Interactive comment on "Depletion of atmospheric gaseous elemental mercury by plant uptake at Mt. Changbai, Northeast China" by Xuewu Fu et al.

Responses to Anonymous Referee #1

RC- Reviewer's Comments; **AC** – Authors' Response Comments

RC: Summary: The study by Fu et el. reports on depletion events of measurements of gaseous elemental mercury (GEM), particulate bound mercury (PBM) and gaseous oxidized mercury (GOM), in a temperate mixed forest at Mt Changbai. Northeast China. Mercury depletion events are a very interesting as well as complicated phenomena and this study help to share light on the mechanism associated with these rapid depletion events occurring in forest in the absence of GOM enrichments. I recommend that this manuscript be published in ACP GMOS Special Issue after the authors address these minor comments.

AC: we would like to acknowledge the anonymous reviewer for dictating the time to read our original manuscript and provide valuable suggestions. These suggestions are very helpful and constructive. We have made careful revision in the revised manuscript based on the reviewer's recommendations. The corrections are shown in blue fonts in the revised manuscript.

Specific comments:

RC: P3, L35; There are three operationally defined Hg Forms. The word operationally is so unnecessary in this context please remove.

AC: we deleted the word of 'operationally' in the revised manuscript.

RC: P3, L37; instead of starting the sentence with because rather use Due to its mild reactivity.

AC: 'Because' was changed to 'Due to' in line 37 on page 3 in the revised manuscript.

RC: P3, L39 the reference Gustin and Jaffe, 2010; Holmes et al. A space is needed between the text. This is a problem throughout the whole manuscript. The authors should have a close look of all the references in text and apply correct format. There is either no space between different references or there is no space after a semi colon or comma.

AC: we carefully read the manuscript and added a space between the citations and wording in the revised manuscript.

RC: P3, L42 GEM residence time in atmosphere is 0.5 – 2 years. Please check this statement as most literature state the GEM residence time as 0.5 – 1 year.

AC: we agree with this comment and change the residence of atmospheric GEM to 0.5-1.0 year in line 42 on page 3 in the revised manuscript.

RC: P3, L44 see comments made at L39

AC: a space between the citations and wording in the revised manuscript.

RC: P3, L57; change small to slow you are referring to cm s-1 which is speed

AC: the wording has been corrected in line 58 on page 3 in the revised manuscript.

RC: P4, L63 be consistent when writing chemical names or formulas "CO2, Ozone, sulfur dioxide.." Choose one format and keep with this throughout.

AC: the names of chemical compounds were uniformed in the revised manuscript.

RC: P4, L84; leaf growing season. When is this and how long was the leaf growing season, 1 month, 7 months. Please be specific with this time period.

AC: we defined the period of leaf-growing season in line 86 on page 4 in the revised manuscript.

RC: P5, L108; above ground level should be (a.g.l) please correct this throughout the manuscript.

AC: the wording of 'agl' has been corrected throughout the revised manuscript.

RC: I would advise the authors to keep the Supplement information to a minimum. Certain aspect mentioned in the paper can be left out. It's very confusing and time wasting that such a big portion of the text is spent on explaining an aspect but yet the graph containing the information is in the S1 section.

AC: we understand the concern of the length of the supplementary information. Most of the information listed in the supplementary information are valuable for the discussion in the main text. We therefore think the information in the supplement should not be removed

It is not clear to us which aspect motioned in the paper can be left out. The aspect regarding the patterns of depletion events, vertical gradient, foliar flux, and isotopic evidence are important in this paper and should not be left out. The key information with word and data was introduced in the main text. These information together with the supplementary figures are expected to make the discussions more clear.

RC: It would also be useful to mention how long (min, hours or days) a DE occurred. Did the authors investigate this. What was the time criteria for a DE. Also, where there any DE outside of May – Sep and if so, how did these DE differ from the May – Sep DE. Where they shorter or longer events. Were there any significant differences in the GEM concentration levels/time if a DE took place outside of the May– Sep window. Was the only criteria for a May – Sep DE that the GEM concentration should be below 0.5 ng/m3. What if the GEM concentration recovered after 10 min was this also classified as a DE. See Brunke et al. and how they classified a DE at Cape Point with time and Hg concentration.

AC: the duration of the depletions were shown in line 219 on page 10 in the revised manuscript. The criteria that define the strong depletions was shown in line 220-221 on page 10 in the revised manuscript.

The question regarding the depletions outside the leaf-growing season is interesting. In the revised manuscript, we discussed the depletions outside the leaf-growing season and compared them with these during leaf-growing season.

The criteria < 0.5 ng m^{-3} was defined for strong GEM depletions. In general, the duration of strong depletions with GEM concentrations < 0.5 ng m^{-3} lasted for more than 0.5 hour.

Responses to Anonymous Referee #1

RC- Reviewer's Comments; **AC** – Authors' Response Comments

RC: This manuscript having the quality to be acceptable for publication.

AC: We would like to thank the anonymous reviewer for their positive comments on our original manuscript. We made several revision in the revised manuscript following the specific recommendations provide by anonymous Referee #1.

Depletion of atmospheric gaseous elemental mercury by plant uptake at Mt. Changbai, Northeast China

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Abstract: There exists observational evidence that GEM can be readily removed from the atmosphere via chemical oxidation followed by deposition in the polar and sub-polar regions, free troposphere, lower stratosphere, and marine boundary layer under specific environmental conditions. Here we report GEM depletions in a temperate mixed forest at Mt. Changbai, Northeast China. The strong depletions occurred predominantly at night during leaf-growing season and in the absence of GOM enrichment (GOM < 3 pg m^{-3}). Vertical gradients of decreasing GEM concentrations from layers above to under forest canopy suggest in situ loss of GEM to forest canopy at Mt. Changbai. Foliar GEM flux measurements showed that the foliage of two predominant tree species is a net sink of GEM at night, with a mean flux of -1.8 ± 0.3 ng m^2 h^{-1} over *Fraxinus mandshurica* (deciduous tree species) and -0.1 ± 0.2 ng m^2 h^{-1} over *Pinus Koraiensis* (evergreen tree species). Daily integrated GEM δ^{202}Hg, Δ^{199}Hg, and Δ^{200}Hg at Mt. Changbai during 8-18 Jul 2013 ranged from -0.34 to 0.91‰, from -0.11 to -0.04‰ and from -0.06 to 0.01‰, respectively. A large positive shift of GEM δ^{202}Hg occurred during the strong GEM depletion events, whereas Δ^{199}Hg and Δ^{200}Hg remained essentially unchanged. The observational findings and box model results show that uptake of GEM by forest canopy plays a predominant role in the GEM depletion at Mt. Changbai forest. Such depletion events of GEM are likely to be a widespread phenomenon, suggesting that the forest ecosystem represents one of the largest sinks (~1930 Mg) of atmospheric Hg at global scale.

1 Introduction

Mercury (Hg) is a persistent toxic air pollutant that is ubiquitously distributed in the atmosphere. There are three major Hg forms in the atmosphere: gaseous elemental mercury (GEM), particulate bound mercury (PBM), and gaseous oxidized mercury (GOM). The sum of GEM and GOM is known as total gaseous mercury (TGM). Due to its mild reactivity, high volatility, low dry deposition velocity and water solubility, GEM is the most abundant form of Hg in the atmosphere (Gustin and Jaffe, 2010; Holmes et al., 2010). The cycling of GEM in the atmosphere is largely depending on either direct dry deposition or chemical oxidation followed by wet and dry deposition. The residence time of GEM in the atmosphere is estimated to be from several months to a year, based on global Hg budget and empirical models (Hedgecock and Pirrone, 2004; Holmes et al., 2006; Holmes et al., 2010; Gustin et al., 2015). Over the past decades, our understanding regarding the sources and sinks of atmospheric Hg has been improved (Strode et al., 2007; Selin et al., 2008; Holmes et al., 2010; Amos et al., 2013). For instance, the discovery of atmospheric mercury depletion events (AMDEs) in polar and sub-polar regions demonstrated that atmospheric GEM can be readily removed from the atmosphere via reactive halogens-induced oxidation, leading to a deposition of up to 300 Mg yr^{-1} to the arctic (Schroeder et al., 1998; Ebinghaus et al., 2002; Lindberg et al., 2002; Steffen et al., 2008). Similar depletion events occurred in the marine boundary layer at middle latitude to a lesser extent (Brunke et al., 2010; Obrist et al., 2011; Timonen et al., 2013). Fast oxidation of GEM by O_3, reactive halogens (e.g., BrO) and OH· in the free troposphere has also been observed (Swartzendruber et al., 2006; Fain et al., 2009; Slemr et al., 2009; Swartzendruber et al., 2009b; Lyman and Jaffe, 2012; Shah et al., 2016). These findings indicate that GEM probably has a much shorter atmospheric residence time under specific environmental conditions. Dry deposition of GEM (V_d) depends on surface characteristics, meteorological variables, biological and chemical conditions of soil and water. V_d over non-vegetated surfaces (e.g., bare soil) and water bodies is typically low (less than 0.03 cm s^{-1}) to counter the emission and re-emission of GEM from these surfaces (Zhang et al., 2009). Therefore soil and water have long been considered a net GEM source (Selin et al., 2007; Holmes et al., 2010). In contrast, strong dry deposition of GEM to vegetated surfaces and wetlands are frequently observed with V_d up to about 2 cm s^{-1} (Zhang et al., 2009), suggesting vegetation is a sink of atmospheric GEM.

Forest represents a dominant terrestrial ecosystem on the Earth and covers an area of ~4×10^7 km^2. It readily removes trace gases such as CO_2, O_3, sulfur dioxide, nitrogen oxides, and aerosols from the atmosphere (Munger et al., 1996; Finkelstein et al., 2000; Zhang et al., 2001; Pan et al., 2011). However, there are ongoing debates regarding whether or not forest is a sink or a source of atmospheric GEM. Previous laboratory studies suggested that foliar exchange of GEM is bi-directional with net deposition occurring at elevated Hg concentration and net emission under typical background concentrations (Hanson et al., 1995; Ericksen and Gustin, 2004; Gustin et al., 2004; Graydon et al., 2006). Lindberg et al. (1998) measured GEM fluxes over a mature deciduous forest using the modified Bowen ratio (MBR) method and suggested that global forest is a net source of GEM with an emission ranging from 850 to 2000 Mg yr^{-1}. Later, the observation of Hg fluxes in a deciduous forest using a relaxed eddy accumulation (REA) method showed seasonal shift in flux with a net deposition of GEM during leaf-growing season (Bash and Miller, 2009). Although the discrepancy in the measured GEM exchanges between forest and atmosphere is partially attributed to the uncertainties of the flux quantification method (Sommar et al., 2013), there is a need to clarify the role of forest ecosystem in the mass budget of atmospheric GEM. A study in Québec, Canada showed that GEM concentrations at a maple forest site are consistently lower than those measured at an adjacent open site (Poissant et al., 2008). Similarly, the lower GEM concentrations observed in leaf-growing season at many forest sites across the Atmospheric Mercury Network (AMNet) in USA (Lan et al., 2012) also suggest forest a net GEM sink. Currently, it is still unclear whether the loss of GEM over forest is caused by direct dry deposition to canopy or chemical conversions of GEM to GOM (Mao et al., 2008).

In this study, we report consistent GEM depletion events during the leaf-growing season (from May to September) in a temperate mixed forest in Northeast China over a time scale of 7 years. Atmospheric Hg speciation, vertical gradient of GEM, foliage/air and soil/air exchange flux of GEM and isotope signatures of GEM samples have also been observed in an intensive campaign to explore the possible mechanisms responsible for the observed GEM depletion.

2 Material and Methods

2.1 Site description

The study site (42º24′0.1″N, 128º 06′25″E, 738 m above sea level) is located in a temperate broadleaf and Korean pine mixed forest on the north slope of Mt. Changbai (Figure S1). The forest is dominated by tree species of *Pinus koraiensis*, *Fraxinus mandshurica*, *Tilia amurensis*, *Acer mono* and *Quercus mongolica*. The height of the forest canopy is 5 - 22 m (mean = 18.3 m) with the heights of mature trees (> 50 y) and young trees (< 20 y) and shrubs ranging from 15 to 22 m and from 5 to 10 m, respectively. Regions to the east and south of the site consist of pristine forest with little anthropogenic influence (Fu et al., 2012). Most of the regional industrial sources are located more than 50 km west of the sampling site (Supplementary Figure S1).

2.2 Atmospheric Hg measurements

From Oct 2008 to Jul 2013 and from Jul 2014 to Dec 2015, TGM concentrations were continuously measured using an automated Hg vapor analyzer (Tekran® 2537, Tekran Inc., Canada). The analyzer has been used extensively for atmospheric TGM measurements worldwide. The analyzer was calibrated automatically every 25 h using the internal Hg^0 permeation source. The permeation rate of the internal source was manually calibrated every 4 - 6 months by using an external Hg vapor source (Tekran® 2505). The sampling inlet was mounted at a height of 24 m above ground level (a.g.l., ~3 m above canopy) by using a 25 m Teflon tube and a 15 m heated Teflon tube. Atmospheric TGM consists of GEM and GOM. Gustin et al. (2013; 2015) proposed that GOM could be transformed to GEM within the uncovered Teflon tubing, which in turn would be transported efficiently through the tubing and quantified by the Tekran analyzer. However, GOM generally constitutes a small portion of TGM (mean of 0.32% on basis of one year of measurements and will not exceed 1% using a three-fold correction factor to adjust GOM concentrations measured by the Tekran® speciated system) (Gustin et al., 2015). Therefore, we interpret the TGM observations as GEM throughout the paper.

GEM, GOM and PBM were measured using the Tekran® 2537/1130/1135 unit (Tekran Inc., Canada) from Jul 2013 to Jul 2014. The sampling inlet was positioned at 4 m a.g.l. in a small clearing plot with tall trees of ~5 m from the system. This system has been widely used and described in detail by many earlier studies (Landis et al., 2002; Lindberg et al., 2002; Lan et al., 2012; Fu et al., 2016b). Briefly, GOM, PBM, and GEM in ambient air were collected onto KCl-coated annular denuder, quartz fiber filter and dual gold cartridges in sequence. This system was programmed to collect GOM and PBM at 1-h intervals at a volumetric flow rate of 10 L min^{-1}. GEM was collected from air samples at 5-min intervals at a volumetric flow rate of 1.0 L min^{-1}. Once collected, Hg is thermally decomposed from each unit and detected by cold vapor atomic fluorescence spectroscopy (CVAFS) as Hg^0. KCl-coated denuder, Teflon coated glass inlet, and impactor plate were replaced bi-weekly and quartz filters were replaced monthly. Denuders and quartz filters were prepared and cleaned before field sampling following the methods in Tekran technical notes. GEM concentrations measured at 4 m a.g.l. in the small clearing plot and at 45 m a.g.l. (~24 m above canopy) did not bias significantly with each other with a mean difference of 0.03 ng m^{-3} (3% of the mean GEM concentration during the study period) (Supplementary Figure S2). The two Tekran instruments used for this comparison were run side by side for 2 days in the laboratory and showed a mean systematic uncertainty of 1.8 ± 1.1% (ranging from 0% to 5.7%). This indicates the measurements at 4 m a.g.l. in the small clearing plot did not significantly underestimate the GEM concentrations of ambient air in the study area. In the study area, GEM also has a fast dry deposition velocity within the forest (more details in sections below), although to a lesser extent compared to atmospheric GOM. We therefore assume that the measurements of GOM in the clearing plot didn't result in significantly biased low GOM concentrations and were representative of ambient air in the study area.

Vertical profile of GEM concentrations at 1 m, 10 m, 24 m, and 45 m a.g.l. within the forest was measured from 10 to 15 Jul 2013 using the Tekran® 2537 analyzer and the Tekran® 1115 Synchronized Multi-Port manifold (Tekran Inc., Canada). The sampling duration of GEM during the vertical gradient measurements was programmed to be 2.5 min, and switching of ports of the manifold was made every 5 min.

The GEM detection limit for 7.5 L samples measured with Tekran® 2537 analyzer as specified by Tekran Instrument Corporation is 0.1 ng m^{-3}. Due to the lack of understanding of the specific forms and calibration standards of GOM, there are uncertainties regarding the GOM measurements (Gustin et al., 2015). Previous studies suggested that GOM measured by the Tekran system could be biased low and a correction factor of 3 should be applied for adjusting GOM concentrations measured by the Tekran system (Gustin et al., 2013; Huang et al., 2013; Gustin et al., 2015; Huang and Gustin, 2015). Tekran® 2537's default integration at low Hg loading (~1 pg per cycle) was reported to have a 25% underestimation of GEM concentration. This could also underestimate GOM concentrations when GOM concentrations were lower than 2 pg m^{-3} (Swartzendruber et al., 2009a). These analytical uncertainties are taken into account for the discussions of GEM depletion mechanism in the Results and Discussion section.

2.3 Foliar GEM exchange

Exchange flux of GEM between leaf and the atmosphere was measured using a new dynamic flux bag method described by Graydon et al. (2006) which is thought to maintain normal physiological function of enclosed foliage. Briefly, a Tedlar® gas sampling bag (~20 L volume, polyvinyl fluoride, DuPont, USA) enclosed living intact leaves, and the foliar GEM flux was obtained via measuring the difference in GEM concentrations at the inlet and outlet of the flux bag. Ambient air was pumped into the flux bag using a Mini Diaphragm vacuum pump (N89 KTDC, KNF, Germany, oil-free, brushless and with diaphragm coated with PTFE). GEM flux was calculated using Equation (1):

$$F = (C_o - C_i) \times Q/A \qquad (1)$$

where *F* is the foliar GEM flux in ng m^{-2} h^{-1}, with positive and negative values representing emission and deposition, respectively, C_o and C_i are the GEM concentrations at the outlet and inlet of the flux bag, respectively, which were measured by the Tekran® 2537 analyzer, *Q* is the flushing flow rate of air through the flux bag (0.5 m^3 h^{-1}), and *A* is the single-sided leaf area enclosed by the flux bag in m^2.

Two tree species, *Fraxinus mandshurica* (deciduous tree species) and *Pinus Koraiensis* (evergreen tree species), were selected for the foliar GEM flux measurement. They are the predominant species in the study area with the basal coverage of the *Fraxinus Mandschurica* and *Pinus Koraiensis* accounting for 26.3% and 27.5% of the total basal area (Dai et al., 2011). Both selected species for flux measurement are mature with a height of ~20 m. The flux bag was installed at the height of 15 m a.g.l.. Foliar GEM fluxes over *Fraxinus mandshurica* and *Pinus Koraiensis* were continuously measured during 16 - 17 and 17 - 18 Jul 2013, respectively, and 24-h continuous flux data were obtained for each species. Mean blank of flux chamber measured before and after the field experiment was -0.02 ± 0.04 ng m^{-2} h^{-1} (n = 24), which was indistinguishable from zero and not used to calibrate the measured fluxes.

2.4 Isotopic Composition of Atmospheric GEM

From 8 to 18 July 2013, GEM samples were collected at 4 m a.g.l. at the study site for Hg isotopes analysis using a chlorine-impregnated activated carbon (CLC) trap (Fu et al., 2014). Atmospheric GEM was collected daily (24-h sampling duration) at a flow rate of 10 LPM. CLC traps collect GEM at > 95% efficiency at the given sampling flow rate (Fu et al., 2014). To remove air particles, a 47-mm diameter Teflon filter (pore size 0.2 μm) was installed at the inlet of CLC trap. The CLC trap was kept warm (50 - 70 °C) during sampling using a silicone rubber heating pads (RadioSpares) to prevent water condensation. The sampling flow rate of CLC traps was regulated via a gas flow meter installed at the outlet of the vacuum pump, and the total sampling volumes of the CLC traps were recorded using a gas meter, calibrated to standard volumes under a standard pressure of 1013 hPa and a standard temperature of 273.14 K using a Bios Defender.

After the completion of field sampling, CLC traps were sealed with silicone stoppers and three successive polyethylene bags and stored in a clean environment until pre-concentration into trap solutions for Hg isotope analysis. GEM collected by CLC traps were preconcentrated into reverse aqua regia solution (*v*/*v*, $2HNO_3/1HCl$) in the laboratory using a double-stage combustion protocol for Hg isotope analysis (Biswas et al., 2008; Sun et al., 2013; Fu et al., 2014). Hg isotope ratios were determined by Nu-Plasma MC-ICP-MS following a previously established method (Yin et al., 2013). Hg isotopic composition is reported in delta notation (δ) in per mil referenced to the bracketed NIST 3133 Hg standard (Blum and Bergquist, 2007):

$$\delta^{xxx}Hg = \left(\frac{\left(\frac{^{xxx}Hg}{^{198}Hg}\right)_{sample}}{\left(\frac{^{xxx}Hg}{^{198}Hg}\right)_{SRM3133}} - 1 \right) \times 1000‰ \qquad (2)$$

Mass independent fractionation (MIF) values are expressed by "capital delta (Δ)" notation (‰), which is the difference between the measured values of $\delta^{199}Hg$, $\delta^{200}Hg$, $\delta^{201}Hg$ and those predicted from $\delta^{202}Hg$ using the kinetic MDF law (Blum and Bergquist, 2007):

$$\Delta^{199}Hg\ (‰) = \delta^{199}Hg - (0.252 \times \delta^{202}Hg) \qquad (3)$$

$$\Delta^{200}Hg\ (‰) = \delta^{200}Hg - (0.502 \times \delta^{202}Hg) \qquad (4)$$

$$\Delta^{201}Hg\ (‰) = \delta^{201}Hg - (0.752 \times \delta^{202}Hg) \qquad (5)$$

The analytical uncertainty of isotopic analysis was obtained by repeated analysis of the UM-Almaden standard. The overall mean values of $\delta^{202}Hg$ and $\Delta^{199}Hg$ for all the UM-Almaden standards were -0.57 ± 0.09 ‰ and -0.03 ± 0.04 ‰ (2SD, n = 12), respectively, consistent with previously reported values (Blum and Bergquist, 2007). In the present study, the analytical uncertainty of CV-MC-ICPMS isotope analysis is the 2SD uncertainty of the UM-Almaden standard, unless the 2SD uncertainty on repeated analysis of the same sample over different analytical sessions is larger.

3 Results and discussion

3.1 Characteristics of depletion events at Mt. Changbai

From Oct 2008 to Dec 2015, we observed 52 strong depletion events with dips of GEM concentrations < 0.5 ng m^{-3}. These depletions occurred predominantly at night during leaf-growing season and generally lasted for 0.5-6 hours (Figure 1). GEM concentrations during the strong depletion events decreased rapidly from background level of ~1.50 ng m^{-3} around noon to < 0.5 ng m^{-3} at night, corresponding to > 65% loss of GEM. It is worth noting that depletions also occurred at night during non-leaf-growing season (Jan-Apr and Oct-Dec). However, depletions of atmospheric GEM during non-leaf-growing season were less pronounced and frequent compared to leaf-growing season with dips of GEM concentrations generally higher than 1.0 ng m^{-3}, which represented 2/3 of the background level at the study site (Figure 1). Figure 2 shows the representative depletion events in summer of 2010 and 2013. Strong depletion of GEM consistently occurred at night. During the 7 - 13 July, 2010 period, a nearly complete depletion occurred with GEM concentrations decreasing from 1.6 - 2.0 ng m^{-3} at noon to nearly zero at night (removal of GEM averaged 1.83 ± 0.35 ng m^{-3} (n = 7)). The daytime peak GEM concentrations for the depletion events during 9 - 23 Jul, 2013 ranged from 1.50 - 2.31 ng m^{-3}, and the lowest GEM concentrations at night were 0.35 - 0.99 ng m^{-3}, yielding an averaged removal of GEM of 1.08 ± 0.23 ng m^{-3} (n = 12).

GOM concentrations during the strong nighttime atmospheric GEM depletion events (n = 10, defined as nighttime dips in GEM concentrations < 0.5 ng m^{-3}) from Jul 2013 to Jul 2014 were typically low (< 3 pg m^{-3} with a mean value of 0.8 pg m^{-3}). This is in contrast to previously characterized GEM depletions in the polar and sub-polar regions, marine boundary layer and free troposphere where depletions of GEM were accompanied by strong GOM enhancements (up to 195 - 1200 pg m^{-3}) (Lindberg et al., 2002; Swartzendruber et al., 2006; Sheu et al., 2010; Obrist et al., 2011; Lyman and Jaffe, 2012). Wind speed was low (mean of 0.1 m s^{-1} during 7 - 13 Jul, 2010 and 0.4 m s^{-1} during 9 - 23 Jul, 2013) during the nighttime depletion events at the study site (Figure 2). Shallow nocturnal boundary layer (NBL, see text in the SI) was frequently developed when the depletion occurred with a mean height of 146 m (74 - 200 m) during 7 - 13 Jul, 2010 and 209 m (57 - 300 m) during 9-23, Jul, 2013 (Figure 2). The low winds and shallow

NBL limited the transport of air masses at the sampling site and facilitated a continuous depletion of GEM in the presence of vegetative uptake of GEM (more details in sections below). During daytime, the surface wind speed and NBL depth increased due to solar heating (Talbot et al., 2005), enabling the downward transport of GEM from upper air, resulting in the increasing GEM concentrations.

Strong depletion of GEM during summer nighttime has also been observed at forest sites in North America (e.g., St. Anicet Maple forest station in Canada, and Piney Reservoir, Huntington Wildlife, Thompson Farm, Kejimkujik National Park, and Stilwell in AMNet, USA) (Mao et al., 2008; Poissant et al., 2008; Lan et al., 2012). Such depletion of GEM in forest ecosystems is likely a widespread phenomenon globally. The depletion at forest sites was different from the atmospheric mercury depletion events (AMDEs) elsewhere. For instance, the AMDEs at Cape Point, coast of South Africa and Dead Sea, Israel are mostly observed during daytime (Brunke et al., 2010; Obrist et al., 2011). The AMDEs in the Polar Regions occur exclusively during Polar sunrise in spring and do not exhibit a well-defined diurnal pattern (Schroeder et al., 1998; Ebinghaus et al., 2002; Lindberg et al., 2002).

3.2 Vertical gradient of GEM observed at Mt. Changbai

A clear vertical gradient of GEM concentrations was observed at the study site, with increasing GEM concentrations with respect to sampling altitude (Figure 3). The average difference in GEM concentrations between 45 m and 1 m (all in a.g.l., $\Delta GEM_{45\text{-}1m}$) was 0.22 ± 0.15 ng m^{-3} (n = 330), ~20% of the mean GEM concentration at 45 m a.g.l.. Average differences in GEM concentrations between 45 m and 24 m ($\Delta GEM_{45\text{-}24m}$), between 24 m and 10 m ($\Delta GEM_{24\text{-}10m}$), and between 10 m and 1 m ($\Delta GEM_{10\text{-}1m}$) were 0.11 ± 0.10, 0.05 ± 0.09, and 0.06 ± 0.11 ng m^{-3} (n = 330), respectively. The observed gradient suggested that the forest at the study site is a net sink for atmospheric GEM, in contrast to the vertical GEM gradients observed in a mature hardwood forest (between 30 and 40 m a.g.l.) in Walker Branch Watershed, Tennessee, USA during daytime, which showed decreasing GEM concentrations with sampling altitude above the forest canopy (Lindberg et al., 1998). This difference might be caused by the different forest structure and elevated emission flux of GEM from forest soil (7.5 ng m^{-2} h^{-1} in Walker Branch Watershed versus 2.8 ng m^{-2} h^{-1} at Mt. Changbai, Supplementary Figure S3) in Walker Branch Watershed (Kim et al., 1995; Lindberg et al., 1998).

The vertical gradients of GEM at Mt. Changbai showed clear diurnal trends (Figure 3 and Supplementary Figure S4). $\Delta GEM_{45\text{-}24m}$ and $\Delta GEM_{24\text{-}10m}$ values were comparably higher at night (means of 0.13 and 0.08 ng m^{-3}, respectively) than those during daytime (mean of 0.09 and 0.02 ng m^{-3}, respectively). The smaller daytime $\Delta GEM_{45\text{-}24m}$ and $\Delta GEM_{24\text{-}10m}$ were a result of weaker dry deposition of GEM to the forest canopy (more discussion later). A strong negative correlation between the $\Delta GEM_{24\text{-}10m}$ and wind speed ($r^2 = 0.55$, $p < 0.01$) also suggested stronger vertical mixing during daytime inhibited the buildup of GEM gradient. The diurnal trend of $\Delta GEM_{10\text{-}1m}$ was opposite to $\Delta GEM_{45\text{-}24m}$ and $\Delta GEM_{24\text{-}10m}$, with larger values during daytime (mean = 0.09 ng m^{-3}) and lower values at night (mean = 0.04 ng m^{-3}).

3.3 Foliage/air exchange flux of GEM

Mean foliar GEM fluxes over *Fraxinus Mandschurica* and *Pinus Koraiensis* were -1.2 ± 0.6 (-2.2 to -0.2 ng m^{-2} h^{-1}) and 0.0 ± 0.4 ng m^{-2} h^{-1} (-0.5 to 2.0 ng m^{-2} h^{-1}), respectively (Figure 4). Mean ambient GEM concentrations during the flux measurements over *Fraxinus Mandschurica* and *Pinus Koraiensis* were 1.42 ± 0.23 and 0.93 ± 0.28 ng m^{-3}, respectively, below the background concentrations of GEM in the Northern Hemisphere (1.5 - 1.7 ng m^{-3}) (Lindberg et al., 2007). The low GEM deposition flux over *Pinus Koraiensis* was partially attributed to the low ambient GEM concentration that weaken the deposition flux (Hanson et al., 1995; Ericksen and Gustin, 2004). The mean deposition fluxes over *Fraxinus Mandschurica* (0.7 ± 0.1 ng m^{-2} h^{-1}) was much greater than the that over *Pinus Koraiensis* (0.0 ± 0.5 ng m^{-2} h^{-1}) given the same GEM (1.0 – 1.4 ng m^{-3}) range (Figure 4), suggesting that GEM deposition flux varies with tree species with deciduous tree species inducing higher deposition compared to evergreen tree species (Millhollen et al., 2006).

The observed foliar GEM fluxes over *Fraxinus Mandschurica* and *Pinus Koraiensis* were within the range of reported values (means = -6 to 3.5 ng m^{-2} h^{-1}) (Ericksen et al., 2003;

Frescholtz and Gustin, 2004; Gustin et al., 2004; Graydon et al., 2006; Poissant et al., 2008; Stamenkovic and Gustin, 2009). A diurnal pattern with higher deposition fluxes at night was observed for both species. The higher deposition flux at night can be attributed to enhanced foliar GEM uptakes. As seen in Figure 4, difference in GEM concentrations between the inlet and outlet stream of the flux bag (corresponding to the net loss of atmospheric GEM to foliage) over *Fraxinus Mandschurica* at night (mean = 1.31 ± 0.23 ng m^{-3}) were much higher compared to daytime (mean = 0.48 ± 0.11 ng m^{-3}). It has been suggested that lower O_3 and higher relative humidity (RH) could facilitate the uptake of GEM by foliage (Lindberg and Stratton, 1998; Stamenkovic and Gustin, 2009). O_3 and RH at the study site showed strong diurnal patterns with decreasing O_3 concentrations and increasing RH at night (Supplementary Figure S5), which may explain the higher deposition fluxes of GEM to foliage at night. Both stomatal and non-stomatal uptakes have been suggested to be responsible for the observed foliage-atmosphere GEM exchange (Zhang et al., 2005; Stamenkovic and Gustin, 2009). Stamenkovic and Gustin (2009) found that GEM deposition flux to foliage remained essentially unchanged whether or not stomata are open. This indicates that non-stomatal route plays an important role in the uptake of GEM by foliage, consistent with the observations in this study. Previous studies suggested that foliar exchange of GEM is bi-directional with foliage emitting GEM at global background air GEM concentrations (Hanson et al., 1995; Ericksen and Gustin, 2004; Graydon et al., 2006). With the GEM concentrations in the range of 0.41 - 1.82 ng m^{-3} during this study, however, net deposition was observed except for *Pinus Koraiensis* during daytime when stoma are open. Net emission of GEM from *Pinus Koraiensis* during daytime could be attributed to the enhanced photochemical reduction and re-emission of previously deposited Hg (GEM, GOM and PBM), Hg in dew water and transpiration stream as well as transpiration of Hg^0 in soil pores (Bishop et al., 1998; Lindberg et al., 1998; Ericksen and Gustin, 2004; Stamenkovic and Gustin, 2009).

The observed foliar GEM fluxes over *Fraxinus Mandschurica* were negatively correlated with the GEM concentrations in the inlet air (Figure 5A), yielding a compensation point of 0.52 ng m^{-3} during daytime and 0.47 ng m^{-3} during nighttime, respectively. No clear correlation between foliar GEM fluxes and ambient GEM concentrations was observed for *Pinus Koraiensis* during daytime. However, a significant negative correlation was observed at night when ambient GEM concentrations were higher than 0.98 ng m^{-3} (Figure 5B), which was likely the compensation point for *Pinus Koraiensis* during nighttime. These observed compensation points were comparatively lower than the values (2 - 3 ng m^{-3}) measured in laboratory studies (Ericksen and Gustin, 2004; Graydon et al., 2006), but consistent with the field observation at St. Anicet Maple forest, Canada (0.53 ng m^{-3}) (Poissant et al., 2008). For *Pinus Koraiensis*, the observed foliar GEM fluxes were not significantly different from zero (mean = -0.1 ± 0.1 ng m^{-2} h^{-1}) at GEM concentrations lower than the compensation point (0.98 ng m^{-3}). A similar conclusion cannot be reached for *Fraxinus Mandschurica* because the ambient GEM concentrations were higher than the respective compensation points during the entire campaign (Figure 5A). This finding is different from previous results that showed net GEM emissions from foliage at ambient GEM concentrations below the compensation points (Hanson et al., 1995; Graydon et al., 2006; Poissant et al., 2008). Based on the field findings, it is likely that the uptake and emission of GEM over the foliage of *Pinus Koraiensis* reached equilibrium during nighttime when the ambient GEM concentrations were below the compensation point.

The total deposition flux of GEM to forest canopy at Mt. Changbai was estimated using Equation (6):

$$\mathrm{F} = \mathrm{LAI} \times \sum_{\mathrm{i}}^{n} (\mathrm{F_i} \times \mathrm{A_i}) \qquad (6)$$

where F is the total deposition flux of GEM in ng m^{-2} h^{-1}, LAI is the mean leaf area index (dimensionless), F_i is the foliar GEM flux of a tree species (i) in ng m^{-2} h^{-1}, and A_i is the relative basal area of a tree species (i) in percentile (Dai et al., 2011). In this study, it is assumed that the measured mean foliar GEM fluxes over *Fraxinus Mandschurica* and *Pinus Koraiensis* are representative of deciduous tree species and evergreen tree species, respectively. The measured mean LAI at Mt. Changbai during leaf-growing season was 5.4.

The total deposition fluxes of GEM to forest canopy at Mt. Changbai during nighttime and daytime are estimated to be 7.3 (V_d of 0.14 cm s^{-1}) and 2.5 ng m^{-2} h^{-1} (V_d of 0.04 cm s^{-1}). We acknowledge that, due to the relatively short field sampling periods for the two selected tree species and the fact that foliar GEM flux may vary with tree species, GEM concentrations and other environmental variables, our estimates may have large uncertainties. Nevertheless, the estimates are generally consistent with the measured deposition flux using Hg accumulated in foliage over time. The mean mass-weighted Hg concentration in litter samples at the study site was 43.0 ± 29.5 ng g^{-1} (Supplementary Table S1). With the annual litterfall of 486 g m^{-2} at the site (Zhou et al., 2014), the Hg deposition flux in litterfall was 20.9 ± 14.3 μg m^{-2} yr^{-1}. Assuming that the plant foliage had a constant uptake rate of Hg in the leaf-growing season, the hourly deposition flux of Hg that end up being contained in litterfall would be 5.7 ng m^{-2} h^{-1}, comparable to the GEM deposition flux calculated from flux bag observations (daily mean: 4.9 ng m^{-2} h^{-1}).

3.4 Mechanisms for the observed GEM depletion

Oxidation of GEM by reactive halogens and O_3 has been proposed to be an important mechanism for GEM depletions observed elsewhere as evidenced by the elevated GOM concentrations (up to 500 - 1200 pg m^{-3}) associated with the GEM depletion events and an inverse correlation between GOM and GEM concentrations (Lindberg et al., 2002; Obrist et al., 2011; Lyman and Jaffe, 2012). Based on modeling assessments, the nighttime loss of GEM in forest areas has been suggested to be caused by dry deposition and chemical oxidation (by O_3, $OH^\cdot$ and NO_3) (Mao et al., 2008). However, the GOM concentrations observed during strong nighttime GEM depletion events at Mt. Changbai were extremely low (< 3 pg m^{-3} with a mean value of 0.8 pg m^{-3}), similar to those observed at other forest sites (Piney Reservoir, Huntington Wildlife, Thompson Farm, Kejimkujik National Park, and Stilwell) in North America (means = 0.5 - 4 pg m^{-3} at summertime night) (Lan et al., 2012). In addition, concentrations of many atmospheric oxidants (e.g., O_3, $OH^\cdot$, NO_3, BrO) at global forest sites were low (Spivakovsky et al., 2000; Yang et al., 2005; Rinne et al., 2012; Hens et al., 2014), which does not support significant conversion of GEM to GOM. Given the environmental condition at Mt. Changbai, the dry deposition flux of GOM was estimated to be 0.034 ng m^{-2} h^{-1}, using the mean nighttime

GOM concentration (0.8 pg m^{-3}) measured during the strong GEM depletion events and reported V_d of GOM (0.1 to 5.9 cm s^{-1} with a mean of 1.2 cm s^{-1}) to forest canopy (Lindberg and Stratton, 1998; Rea et al., 2000; Zhang et al., 2012). Even with a correction factor of 3 to account for the potential under-estimation of GOM concentration by the Tekran® speciation system (Gustin et al., 2013; Huang et al., 2013; Gustin et al., 2015), the deposition flux contributed by GOM is 0.1 ng m^{-2} h^{-1}. Assuming that all GOM was formed through in situ oxidation of GEM, the chemical pathway would contribute to merely 1.4% of the measured deposition flux of GEM to forest canopy during the nighttime depletion events.

Measurements of GEM isotopic composition also provided insight into the mechanisms responsible for the GEM depletion at Mt. Changbai. δ^{202}Hg, Δ^{199}Hg, and Δ^{200}Hg of the daily GEM samples from 8 to 18 Jul 2013 were -0.34 to 0.91‰, -0.11 to -0.04‰ and -0.06 to 0.01‰, respectively (n=10, Figure 6, Supplementary Table S2). These are consistent with the observations in the Great Lakes region, Barrow, Alaska, Pensacola, FL and Wisconsin forest in USA and the Pic du Midi Observatory in France ($\delta^{202}Hg_{GEM}$ = -0.12 to 1.43‰, $\Delta^{199}Hg_{GEM}$ = -0.31 to -0.01‰, Δ^{200}Hg = -0.11 to 0.1‰) (Gratz et al., 2010; Sherman et al., 2010; Demers et al., 2013; Demers et al., 2015; Fu et al., 2016a). A large positive $\delta^{202}Hg_{GEM}$ shift was associated with strong GEM depletions; whereas $\Delta^{199}Hg_{GEM}$ and $\Delta^{200}Hg_{GEM}$ remained unchanged. The $\delta^{202}Hg_{GEM}$ was up to 0.91‰ during the most pronounced depletion event (on 13 Jul 2013, daily mean GEM of 0.91 ng m^{-3}), 1.05‰ higher than the values at the beginning and end of the sampling period (on 9 and 17 Jul 2013, mean GEM = 1.57 - 1.60 ng m^{-3}, mean $\delta^{202}Hg_{GEM}$ = -0.14‰). The $\delta^{202}Hg_{GEM}$ values were anti-correlated with GEM concentrations ($r^2 = 0.58$, $p < 0.01$), whereas no clear relationship can be established between Δ^{199}Hg, and $\Delta^{200}Hg_{GEM}$ values and atmospheric GEM concentrations (p values for both > 0.05). The lower $\delta^{202}Hg_{GEM}$ values at the beginning and end of the sampling period were likely representative of the regional background $\delta^{202}Hg_{GEM}$ signatures as the mean GEM concentrations of the two samples (1.46-1.60 ng m^{-3}, Supplementary Table S2) were close to the long-term GEM mean concentration at Mt. Changbai, whereas the positive $\delta^{202}Hg_{GEM}$ shifts during 11 - 15 Jul 2013 were most likely due to the uptake of GEM by forest foliage which has been known to induce mass dependent fractionation (MDF, δ^{202}Hg signature) and negligible MIF (Δ^{199}Hg, Δ^{200}Hg signatures) of Hg isotopes (Demers et al., 2013; Enrico et al., 2016). MDF and MIF of Hg isotopes caused by GEM oxidation have not been well characterized. Studies observed both significant MDF and MIF of Hg isotopes during aqueous- and gas-phase chemical oxidation of elemental Hg (Stathopoulos, 2014; Sun et al., 2016). Our study at Pic du Midi, France (2877 m above sea level) also observed clear shifts of $\delta^{202}Hg_{GEM}$ and $\Delta^{199}Hg_{GEM}$ during oxidation of GEM to GOM, indicating both MDF and MIF could occur during 'net oxidation' of GEM in the ambient air (Sonke *et al.*, manuscript under preparation). Therefore, we conclude foliar uptake of GEM played a predominant role in the GEM depletion at Mt. Changbai.

To answer the question whether or not GEM dry deposition to forest canopy alone can explain the GEM depletion at Mt. Changbai, the forced change of GEM concentrations by canopy uptake at the sampling height of 24 m a.g.l. under a typical NBL height of 100 m was simulated using a box model (see text in the SI). The box model results suggest that complete GEM depletions can be achieved by canopy uptake alone in the presence of shallow NBL and low vertical turbulent diffusivity (Figure 7). With a dry deposition GEM flux of 7.3 ng m^{-2} h^{-1} (section 3.3) and turbulent diffusivity of 0.1 - 1.0 cm s^{-1} at night (Figure S6), the model predicted that GEM concentrations can be decreased to nearly 0 ng m^{-3} (Figure 7). Depletion cannot occur during daytime mainly due to the low dry deposition flux (~2.5 ng m^{-2} h^{-1}), high vertical turbulent diffusivity (1 - 100 cm s^{-1}) and absence of shallow NBL (Figure 7). The GEM depletion event at Mt. Changbai showed a clear seasonal trend with the depletion occurring more frequently and pronouncedly during leaf-growing season (Figure 1). This can be attributed to: (1) seasonal LAI changes (Figure S7.A), (2) lower wind speed during leaf-growing season (Figure S7.B), and (3) the wind direction that inhibited the transport of polluted air from anthropogenic source regions (90º-202º, natural preserve areas without significant local and regional sources) during leaf-growing season (Figure S7.C). LAI is much higher during leaf-growing season compared to non-leaf-growing season (<2, Figure S8.A) (Shi et al., 2008). Higher LAI values indicate higher dry deposition fluxes of GEM to forest canopy. The low wind speed facilitated the buildup of shallow NBL.

4 Conclusions and implications for the global atmospheric Hg cycling

Strong depletions of atmospheric GEM were consistently observed during leaf-growing season in Mt. Changbai forest, Northeast China. The depletions occurred exclusively at night in the absence of GOM enrichments. This is in contrast to previously characterized GEM depletions in the polar and sub-polar regions, marine boundary layer and free troposphere where depletions of GEM were mainly caused by fast chemical oxidation of GEM to GOM followed by deposition. The measurements of GEM vertical gradients, foliar GEM fluxes, atmospheric speciated Hg and ambient GEM isotope compositions suggest foliar uptake of GEM played a predominant role in the GEM depletion at Mt. Changbai.

Forests cover ~30% (~40 million km^2) of the Earth's land surface. There is a need to quantitatively assess the role of global forest in global Hg cycling. Tables S3, S4, and S5 summarize the published data of litterfall fluxes at 68 forest sites, throughfall fluxes at 23 forest sites, and emissions from forest floors at 31 forest sites in North America, Europe, Asia, and South America. For the regions (Africa and Oceania) that lack observational data, it is assumed that that the median values of the published data are representative. There has not been reliable data on Hg emission from forest canopies via evapotranspiration. We therefore use the observed xylem Hg concentrations and total evapotranspiration from the global forests to estimate Hg emissions from this sector (Bishop et al., 1998; Baldocchi and Ryu, 2011).

Using a mass balance approach, we estimated that global inputs of Hg via litterfall and throughfall were 1,232 and 1,338 Mg yr^{-1}, respectively. Hg emissions via the evasion from soil and plant evapotranspiration were 381 and 260 $Mgyr^{-1}$, respectively. Combining the source and sink terms, the global forest ecosystem represents a net sink of ~1,930 Mg yr^{-1} of atmospheric Hg. The value is much larger than the estimate of Hg uptake by forest above-ground biomass (Obrist, 2007). The estimate by Obrist (2007) did not include deposition flux by throughfall; and the Hg concentration in biomass used in the study was 2 - 10 times lower than the measured Hg contents in North America, Europe, China and South America (Lindberg et al., 2007; Obrist, 2007; Risch et al., 2012; Teixeira et al., 2012; Fu et al., 2015). Our estimate is comparable to the upper limit of atmospheric Hg deposition to terrestrial ecosystem predicted by modeling studies (800-1900 Mg) (Mason and Sheu, 2002; Holmes et al., 2010; Driscoll et al., 2013). This implies that forest ecosystem may be the largest sink of atmospheric Hg in the terrestrial ecosystems, whereas other terrestrial ecosystems may represent net sources.

Supporting Information:

Descriptions of the simulation of NBL, turbulent diffusivity and the box model are shown in supplementary text. The location of the Mt. Changbai forest, GEM concentrations at 4 m a.g.l. in a small clearing plot and 24 m and 45 m a.g.l., diurnal trends in vertical GEM gradient, soil/air GEM flux, diurnal variations of meteorological parameters, turbulent diffusivity and seasonal variations in LAI, wind speed and wind direction at Mt. Changbai forest are shown in Figure S1-S7. Littefall Hg concentrations and litter mass at Mt. Changbai forest, isotopic composition of atmospheric GEM as well as compiled litterfall and throughfall Hg deposition fluxes, and forest soil/air GEM fluxes over the global forests are shown in Table S1-S5.

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Figure 1. Atmospheric 5-min GEM concentrations at Mt. Changbai from Oct 2008 to Dec 2015 (leaf-growing season is marked as the shaded area).

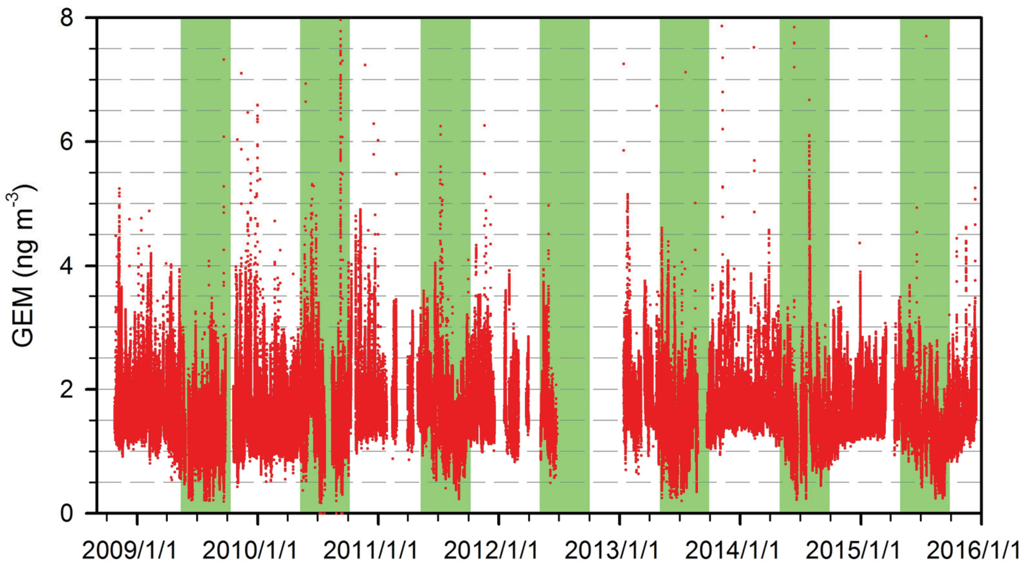

Figure 2. Time series of (A) GEM (5-min mean) and meteorological parameters from 3 to 19 July 2010 and (B) speciated atmospheric Hg (GEM, GOM, and PBM) and meteorological parameters 8 to 24 July 2013 (nighttime is marked as the shaded area).

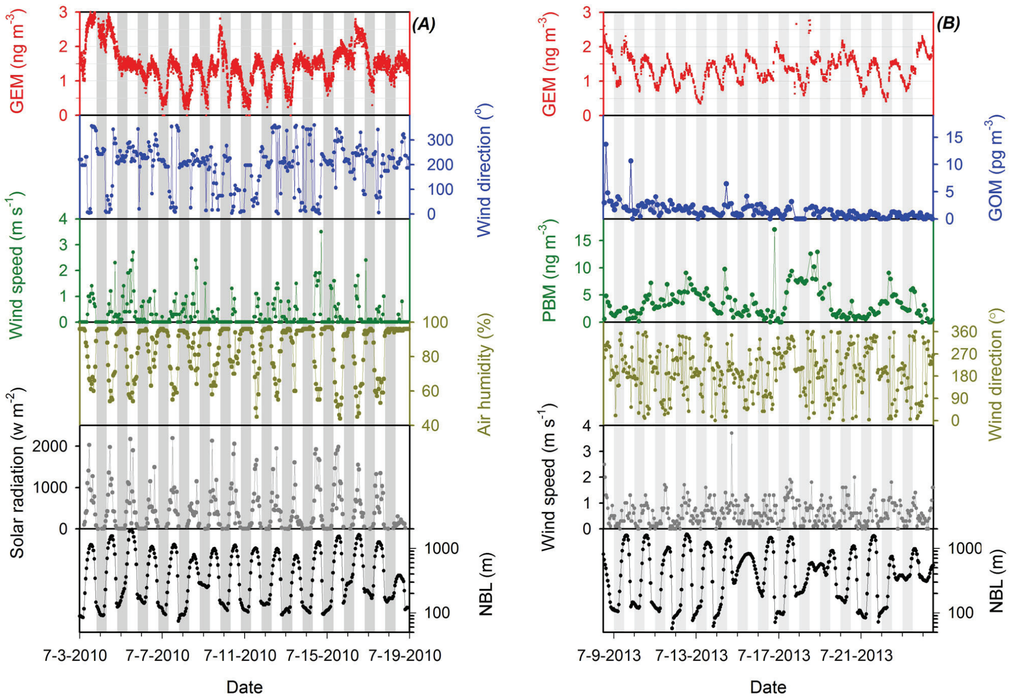

Figure 3. Diurnal variations of GEM concentrations at different height and metrological parameters in Mt. Changbai forest from 10 to 15 July 2013 (nighttime is marked as shaded area).

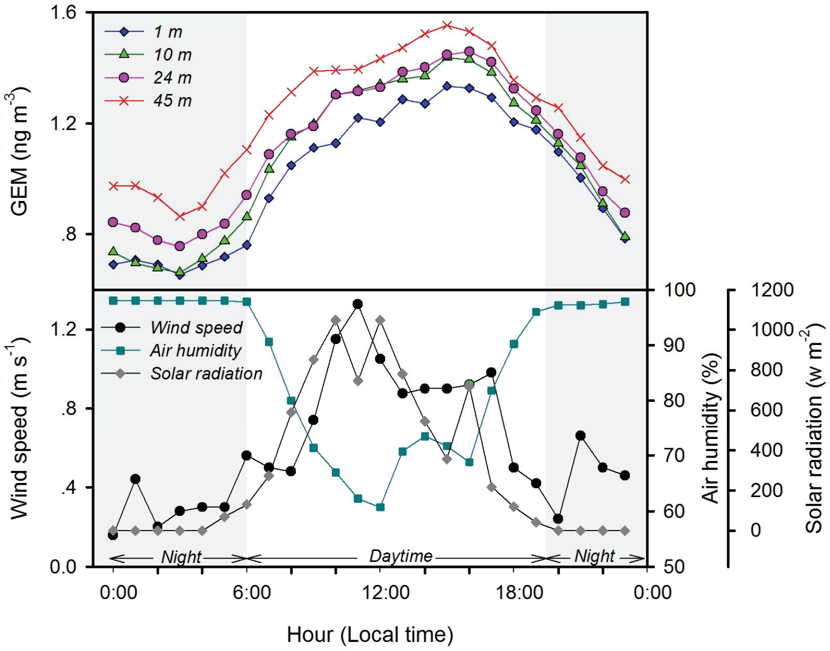

Figure 4. Foliar Hg flux over *Fraxinus Mandschurica* and *Pinus Koraiensis*, inlet and outlet GEM concentrations from flux bag and meteorological parameters at Mt. Changbai in July 2013 (nighttime is marked as the shaded area).

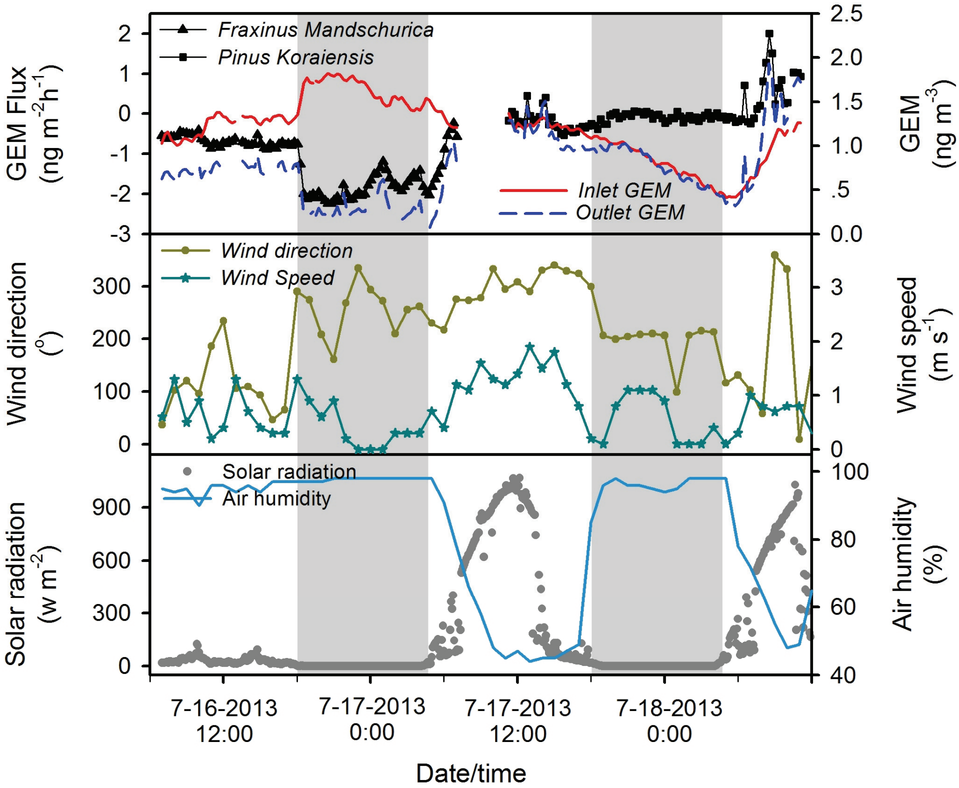

Figure 5. Daytime and nighttime correlations between atmospheric GEM concentrations and foliar GEM fluxes over (A) *Fraxinus Mandschurica* and (B) *Pinus Koraiensis*.

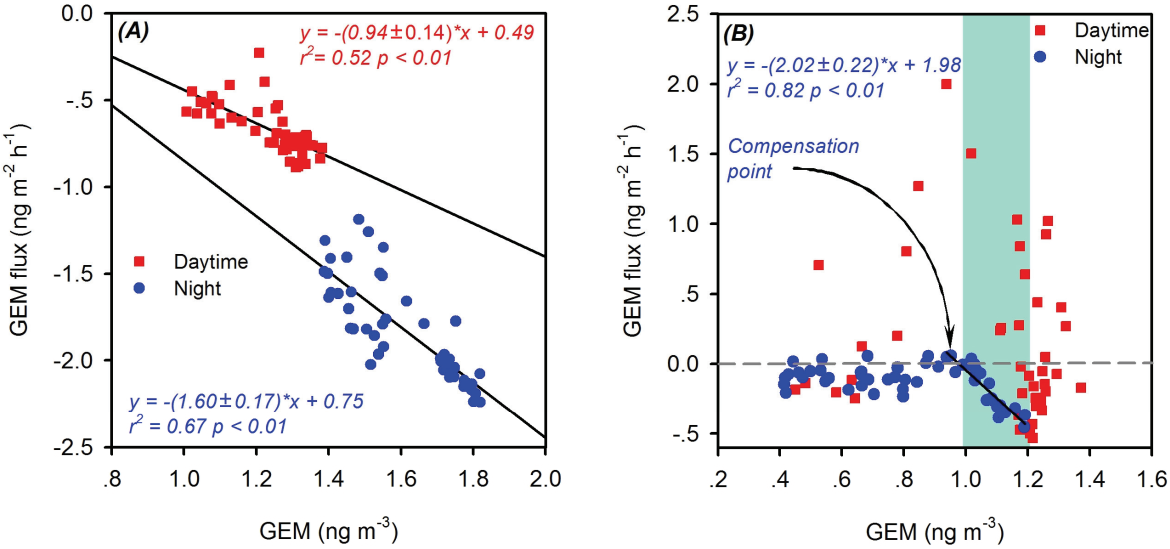

Figure 6.Temporal variation in (A) atmospheric GEM concentrations and (B) δ^{202}Hg, Δ^{199}Hg and Δ^{200}Hg values of daily integrated atmospheric GEM from 9 to 18 July 2013.

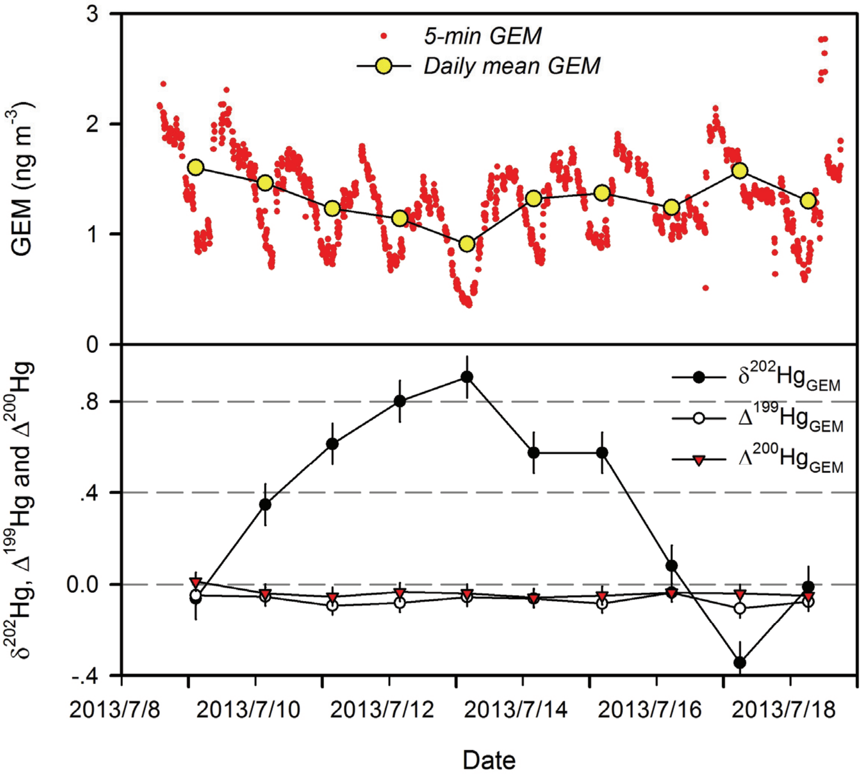

Figure 7. Modeling predicted variations of GEM concentration at the height of 24 m a.g.l. with dry position fluxes of GEM to forest canopy and vertical turbulent diffusivity under a typical NBL height of 100 m.