.*, 24(4):193-197, doi:10.13870/j.cnki.stbcxb.2010.04.048, 2014.

P35867, L27, add the reference: (Ma et al., 2015)

P35868, L1-3, add the reference: (Lee et al., 2000; Larssen et al., 2008; Ericksen et al., 2003)

Clarification issues:

A1: According to the advice of the referee, we made the following changes in 2.1:

“There are four seasons in Chongqing, spring (March to May), summer (June to August), autumn (September to November), and winter (December to February), in which July, August and September belong to the dry season, and the other nine months belong to the wet season.”

A2: Section 2.2.4 had been condensed as follows according to the suggestion.

“A dynamic flux chamber (DFC) in series with Tekran 1110 synchronized dual-port sampling unit and Tekran automated Hg analyzer (2537X) were used to measure the emission rates of Hg⁰(Fu et al., 2010). The special DFC method was described in detail at Ma et al (2013). Hg emission fluxes were calculated by the equation below (Ma et al., 2013):

$$F = (C_{out} - C_{in}) \times Q/A$$

A3:P35862, L6-14:

According to the advice of the referee, we removed the second paragraph in section 2.2.3.

A4: P30863, L12:

This issue is also proposed by the referee #1.

We are sure that it is no problem based on the following reasons.

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.

A5: P30864, L4:

Higher canopy density may be one of the reasons of higher THg and MeHg concentrations in throughfall. We believed it was one of the important reasons and proposed it in the manuscript.

So, we made no change in the coming version for this question.

A6:P30865 L23:

The precipitation samples were collected at an open-air site near a meteorological station, which belong 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 in Ma et al., 2015) 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 installed in an unshadowed field at the sampling sites.

A7: Thanks for the good suggestion. We'll add the detailed month in fig.2 in the coming version.

A8: Here we want to say that the input fluxes through throughfall accounted for nearly 40% (exactly 42.9%) of the total Hg inputs, which is lower than that through litterfall. We will use the exact number (42.9%) or add the word “nearly” before 40% in the coming version.

A9: Thanks for your suggestion, but we discussed this question and believed that not changing the place was better.

A10: Good suggestion! We'll add the definitions of DHg, PHg, DMgHg, PMeHg in table 1. As for the analytical methods, we'll add the reference in Material and Methods.

A11: Very good suggestion! We'll add a column header, “total”, for the last two values (5148.7, 20 192.6) in table 2 in the coming version.

A12: Very good suggestion! We'll add the meaning of the three *O*s in Table 2 and the main body (Materials and Methods) in the coming version.

A13: Very good suggestion! The method to calculate “Volume-weighted mean concentrations” was introduced in A3. And we'll add this introduction in the coming version.

A14: Very good suggestion! Combing with the question 12, we will delete the introduction of *O*s here and define it in Materials and Methods. But we do think Line 18-22 is reasonable, so we think it should not be deleted.

For Editorial comments/suggestions:

Thanks very much for the good suggestions on English expressions! Under the help of an English teacher from the USA in our university, the English in this paper has changed a lot and we think it can meet the requirement of ACP.

A1: The awkward word choices, such as obvious, obviously, formerly, fishes, whole etc. had been modified.

P35858, Line 7, obviously→ remarkably; “formerly” is deleted; “fishes” → “fish”.

P35859, Line 11, “whole” is deleted; Line 22, “whole” is deleted;

P35860, Line 20, “whole” is deleted;

P35861, Line 4, “whole” is deleted; Line 16, “whole” is deleted; line 17: “during the whole year” → “during the study period”

P35865, Line 9, “It was obviously that” → “Obviously,”

A2: The awkward phrases, such as “Hg as a gas phase can travel; higher data; bound up; One of the possible reasons perhaps was; Normally it was supposed; mean average, and so on had been modified.

P35858, line 17: “Hg as a gas phase” → “gaseous Hg”

P35864, line 21, “with higher data appearing in wet-season” → “with higher fluxes appearing in wet-season”

P35865, line 15-17, “were probably bound up with” → “probably related with”

P35866, line 26-27, “One of the possible reasons perhaps was” → “Perhaps the primary reason lies that”

P35867, line 13, “Normally it was supposed that” → “It was supposed that”

P35869, line 5-7: “mean average” → “average”.

A3: Awkward sentences:

P35858, line 24-26: “Hg transformation processes in the forest is considered as a vital part of global Hg cycling and possible climate changes.” →

“Hg transformation processes in the forest is considered as a vital part of global Hg cycling.”

P35860, line 7-8: “The study area has . . . , which means that this area has . . .”; →

“The study area has a subtropical monsoon climate, with abundant rainfall every year.”

P35861, line 6-8: “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.” →

“The stream/runoff was collected at the edge of the forest catchment. For the water yield of the stream/runoff, it was monitored in the outlets of the forest catchment by the local hydrological departments.”

P35864, Line 23-24:

“It is, however, still in September and October that a higher throughfall flux is observed.” →

“However, it is still in September and October that a higher throughfall flux is observed.”

P35867, Line 11-12:

“and thus only several data were observed with Hg deposition in the night” →

“At Mt. Simian, the estimated net GEM fluxes were released from soils during the warm season (spring, summer and fall) and slightly volatilized during the cold season (winter). Hg deposition was only observed in several nights of the cold season during the study period.”

P35868, Line 1-2:

“Numerous studies showed that the remote forest already considered the forested catchments as filters between atmosphere and hydrosphere.” →

“Numerous studies showed that the catchments of remote forest was regarded as filters between atmosphere and hydrosphere.”

P35869, Line 15-16:

“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.” →

“The ultimate fate of Hg in the terrestrial ecosystem may depend upon its delivery and incorporation into the forest floor.”

P35869, Line 22-23:

“An amount of the atmospherically deposited THg was released through Hg⁰ emission at a rate of 18.6 μgm⁻² yr⁻¹.” →

“A majority of atmospherically deposited THg was released through Hg⁰ at a rate of 18.6 μg m⁻² y⁻¹.”

A4: For contradicting statements:

P35865, Line 2-10:

“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.”

Yes, it is contradicted. We are sorry for the clerical error! “It was obviously that the THg fluxes at Mt. Simian were higher than other sites at home and abroad.” will be changed to “Obviously, the THg fluxes at Mt. Simian were higher than other sites abroad.” in the coming version.

A5: For incorrect statements

P35866, Line 23-26:

“Unlike some other studies, in which average fluxes of Hg in spring (12.25.1 ngm⁻² h⁻¹) were slightly lower than that in summer (14.24.7 ngm⁻² h⁻¹)”; the seasonal fluxes reported here seem to be yours.

Yes, it is an incorrect use of clause. It will be changed to “The average fluxes of Hg in spring (12.25.1 ngm⁻² h⁻¹) were slightly lower than that in summer (14.24.7 ngm⁻² h⁻¹), which was different from other studies (Larssen et al., 2008; Fu et al., 2010).”

A6: The word “and” may not be used to start a sentence.

We made the following changes:

P35866, Line 8: “And it is also...” → “It is also...”

P35870, Line 11: “And higher wet deposition...” → “Higher wet deposition...”

P35860, Line 14, “And it is also.....” → “Moreover, it is also....”

A7: Pg 35861, L3, “through” had been replaced by “throughfall”. Thanks a lot!

A8: Pg 35866, L4, “were shown in Table 1”. → “are shown in Table 1”.

A9: Pg 35868, L19, “export mass of THg through stream water was 7.23 μgm⁻² yr⁻¹”. → “export flux of THg through stream water was 7.23 μgm⁻² yr⁻¹”

A10: Pg 35870, L19, “Compared the ratios of output flux with other places, the higher output flux may be greatly...” → “Compared the ratios of output to input flux with other places, the higher ratios may be greatly...”

A11: 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.

P35870, Line 3-4: “982.2 times” → “982 times”; “337.6 times” → “338 times”

A12: 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.

Thanks a lot for this suggestion. We had separate 3.3 to two paragraphs, and 3.4 to three paragraphs based on the logical relationship.

Thanks a lot for the thorough check to our manuscript, which help us a lot!

Ma Ming