Reply to Comments from Reviewer #2

We thank the editor and reviewers' comments which help us to improve the manuscript. We have carefully revised our manuscript following the reviewers' comments. A point-to-point response is given below. The reviewers' comments are in black and our replies are in blue.

To reviewer

Comment #1:

This manuscript reviewed the mercury fluxes, budgets and pools in forest ecosystems in China, however, the authors did not do a good systemic summary for all the current research. For example, the underground flow was not considered into the budget.

Response:

We have revised the manuscript by around and around and revised all the comments. First, the fluxes from the underground flow were estimated in the manuscript and was considered into the budget, showing Section 3.1, Section 4 and Fig. 6.

The underground flow considered into the budget is revised as below:

"The direct measurements of THg in underground runoffs were not conducted in any forests of China, but they played important roles in the THg export from forests due to both of the amounts and THg concentrations usually higher than those of surface runoffs in subtropical forests (Liu, 2005; Luo et al., 2015b). Several studies have measured THg concentrations in solutions of soil profiles in subtropical forest of Tieshanping, which was averaged 21.8 ng L^{-1} and ranged from 1.98 to 60 ng L^{-1} (Wang et al., 2009; Zhou et al., 2015; Luo et al., 2015b). The observed THg concentrations of soil solution was higher than those in five Swiss forest soils, and the reason may be due to higher THg loads and soil THg content in this Chinses forest. Although no studies directly measured the export flux of THg via underground runoff, we roughly estimated the flux based on the THg in soil solutions and runoff amount in Tieshanping forest, which is 6.0 μg m^{-2} yr^{-1}; therefore, the total Hg output by runoffs as the sum of Hg output by surface runoff (3.5 μg m^{-2} yr^{-1}) and underground runoff (6.0 μg m^{-2} yr^{-1}) was 9.5 μg m^{-2} yr^{-1}." and "Based on our measured THg concentrations in soil solution (9.2 ng L^{-1}, our unpublished data) and the amounts of underground runoffs in the three stands (Wang et al., 2012), the export fluxes by underground runoffs were estimated."

See the revised manuscript, line 374-283 and 508-510, and Figure 6.

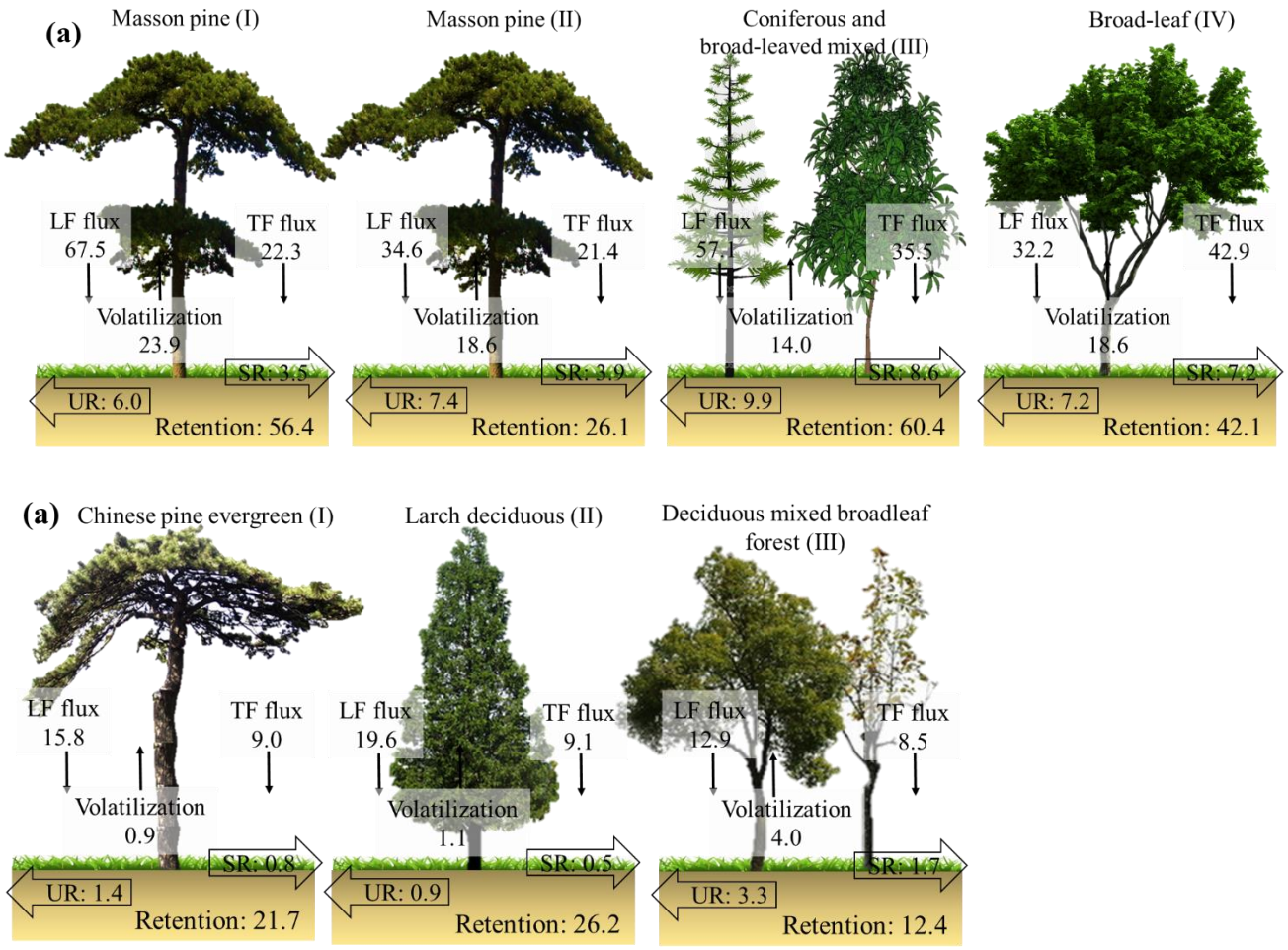

Fig. 6. Total mercury budgets (μg m^{-2} yr^{-1}) at the four subtropical forests of Tieshanping (I), Qianyanzhou (II), Mt. Gongga (III) and Mt. Simian forests (IV) (a) and three temperate forest stands of Mt. Dongling (I-III) (b). LF, TF, SR and UR represent litterfall, throughfall, surface runoff and underground runoff fluxes, respectively. Data are from Zhou et al. (2016a, c), Luo et al. (2015b), Wang et al. (2009), Luo et al. (2015a), Fu et al. (2010a), Ma et al., 2016.

Comment #2:

There are numerous typographical errors and poor sentence structure throughout the paper.

Response:

We have revised the typographical errors and poor sentence structure throughout the paper by us and our colleagues. We hope the revised grammar and sentence structure meet the publish standard.

Comment #3:

Tieshanping showed a high Hg pool, why? , due to its location or being influenced by human activities.

Response:

The high Hg pool in Tieshanping is due to it located near the larger city of Chongqing, which has emitted large amount of atmospheric Hg annually and showed as "The much higher Hg input at Tieshanping forest is due to it located near the center of Chongqing City (20 km), the annual atmospheric emissions of which just from coal combustion was 4.97 t (Wang et al., 2006) and Hg pollution was regarded as major environmental burdens in Chongqing (Yang et al., 2009). The large mercury emission resulted in much higher Hg deposition fluxes not only in the urban areas but also in the suburban areas (Ma, 2015; Wang et al., 2009, 2014)." in line 311-315.

Comment #4:

Line 29, Chinses??

Response:

It has changed to "Chinese" in line 179.

Comment #5:

Line 43, change are to is

Response:

It has changed to "is" accordingly in line 195.

Comment #6:

Line 50, change sinks to sink

Response:

It has changed accordingly in line 202.

Comment #7:

Line 120, change plays to play

Response:

It has changed accordingly in line 278.

Comment #8:

Line 136, change resulted to result; which is closed to the large Hg mine of Wanshan, I think your mean is that the litterfall Hg concentration at Leigong is closed to the Hg concentration of litterfall in Wanshan?? If like this, please reorganize this sentence.

Response:

We have changed the sentence as "Although Mt. Leigong was relatively isolated from anthropogenic activities with lower GOM, PBM, precipitation and throughfall Hg concentrations, GEM could undergo long-range transport from emission sources and the GEM concentration was 2.80 ng m−3 in Mt. Leigong that is about 170 km to the large Hg mine of Wanshan (Fu et al., 2010a). The relatively higher GEM concentration resulted in elevated litterfall Hg concentrations." in line 291-294.

Comment #9:

Line 152, change we to it

Response:

It has changed accordingly in line 307.

Comment #10:

Line 154, change was to were

Response:

It has changed accordingly in line 310.

Comment #11:

Line 157, change ranging to range

Response:

It has changed accordingly in line 317.

Comment #12:

Line 209, change has to have

Response:

It has changed accordingly in line 366.

Comment #13:

Line 214, change depends to depend

Response:

It has changed accordingly in line 371.

Comment #14:

Line 223, only one sites??

Response:

It has changed to “only one site” in line 390.

Comment #15:

Line 239, change were to was

Response:

It has changed accordingly in line 404.

Comment #16:

Line 247, change showed to show

Response:

It has changed accordingly in line 412.

Comment #17:

Line 277, humidity??

Response:

It has changed to “humidity” in line 442.

Comment #18:

Line 287, change were to was

Response:

It has changed accordingly in line 451.

Comment #19:

Line 306, add were after forests

Response:

It has added in line 470.

Comment #20:

Line 439, change was to were

Response:

It has revised accordingly.

Mercury fluxes, budgets and pools in forest ecosystems of China: A critical review

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Abstract: Mercury (Hg) accumulation and retention in forest ecosystems play a key role in global biogeochemical cycling of Hg. Especially in China, forests are suffering highly elevated Hg loads. Numerous studies have been conducted to characterize the fluxes and pools of Hg in the terrestrial forests in China during the past decade, which provide insights into spatial distributions and estimate the Hg mass balance in forests through observations at widely diverse subtropical and temperate locations. In this paper, we present a comprehensive review of the research status of forest Hg in China to characterize the Hg budgets and pools. Averaged total Hg (THg) inputs at remote forests and rural & suburban forests in China are about 2 to 4-fold and 2.5 to 5-fold higher than the observed values in Europe and North America, respectively. The highly elevated THg inputs are mainly derived from the elevated atmospheric Hg concentrations. Additionally, production of litterfall biomass is showed to be an important influential factor raising the high Hg inputs at subtropical forests. Compared to the input, THg outputs from the forest ecosystems are relative small, which results in large amount of Hg resided in the forest soils. The annual THg retentions range from 26.1 to 60.4 μg m^{-2} at subtropical forests and from 12.4 to 26.2 μg m^{-2} at temperate forests of China, which are about 3.8- to 7.9-fold and 1.2 to 2.8-fold higher compared to those in North America. Given the large areal coverage, THg retention in forest is appropriately 69 t yr^{-1} in China and is much high than that in global scale estimated by models. The much higher THg retention has elevated the THg pools in Chinese subtropical forests, which poses a threat for Hg pulses remitted back to the atmosphere and additional ecological risks in the forest. The current study has implication for the role of China forests in the global Hg biogeochemical cycle and the optimization of atmospheric Hg transport and deposition models.

Keywords: Trace metals; Atmospheric deposition; Input–output; Storage; Risk assessment

1. Introduction

Mercury (Hg) is considered as a highly toxic heavy metal due to its biogeochemical properties and its toxicity that can affect the health of human and ecosystems (Kojta et al., 2015; Falandysz et al., 2015a; Zhou et al., 2015b; Du et al., 2016). Unlike other heavy metals, atmospheric Hg may exist three operational Hg forms: gaseous elemental Hg (GEM); gaseous oxidized Hg (GOM, also known as reactive gaseous Hg); and particulate-bound Hg (PBM) (Lindberg and Stratton, 1998; Fu et al., 2015). Due to its strong stability and low water solubility, GEM is the most abundant (more than 90%) in the atmosphere and has long resistance time of 2.7 months–2 years, which can be transported globally and deposited to the remote eco-environment (Zhou et al., 2017a; Horowitz et al., 2017). Atmospheric Hg deposition to terrestrial and aquatic ecosystems plays a significant role in the global biogeochemical cycling of Hg (Zhou et al., 2103a; Blackwell and Driscoll, 2015a). Consequently, an understanding of how Hg is transported, deposited and circled the globe is significant for a full understanding and quantifying of Hg biogeochemical cycles (Fisher and Wolfe, 2012; Amos et al., 2013).

Atmospheric Hg is nearly the exclusive source of Hg in forest biomass due to the limitation of root uptake (Grigal, 2003). The forest canopy is a major receptor of Hg in terrestrial forest ecosystems, which can absorb Hg through stomatal uptake of GEM, and adsorb PBM and GOM onto foliage surface (Fu et al., 2015). Therefore, atmospheric deposition by litterfall and throughfall is the largest input of Hg to forested watersheds that are not affected by natural geologic or point sources (Blackwell and Driscoll, 2015b; Zhou et al., 2016b; Wang et al., 2016a). Forest ecosystems are considered as a large sink of atmospheric Hg and an active pool of Hg, which is a vital part in the global Hg cycle (Friedli et al., 2007; Wang et al., 2016a; Ma et al., 2016). Additionally, the large amounts of Hg inputted to the forest are sequestrated in the vegetation and soils, and have produced ecological risks on the bioaccumulation of Hg in the internal forest. For example, Hg sequestrated in the forest soil are considered as potential sources of both total Hg (THg) and methylmercury (MeHg) to downstream aquatic ecosystems (Selvendiran et al., 2008; Ma et al., 2015). Moreover, Hg in the forest soil and biomass can be directly used by forest animals that may be highly vulnerable to the increasing Hg loads (Rimmer et al., 2010; Zhou et al., 2016a).

Dynamic and budget studies quantifying Hg flux and pool in the terrestrial forests are necessary for investigating status of Hg inputs to, retention within, and output from forest ecosystems. Many studies have improved our knowledge of current Hg pools and fluxes (Grigal et al., 2000; Grigal, 2003; Larssen et al., 2008; Ma et al., 2015). However, terrestrial forest has constantly been underestimated as sink for atmospheric Hg on a global scale (Wang et al., 2016b; Obrist, 2007). Previous reviews were mainly focused on the atmospheric Hg concentrations (Fu et al.,

2015), atmospheric Hg depositions (Wang et al., 2016a; Wright et al., 2016) and air–surface fluxes (Zhu et al., 2016); however, no studies aimed at the Hg budgets and quantified the Hg retention in the forest ecosystems. Agnan et al. (2016) suggested that the earth's surface contributed to half of the global natural emissions (607 Mg yr^{-1}); however, the estimated value had a large uncertainty ranges between −513 to 1353 Mg yr^{-1}, due to what degree forests are net sinks or sources of GEM. China, the largest emitting country of anthropogenic Hg source, has done quite a lot of works to positioning the role of forests in the regional- and global-scale Hg biogeochemical cycles. In order to provide a better understanding of current knowledge with respect to forest Hg in China and quantify the forest act as net sinks or sources of GEM, we comprehensively review the forest Hg data in China to estimate the Hg mass balance in forests based on the observations. The important ecological risks of Hg accumulation and storage in forest are also presented. The Hg budgets in forests partly help dissolve the question: what degree the ecosystems are net sinks or sources of atmospheric Hg. The implications and future research needs for further understanding of forest Hg in China are also presented.

2. Processes of Hg input

2.1. Wet input

The THg and MeHg input fluxes by precipitation, throughfall and litterfall in forested area of China are showed in Table 1. The averaged THg and MeHg concentrations in precipitation sampled via wet-only precipitation sampling device at remote forests were 4.5 ng L^{-1} (n = 4, range from 3.0 to 7.4 ng L^{-1}) and 0.06 ng L^{-1} (n = 2, range from 0.04 to 0.08 ng L^{-1}) , respectively. Prospectively, the mean THg and MeHg concentrations in bulk precipitation samples at remote forests of China were 12.5 ng L^{-1} (n = 3, range from 9.9 to 14.2 ng L^{-1}) and 0.16 ng L^{-1} (n = 1), which were much higher than those collected by wet-only precipitation sampling devices (Table 1). Although the PBM and GOM in remote forests were relatively lower, dry deposition of PBM and GOM can also contribute to the elevation of Hg concentrations in bulk precipitation. At rural & suburban forests, the THg and MeHg concentrations were much higher in wet-only precipitation, with the average concentration range from 10.9 to 32.3 ng L^{-1} (n = 5, mean = 18.1 ng L^{-1}) and range from 0.20 to 0.24 ng L^{-1} (n = 2, mean = 0.22 ng L^{-1}), respectively. Wet-only input fluxes of THg and MeHg were comparable and ranged from 5.4 to 6.1 μg m^{-2} yr^{-1} (n = 4, mean = 5.8 μg m^{-2} yr^{-1}) and 0.06 to 0.14 μg m^{-2} yr^{-1} (n = 2, mean = 0.10 μg m^{-2} yr^{-1}) at remote sites, and ranged from 14.4 to 29 μg m^{-2} yr^{-1} (n = 5, mean = 18.1 μg m^{-2} yr^{-1}) and 0.26 to 0.36 μg m^{-2} yr^{-1} (n = 2, mean = 0.31 μg m^{-2} yr^{-1}) at rural & suburban forests, respectively (Table 1). THg concentrations in precipitation and corresponding wet deposition fluxes at rural &

suburban forested areas were elevated compared to those in North America and Europe, but the concentrations and fluxes at remote forests were in the lower range of those obtained from remote forested areas in North America and Europe (Choi et al., 2008; Graydon et al., 2008).

Previous studies suggested that THg in rainwaters was originated from the scavenging of PBM and GOM in the atmosphere (Zhou et al., 2013a). Additionally, Fu et al. (2015) reviewed the THg fluxes in China and observed significant correlations between rainwater THg concentrations and GOM as well as PBM concentrations at urban, suburban and remote areas. However, THg concentrations in precipitations were not significantly correlated with the three Hg forms of GEM, PBM and GOM in the forested areas of China (n= 10, 4 and 4; T test, $p > 0.05$ for all). The reason may be that reduced PBM and GOM in forested areas resulted in low scavenging during wet deposition events (Seigneur et al., 2004). One the other hand, the vast majority of forest was at high altitude with low-level clouds, which limited the scavenging height and reduced the washout efficiency.

2.2. Throughfall and litterfall input

Throughfall and litterfall depositions are the two major pathways for Hg delivery to forest floor. Throughfall is rainfall that delivers to the forest floor after interacting with the forest canopy, which can wash off a large portion of the PBM and GOM deposited to forest leaves (Rea et al., 2000), resulting in higher THg and MeHg concentrations compared to those in precipitation. There are many factors influencing THg concentrations and depositions by throughfall, including canopy type (Demers et al., 2007), meteorological conditions (Blackwell and Driscoll, 2015b) and sample locations (Luo et al., 2015a). In addition, THg concentrations in precipitations also significantly affected these in throughfall duo to similar source in both aqueous, which showed a significant positive correlation (n = 9, r^2=0.92, $p<0.01$). The THg concentrations were ranged from 8.9 to 40.2 ng L^{-1} (n = 3, mean = 28.6 ng L^{-1}) at remote forests and ranged from 20.1 to 69.7 ng L^{-1} (n = 6, mean = 42.5 ng L^{-1}) at rural & suburban forests, which averaged 2.6- and 2.0-fold compared to the corresponding THg concentrations in precipitation (Table 1).

The mean THg depositions by throughfall were 36.3 μg m^{-2} yr^{-1} (rang of 10.5−57.1 μg m^{-2} yr^{-1}) at remote forests and 42.5 μg m^{-2} yr^{-1} (rang of 21.8−71.3 μg m^{-2} yr^{-1}) at rural & suburban forests, respectively. The means of THg inputs are 2–3 times and 4–6 times higher than those of the European values (mean = 19.0 μg m^{-2} yr^{-1}) and the North America values (mean = 9.3 μg m^{-2} yr^{-1}), the ranges of which were between 12.0 and 40.1 μg m^{-2} yr^{-1} and between 2.07 and 25.4 μg m^{-2} yr^{-1}, respectively (Fig. 4). At forests of China, throughfall contributed higher Hg inputs than those of wet inputs, with throughfall ranging from about 1.7 to 2.5 times the wet input (Fig. 1). However, these were

different with the North America forests, where throughfall Hg inputs were found to be lower than wet-only depositions in deciduous forests, but to be higher than wet-only depositions in coniferous forests (Wright et al., 2016).

Litterfall Hg inputs have been confirmed to be the other important pathway trapping atmospheric Hg to the forest floor via senesced leaves, needles, twigs, and branches, and other plant tissues. Concentrations of Hg in litterfall could be affected by many factors, such as tree species, lifespan, and environmental factors (e.g., solar irradiation, air temperature, altitude, etc.) (Blackwell and Driscoll, 2015b; Zhou et al., 2017a). However, atmospheric Hg concentrations play the most important role in Hg concentrations in litterfall, and Hg concentrations in atmosphere were deemed to be a good indicators of leaf Hg contents in forest areas (Fay and Gustin, 2007; Niu et al., 2011). Based on the available atmospheric total gaseous Hg (TGM) or GEM concentrations and litterfall Hg concentrations in 11 forested areas and 14 pairs of datasets in China, annual mean atmospheric TGM/GEM concentrations were significantly correlated with the THg concentrations in litterfall samples (Fig. 2). The significant correlation might verify that foliage can effectively trap Hg from the atmosphere by accumulation Hg through stomatal uptake of GEM (Fay and Gustin, 2007; Fu et al., 2010a, b; Zhou et al., 2017b). The mean THg and MeHg concentrations in litterfall at remote sites ranged from 12.6 to 135.1 ng g^{-1} (mean = 54 ng g^{-1}, n = 12) and from 0.28 to 0.48 ng g^{-1} (mean = 0.38 ng g^{-1}, n = 2), respectively (Table 1). Such litterfall THg and MeHg concentrations were higher in rural & suburban areas, with mean concentration range of 25.8 to 176.1 ng g^{-1} (mean = 61.2 ng g^{-1}, n = 5) and 0.21 to 0.84 ng g^{-1} (mean = 0.52 ng g^{-1}, n = 4), respectively. THg and MeHg concentrations in litterfall at rural & suburban areas of China were higher than those in North America and Europe, but litterfall concentrations of THg and MeHg at remote areas were compared those observed in North America and Europe, except in Mt. Leigong, Guizhou Provence (Table 1, Fig. S1). Although Mt. Leigong was relatively isolated from anthropogenic activities with lower GOM, PBM, precipitation and throughfall Hg concentrations, GEM could undergo long-range transport from emission sources. The GEM concentration was 2.80 ng m^{-3} in Mt. Leigong that is about 170 km to the large Hg mine of Wanshan (Fu et al., 2010a). The relatively higher GEM concentration resulted in elevated litterfall Hg concentrations.

Mean THg inputs by litterfall from 20 forests in China (41.8 μg m^{-2} yr^{-1}) were approximately 2 to 3 times higher than those in Europe over 11 sites (14.2 μg m^{-2} yr^{-1}) and more than 3 times higher than those in North America over 37 sites (12.9 μg m^{-2} yr^{-1}) (Fig. 4). Since litterfall THg inputs to terrestrial ecosystems are estimated by multiplying the biomass and corresponding THg content in litterfall, both of them could influence the input fluxes. Therefore, compared to North America and Europe, higher TGM or GEM concentrations in rural & suburban forests of China resulted in the elevated litterfall Hg concentrations and corresponding higher fluxes in China. However, it should be

noted that the litterfall biomass productions in forests of China (565 ± 450 g m^{-2} yr^{-1}) were more than 2-fold higher than those observed in North America and Europe (200 ± 145 g m^{-2} yr^{-1}). The regional differences of litterfall Hg inputs to forest ecosystems was primarily resulted by the factor of litterfall biomasses rather than litterfall Hg concentrations, as evidenced by the much stronger correlation between litterfall Hg input fluxes and litter biomass productions than that with litterfall THg concentrations (Fig. 3a and b).

The total Hg input as the sum of Hg input by litterfall and throughfall (i.e., input flux by litterfall + input flux by throughfall) to forests were ranged from 47.7 to 291.3 μg m^{-2} yr^{-1} (n=11 from 9 forests) in China (Fig. 1). Here, it should be noted that the highest Hg deposition (291.3 μg m^{-2} yr^{-1}) was observed at Tieshanping forest from March 2005 to March 2006 (Wang et al., 2009); however, due to overestimation of litterfall biomass, the measured Hg fluxes were more than 3 times the recent studies by Luo et al. (2015a) in 2010–2011 and Zhou et al. (2017b) in 2014 –2015. The much higher Hg input at Tieshanping forest is due to it located near the center of Chongqing City (20 km), the annual atmospheric emissions of which just from coal combustion was 4.97 t (Wang et al., 2006) and Hg pollution was regarded as major environmental burdens in Chongqing (Yang et al., 2009). The large mercury emission resulted in much higher Hg deposition fluxes not only in the urban areas but also in the suburban areas (Ma, 2015; Wang et al., 2009, 2014). If we use the updated Hg inputs fluxes by Luo et al. (2015b) at Tieshanping forest, the annually mean total Hg input flux was 73.9 μg m^{-2} yr^{-1} (n=10) in China. Hg input to forest floor via litterfall was substantially comparable or greater than the throughfall input and the litterfall to throughfall input ratios range from 0.33 to 6.59 (mean= 2.14), indicating that Hg input via litterfall surpassed that by throughfall and become the major pathway of Hg input to forests in China. The observed ratios in forest ecosystem of China were much greater than those observed in North America and Europe. Ratios of litterfall Hg input to throughfall Hg input to forest ecosystems were in the range of 0.27 to 1.56 (mean=0.89; n = 9) in Europe (Schwesig and Matzner, 2000; Hultberg et al., 1995; Iverfeldt et al.,1991; Larssen et al., 2008; Lee et al., 2000; Munthe et al., 1995, 1998; Schwesig and Matzner, 2001), and in the range of 0.60 to 4.13 in North America (mean = 1.37; n = 16) (Blackwell and Driscoll, 2015b; Choi et al., 2008; Demers et al., 2007; Kalicin et al., 2008; Kolka, 1999; Grigal et al., 2000; Lindberg et al., 1994; Fisher and Wolfe, 2012; Rea et al., 1996, 2001; Johnson, 2002; Johnson et al., 2007; Nelson et al., 2007; St. Louis et al., 2001; Graydon et al., 2008), which was about 2.4 to 1.6 times lower compared to the ratios observed in China. The reason is the much higher litterfall biomass production in forest of China as we stated above.

Additionally, more than 90% of Hg in litterfall biomass is considered to be uptake from atmosphere, and throughfall can wash off most of the PBM and GOM on the leaf surface by previous dry depositions; therefore,

litterfall and throughfall Hg inputs could be a good indicator of TGM dry deposition to forest ecosystems (Zhou et al., 2013a; Fu et al., 2015). Considering dry Hg input in a forest ecosystem as the difference between total Hg input and wet Hg input (dry Hg input = total Hg input – wet Hg input), more than 80% of total Hg inputs were from dry inputs in forests of China, which was higher than those in North America and Europe (70%) but lower than those in Brazil (85%) (Wang et al., 2016).

Higher dry and wet depositions resulted in higher total Hg inputs to Chinese forests, which averaged 78.4 μg m^{-2} yr^{-1} at remote forests and 106.5 μg m^{-2} yr^{-1} at rural & suburban forests, and ranged from 47.7 to 119.5 μg m^{-2} yr^{-1} (n= 5) and from 56.0 to 291.3 μg m^{-2} yr^{-1} (n= 6), respectively. We have also reviewed the THg inputs by throughfall and litterfall in the Europe and North America (Fig. 4), and the results showed that THg inputs were significantly lower than those observed in China ($p<0.05$ for Europe and $p<0.01$ for North America). Mean THg input was about 39.2 μg m^{-2} yr^{-1} (n= 9) in the Europe, which was about 2.0- and 2.5-fold lower than that observed at remote forests and rural & suburban forests in China. Even lower THg input was found in the North America (20.2 μg m^{-2} yr^{-1}, n= 17) and was about 4- and 5-fold lower than that at remote forests and rural & suburban forests in China.

3. Processes of Hg output

3.1. Exports from surface runoff and underground runoff

The dominate pathways of Hg output from forest catchments were runoffs and soil-atmosphere exchange fluxes. The output fluxes of THg and MeHg via surface runoff measured in China are showed in Table 2. The mean THg and MeHg concentrations in surface runoffs ranged from 2.3 to 17.2 ng L^{-1} (mean = 6.0 ± 4.1 ng L^{-1}, n = 11) and from 0.2 to 0.25 ng L^{-1} (mean = 0.23 ng L^{-1}, n = 2), respectively. Comparing to the THg (40.5± 19.6 ng L^{-1}) and MeHg (0.32 ng L^{-1}) in throughfall, the corresponding Hg concentrations in surface runoffs were seemed much lower, which was consistent with the general concept that atmospherically deposited Hg accumulates in soils rather than being directly transported to streams (Larssen et al., 2008). The export fluxes of THg via surface runoffs and/or stream waters ranged from 3.0 to 8.6 μg m^{-2} yr^{-1} (mean = 4.8 ± 2.6 μg m^{-2} yr^{-1}, n = 6). Luo et al. (2014) collected 117 stream water samples in China, including 42 streams from 9 sites in the northeastern forests and 75 streams from 16 sites in the southern forests, and the result showed that THg concentration was higher in northeastern forests (17.2 ± 11.0 ng L^{-1}) than that in the southern forests (6.2 ± 6.4 ng L^{-1}). The THg concentrations in stream water were positively correlated to DOC concentrations, suggesting that the DOC may facilitate the Hg mobility. Due to cool and dry climate in northern forests, litter decomposed more slowly and resulted in deeper litter and organic layers

than those in southern forests (Zhou et al., 2015a, 2017a). Therefore, soil erosion in northern forests with higher DOC in stream waters resulted in higher THg concentrations.

No statistically significant correlations were showed between THg concentrations in stream water and throughfall (r^2 = 0.00, p>0.05, n = 9), and between throughfall Hg inputs and stream water exports (r^2 = 0.03, p>0.05, n = 6), implying that THg output from stream water was regulated directly by processes other than current deposition input in these forested catchments. However, THg export fluxes via runoff and/or stream waters were significantly correlated with THg concentrations in surface soils (organic layer or top 10 cm) (r^2 = 0.52, p < 0.05, Fig. S2). Higher THg depositions have resulted in much higher soil THg concentrations at forest sites of China. Although soils in forests have been suggested as filters between throughfall and stream waters, but THg in stream waters also can desorb from soils (Xue et al., 2013). Yin et al. (1997) suggested that higher Hg concentrations in the water of prefiltration and soils both could be resulted in higher Hg concentrations in the leachate. Therefore, higher soil Hg contents caused by higher deposition at forests of China caused high Hg concentrations in the stream water. Since the adsorption and desorption of THg in soils cloud also depend on other factors, including the soil physical and chemical properties (pH, organic matter, consistency) and leachate properties (pH, dissolved organic matter, salinity) (Yin et al., 1997; Xue et al., 2013), the deduction may have large uncertainties.

The direct measurements of THg in underground runoffs were not conducted in any forests of China, but they played important roles in the THg export from forests due to both of the amounts and THg concentrations usually higher than those of surface runoffs in subtropical forests (Liu, 2005; Luo et al., 2015b). Several studies have measured THg concentrations in solutions of soil profiles in subtropical forest of Tieshanping, which was averaged 21.8 ng L^{-1} and ranged from 1.98 to 60 ng L^{-1} (Wang et al., 2009; Zhou et al., 2015; Luo et al., 2015b). The observed THg concentrations of soil solution was higher than those in five Swiss forest soils, and the reason may be due to higher THg loads and soil THg content in this Chinses forest. Although no studies directly measured the export flux of THg via underground runoff, we roughly estimated the flux based on the THg in soil solutions and runoff amount in Tieshanping forest, which is 6.0 μg m^{-2} yr^{-1}; therefore, the total Hg output by runoffs as the sum of Hg output by surface runoff (3.5 μg m^{-2} yr^{-1}) and underground runoff (6.0 μg m^{-2} yr^{-1}) was 9.5 μg m^{-2} yr^{-1}.

3.2. Export of soil-atmosphere exchange fluxes

Table 3 shows the statistical summary of soil-atmosphere Hg exchange fluxes and associated site information in the 30 forest sites. Mean soil-atmosphere Hg exchange fluxes at remote forests were in the range of 1.6–4.77 ng m^{-2}

388 hr^{-1} (mean = 3.3 ± 3.4 ng m^{-2} hr^{-1}, n = 12), and those at rural & suburban forests were significantly higher (T test, p
389 < 0.05) and ranged from –0.8 to 17.8 ng m^{-2} hr^{-1} (mean = 8.3 ± 7.1 ng m^{-2} hr^{-1}, n = 18). Generally, soil-atmosphere
390 Hg exchange fluxes are bi-directional. Nevertheless, only one site showed overall net deposition of −0.8 ng m^{-2} hr^{-1}
391 in the wetland of Tieshanping forest and the other forest soils showed overall net emissions in China.

392 Many studies have identified factors that correlate with the magnitude and direction of soil-atmosphere Hg
393 exchange fluxes, including atmospheric and soil physicochemical properties. The well-known factors studied in the
394 previous researches influencing soil-atmosphere Hg exchange fluxes included substrate Hg concentration, air and
395 soil temperature, measurement methodology, as well as environmental variables (e.g. forest type, terrain type and
396 soil cover). The most commonly promoting Hg^0 production is solar radiation that is reported with positive correlations
397 in all the studied forests in China (n = 30). The relationship is mainly attributed to photochemical reduction of soil-
398 bound Hg, which converts soil Hg^{2+} to volatile Hg^0 (Xin et al., 2007; Zhou et al., 2017b). Photo-reduction is a major
399 driver of Hg^0 generation and evasion from soils (Choi and Holsen, 2009; Zhou et al., 2015a, 2017b), although other
400 abiotic and biotic processes also resulted in translation of Hg^{2+} to Hg^0 production, including reduction by humic acids
401 (Allard and Arsenie, 1991) and iron oxides under anoxic conditions (Lin and Pehkonen, 1997) as well as reduction
402 by microorganisms (Agnan et al., 2016) and/or microbial exudates (Poulain et al., 2004, 2007). Additionally, other
403 important correlation was identified with soil or air temperature, which is also significantly correlated to the Hg^0
404 production and observed with soil-atmosphere Hg flux in all the forests in China (n=30). Soil temperature was
405 generally stimulated directly to activation energy of Hg^0 (Gustin et al., 1997; Edwards and Howard, 2013) or
406 stimulation Hg^0 evasion by action of soil microorganism activity (Pannu et al., 2014).

407 Agnan et al. (2016) showed that substrate Hg concentration was significantly correlated with soil-atmosphere Hg
408 fluxes across Hg-enriched sites by large global data set (n = 538), but an apparent lack of correlation between substrate
409 Hg concentrations and soil-atmosphere Hg fluxes across all background soils (n = 307) that defined as substrate Hg
410 concentrations ≤ 300 ng g^{-1} and atmospheric Hg^0 concentrations ≤ 3 ng m^{-3}. Across all vegetation-covered soils
411 (forest and wetland) of China, the correlation between soil Hg concentrations and soil-atmosphere exchange fluxes
412 also did not show significantly across the entire database (r^2 = 0.02, p > 0.05, n = 25), which was consistent with the
413 global database set in background soils (Agnan et al., 2016). The lack of correlation between substrate Hg
414 concentrations and soil-atmosphere Hg fluxes may indicate either little control of soil Hg content on the exchange
415 fluxes across forested areas, or that other parameters prevailed over the effects of soil Hg content. Alternatively, forest
416 areas showed a much narrower range of soil Hg content compared to Hg-enriched substrates, which influenced the

fluxes inconspicuously. However, Zhou et al. (2016c) reported strongly positive correlations between soil Hg contents and fluxes at individual forest of Tieshanping subtropical forest (r^2=0.97, p<0.001) due to the sampling locations that were nearby and have similar other environmental factors.

According to the two-resistance exchange interface model, the exchange fluxes are caused by the gradient of Hg^0 concentrations on both interfaces (Zhang et al., 2002); therefore, high Hg^0 concentrations in the atmosphere will reduce the potential of Hg^0 produced in the soil and diffusion to atmosphere. Laboratory and filed simulation studies showed that elevated atmospheric Hg concentrations significantly inhibited soil Hg volatilizations (Hanson et al., 1995; Poulain et al., 2004; Zhou et al., 2017b). Atmospheric compensation point for Hg^0 flux was firstly presented by Hanson et al. (1995), which is the atmospheric Hg concentration at which no net flux occurs between soil and air (flux to be 0). A previous study using the global database set in background areas showed significant correlation between atmospheric Hg and soil-atmosphere exchange fluxes ($p < 0.001$, $n = 263$) (Agnan et al., 2016). In contrast, based on the database combining all forest-covered soils in China, correlation between atmospheric Hg concentrations and soil-atmosphere exchange fluxes was not significant ($r^2 = 0.05$, $p > 0.05$, $n = 28$), which was inconsistent to the concept of the compensation point. The no correlation was contributed to the variations of environmental factors and Hg emissions at forest sites that resulted in a different buildup of GEM/TGM near the surface in the boundary layer. Thus, high soil emissions caused high GEM/TGM concentrations and not vice versa via a control of air GEM/TGM concentrations on soil-atmosphere exchange fluxes. However, in individual forests, studies showed that compensation points at subtropical forests were in the range of 3.89–6.90 ng m^{-3} in Tieshanping forest stands (Du et al., 2014; Zhou et al., 2016c) and 7.75 ng m^{-3} in Qianyanzhou forest (Luo et al., 2015a), which were much higher than that calculated according to the global database in background sites (2.75 ng m^{-3}, Agnan et al., 2016). Higher compensation points observed in China also imply that natural surface contribute larger atmospheric Hg pools in China.

Additionally, studies have observed predictable influences of environmental variables on Hg^0 exchange across multiple forests when using consistent measurement methodology, such as significant correlations with air relative humidity (Ma et al., 2013, 2015; Du et al., 2014; Luo et al., 2015a). However, it should be noted that the correlation between air humidity and air temperature were also observed, indicating that air temperature may control the air and soil humility. Furthermore, soil moisture stimulated soil Hg emissions at Qianyanzhou and Zhuzhou forests (Luo et al., 2015a; Du et al., 2014) but reduced emissions at Tieshanping forest stands (Du et al., 2014; Zhou et al., 2016c). Previous studies suggested that soil moisture contributed to TGM flux had optimum interval and should be under

intermediate conditions, neither under fairly dry nor very wet (Lin et al., 2010; Pannu et al., 2014; Obrist et al., 2014; Zhou et al., 2017b), which can elucidate the different correlations at different forest ecosystems.

Fig. 5 shows the seasonal variations of soil-atmosphere Hg exchange fluxes at forest areas in China. The variations can be classified into two distinct types: evergreen forest and deciduous forest. At evergreen forests, the mean exchange fluxes in warm seasons (summer and spring) were relative higher than those in cold seasons (winter and autumn, T test: $p < 0.05$ for all). Solar radiation over the forest canopy was much higher in the warm seasons, but the branches and leaves were also luxuriant, so soils received similar sunlight with other seasons at the subtropical evergreen forests (Ma et al., 2013). Therefore, elevated soil-atmosphere Hg exchange fluxes in warm seasons under the evergreen canopy were mainly caused by the increasing soil/air temperature. In contrast, in the deciduous forests, such as larch, mixed broadleaf forest and wetland in Mt. Dongling, the means of soil-atmosphere Hg exchange fluxes were significantly higher in cold seasons (leaf-off period) than that in the other seasons (T test: $p < 0.01$). Solar radiation was the maximum amount reaching the forest floor during leaf-off periods in winter, which was approximately 300 W m^{-2} and promoted Hg^0 production. Whereas during leaf-on periods in summer, the maximum solar radiation at the forest floor was only about 116 W m^{-2}.

In summary, our results suggested that soil-atmosphere Hg exchange fluxes are highly dependent on temperature at the evergreen forests, which increased the rate of reduction of Hg^{2+} by thermal processes, biological activities and stimulating Hg^0 evasion (Poissant et al., 1998; Zhang et al., 2001; Choi and Holsen, 2009). In the deciduous forests, the fluxes were similar to evergreen forests during leaf-on periods, whereas the exchange fluxes are dependent on solar radiation during leaf-off periods because that can directly reach to the forest floor. Although soil received direct solar radiation at forests in north China during leaf-off periods that can be lasted for about half a year (November to April), the exchange fluxes displayed a spatial pattern with significantly lower fluxes in the temperate zones in north China than those at subtropical zones in south China (T test, $p<0.01$) due to lower temperature at temperate zones. Additionally, the remote forests in the temperate zones in north China had similar exchange fluxes to Europe and North America, due to similar forest type, soil properties, TGM concentrations and environmental factors at those forests. However, the fluxes at subtropical zones of remote, rural & suburban forests were generally higher compared to those observed in North America, Europe and South America. The reason may be that forest soils at these areas have higher THg concentrations and receive more solar radiation and causing higher temperature than those at boreal and temperate forests in Europe and North America.

4. Hg budgets

The ultimate fate of Hg deposited to the forest ecosystem may depend on its delivery and incorporation into the forest floor. Input of THg to the forest fields included net throughfall and litterfall depositions and output pathway from the forest ecosystem included runoff outflow and soil Hg emission back to atmosphere (St. Louis et al., 2001; Fu et al., 2010a). A synthesis of Hg input into and output from forests, we conclude the Hg retentions in forest soils in four subtropical forests in south China, including Tieshanping forest, Mt. Gongga, Mt. Simian and Qianyanzhou forest (Fig. 6a). To identify how the Hg retention in the temperate forests in north China, we have also estimated the budgets in three forest stands at Mt. Dongling in north China (Fig. 6b).

Due to no studies estimated the THg export by underground runoff in China, the underground runoff fluxes in the four subtropical forests in south China was estimated according to the runoff amounts and THg concentrations. The runoff amount was estimated to 25% rainfall amount (Liu et al., 2005) and THg concentration in runoff was estimated to same as that in Tieshanping due to similar soil THg concentrations in these areas. The estimated export fluxes by underground runoffs were ranged 6.0 to 9.9 μg m^{-2} yr^{-1} in the four forests. Base on the budget calculation, the THg retention (= throughfall + litterfall – runoff outflow (surface and underground) – soil-atmosphere exchange fluxes) at the subtropical forests ranged from 26.1 to 60.4 μg m^{-2} yr^{-1}, accounted for ranging from 46.6% to 62.8% of THg inputs (Fig. 6a). Evasion of Hg from forest soil was the dominated pathway of THg outputs from the forest compared to runoff outflow. By comparison, the annual loading of THg to subtropical forests in China were much higher compared to some forest catchments in Europe and North America (Larssen et al., 2008; Grigal et al., 2000). Since atmospheric Hg distributions at subtropical areas indicated rural to suburban areas suffered heavy regional Hg emissions from industrial and urban areas (Fu et al., 2015), we infer anthropogenic emissions caused the elevated loading of Hg to subtropical forests in China.

In a study on Hg input at a remote temperate forest ecosystem in Mt. Changbai, northeastern China, THg concentrations in throughfall was approximately 17 ng L^{-1} (Wan et al., 2009a). The forest types at Mt. Changbai were similar to Mt. Dongling in Beijing: mixed forest (600–1100 m a.s.l.), coniferous forest (1100–1700 m a.s.l.), and mountain birch zone (1700–2000 m a.s.l.). Additionally, the TGM concentrations were between 1.60 ± 0.51 ng m^{-3} and 3.58 ± 1.78 ng m^{-3} (Wan et al., 2009b; Fu et al., 2012), which were comparable with the concentration of 2.5 ± 0.5 ng m^{-3} at Mt. Dongling (Zhou et al., 2017a). If we hypothesized the THg concentration in throughfall at Mt. Dongling was also similar to that in Mt. Changbai and throughfall amount were estimated through the mean interception of water-holding capacity of canopy measured by Fei et al. (2011). The estimated inputs of THg

deposition were ranged from 21.40 to 28.73 μg m^{-2} yr^{-1} at Mt. Dongling. As forest types in Mt. Dongling and Changbai are similar, the forest soil types are also similar, which are both mountain brown forest soil (Wang et al., 2013; Zhou et al., 2017a). Therefore, we also referred the Hg concentrations in runoff (5.75 ng L^{-1}) at Mt. Changbai (Wang et al., 2013) and runoff volume were used a previous study in the three stands at Mt. Dongling (Fei et al., 2011). Based on our measured THg concentrations in soil solution (9.2 ng L^{-1}, our unpublished data) and the amounts of underground runoffs in the three stands (Wang et al., 2012), the export fluxes by underground runoffs were estimated. Studies in the Chinese pine plantation, larch plantation and mixed broad-leaved forest found that the annual emission by soil volatilization measured by dynamics chamber and were from 0.87 to 4.03 μg m^{-2} yr^{-1} (Zhou et al., 2016c), and the total Hg outputs of which were 3.1, 2.5 and 9.0 μg m^{-2} yr^{-1}, respectively. Therefore, the annual net retention Hg from the atmosphere was 21.7 μg m^{-2} yr^{-1} for Chinese pine plantation, 26.2 μg m^{-2} yr^{-1} for larch plantation and 12.4 μg m^{-2} yr^{-1} for mixed broad-leaved forest in north China. The ratios of THg retentions to the THg inputs were much higher than these at subtropical forests (T test, $p<0.05$), which accounted for 57.9% to 91.3% of THg deposition. However, it should be noted that the Hg input by throughfall and output by runoff have relative greater uncertainties, so the Hg budget in the temperate forest is roughly estimated in the current study.

The THg retention at subtropical forests in south China were about 2.5 times these at temperate forests in north China. If we hypothesis the total input fluxes of Hg were 20.2 and 39.2 μg m^{-2} yr^{-1} and output were 11.3 μg m^{-2} yr^{-1} (8.6 for soil emission flux, 2.7 for runoff flux) and 8.8 μg m^{-2} yr^{-1} (soil emission flux: 6.7, outflow flux: 2.1) for North America and Europe, respectively, according to the average fluxes for each item, the calculated retention were 8.9 and 30.4 μg m^{-2} yr^{-1}, respectively. The THg retention at subtropical forests was higher compared to these in North America (3.8 to 7.9 folds) and Europe, and the retention in the temperate forest was lower compared to those in the Europe but higher compared to those in North America (1.2 to 2.8 folds).

5. Hg storage and risk assessment

5.1. Hg storage in soils

Highly elevated THg contents in forest top soils were mostly likely originated from atmospheric depositions via litterfall and throughfall, whereas very limited source was originated from geological sources (Obrist et al., 2011). Table S1 summarizes all studies of soil Hg concentrations and pools at forests of China from the literature. However, it is should be note that the attempts to compare soil Hg concentrations and pools with the data from each other and some other studies are facing difficulties, because these studies either reported the amounts of THg accumulated in

different horizons or calculated THg pools stored in soil profiles of different depths, which were inconsistent with each other.

Declining Hg concentrations with soil depth are generally observed in organic to mineral layers and did not vary in the lower mineral soils from all the soil profiles in Chinese forests. Highest THg concentrations observed in litter and upper soils are indicative of Hg sorption from atmospheric deposition to upper soil horizons. As organic soils are net traps of deposited atmospheric Hg and topsoil concentrations reflect recent Hg depositions from the atmosphere, we concluded THg concentrations from topsoil (most in the organic horizons) in the Fig. S3. The soil THg concentration at remote forests averaged 150 ng g^{-1} and the median concentration was 104 ng g^{-1}, ranging from 59 to 353 ng g^{-1} (n = 18). The concentrations were slight higher than those observed in remote areas of North America, which were generally less than 150 ng g^{-1} for surface soils (Larssen et al., 2008; Obrist et al., 2011). The THg concentrations at rural & suburban forests were much higher than these observed at remote forests, which ranged from 76 to 332 ng g^{-1} (mean: 198 ng g^{-1}; median: 196 ng g^{-1}). This is in a good agreement with the elevated atmospheric Hg concentrations and higher loading of Hg in at rural & suburban forests of China, which can be proved by the significant correlation between Hg retentions and soil THg concentrations (r^2=0.62, $p< 0.05$, n=7). Predictably, higher THg depositions and soil THg concentrations has resulted higher THg pools in forest soils. For example, in the remote forests of Mt. Gongga and Mt. Ailao, the THg storage were up to 152.3 and 191.3 mg m^{-2} in the soil profiles of 90 and 80-cm depth, which were much higher than these in the upland forest of central Adirondack Mountain of USA and (64 mg m^{-2} in 0–90 cm depth) (Selvendiran et al., 2008) and upland forest of Steinkreuz, Germany (19 mg m^{-2} in 0–60 depth) (Schwesig and Matzner, 2000). However, THg storage in forest soils of temperate forests and Tibet Plateau with relative lower atmospheric Hg deposition (Zhou et al., 2017a; Gong et al., 2014), were comparable to that in North America and Europe.

5.2. Hg storage in biomass

Vegetation is known to exert significant influence the dynamics of Hg in the forest ecosystem including atmospheric Hg input and output in the terrestrial ecosystem (Ma et al., 2016; Zhou et al., 2016a). Two studies investigated the Hg distribution in the tissues of vegetation at the subtropical forest (Tieshanping forest, Zhou et al., 2016) and temperate forest (Mt. Dongling, Zhou et al., 2017a) and highest THg concentrations are observed in the O horizons compared to THg in the other biomass, because organic matter was enhanced during natural processes of litterfall decomposition and transformation, in which organic matter binding Hg compounds are usually more

562 stabilized via complexing, humification and adsorption to clay minerals (Demers et al., 2007; Zhou et al., 2017a).
563 Sequentially, relative higher THg was observed in the litterfall and leaf due to canopy leaf can effectively capture Hg
564 in atmosphere, which can uptake Hg by stomata (Fu et al., 2015).

565 Root is contacted with mineral soil directly, likely to higher concentration than that of aboveground wood (Grigal,
566 2003). THg concentrations in roots of Norway spruce in southern Sweden were 40 ng g^{-1} (Munthe et al., 1998), which
567 was much lower than that in the root of Masson pine in southwestern China (71 ng g^{-1}, Zhou et al., 2016a) due to
568 large THg loading in this area. Only Zhou et al. (2016) estimated the THg pools in roots that accounted for about 34%
569 of the overstory THg pools. Bole wood had the largest biomass of vegetation in the forest, but lowest THg
570 concentrations were observed. A previous study suggested that the source of the THg in wood was translocated from
571 foliage (Barghigiani et al., 1991). Concentrations of Hg were positively correlated in 11 pairs of leaf and adjacent
572 bole wood samples of different tree species at forests of China (Fig. S4). It is reasonable for their correlation because
573 leaf and bole wood are both exposed, one directly and the other indirectly to the same atmospheric pool of Hg.
574 However, no significant correlation was observed between THg concentrations in bark and bole wood or leaf,
575 probably due to that the THg accumulation rates were differed in the barks of different tree species.

576 THg concentrations of each component at the suburban forest of Tieshanping at subtropical zone was much higher
577 than those at the remote forest of Mt. Dongling at temperate zone. Accordingly, much higher THg pool of 103.5 mg
578 m^{-2} showed in suburban forest of Tieshanping than that of 7.3–10.8 mg m^{-2} in remote forest of Mt. Dongling (Fig.
579 S5). The THg pools in North America were much lower than those at subtropical forest of China and comparable to
580 those at temperate forest of China (Friedli et al., 2007; Obrist et al., 2009; Richardson et al., 2013). Nonetheless, soil
581 THg pools accounted for over 90% of the total ecosystem Hg pools forests around the world.

582

583 5.3. Risk assessment

584 The studies summarized in this review showed significant inputs and retention of Hg in forest ecosystems in
585 China. The apparent accumulation and storage of THg may present an important ecological risk. Firstly, the Hg in
586 forest soil could be re-emitted back to the atmosphere. Organic matter has a high binding ability of Hg in forest
587 surface soils, but the Hg bonded organic carbon would probably be released to the environment as the decomposition
588 of organic matter occurs. Studies on climate change showed that the accelerated global warming would accelerate
589 the decomposition of organic carbon (Schimel et al., 1994), which could probably accelerate Hg emission from soil
590 (Obrist, 2007; Fu et al., 2010a). Additionally, the increasing of global temperature would aggravate the occurrence

of potential fires and causing large pulses of Hg to the global atmospheric pool (Zhou et al., 2016a, 2017a).

Secondly, the Hg retention in the forest soils would accumulate through food webs, threatening the balance of forest ecosystems (Rimmer et al., 2010). However, the relevant studies in China were rare. Many studies showed that mushroom had high accumulation ability of THg and MeHg from substrate (like soil, litter and wood) and strong translocation to the fruiting bodies (Falandysz et al., 2015a, b, 2016). Studies in southwestern China showed that THg concentrations in the Fungi *Boletus* species and genus *Leccinum* species were up to 3500–4800 ng g^{-1} (mean 42000 ng g^{-1}) and 4900–22000 ng g^{-1} (10900 ng g^{-1}) dry matter, respectively (Falandysz et al., 2015a, b). Similarly, a study in Poland also showed efficient accumulation of THg in the *Leccinum* mushrooms, but the average Hg concentrations being an order of magnitude lower because of lower concentrations of THg in surface forest soil of Poland. Although some lowly cumulative species of mushroom were observed in the subtropical forests (Kojta et al., 2015), mushroom is an important food item in southwestern China, and high rates of consumption can deliver relatively high doses of Hg to local human beings (Kojta et al., 2015; Falandysz et al., 2015a, b, 2016). The chronic dietary intake (CDI, μg kg^{-1} bw day^{-1}) of Hg depends on both the mushroom Hg concentrations (C) and the daily intake rates (IR), which are widely used to predict the exposure level of humans to trace elements(Du et al., 2016; Zhou et al., 2018). According to the Exposure Factors Handbook of the US Environmental Protection Agency, the CDI can be calculated as

$$CDI = \sum (C \times IR)/BW \quad (1)$$

where BW (kg) is body weight. The IR was assumed as 43 g day^{-1} and the bw was assumed as 60 kg for Chinese residents according to the previous studies in Yunnan province (Kojta et al., 2015).

According to the *CDI* of mushroom consumptions, a Hazard Quotient (*HQ*) indicating the non-carcinogenic health risk during a lifetime can be calculated by dividing the *CDI* by the toxicity threshold value of the reference dose (*RfD*).

$$HQ = CDI/RfD \quad (2)$$

The recommended *RfD* of Hg by Joint Food and Agriculture Organization (FAO)/WHO Expert Committee on Food Additives is 0.57 μg kg^{-1} bw day^{-1} (JECFA 2010). When the *HQ* is ≤ 1, the adverse health effects are unlikely experienced, whereas the value > 1 indicates potential non-carcinogenic effects. Based on the averaged Hg concentrations in the mushrooms from five studies in subtropical forests of China, all the values of HQ showed > 1, demonstrating a much higher non-carcinogenic risk to consumers caused by daily intake of Hg through mushroom ingestions (Table S2). The result suggested the need for greater focus on the adverse health effects induced by Hg on

the consumers in China.

Additionally, the ecological stress to forest insect were investigated in an suburban forest (Tieshangping) in China, which showed that insect living in the soil has two to three orders of magnitude higher THg accumulation than that living on the plant due to large Hg pools in the forest soils (Zhou et al., 2016a). Although animals in the high position of the food chain were not studied in forest of China, Rimmer et al. (2010) showed that food web reflected the transfer of Hg from lower to higher trophic levels with a resulting increase in Hg burden. Therefore, we can infer that Hg will be seriously bioaccumulated along the food chain and pose risk to the local creatures by physiological toxicity.

6. Environmental implication and research needs

The large THg retention of in the forest ecosystem suggested strong adsorption and absorption of Hg by vegetation that was underestimated by global modeling of previous studies. If we roughly estimated the THg deposition at forests of China using the average THg depositions (92.45 μg m^{-2} yr^{-1}) by present studies and the forest area (2.08×10^{12} m^2) in 2015, the THg deposition would be 192.3 t yr^{-1} in forest areas of China. GEOS-Chem model estimates the mean dry deposition of 12.3 mg m^{-2} yr^{-1}, which converted to the total Hg deposition in China is <121.0 t yr^{-1} (Wang et al., 2014). Given that more than 80% of the THg deposition was from dry deposition, the THg dry deposition was 153.8 t yr^{-1} in forest ecosystems of China, which is even higher than the total Hg deposition in the whole mainland China. Therefore, a large underestimation compared to the observation-based estimate just from forest areas of China in this study. Therefore, future model studies should consider the THg dry deposition in forested areas individually.

Hg sequestrated in forest litters and surface soil by legacy Hg retention can be quickly volatilized to the atmosphere by soil-atmosphere exchanges. Recent global Hg models suggested that soils not only act as net sinks but also as net sources for atmospheric Hg in global Hg cycling (Amos et al., 2013), and the role of forest ecosystems as atmospheric Hg sink or a source are existing confliction (Lindberg et al., 1991, 1998; Pirrone et al., 2010). Using the global database of terrestrial surface−atmosphere fluxes, forest ecosystems appear a net deposition of 59 t yr^{-1}, but the estimation existed large uncertainties and ranged (37.5th−62.5th percentiles) from a deposition of 727 t yr^{-1} to an emission of 703 t yr^{-1} (Agnan et al., 2016). Base on the field observations of THg retention in Chinese forests, the THg retention in forest soils was 69 t yr^{-1} just in China, which was much higher than the global data of 59 t yr^{-1} (Agnan et al., 2016). Such difference is mainly resulted from the variation of reported atmospheric Hg uptake by foliage and the limited geospatial representation of available data (Wang et al, 2016; Zhu et al., 2016; Agnan et al., 2016). Thus, more studies should be conducted to character the whole-ecosystem fluxes and to question to what

649 degree the ecosystems are net sinks or sources of atmospheric Hg.

650 To better assess the role of forest ecosystems in the global Hg cycling, it is also essential to understand the THg
651 pools in the branches, stems and roots that can be translated from the atmosphere by the foliage uptake. A previous
652 study estimated that approximately 139 t yr^{-1} Hg was stored in bole woods (Obrist et al., 2007). However, there is no
653 study exactly quantifying the amount of Hg translocation after Hg uptake by leaves, and the THg storage in biomass
654 are scarce and need more data. Further studies concerning the transformation and migration processes after vegetation
655 uptake will benefit to constrain atmospheric Hg sink in forest ecosystems.

656 In addition, the large "active" soil pool at forests is a potential short-term and long-term source of THg and MeHg
657 to downstream aquatic ecosystems (Selvendiran et al., 2008; Ma et al., 2015). However, there is no study reporting
658 the accumulation of THg and MeHg in aquatic ecosystem after output from the forest ecosystem. The processes of
659 Hg methylation, transformation and translocation may be different from those in North America and Europe because
660 of the larger Hg deposition and storage in China, which requires further investigation when more data become
661 available.

662

663 **7. Conclusions**

664 As the largest energy consumer and anthropogenic Hg emission in the world, much attention has been paid to
665 characterize the behavior of Hg in China. Forests are regarded as large pools of Hg in the global Hg cycle. In this
666 paper, an integrated review of the knowledge reported in peer-reviewed literature is provided. Hg deposition and
667 pools have been found to be substantially elevated in both remote, rural & suburban forests of China compared to
668 those observed in North America and Europe. A strong spatial variation in Hg pools was observed, with high storage
669 related to regional atmospheric Hg concentrations in southern China. The large Hg storage in the forests pose a
670 serious threat for large pluses to the atmospheric Hg during accelerated organic matter decomposition and potential
671 wildfires, and additional ecological stress to forest animals. However, very few studies are attempted to study the
672 ecological risk of Hg in the forest ecosystem in China, which are suffering highly Hg depositions.

673 The forests play important roles in the geochemical cycles of Hg in China. According to the budget calculation,
674 the THg retention ranged from 26.1 to 60.4 μg m^{-2} yr^{-1} at the subtropical forests in southern China, accounted for
675 ranging from 46.6% to 62.8% of THg inputs, and ranged from 12.4 to 26.2 μg m^{-2} yr^{-1} at the temperate forests in
676 northern China. The Hg retention and storage pools in at the subtropical forests were much higher than those in North
677 America, but those in the temperate forests were comparable to Europe and North America. The result of the current

review may answer the question to what degree the ecosystems are net sinks or sources of atmospheric Hg in China. However, further studies are needed to accurately quantify Hg budgets and retentions of Hg in different forests ecosystems in China, as well as the atmospheric Hg budget in China.

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Reference:

Agnan, Y., Le Dantec, T., Moore, C. W., Edwards, G. C., and Obrist, D.: New constraints on terrestrial surface–atmosphere fluxes of gaseous elemental mercury using a global database, Environ. Sci. Technol., 50(2), 507–524, 2016.

Allard, B., and Arsenie, I.: Abiotic reduction of mercury by humic substances in aquatic system – an important process for the mercury cycle, Water Air Soil Poll., 56, 457–464, 1991.

Almeida, M. D., Marins, R. V., Paraquetti, H. H. M., Bastos, W. R. and Lacerda, L.: D. Mercury degassing from forested and open field soils in Rondônia, Western Amazon, Brazil, Chemosphere, 77 (1), 60− 66, 2009.

Amos, H. M., Jacob, D. J., Streets, D. G., and Sunderland, E. M.: Legacy impacts of all-time anthropogenic emissions on the global mercury cycle, Global Biogeochem. Cyc., 27(2), 410–421, 2013.

Barghigiani, C., Ristori, T., and Bauleo, R.: Pinus as an atmospheric Hg biomonitor, Environ. Technol., 12, 1175–1181, doi:10.1080/09593339109385118, 1991.

Blackwell, B. D., and Driscoll, C. T.: Deposition of mercury in forests along a montane elevation gradient, Environ. Sci. Technol., 49, 5363–5370, 2015b.

Blackwell, B. D., and Driscoll, C. T.: Using foliar and forest floor mercury concentrations to assess spatial patterns of mercury deposition. Environ. Pollut., 202, 126-134, 2015a.

Bushey, J. T., Nallana, A. G., Montesdeoca, M. R., and Driscoll, C. T.: Mercury dynamic of a northern hardwood canopy, Atmos. Environ., 42, 6905-6914, 2008.

Carpi, A., and Lindberg, S. E.: Application of a Teflon TM dynamic flux chamber for quantifying soil mercury flux: tests and results over background soil, Atmos. Environ., 32 (5), 873−882, 1998.

Carpi, A.; Fostier, A. H.; Orta, O. R.; dos Santos, J. C.; and Gittings, M. Gaseous mercury emissions from soil following forest loss and land use changes: field experiments in the United States and Brazil, Atmos. Environ. 2014, 96, 423−429.

Chen, C., Wang, H. H., Zhang, W., Hu, D., Chen, L., and Wang, X. J.: High-resolution inventory of mercury emissions from biomass burning in China for 2000–2010 and a projection for 2020, J. Geophys. Res.–Atmos., 118 (21), 12248−12256, 2013.

Choi, H. D., and Holsen, T. M.: Gaseous mercury emissions from unsterilized and sterilized soils: The effect of temperature and UV radiation, Environ. Pollut., 157, 1673–1678, 2009.

Choi, H. -D., Sharac, T. J., and Holsen, T. M.: Mercury deposition in the Adirondacks: A comparison between precipitation and throughfall, Atmos. Environ., 42, 1818−1827, 2008.

Demers, J. D., Driscoll, C. T., Fahey, T. J., and Yavitt, J. B.: Mercury cycling in litter and soil in different forest types in the Adirondack Region, New York, USA, Ecol. Appl., 17, 1341–1351, 2007.

Du B.: Field measurement of soil mercury emission flux in forest. Master's dissertation, Tsinghua University, Beijing, China, 1–112, 2014. (in Chinese).

Du, B., Li, P., Feng, X., Qiu, G., Zhou, J., and Maurice, L.: Mercury exposure in children of the Wanshan mercury mining area, Guizhou, China, Inter. Int. J. Environ. Res. Public Health 13, 1107, 2016.

Edwards, G. C., and Howard, D. A.: Air-surface exchange measurements of gaseous elemental mercury over naturally enriched and background terrestrial landscapes in Australia, Atmos. Chem. Phys., 13, 5325–5336, doi:10.5194/acp-13-5325-2013, 2013.

Ericksen, J. A., Gustin, M. S., Xin M., Weisberg, P. J., and Femandez, G. C. J.: Air–soil exchange of mercury from background soils in the United States, Sci. Total Environ., 366, 851–863, 2006.

Falandysz, J., and Drewnowska, M.: Distribution of mercury in amanita fulva (schaeff.) secr. mushrooms: accumulation, loss in cooking and dietary intake, Ecotox. Environ. Safe., 115, 49–54, 2015a.

Falandysz, J., Saba, M., Liu, H. G., Li, T., Wang, J. P., Wiejak, A., Zhang, J., Wang, Y. Z., and Zhang, D.: Mercury in forest mushrooms and topsoil from the Yunnan highlands and the subalpine region of the Minya Konka summit in the eastern Tibetan plateau, Environ. Sci. Pollut. Res., 23(23), 1–12, 2016.

Falandysz, J., Zhang, J., Wang, Y., Krasińska, G., Kojta, A., Saba, M., Shen, T., Li, T., and Liu, H.: Evaluation of the mercury contamination in mushrooms of genus leccinum from two different regions of the world: accumulation, distribution and probable dietary intake, Sci. Total Environ., 537, 470–478, 2015b.

Fay, L., and Gustin, M.: Assessing the influence of different atmospheric and soil mercury concentrations on foliar mercury concentrations in a controlled environment, Water Air Soil Poll., 181, 373–384, 2007.

Fei, M. O., Xuyong, L. I., Shuxia, H. E., and Wang, X.: Evaluation of soil and water conservation capacity of different forest types in Dongling Mountain, Acta Ecologica Sinica, 31(17), 5009−5016, 2011. (in Chinese with English abstract)

Fisher, L. S., and Wolfe, M. H.: Examination of mercury inputs by throughfall and litterfall in the Great Smoky Mountains National Park, Atmos. Environ., 47, 554−559, 2012.

Friedli, H. R., Radke, L. F., Payne, N. J., Mcrae, D. J., Lynham, T. J., and Blake, T. W.: Mercury in vegetation and organic soil at an upland boreal forest site in Prince Albert National Park, Saskatchewan, Canada, J. Geophys. Res., 112, G01004. http://dx.doi.org/10.1029/2005JG000061, 2007.

Fu, X. W., Feng, X. B., Zhu, W. Z., Wang, S. F., and Lu, J. L.: Total gaseous mercury concentrations in ambient air in the eastern slope of Mt. Gongga, South-Eastern fringe of the Tibetan plateau, China, Atmos. Environ., 42, 970–979, doi:10.1016/j.atmosenv.2007.10.018, 2008a.

Fu, X. W., Feng, X. B., Zhu, W. Z., Zheng, W., Wang, S. F., and Lu, J. Y.: Total particulate and reactive gaseous mercury in ambient air on the eastern slope of the Mt. Gongga area, China, Appl. Geochem., 23, 408–418, doi:10.1016/j.apgeochem.2007.12.018, 2008b.

Fu, X. W., Feng, X., Dong, Z. Q., Yin, R. S., Wang, J. X., Yang, Z. R., and Zhang, H.: Atmospheric gaseous elemental mercury (GEM) concentrations and mercury depositions at a high-altitude mountain peak in south China, Atmos. Chem. Phys., 10, 2425–2437, doi:10.5194/acp-10-2425-2010, 2010a.

Fu, X. W., Feng, X., Shang, L. H., Wang, S. F., and Zhang, H.: Two years of measurements of atmospheric total gaseous mercury (TGM) at a remote site in Mt. Changbai area, Northeastern China, Atmos. Chem. Phys., 12, 4215–4226, doi:10.5194/acp-12-4215-2012, 2012.

Fu, X. W., Feng, X., Zhu, W., Rothenberg, S., Yao, H., and Zhang, H.: Elevated atmospheric deposition and dynamics of mercury in a remote upland forest of southwestern China, Environ. Pollut., 158, 2324–2333, doi:10.1016/j.envpol.2010.01.032, 2010a.

Fu, X. W., Zhang, H., Yu, B., Wang, X., Lin, C. -J., and Feng, X. B., Observations of atmospheric mercury in China: a critical review, Atmos. Chem. Phys., 15, 9455–9476, doi:10.5194/acp-15-9455-2015, 2015.

Fu, X., Feng, X., and Wang, S.: Exchange fluxes of hg between surfaces and atmosphere in the eastern flank of mount gongga, sichuan province, southwestern China, Journal of Geophysical Research–Atmospheres, 113(D20), 253–270, 2008c.

Fu, X., Xu, Y., Lang, X., Zhu, J., Zhang, H., Yu, B., Yan, H., Lin, C. -J., and Feng, X. B.: Atmospheric wet and litterfall mercury deposition in typical rural and urban areas in China, Atmos. Chem. Phys., 16(18), 11547–11562, 2016.

Gong, P., Wang, X. -P., Xue, Y. -G., Xu, B. -Q., and Yao, T. -D.: Mercury distribution in the foliage and soil profiles of the Tibetan forest: Processes and implications for regional cycling, Environ. Pollut., 188, 94–101, 2014.

Graydon, J. A., St. Louis, V. L., Hintelmann, H., Lindberg, S. E., Sandilands, K. A., Rudd, J. W. M., Kelly, C. A., Hall, B. D., and Mowat, L. D.: Long-term wet and dry deposition of total and methyl mercury in the remote boreal ecoregion of Canada, Environ. Sci. Technol., 42, 8345–8351, 2008.

Grigal, D.F., Kolk, R. K., Fleck, J. A., and Nater, E. A.: Mercury budget of an upland-peatland watershed, Biogeochemistry, 50, 95–109, 2000.

Grigal, D.F.: Mercury sequestration in forests and peatlands: A review, J. Environ. Qual., 32, 393–405, doi:10.2134/jeq2003.3930, 2003.

Gustin, M. S., Taylor, G. E., and Maxey, R. A.: Effect of temperature and air movement on the flux of elemental mercury from substrate to the atmosphere, J. Geophys. Res.–Atmos., 102, 3891–3898, 1997.

Hanson, P. J., Lindberg, S. E., Tabberer, T. A., Owens, J. G., and Kim, K. H.: Foliar exchange of mercury-vapor-evidence for a compensation point, Water Air Soil Poll., 80, 373–382, 1995.

Hartman, J. S., Weisberg, P. J., Pillai, R.; Ericksen, J. A., Kuiken, T., Lindberg, S. E., Zhang, H.; Rytuba, J. J., and Gustin, M. S.: Application of a rule-based model to estimate mercury exchange for three background biomes in the continental United States, Environ. Sci. Technol., 43 (13), 4989−4994, 2009.

Horowitz, H. M., Jacob, D. J., Zhang, Y., Dibble, T. S., Slemr, F., Amos, H. M., Schmidt, J. A., Corbitt, E. S., Marais, E. A., and Sunderland, E. M.: A new mechanism for atmospheric mercury redox chemistry: implications for the global mercury budget, Atmos. Chem. Phys., 17, 6353-6371, https://doi.org/10.5194/acp-17-6353-2017, 2017.

Hultberg, H., Munthe, J., and Iverfeldt, Å.: Cycling of methylmercury and mercury – Responses in the forest roof catchment to three years of decreased atmospheric deposition, Water Air Soil Poll., 80, 415–424, 1995.

Iverfeldt, Å.: Mercury in forest canopy throughfall water and its relation to atmospheric deposition, Water Air Soil Poll., 56, 553–564, 1991.

JECFA, (Joint FAO/WHO Expert Committee on Food Additives). (2010). Joint FAO/WHO food standards programme, committee of the codex alimentarius commission, 33rd session. Geneva, Switzerland, July 5–9.

Johnson, K. B.: Fire and its effects on mercury and methylmercury dynamics for two watersheds in Acadia National Park, Maine, MSc. Thesis, the University of Maine, Maine, 73 pp., 2002.

Johnson, K. B., Haines, T. A., Kahl, J. S., Norton, S. A., Amirbahman, A., and Sheehan, K. D.: Controls on mercury and methylmercury deposition for two watersheds in Acadia National Park, Maine, Environ, Monit. Assess., 126, 55–67, 2007.

Juillerat, J. I., Ross, D. S., and Bank, M. S.: Mercury in litterfall and upper soil horizons in forested ecosystems in Vermont, USA, Environ. Toxicol. Chem., 31, 1720–1729, 2012.

Kalicin, M. H., Driscoll, C. T., Yavitt, J., Newton, R., and Munson, R.: The Dynamics of Mercury in Upland Forests of the Adirondack Region of New York, in: Mercury in Adirondack wetlands, lakes and terrestrial systems (MAWLTS), New York State Energy Research and Development Authority, New York, 8-1-8-15, 2008.

Kojta, A. K., Zhang, J., Wang, Y., Li, T., Saba, M., and Falandysz, J.: Mercury contamination of fungi genus xerocomus in the Yunnan province in China and the region of Europe, J. Environ. Sci. Heal. A, 50(13), 1342–1350, 2015.

Kolka, R. K., Nater, E. A., Grigal, D. F., and Verry, E. S.: Atmospheric inputs of mercury and organic carbon into a forested upland/bogwatershed, Water Air Soil Poll., 113, 273–294, 1999.

Kuiken, T., Gustin, M., Zhang, H., Lindberg, S. and Sedinger, B.: Mercury emission from terrestrial background surfaces in the eastern USA. II: Air/surface exchange of mercury within forests from South Carolina to New England, Appl. Geochem., 23 (3), 356−368, 2008a.

Kuiken, T., Zhang, H., Gustin, M., and Lindberg, S.: Mercury emission from terrestrial background surfaces in the eastern USA. Part I: Air/surface exchange of mercury within a southeastern deciduous forest (Tennessee) over one year, Appl. Geochem., 23 (3), 345−355, 2008b.

Kyllönen, K., Hakola, H., Hellen, H., Korhonen, M. and Verta, M.: Atmospheric mercury fluxes in a Southern boreal forest and wetland, Water, Air, Soil Pollut., 223 (3), 1171−1182, 2012.

Lang, X., Mercury in atmospheric precipitation and litterfall in Mt. Ailao and Mt. Damei, Master's dissertation, Guizhou University, Guiyang, China, 1–78, 2014 (in Chinese).

Larssen, T., de Wit, H., Wiker, M., and Halse, K.: Mercury budget of a small forested boreal catchment in southeast Norway, Sci. Total Environ., 404, 290–296, doi:10.1016/j.scitotenv.2008.03.013, 2008.

Lee, Y. H., Bishop, K. H., and Munthe, J.: Do concepts about catchment cycling of methylmercury and mercury in boreal catchments stand the test of time? Six years of atmospheric inputs and runoff export at Svartberget, northern Sweden, Sci. Total Environ., 260, 11–20, 2000.

Lin, C. J., Gustin, M. S., Singhasuk, P., Eckley, C., and Miller, M.: Empirical models for estimating mercury flux from soils, Environ. Sci. Technol., 44, 8522–8528, 2010.

Lin, C.-J. and Pehkonen, S. O.: Aqueous free radical chemistry of mercury in the presence of iron oxides and ambient aerosol, Atmos. Environ., 31, 4125–4137, 1997.

Lindberg, S. E. and Stratton, W. J., Atmospheric mercury speciation: Concentrations and behaviour of reactive gaseous mercury in ambient air, Environ. Sci. Technol., 32, 49–57, 1998.

Lindberg, S. E., Brooks, S., Lin, C.-J., Scott, K. J., Landis, M. S., Stevens, R. K., Goodsite, M., and Richter, A.: Dynamic Oxidation of Gaseous Mercury in the Arctic Troposphere at Polar Sunrise, Environ. Sci. Technol., 36, 1245-1256, 2002.

Lindberg, S. E., Owens, J. G., and Stratton, W. J., Application of throughfall methods to estimate dry deposition of mercury, in: Mercury as a global pollutant, Huckabee, J. and Watras, C. (Eds.), Lewis Publications, 261–272, 1994.

Lindberg, S. E., Turner, R. R., Meyers, T. P., Taylor, G. E., and Schroeder, W. H.: Atmospheric concentrations and deposition of Hg to a deciduous forest atwalker branch watershed, Tennessee, USA, Water Air Soil Poll., 56(1), 577–594, 1991.

Lindberg, S. E.: In Global and Regional Mercury Cycles: Sources, Fluxes and Mass Balances; Baeyens, W., Ebinghaus, R., Vasiliev, O., Eds.; NATO-ASI-Series, Vol. 21; Kluwer Academic Publishers: Dordrecht, The Netherlands, pp 359-380, 1996.

Liu, H.: Dynamics of soil properties and the effects factors among secondary successive communities in Mt. Jinyun, Doctor's dissertation, Southwest Agricultural University, 2005 (in Chinese with English abstract).

Luo Y.: Mercury input, output and transport in forest ecosystems in southern China, Doctor's dissertation, Tsinghua Universit, Beijing, China, 1–112, 2015a (in Chinese).

Luo, Y., Duan, L., Wang, L., Xu, G., Wang, S., and Hao, J.: Mercury concentrations in forest soils and stream waters in northeast and south China, Sci. Total Environ., 496, 714–720, 2014.

Luo, Y., Duan, L., Xu, G., and Hao, J.: Inhibition of mercury release from forest soil by high atmospheric deposition of Ca2+ and SO42-. Chemosphere, 134, 113-119, 2015b.

Ma, M., Wang, D., Du, H., Sun, T., Zhao, Z., Wang, Y., and Wei, S.: Mercury dynamics and mass balance in a subtropical forest, southwestern China, Atmos. Chem. Phys., 16, 4529–4537, 2016

Ma, M., Wang, D., Sun, R., Shen, Y., and Huang, L.: Gaseous mercury emissions from subtropical forested and open field soils in a national nature reserve, southwest China, Atmos. Environ., 64, 116–123, 2013.

Ma, M., Wang, D., Sun, T., Zhao, Z., and Du, H.: Forest runoff increase mercury output from subtropical forest catchments: an example from an alpine reservoir in a national nature reserve (southwestern China), Environ. Sci. Pollut. Res., 22(4), 2745–2756, 2015.

Ma, M.: Mercury inputs, outputs, and sources under the forest canopy in typical subtropical forest ecosystem of southwest China, PhD Dissertation, Southwest University, Chongqing, China, 1–116, 2015 (in Chinese).

Magarelli, G., and Fostier, A. H.: Quantification of atmosphere - soil mercury fluxes by using a dynamic flux chamber: application at the Negro river basin, amazon, Química Nova, 28(6), 968-974, 2005.

Munthe, J. Hultberg, H., and Iverfeldt, A.: Mechanisms of deposition of methylmercury and mercury to coniferous forests, Water Air Soil Poll., 80, 363–371, 1995.

Munthe, J., Pleijel, K., Iverfeldt, A., Kruger, O., and Petersen, G.: Atmospheric deposition of mercury in the Nordic countries at, different scenarios of reduced anthropogenic emissions in Europe, IVL Rapport B, 1998.

Nelson, S. J., Johnson, K. B., Kahl, J. S., Haines, T. A., and Fernandez, I. J.: Mass balances of mercury and nitrogen in burned and unburned forested watersheds at Acadia National Park, Maine, USA, Environ, Monit. Assess, 126, 69–80, 2007.

Niu, Z., Zhang, X., Wang, Z., and Ci, Z.: Mercury in leaf litter in typical suburban and urban broadleaf forests in China, J. Environ. Sci., 23(12), 2042-2048, 2011.

Obrist, D., Johnson, D. W., and Lindberg, S. E.: Mercury concentrations and pools in four Sierra Nevada forest sites, and relationships to organic carbon and nitrogen, Biogeosciences 6, 765–777, 2009.

Obrist, D., Johnson, D. W., Lindberg, S. E., Luo, Y., Hararuk, O., Bracho, R., Battles, J. J., Dail, D. B., Edmonds, R. L., Monson, R. K., Ollinger, S. V., Pallardy, S. G., Pregitzer, K. S., and Todd, D. E.: Mercury distribution across 14 U.S. forests. Part I: spatial patterns of concentrations in biomass, litter, and soils, Environ. Sci. Technol., 45, 3974–3981, 2011.

Obrist, D., Pokharel, A. K., and Moore, C.: Vertical profile measurements of soil air suggest immobilization of gaseous elemental mercury in mineral soil, Environ. Sci. Technol., 48 (4), 2242−2252. 2014.

Obrist, D.: Atmospheric mercury pollution due to losses of terrestrial carbon pools? Biogeochemistry 85, 119–123, 2007.

Obrist, D.: Mercury distribution across 14 U.S. Forests. Part II: Patterns of methyl mercury concentrations and areal mass of total and methyl mercury, Environ. Sci. Technol., 46, 5921–5930, 2012.

Pannu, R., Siciliano, S. D., and O'Driscoll, N. J.: Quantifying the effects of soil temperature, moisture and sterilization on elemental mercury formation in boreal soils, Environ. Pollut., 193, 138–146, 2014.

Pirrone, N., Cinnirella, S., Feng, X., Finkelman, R. B., Friedli, H. R., Leaner, J., Mason, R., Mukherjee, A. B., Stracher, G. B., Streets, D. G., and Telmer, K.: Global mercury emissions to the atmosphere from anthropogenic and natural sources, Atmos. Chem. Phys., 10 (13), 5951−5964, 2010.

Poissant, L., and Casimir, A.: Water-air and soil-air exchange rate of total gaseous mercury measured at background sites, Atmos. Environ., 32 (5), 883−893, 1998.

Poissant, L., Pilote, M., Constant, P., Beauvais, C., Zhang, H. H., and Xu, X.: Mercury gas exchanges over selected bare soil and flooded sites in the bay St. François wetlands (Québec, Canada), Atmos. Environ., 38(25), 4205-4214, 2004.

Poulain, A. J., Lalonde, J. D., Amyot, M., Shead, J. A., Raofie, F., and Ariya, P. A.: Redox transformations of mercury in an Arctic snowpack at springtime, Atmos. Environ., 38, 6763–6774, 2004.

Poulain, A. J., Roy, V., and Amyot, M.: Influence of temperate mixed and deciduous tree covers on Hg concentrations and photoredox transformations in snow, Geochim. Cosmochim. Acta, 71 (10), 2448−2462, 2007.

Rea, A. W., Keeler, G. J., and Scherbatskoy, T.: The deposition of mercury in throughfall and litterfall in the Lake Champlain watershed: A short-term study, Atmos. Environ., 30 (19), 3257−3263, 1996.

Rea, A. W., Lindberg, S. E., and Keeler, G. J.: Assessment of dry deposition and foliar leaching of mercury and selected trace elements based on washed foliar and surrogate surfaces, Environ. Sci. Technol., 34, 2418−2425, 2000.

Rea, A. W., Lindberg, S. E., and Keeler, G. J.: Dry deposition and foliar leaching of mercury and selected trace elements in deciduous forest throughfall, Atmos. Environ., 35, 3453–3462, doi:10.1016/S1352-2310(01)00133-9, 2001.

Richardson, J. B. and Friedland, A. J.: Mercury in coniferous and deciduous upland forests in northern New England, USA: implications of climate change, Biogeosciences, 12, 6737–6749, doi:10.5194/bg-12-6737-2015, 2015.

Richardson, J. B., Friedland, A. J., Engerbretson, T. R., Kaste, J. M., and Jackson, B. P.: Spatial and vertical distribution of mercury in upland forest soils across the northeastern United States, Environ. Pollut., 182 (6), 127−134, 2013.

Rimmer, C. C., Miller, E. K., Mcfarland, K. P., Faccio, S. D., Strong, A. B., Taylor, R. J., and Faccio, S. D.: Mercury bioaccumulation in a terrestrial food web of a montane forest, Ecotoxicology 19, 697–709. http://dx.doi.org/10.1007/s10646-009-0443-x, 2010.

Risch, M. R., DeWild, J. F., Krabbenoft, D. P., Kolka, R. K., and Zhang, L.: Litterfall mercury dry deposition in the eastern USA, Environ. Pollut., 161, 284–290, 2012.

Schimel, D. S., Braswell, B. H., Holland, E. A., McKeown, R., Ojima, D. S., Painter, T. H., Parton, W. J., and Townsend, A. R.: Climatic, edaphic and biotic controls over storage and turnover of carbon in soils, Global Biogeochem. Cy., 8, 279–293, 1994.

Schroeder, W. H., Munthe, J., and Lindqvist, O.: Cycling of mercury between water, air, and soil compartments of the environment, Water, Air, Soil Pollut., 48 (3−4), 337−347, 1989.

Schwesig, D. and Matzner, E.: Dynamics of mercury and methylmercury in forest floor and runoff of a forested watershed in Central Europe, Biogeochemistry, 53, 181–200, 2001.

Schwesig, D. and Matzner, E.: Pools and fluxes of mercury and methylmercury in two forested catchments in Germany, Sci. Total Environ., 260, 213–223, 2000.

Seigneur, C., Vijayaraghavan, K., Lohman, K., Karamchandani, P., and Scott, C.: Global source attribution for mercury deposition in the United States, Environ. Sci. Technol., 38, 555–569, 2004.

Selvendiran, P., Driscoll, C. T., Montesdeoca, M. R., and Bushey, J. T.: Inputs, storage, and transport of total and methyl mercury in two temperate forest wetlands, J. Geophys. Res., 113, G00C01. http://dx.doi.org/10.1029/2008JG000739, 2008.

Sheehan, K. D., Fernandez, I. J., Kahl, J. S., and Amirbahman, A.: Litterfall mercury in two forested watersheds at Acadia National Park, Maine, USA, Water Air Soil Poll., 170, 249–265, 2006.

St. Louis, V. L., Rudd, J. W. M., Kelly, C. A., Hall, B. D., Rolfhus, K. R., Scott, K. J., Lindberg, S. E., and Dong, W.: Importance of the forest canopy to fluxes of methylmercury and total mercury to boreal ecosystems, Environ. Sci. Technol., 35, 3089–3098, 2001.

Wan, Q., Feng, X. B., Lu, J. L., Zheng, W., Song, X. J., Han, S. J., and Xu, H.: Atmospheric mercury in Changbai Mountain area, northeastern China I. The seasonal distribution pattern of total gaseous mercury and its potential sources, Environ. Res., 109, 201–206, DOI 10.1016/j.envres.2008.12.001, 2009b.

Wan, Q., Feng, X. B., Lu, J., Zheng, W., Song, X. J., Li, P., Han, S. J., and Xu, H.: Atmospheric mercury in Changbai Mountain area, northeastern China II. The distribution of reactive gaseous mercury and particulate mercury and mercury deposition fluxes, Environ. Res., 109, 721–727, doi:10.1016/j.envres.2009.05.006, 2009a.

Wang, D. Y., He, L., Shi, X. J., Wei, S. Q., and Feng, X. B.: Release flux of mercury from different environmental surfaces in Chongqing, China, Chemosphere, 64, 1845–1854, doi:10.1016/j.chemosphere.2006.01.054, 2006.

Wang, L., Wang, S. X., Zhang, L., Wang, Y. X., Zhang, Y. X., Nielsen, C., McElroy, M. B., and Hao, J. M.: Source apportionment of atmospheric mercury pollution in China using the GEOS-Chem model, Environ. Pollut., 190, 166–175, doi:10.1016/j.envpol.2014.03.011, 2014.

Wang, S., Xing, D., Wei, Z., and Jia, Y.: Spatial and seasonal variations in soil and river water mercury in a boreal forest, Changbai Mountain, Northeastern China, Geoderma, 206(206), 123−132, 2013.

Wang, X., Bao, Z., Lin, C. J., Yuan, W., and Feng, X.: Assessment of global mercury deposition through litterfall. Environ. Sci. Technol., 50(16), 8548-8557, 2016a.

Wang, X., Yuan, W., and Feng, X.: Global review of mercury biogeochemical processes in forest ecosystems. Prog. Chem., 29(9): 970-980, doi:10.7536/PC170343, 2017.

Wang, X., Lin, C., Lu, Z., Zhang, H., Zhang, Y., and Feng, X.: Enhanced accumulation and storage of mercury on subtropical evergreen forest floor: implications on mercury budget in global forest ecosystems, J. Geophys. Res. Biogeo., 121, 2016b.

Wang, Y. M., Peng, Y. L., Wang, D. Y., and Zhang, C.: Wet deposition fluxes of total mercury and methylmercury in core urban areas, Chongqing, China, Atmos. Environ., 92, 87–96, doi:10.1016/j.atmosenv.2014.03.059, 2014.

Wang, Y.: Study on eco-hydrological process to Land use/forest cover change of small typical watersheds in Beijing mountain area, Doctor's dissertation, Beijing Forestry University, 2012 (in Chinese with English abstract).

Wang, Z. W., Zhang, X. S., Xiao, J. S., Zhijia, C., and Yu, P. Z.: Mercury fluxes and pools in three subtropical forested catchments, southwest China, Environ. Pollut., 157, 801–808, doi:10.1016/j.envpol.2008.11.018, 2009.

Wright, L. P., Zhang, L., and Marsik, F. J.: Overview of mercury dry deposition, litterfall, and throughfall studies, Atmos. Chem. Phys., 16(21), 1–46, 2016.

Xiao, Z. F., Munthe, J., W. H. S., and Lindqvist, O.: Vertical fluxes of volatile mercury over forest soil and lake surfaces in Sweden, Tellus B, 43(3), 267–279, 1991.

Xiao, Z., Sommar, J., Lindqvist, O., and Giouleka, E.: Atmospheric mercury deposition to grass in southern Sweden, Sci. Total Environ., 213(213), 85-94, 1998.

Xin, M. and Gustin, M. S.: Gaseous elemental mercury exchange with low mercury containing soils: Investigation of controlling factors, Appl. Geochem., 22, 1451–1466, 2007.

Xue, T., Wang, R. Q., Zhang, M. M., and Dai, J. L.: Adsorption and desorption of mercury (II) in three forest soils in Shandong province, China, Pedosphere, 23(2), 265–272, 2013.

Yang, Y. K., Chen, H., and Wang, D. Y.: Spatial and temporal distribution of gaseous elemental mercury in Chongqing, China. Environ. Monit. Assess. 156, 479-489, 2009.

Yin, Y., And, H. E. A., Huang, C. P., Sparks, D. L., and Sanders, P. F.: Kinetics of mercury (ii) adsorption and desorption on soil, Environ. Sci. Technol., 31(2), 496–503, 1997.

Zhang, H. and Lindberg, S. E.: Sunlight and iron (III)-induced photochemical production of dissolved gaseous mercury in freshwater, Environ. Sci. Technol., 35, 928–935, 2001.

Zhang, H., Fu, X., Lin, C. J., Shang, L., Zhang, Y., Feng, X., and Lin, C.: Monsoon-facilitated characteristics and transport of atmospheric mercury at a high-altitude background site in southwestern China, Atmos. Chem. Phys., 16(20), 1-36, 2016.

Zhang, H., Lindberg, S. E., Barnett, M. O., Vette, A. F., and Gustin, M. S.: Dynamic flux chamber measurement of gaseous mercury emission fluxes over soils. Part 1: simulation of gaseous mercury emissions from soils using a two-resistance exchange interface model, Atmos. Environ., 36, 835–846, 2002.

Zhang, H., Lindberg, S. E., Marsik, F. J., and Keeler, G. J.: Mercury air/surface exchange kinetics of background soils of the Tahquamenon River watershed in the Michigan Upper Peninsula, Water, Air, Soil Pollut., 126 (1−2), 151−169, 2001.

Zhou, J., Feng, X., Liu, H., Zhang, H., Fu, X., Bao, Z., Wang, X., and Zhang, Y.: Examination of total mercury inputs by precipitation and litterfall in a remote upland forest of southwestern China, Atmos. Environ., 81, 364–372, doi:10.1016/j.atmosenv.2013.09.010, 2013a.

Zhou, J., Lang, X., Du, B., Zhang, H., Liu, H., Zhang, Y., and Shang L.: Litterfall and nutrient return in moist evergreen broad-leaved primary forest and mixed subtropical secondary deciduous broad-leaved forest in China, Eur. J. Forest Res., 135(1), 77–86, 2016b.

Zhou, J., Liang, J., Hu, Y., Zhang, W., Liu, H., You, L., Zhang, W., Gao, M., Zhou, J.: Exposure risk of local residents to copper near the largest flash copper smelter in china. Sci. Total Environ., 630, 453-461, 2018.

Zhou, J., Liu, H., Du, B., Shang, L., Yang, J., and Wang, Y.: Influence of soil mercury concentration and fraction on bioaccumulation process of inorganic mercury and methylmercury in rice (Oryza sativa L.), Environ. Sci. Pollut. Res., 22, 6144–6154, doi:10.1007/s11356-014-3823-6, 2015b.

Zhou, J., Wang, Z., Sun, T., Zhang, H., and Zhang, X.: Mercury in terrestrial forested systems with highly elevated mercury deposition in southwestern China: The risk to insects and potential release from wildfires, Environ. Pollut., 212, 188–196, doi:10.1016/j.envpol.2016.01.003, 2016a.

Zhou, J., Wang, Z., Zhang, X., and Chen, J.: Distribution and elevated soil pools of mercury in an acidic subtropical forest of southwestern China, Environ. Pollut., 202, 187–195, doi:10.1016/j.envpol.2015.03.021, 2015a.

Zhou, J., Wang, Z., Zhang, X., and Gao, Y.: Mercury concentrations and pools in four adjacent coniferous and deciduous upland forests in Beijing, China, J. Geophys. Res.–Biogeo., 2017a.

Zhou, J., Wang, Z., Zhang, X., and Sun, T.: Investigation of factors affecting mercury emission from subtropical forest soil: a field controlled study in southwestern China, J. Geochem. Explor., 176, 128–135, 2017b.

Zhou, J.: Atmospheric mercury deposition disciplines and its influencing factors in background area of Mt. Ailao in Yunnan, Master's dissertation, Guizhou University, 2013b (in Chinese with English abstract).

975 Zhou, J.: Soil–atmosphere mercury fluxes and mercury pools in typical forest of China, Doctor's dissertation, University of Chinese
976 Academy of Sciences, 2016c (in Chinese with English abstract).
977 Zhu, W., Lin, C. J., Wang, X., Sommar, J., Fu, X., and Feng, X.: Global observations and modeling of atmosphere-surface exchange of
978 elemental mercury: a critical review, Atmos. Chem. Phys., 16(7), 4451–4480, 2016.
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980 **Table 1.** Hg concentrations (ng L^{-1} or ng g^{-1}) and deposition fluxes (μg m^{-2} yr^{-1}) in precipitation, throughfall, and
981 litterfall in China.

Site	Forest type	Altitude (m a.s.l)	Location Type	Study period	Samples	Concentration		Deposition flux		References
						THg	MeHg	THg	MeHg	
Mt. Ailao, Yunnan	Subtropical evergreen broadleaf mixed	2500	Remote	06/2011–05/2012	Precipitation	3.0	0.08	5.4	0.14	Zhou et al., 2013a, b
					Litterfall	54.0	0.28	71.2	0.36	
Mt. Ailao, Yunnan	Subtropical evergreen broadleaf mixed	2500	Remote	2011–2014	Precipitation	4.9		4.9		Wang et al., 2016
					Litterfall	43–66		75.0		
					Throughfall	22.9		20–30		
Mt. Leigong, Guizhou	Subtropical deciduous broadleaf mixed	2178	Remote	05/2008–05/2009	Precipitation	4.0	0.04	6.1	0.06	Fu et al., 2010a
					Throughfall	8.9	0.1	10.5	0.12	
					Litterfall	91.0	0.48	39.5	0.28	
Mt. Leigong, Guizhou	Subtropical deciduous broadleaf mixed	1680	Remote	03/2005–02/2006	Precipitation	12.9		16.8		Wang et al., 2009
					Throughfall	36.7		41.2		
					Litterfall	135.1		78.3		
Mt. Damei, Zhejiang	Subtropical deciduous broadleaf mixed	550	Remote	08/2012–08/2014	Precipitation	3.7		6.0		Fu et al., 2016
				08/2012–07/2013	Litterfall	42.3		23.1		
Mt. Gongga, Sichuan	Subtropical evergreen broadleaf	1640	Remote	01–12/2006	Precipitation*	9.9		9.1		Fu et al., 2008b
Mt. Gongga, Sichuan	Subtropical evergreen broadleaf	3000	Remote	05/2005–04/2006	Precipitation*	14.2	0.16	26.1	0.30	Fu et al., 2010b
					Throughfall	40.2	0.3	57.1	0.43	
					Litterfall	35.7		35.5		
Mt. Changbai, Jilin	Temperate broadleaf and pine mixed	750	Remote	08/2005–07/2006	Precipitation*	13.4		8.4		Wan et al., 2009a
					Throughfall	9.0		24.9		
Mt. Changbai, Jilin	Temperate broadleaf and pine mixed	736	Remote	08/2011–08/2014	Precipitation	7.4		5.6		Fu et al., 2016
					Litterfall	47		22.8		
Mt. Dongling, Beijing	Temperate Chinese pine evergreen	1100	Remote	09–11/2015	Litterfall	39.8		15.8		Zhou et al., 2017a
	Temperate larch deciduous				Litterfall	63.3		19.6		
	Temperate oak deciduous				Litterfall	46.5		14.1		
	Temperate mixed deciduous				Litterfall	45.3		12.9		
Linzhi, Tibetan	Subtropical evergreen coniferous	3200	Remote	8/2008	Litterfall	12.6		4.2		Gong et al., 2014
China (22 sites)	Suburban evergreen broadleaf		Suburban		Litterfall	50.8		17.9		Niu et al., 2011
	Suburban deciduous broadleaf				Litterfall	25.8		8.73		
Tieshanping, Chongqing	Subtropical evergreen coniferous	500	Suburban	03/2005–03/2006	Precipitation	32.3		29.0		Wang et al., 2009
					Throughfall	69.7		71.3		
					Litterfall	105		220		
Tieshanping, Chongqing	Subtropical evergreen coniferous	500	Suburban	2010–2011	Throughfall	69		67.5		Luo et al., 2015a
					Litterfall	115		22.3		
Tieshanping,	Subtropical evergreen coniferous	500	Suburban	04/2014–	Litterfall	85	0.21	40.51	0.10	Zhou et al.,

Chongqing	Subtropical evergreen broadleaf			03/2015	Litterfall	89	0.23	90.85	0.34	2016c
Mt. Jinyun, Chongqing	Subtropical evergreen broadleaf	900	Rural	03/2012–02/2013	Precipitation	11.9	0.20	15.9	0.26	Ma et al., 2015
					Throughfall	20.1	0.55	21.8	0.60	
					Litterfall	104.5	0.84	43.5	0.27	
Mt. Simian, Chongqing	Subtropical evergreen broad-leaf	1394	Rural	03/2012–02/ 2013	Precipitation	10.9	0.24	15.45	0.36	Ma et al., 2016
					Throughfall	24.04	0.33	32.17	0.45	
					Litterfall	106.7	0.79	42.89	0.32	
Qianyanzhou, Jiangxi	Subtropical evergreen coniferous	60	Rural	11/2013–12/2014	Precipitation	23		14.4		Luo et al., 2015a
					Throughfall	42		34.6		
					Litterfall	42.9		21.4		
Huitong, Hunan	Subtropical evergreen coniferous	335	Rural	4/2013–12/2014	Precipitation	12.5		15.9		Luo et al., 2015a
					Throughfall	29.9		27.8		
					Litterfall	176.1		33.6		
Luchonguan, Guizhou	subtropical broad-leaf and coniferous	1360	Urban	01/2005–01/2006	Throughfall	43.6		49.0		Wang et al., 2009

984 **Table 2.** Hg concentrations (ng L^{-1} or ng g^{-1}) and export fluxes (μg m^{-2} yr^{-1}) in stream water/runoff in China.

Site	Forest type	Altitude (m a.s.l)	Location Type	Study period	THg concentration	THg export flux	References
Northeast China	Temperate evergreen/deciduous coniferous and broadleaf	442 ± 324	Remote and rural		17.2±11.0		Luo et al. 2014
South China	Subtropical evergreen conifers/mixed broad-leaved	548 ± 295	Remote and rural		6.2 ±6.4		Luo et al. 2014
Mt. Leigong, Guizhou	Subtropical deciduous broadleaf mixed forest	1680	Remote	03/2005–02/2006	4.3±2.5	3.0	Wang et al., 2009
Mt. Changbai, Jilin	Temperate broadleaf and pine mixed	750	Remote	04/2009, 09/2009	5.5 ± 4.1		Wang et al., 2013
Tieshanping, Chongqing	Subtropical evergreen coniferous	500	Suburban	03/2005–03/2006	6.2 ±3.5	3.5	Wang et al., 2009
Tieshanping, Chongqing	Subtropical evergreen coniferous	500	Suburban	04/2014	3.1 ± 1.2		Zhou et al., 2015a
Luchongguan, Guizhou	Subtropical broad leave-coniferous mixed subtropical	1360	Urban	01/2005–01/2006	8.9± 6.7	4.5	Wang et al., 2009
Mt. Gongga, Sichuan	Subtropical evergreen broadleaf	3000	Remote	05/2005–04/2006	3.5±0.9	8.6	Fu et al., 2010a
Mt. Simian, Chongqing	Subtropical evergreen broad-leaf	1394	Rural	03/2012–02/ 2013	3.9 ± 2.0	7.23	Ma et al., 2016
Huitong, Hunan	Subtropical evergreen coniferous	335	Rural	4/2013–12/2014	4.9	2.03	Luo et al., 2015a
Qianyanzhou, Jiangxi	Subtropical evergreen coniferous	60	Rural	11/2013–12/2014	2.3		Luo et al., 2015a

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Table 3. Soil-atmosphere Hg exchange fluxes (ng m^{-2} hr^{-1}), soil Hg concentrations and surface TGM concentrations (ng m^{-3}) in atmosphere in forested areas of China and other regions.

Locations	Forest Type	Altitude	Location Type	Study period	Soil Hg	Surface TGM	Flux	References
Mt. Dongling, Beijing (Temperate)	Chinese Pine	1050	Remote	07/2015–05/2016	88	2.2±1	0.01±2.6	Zhou et al. 2016c
	Larch	1020	Remote	07/2015–05/2016	69	2.3±1	0.12±1.28	Zhou et al. 2016c
	Mixed broadleaf forest	1250	Remote	07/2015–05/2016	54	2.4±1	0.46±1.36	Zhou et al. 2016c
	Wetland	1150	Remote	07/2015–05/2016	154	2.5±1.1	3. 6±6.8	Zhou et al. 2016c
Mount Gongga, Sichuan (Subtropical)	Shrub	2350	Remote	21– 22/08/2006	90	3.6±1.3	6.6±4.2	Fu et al., 2008
	Broadleaf Forest	1220	Remote	27– 29/08/2006	60	3.7±0.5	5.7±4.7	Fu et al., 2008
	Broadleaf Forest	1650	Remote	17–18/08/ 2006	110	2.3±0.4	9.3±4.3	Fu et al., 2008
	Broadleaf Forest	2140	Remote	19– 21/08/ 2006	180	2.3±0.3	7.7±3.4	Fu et al., 2008
	Broadleaf Forest	2500	Remote	30–31/08/2007	160	2.0±0.6	0.5±1.8	Fu et al., 2008
	Pine forest	3050	Remote	31/08–1/09/2008	80	1.6±0.6	2.9±2	Fu et al., 2008
Mount Gongga, Sichuan (Subtropical)	Evergreen broadleaf	3000	Remote	17/8/2006–1/9/2013		4.7	1.6	Fu et al., 2010a
Mt. Simian, Chongqing (Subtropical)	Evergreen broadleaf	1394	Rural	19/8/2003	174	19.9±8.6	7.7±3.9	Wang et al., 2006
Mt. Jinyun, Chongqing (Subtropical)	Evergreen broadleaf	900	Rural	9/15/2003	137	9.9±1.8	3.4±1.5	Wang et al., 2006
Mt. Gele, Chongqing (Subtropical)	Evergreen broadleaf	600	Rural	6/1/2003	196	14.1±3	8.4±2.5	Wang et al., 2006
Mt. Jinyun, Chongqing (Subtropical)	Mixed broadleaf-conifer	900	Rural	4/2012–1/2013			14.2±10.9	Ma et al., 2015
	Shrub	900	Rural	5/2012–1/2013			16.9±13.3	Ma et al., 2015
	Bamboo	900	Rural	4/2012–2/2013			17.8±14.2	Ma et al., 2015
	Deciduous broadleaf	900	Rural	4/2012–2/2013			12.2±10.7	Ma et al., 2015
Mt. Jinyun, Chongqing (Subtropical)	Mixed broadleaf-conifer	900	Rural	4/2011–3/2012	140		14.2±6.7	Ma et al., 2014
Mt. Simian, Chongqing (Subtropical)	Deciduous broadleaf	1394	Rural	3/2012–2/2013	161		12.12±10.7	Ma et al., 2016
Qianyanzhou, Jiangxi (Subtropical)	Evergreen coniferous	60	Rural	11/2013–12/2014	101	3.6	2.1	Luo et al., 2015a
Tieshanping, Chongqing (Subtropical)	Masson pine	500	Suburban	03/2014–01/2015	219	3.6±1.3	2.76±3.85	Zhou et al. 2016c
	Masson pine	500	Suburban	03/2014–01/2015	264	3.8±1.3	3.52±4.18	Zhou et al. 2016c
	Camphor	500	Suburban	03/2014–01/2015	156	3.3±1.4	0.18±2.24	Zhou et al. 2016c
	Wetland	500	Suburban	03/2014–01/2015	96	4.9±2	–0.8±5.05	Zhou et al. 2016c
Tieshanping, Chongqing (Subtropical)	Masson pine	500	Suburban	09/2012–07/2013	294	5.2±2	0.3±0.8	Du et al., 2014
Nanhu, Changchun (Temperate)	Temperate		Urban		143		7.6	Fang et al., 2003
Jingyuetan, Changchun (Temperate)	Temperate		Urban		136		3.3	Fang et al., 2003

Zhuzhou, (Subtropical)	Hunan	Mixed conifer	broadleaf-	Contaminated	09/2012–03/2014	3190	13.8	15.3±2.8	Du et al., 2014

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Figure captions:

Fig. 1. Contributions to the Hg input fluxes (μg m^{-2} yr^{-1}) to forests from precipitation, throughfall, litterfall and total inputs (throughfall + litterfall) in China.

Fig. 2. Relationship analysis between the GEM or TGM concentrations verses the litterfall Hg concentrations for field trap measurements.

Fig. 3. Correlations between litterfall deposition fluxes of Hg and (a) mass-weighted mean (MWM) Hg concentrations in litterfall, (b) litterfall biomass.

Fig. 4. Box chart for Hg inputs to forest ecosystems in China, Europe and North America.

Fig. 5. Box chart for soil-atmosphere Hg exchange fluxes in deciduous and evergreen forest ecosystems in China (CHI, including four seasons), North America (NA), Europe (Eur) and Brazil (Bra).

Fig. 6. Total mercury budgets (μg m^{-2} yr^{-1}) at the three temperate forest stands of Mt. Dongling (a) and four subtropical forests of Tieshanping, Qianyanzhou, Mt. Gongga and Mt. Simian forests.

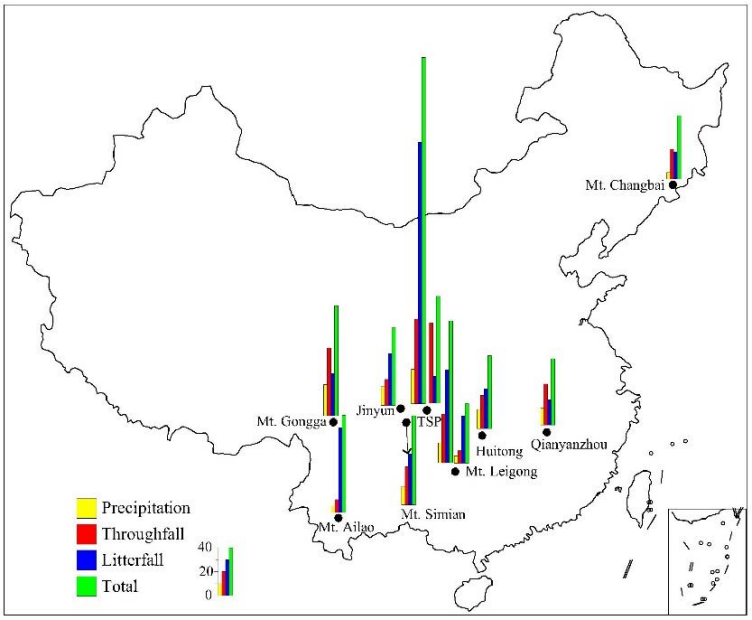

Fig. 1. Contributions to the Hg input fluxes (μg m^{-2} yr^{-1}) to forests from precipitation, throughfall, litterfall and total inputs (throughfall + litterfall) in China. Mt. Ailao, Mt. Leigong, Mt. Gongga and Mt. Changbai are regarded as remote sites and Mt. Jinyun, Mt. Simian, Qianyanzhou, Huitong and Tieshanping (TSP) are regarded as suburban and rural sites.

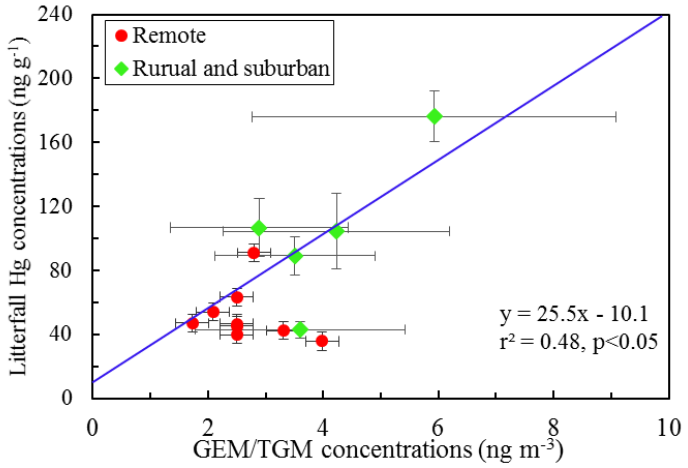

Fig. 2. Relationship analysis between the GEM or TGM concentrations verses the litterfall Hg concentrations for field trap measurements. Data were from Mt. Ailao (Zhou et al., 2013a; Zhang et al., 2015), Mt. Leigong (Fu et al., 2010a, b), Mt. Damei (Lang et al., 2015; Yu et al., 2015), Mt. Gongga (Fu et al., 2008a, b), Mt. Changbai (Fu et al., 2016, 2014), Mt. Dongling (Zhou et al., 2017a), Mt. Jinyun (Ma et al., 2015), Mt. Simian (Ma et al., 2016), Qianyanzhou (Luo et al., 2015), Huitong (Luo et al., 2015) and Tieshanping (Zhou et al., 2016a).

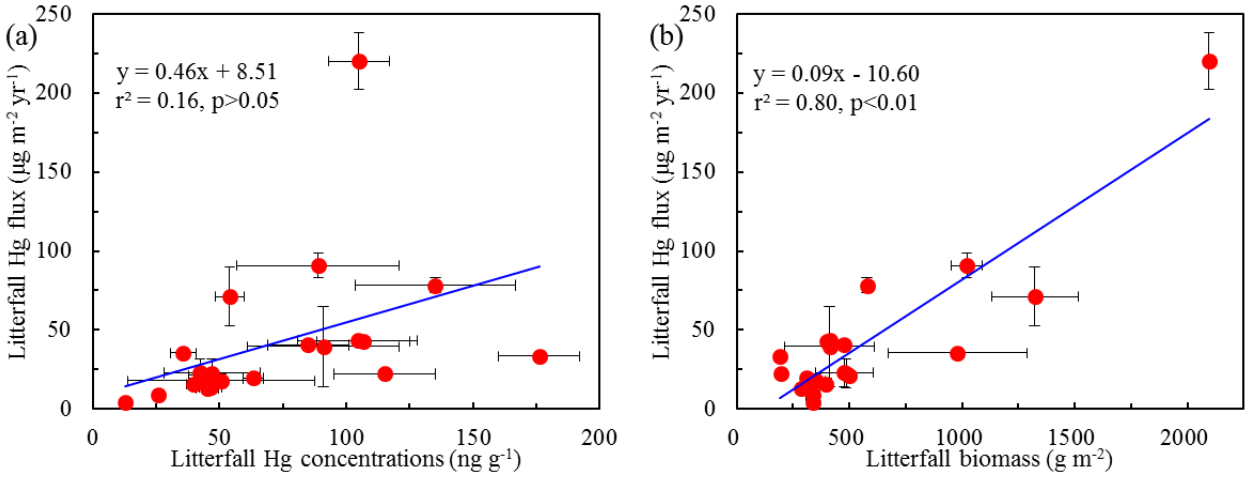

Fig. 3. Correlations between litterfall deposition fluxes of Hg and (a) mass-weighted mean (MWM) Hg concentrations in litterfall, (b) litterfall biomass. Data are from Zhou et al., 2013a, 2016a, 2017a; Fu et al., 2010a, b, 2016; Luo et al., 2015a, b; Wang et al., 2009; Gong et al., 2014; Niu et al., 2011; Ma et al., 2015, 2016; Luo et al., 2015a.

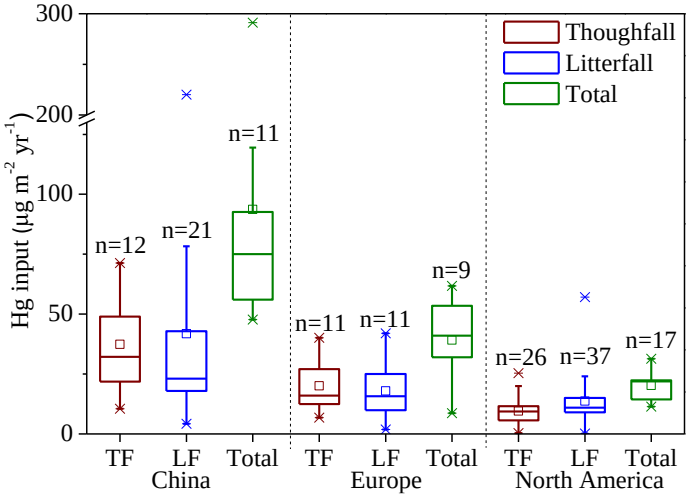

Fig. 4. Box chart for Hg inputs to forest ecosystems in China, Europe and North America. "TF" is the throughfall; "LF" is the litterfall; "Total" is the total Hg input (throughfall + litterfall) to the forest ecosystem. Data are from Hultberg et al., 1995; Iverfeldt et al.,1991; Larssen et al., 2008; Lee et al., 2000; Munthe et al., 1995, 1998; Schwesig and Matzner, 2000, 2001; Xiao et al., 1998; Blackwell and Driscoll, 2015a, b; Bushey et al., 2008; Choi et al., 2008; Demers et al., 2007; Kalicin et al., 2008; Kolka, 1999; Grigal et al., 2000; Lindberg et al., 1994, 1996; Fisher and Wolfe, 2012; Friedli et al., 2007; Rea et al., 1996, 2000, 2001; Johnson, 2002, Johnson, et al., 2007; Nelson et al., 2007; St. Louis et al., 2001; Graydon et al., 2008; Juillerat et al., 2012; Obrist et al., 2012; Richardson and Friedland, 2015; Risch et al., 2012; Sheehan et al., 2006; Selvendiran et al., 2008; Zhou et al., 2013a, 2016c, 2017a; Zhang et al., 2015; Lang et al., 2015; Yu et al., 2015; Fu et al., 2008a, b, 2010a, b, 2016, 2014; Ma et al., 2015, 2016; Luo et al., 2015.

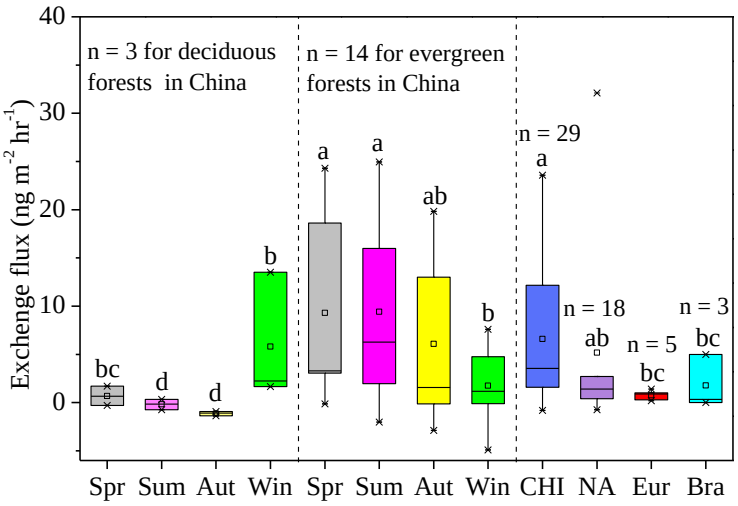

Fig. 5. Box chart for soil-atmosphere Hg exchange fluxes in deciduous and evergreen forest ecosystems in China (CHI, including four seasons), North America (NA), Europe (Eur) and Brazil (Bra). "Spr" is spring; "Sum" is summer; "Aut" is autumn; "Win" is winter. The post hoc tests (Tukey's HSD) were performed at 5% significance level. Data for deciduous forest in China are from Zhou et al. 2016c; for evergreen forests are from Du et al., 2014; Fu et al., 2008c, 2010a; Wang et al., 2006; Ma et al., 2014; Ma et al., 2016; Luo et al. (2015a); Fang et al., 2003; Zhou et al. 2016c; for North America are from Ericksen et al., 2006, Hartman et al., 2009, Carpi and Lindberg, 1998; Kuiken et al., 2008a, b; Lee et al. 2000; Lindberg et al., 1998, 2002; Poissant et al., 2004; Poissant and Casimir, 1998; Carpi et al., 2014; Choi and Holsen, 2009; Zhang et al., 2001; Schroeder et al., 1989; for Europe are from Xiao et al., 1991; Kyllönen et al., 2012; Lindberg et al., 1998; from Brazil are from Almeida et al., 2009; Carpi et al., 2014; Magarelli and Fostier, 2005.

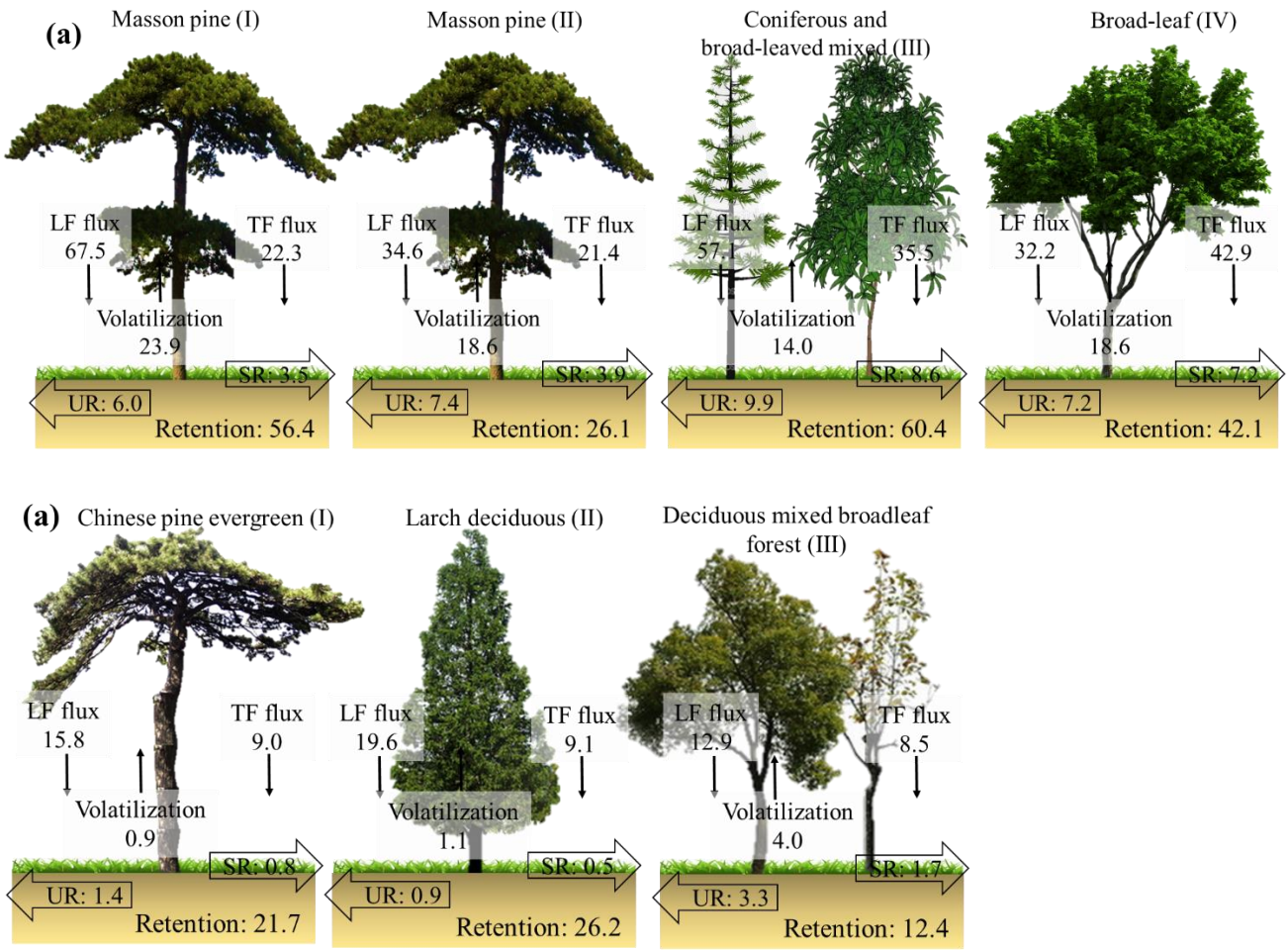

Fig. 6. Total mercury budgets (μg m^{-2} yr^{-1}) at the four subtropical forests of Tieshanping (I), Qianyanzhou (II), Mt. Gongga (III) and Mt. Simian forests (IV) (a) and three temperate forest stands of Mt. Dongling (I-III) (b). LF, TF, SR and UR represent litterfall, throughfall, surface runoff and underground runoff fluxes, respectively. Data are from Zhou et al. (2016a, c), Luo et al. (2015b), Wang et al. (2009), Luo et al. (2015a), Fu et al. (2010a), Ma et al., 2016.