Supporting Information:

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

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1 Supplementary Text

1.1 Nocturnal boundary layer

The Nocturnal boundary layer (NBL) was calculated by Weather Research and Forecasting Model (WRF) 3.5 with two-way nested runs. The spatial resolution for course domain is 30 km with 100×100 grid cells, and for the nested domain is 10 km with 30×30 grid cells. As our studied site is 100-200 km away from Sea of Japan, we chose the MYJ scheme for NBL as earlier studies suggested that MYJ scheme was as first choice for marine atmospheric boundary layer simulations without *a priori* information of atmospheric stability in the region of interest (Huang et al., 2013;Krogsaeter and Reuder, 2015). For other parameterizations, we selected Kain and Fritsch cumulus scheme for cumulus parameterization, Lin (Purdue) scheme for microphysics options RRTM scheme for Radiation Physics Options.

1.2 Simulations of atmospheric GEM at Mt. Changbai forest using a box model

A box model was applied to estimate the GEM concentration at the height of 24 m agl. Based on the measured characteristics of the GEM depletion events, the model assumes that vegetative uptake (in terms of dry deposition flux) is the only pathway for the GEM removal and chemical transformation is not included in the box modeling. A sensitivity analysis was performed on three parameters: (1) dry deposition flux (0-10 ng m^{-2} h^{-1}, the range of measured deposition flux using flux bags), (2) turbulent diffusivity of the atmosphere (0.1-10 cm s^{-1}, typical value under low wind condition), and (3) a typical nocturnal boundary layer height (100 m agl). GEM concentration above the stable nocturnal boundary layer was assigned to 1.56 ng m^{-3}, the mean observed at 45 m during daytime when the vertical mixing is strong. The flux of vegetative uptake (F_C) and the resulted concentration gradient was calculated based on the on the algorithm:

$$F_C^{AGM} = -\underbrace{\frac{\kappa \cdot u_*}{\Phi_H(\varsigma_1)}}_{K_C} \cdot \frac{\partial C}{\partial \ln(z)} \qquad (1)$$

where k is von Kármán constant (~0.41), u_* is the friction velocity (m s^{-1}), $\psi_H(\varsigma_1)$ is the integrated universal function for sensible heat to correct for deviations from the ideal logarithmic profile, K_C term is the turbulent diffusivity (m s^{-1}), C is the concentration parameter for GEM concentration (ng m^{-3}), z is the height parameter (m).

1.3 The turbulent diffusivity (K_c)

The flux-gradient approach (Kaimal and Finnigan, 1994) expresses a scalar flux (F, e.g. ng m^{-2} s^{-1}) as the product between the turbulent diffusivity (K_C, m^2 s^{-1}) and a concentration gradient ($\partial c / \partial z$, ng m^{-4}) assuming that measurements are made within a vertical layer of constant flux that forms over homogeneous terrain:

$$F = -K_C \cdot \frac{\partial c}{\partial z} = -\frac{u_* \cdot \kappa \cdot (z-d)}{\phi_H(\varsigma)} \cdot \frac{\partial c}{\partial z} = -\frac{u_* \cdot \kappa}{\phi_H(\varsigma)} \cdot \frac{\partial c}{\partial \ln z} \quad (2),$$

where, u_* is the friction velocity (m s^{-1}), κ is the von Kármán constant (taken as 0.4), $\phi_H(\varsigma)$ is the diabatic influence function for heat (parameterized as a function of $\varsigma = (z-d)/L$, where L is the Obukhov length), whereas z and d are the measurement and (canopy) displacement height (m) respectively. An empirical form of $\phi_H(\varsigma)$ is $0.95/\sqrt{(1-11.6\varsigma)}$ and $0.95 + 7.8\varsigma$ for unstable ($\varsigma < 0$) and stable ($2 > \varsigma \geq 0$) atmospheric conditions respectively (Foken, 2008).

For flux-gradient measurements made within the roughness sublayer above the canopy height (i.e. $h_{canopy} < z < z_*$), Eq. 1 is not valid (*underestimates* the magnitude of scalar flux) and requires further correction following e.g. Garratt (Garratt, 1992) and Simpson et al. (Simpson et al., 1998):

$$F = -\frac{u_* \cdot \kappa \cdot (z-d)}{\phi_h(\varsigma) \cdot \phi_*(z/z_*)} \cdot \frac{\partial c}{\partial z} \quad (3)$$

In contrast to $\phi_H(\varsigma)$, the additional correction function $\phi_*(z/z_*)$ in Eq. 2 is independent of stability. A common type of parameterization for ϕ_* is $\exp\left[-0.7\left(1-\frac{z}{z_*}\right)\right]$ (Garratt, 1992). In-turn, the upper limit of the roughness sublayer (z_*) can be estimated by $2 \cdot h_{canopy} - d$ (Raupach, 1994) .

Figure S1. Map showing the location of Mt. Changbai forest and coal fired power plants in Northeast Asia.

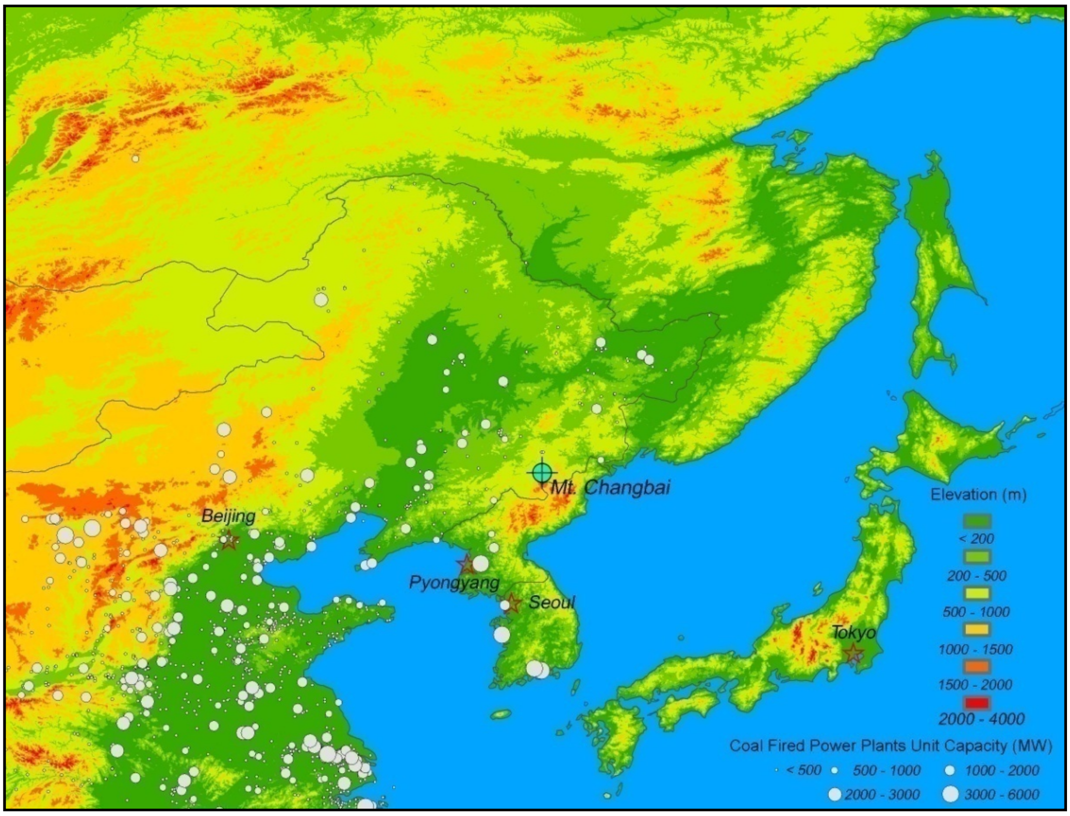

Figure S2. GEM at 4 m agl in the clear spot measured by the Tekran speciation unit, 24 m agl (~3 m above forest canopy, long-term GEM sampling site) and 45 m agl (~24 m above forest canopy) measured by the Tekran 2537 from 10 to 14 July 2013.

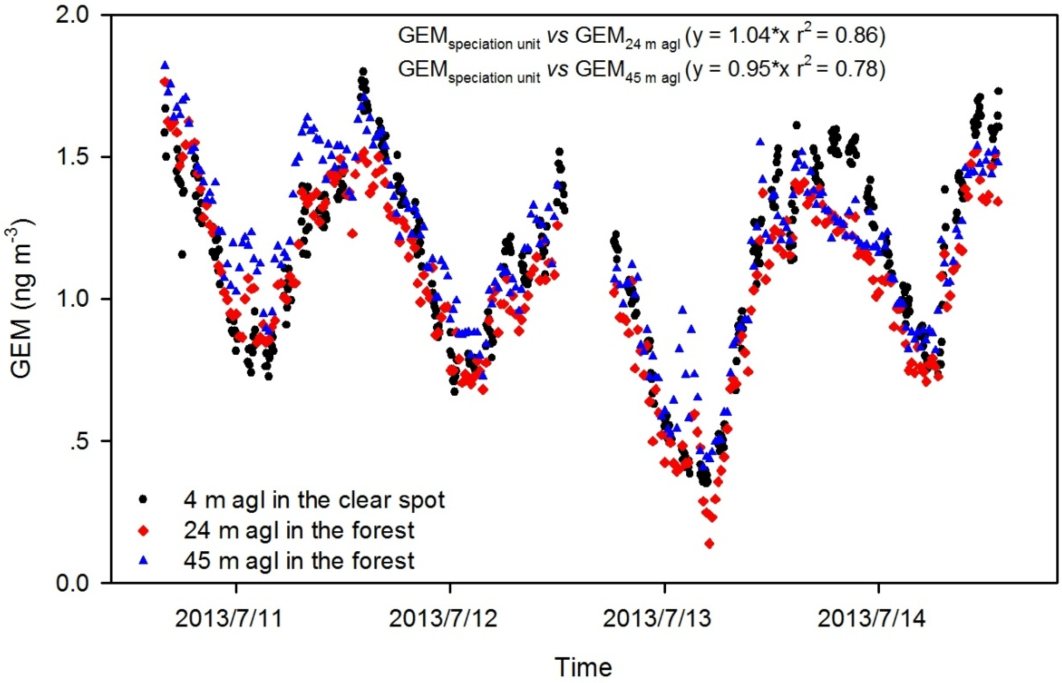

Figure S3. Time series of soil/air GEM flux and meteorological parameters in Mt. Changbai forest in July 2013.

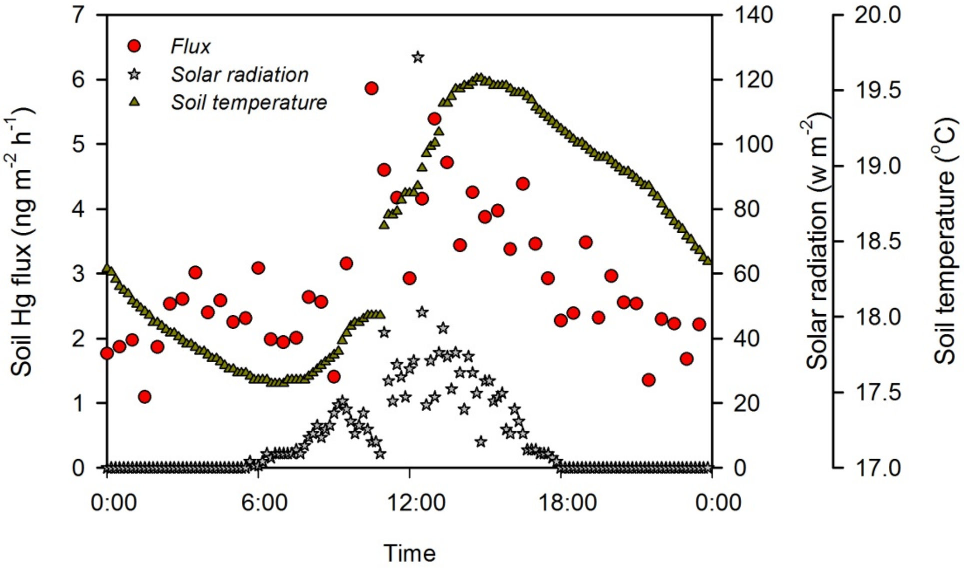

Figure S4. Diurnal trends in vertical gradient of GEM concentrations between the height of 45-24 m, 24-10 m and 10-1m in Mt. Changbai forest from 10 to 15 July 2013.

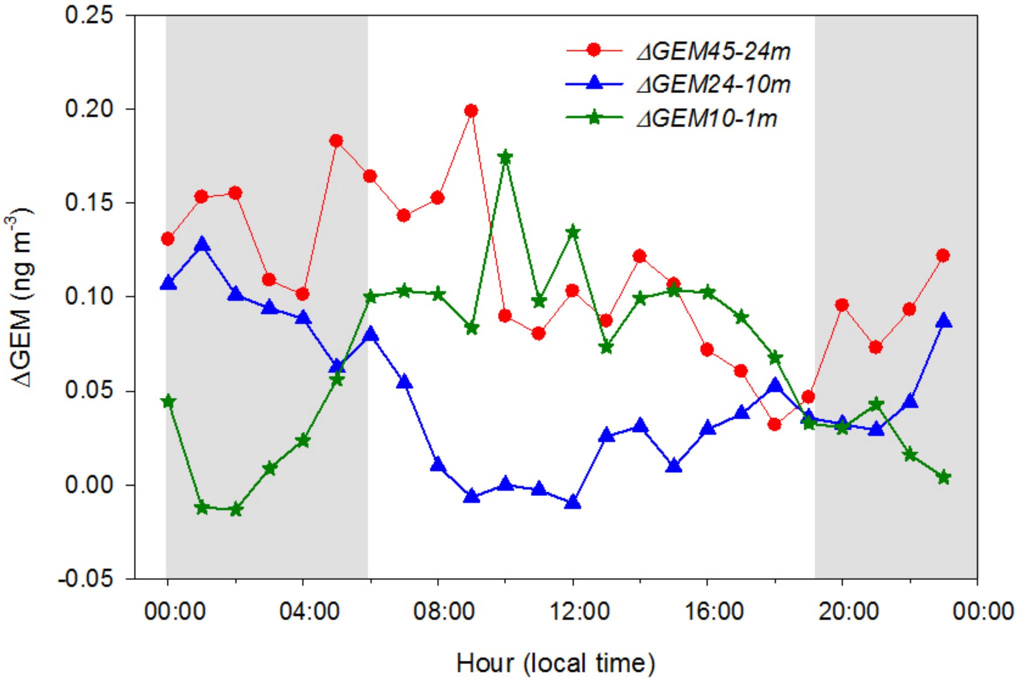

Figure S5. Diurnal variations of air temperature (A), wind speed (B), relative humidity (RH, C) and ozone concentrations (D) in Mt. Changbai forest in leaf-growing season from October 2008 to December 2014.

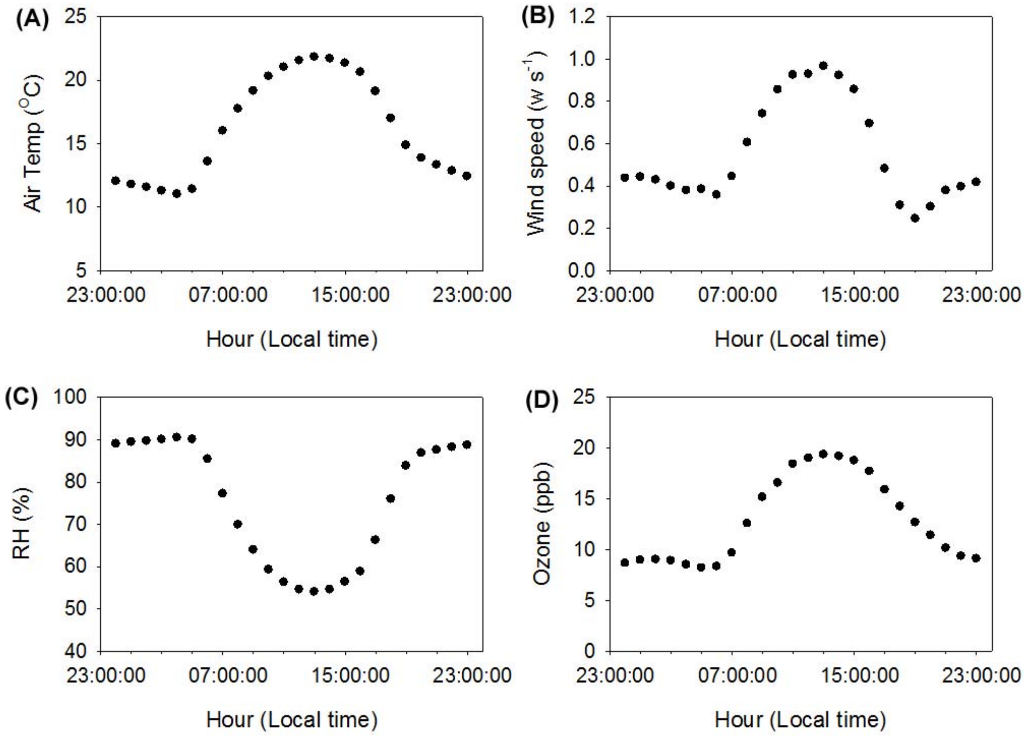

Figure S6. Temporal variation of the turbulent diffusivity at Mt. Changbai forest from 10 to 19 July 2013

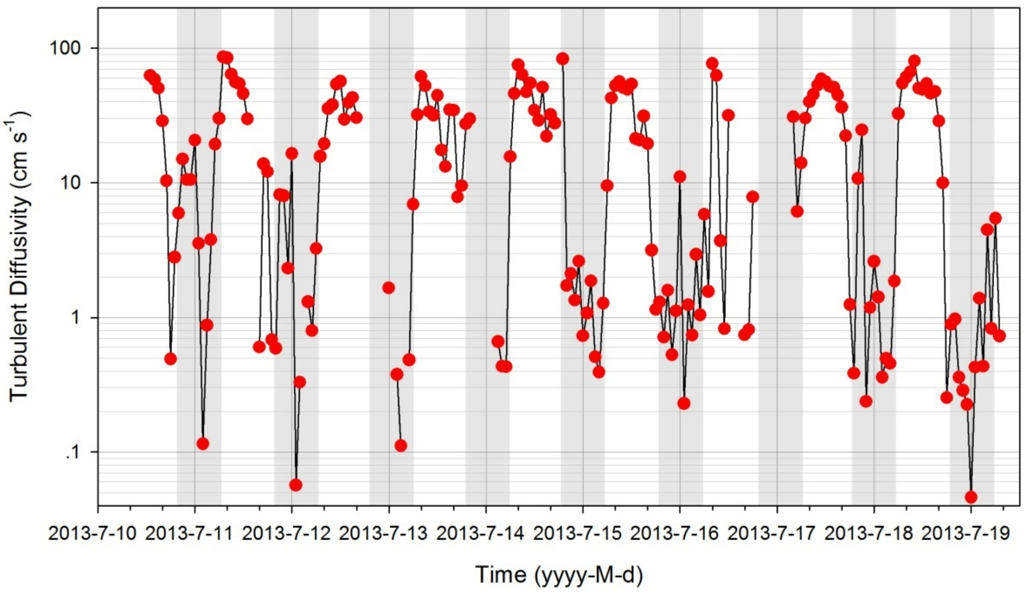

Figure S7. Wind frequency distributions in leaf-growing season and non-leaf-growing season from Aug 2009 to Jul 2013 (A), monthly mean wind speed from Aug 2009 to Jul 2013 (B), and Leaf areas index in leaf-growing season during 2003-2005(C) (Shi et al., 2008).

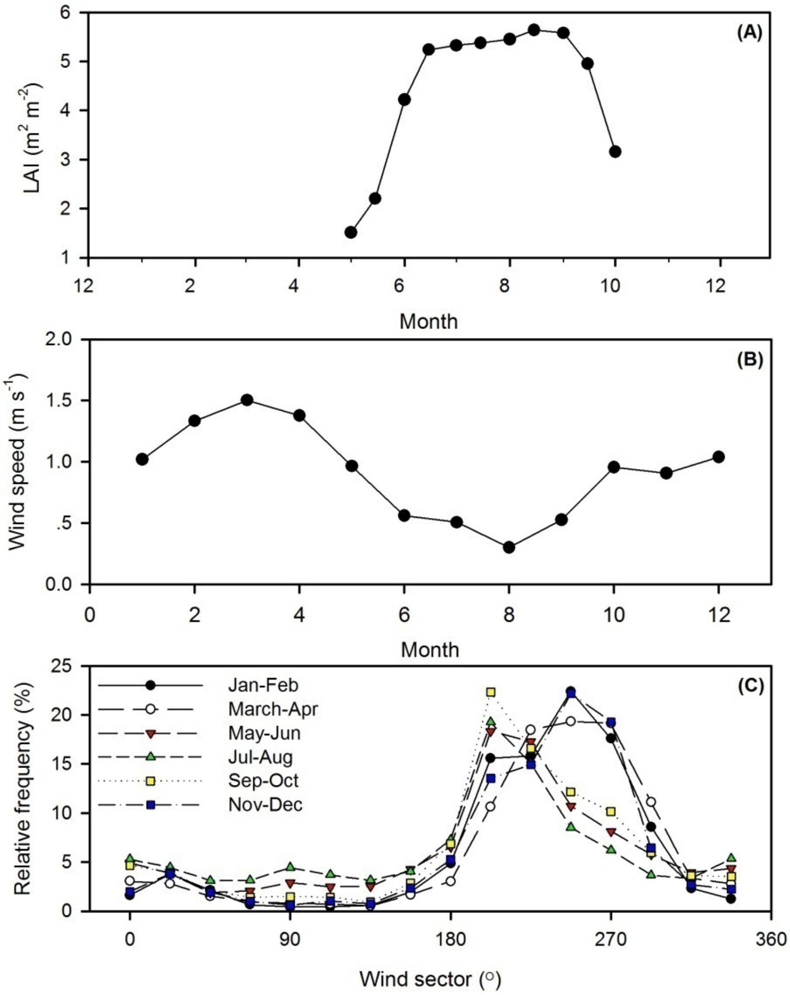

Table S1. Statistical summary of litterfall Hg concentrations and litter mass in Mt. Changbai forest

Litterfall collection site	Time	Species	Concentration (ng g^{-1})	Litter mass (g m^{-2})	Mass-weight concentration (ng g^{-1})
Collector-1	2013-09	*Pinus koraiensis*	44.5	51.70	74.8
		Acer pseudo-sieboldianum	138.5	24.95	
		Quercus mongolica	170.4	35.60	
		Fraxinus mandshurica	44.5	51.70	
		Tilia amurensis	138.5	24.95	
		Others	170.4	35.60	
Collector-1	2013-10	*Acer mono*	60.2	1.8	60.2
Collector-2	2013-09	*Pinus koraiensis*	40.4	6.48	39.7
		Acer pseudo-sieboldianum	46.8	5.38	
		Quercus mongolica	31.2	9.49	
		Acer mono	58.0	4.81	
		Fraxinus mandshurica	34.5	18.54	
		Tilia amurensis	45.7	52.12	
		Others	36.80	91.16	
Collector-2	2013-10	*Pinus koraiensis*	34.1	4.4	53.3
		Quercus mongolica	30.3	8.8	
		Acer mono	70.7	5.8	
		Others	81.3	6.6	
Collector-3	2013-09	*Acer ginnala Maxim*	38.1	5.8	31.1
		Pinus koraiensis	29.4	19.0	
		Acer pseudo-sieboldianum	49.6	4.6	
		Quercus mongolica	32.5	10.8	
		Acer mono	46.9	6.0	
		Fraxinus mandshurica	23.4	1.5	
		Tilia amurensis	40.4	19.6	
		Others	29.8	283.3	
Collector-3	2013-10	*Pinus koraiensis*	21.7	1.5	35.8
		Quercus mongolica	42.3	3.2	
Collector-4	2013-09	*Pinus koraiensis*	31.4	70.6	34.2
		Acer pseudo-sieboldianum	47.4	2.7	
		Quercus mongolica	26.3	4.1	
		Acer mono	50.9	15.5	
		Tilia amurensis	43.0	11.8	
		Others	32.8	121.1	
Collector-4	2013-10	*Pinus koraiensis*	40.1	19.4	64.4
		Quercus mongolica	46.0	21.7	
		Others	100.9	23.9	
		Overall mean			**43.0±29.5**

Table S2. Concentration of atmospheric speciated Hg (GEM, PBM, and GOM) and isotopic composition of atmospheric GEM at Mt. Changbai forest

Sample ID	Sampling period	GEM conc.	PBM conc.	GOM conc.	$\delta^{202}Hg$	$\delta^{202}Hg$	$\Delta^{199}Hg$	$\Delta^{199}Hg$	$\Delta^{200}Hg$	$\Delta^{200}Hg$	$\Delta^{201}Hg$	$\Delta^{201}Hg$
		ng m^{-3}	pg m^{-3}	pg m^{-3}	‰	2σ, ‰	‰	2σ, ‰	‰	2σ, ‰	‰	2σ, ‰
GEM-1	2013/7/8 13:40-2013/7/9 15:30	1.60	2	4	-0.06	0.09	-0.05	0.04	0.01	0.04	-0.01	0.06
GEM-2	2013/7/9 15:30-2013/7/10 15:30	1.46	2	2	0.35	0.09	-0.05	0.04	-0.04	0.04	-0.12	0.06
GEM-3	2013/7/10 15:30-2013/7/11 15:45	1.23	4	2	0.61	0.09	-0.09	0.04	-0.05	0.04	-0.08	0.06
GEM-4	2013/7/11 15:45-2013/7/12 15:50	1.14	5	2	0.80	0.09	-0.08	0.04	-0.03	0.04	-0.06	0.06
GEM-5	2013/7/12 15:50-2013/7/13 16:00	0.91	6	1	0.91	0.09	-0.06	0.04	-0.04	0.04	-0.07	0.06
GEM-6	2013/7/13 16:00-2013/7/14 15:40	1.32	4	2	0.58	0.09	-0.06	0.04	-0.06	0.04	-0.05	0.06
GEM-7	2013/7/14 15:40-2013/7/15 17:10	1.37	2	2	0.58	0.09	-0.08	0.04	-0.05	0.04	-0.04	0.06
GEM-8	2013/7/15 17:15-2013/7/16 17:30	1.24	2	2	0.08	0.09	-0.04	0.04	-0.04	0.04	-0.04	0.06
GEM-9	2013/7/16 17:30-2013/7/17 18:10	1.57	5	1	-0.34	0.09	-0.11	0.04	-0.04	0.04	-0.06	0.06
GEM-10	2013/7/17 18:10-2013/7/18 18:35	1.30	8	1	-0.01	0.09	-0.08	0.04	-0.05	0.04	-0.04	0.06

Table S3. A statistical summary of reported litterfall Hg deposition fluxes and estimated annual litterfall Hg deposition of Hg over the world.

Region	Litterfall Hg deposition flux ($\mu g\ m^{-2}\ yr^{-1}$)			Forest area (km^2)	Estimated litterfall deposition ($Mg\ yr^{-1}$)	Reference
	Range	Median	N			
Asia	20.9-220	37.5±76.1	6	5,775,220	217	(Wang et al., 2009;Fu et al., 2010a;Fu et al., 2010b;Zhou et al., 2013)
North America	3.8-30.9	13.9±5.6	47	6,847,010	95	(Lindberg, 1996;Rea et al., 1996;Grigal et al., 2000;St Louis et al., 2001;Sheehan et al., 2006;Demers et al., 2007;Bushey et al., 2008;Fisher and Wolfe, 2012;Juillerat et al., 2012;Risch et al., 2012;Benoit et al., 2013)
Europe (including Russia)	2.7-25.2	14.2±8.9	6	10,156,300	144	(Iverfeldt, 1991;Munthe et al., 1995;Lee et al., 2000;Schwesig and Matzner, 2000;Lindberg et al., 2007;Larssen et al., 2008)
South America	43.0-184	60.0±49.0	9	9,436,410	566	(Roulet et al., 1998;Fostier et al., 2003;Mélières et al., 2003;Silva-Filho et al., 2006;Teixeira et al., 2012)
Africa				6,164,310	159*	Lack of observational data
Oceania				1,951,370	50*	Lack of observational data
Global total					**1232**	

(Estimated litterfall deposition: * indicates the values were calculated using the global median litterfall Hg deposition flux and the forest area in these regions)

Table S4. A statistical summary of reported throughfall Hg deposition fluxes and estimated annual throughfall Hg deposition of Hg over the world

Region	Throughfall Hg deposition flux (μg m^{-2} yr^{-1})			Forest area (km^2)	Estimated throughfall deposition (mg yr^{-1})	Reference
	Range	Median	N			
Asia	10.5-71.3	36.8±29.9	4	5,775,220	213	(Wan et al., 2009;Wang et al., 2009;Fu et al., 2010a;Fu et al., 2010b)
North America	3.8-30.9	11.8±3.3	10	6,847,010	81	(Lindberg, 1996;Rea et al., 1996;Grigal et al., 2000;St Louis et al., 2001;Sheehan et al., 2006;Choi et al., 2008;Fisher and Wolfe, 2012)
Europe (including Russia)	6.8-39.0	15.2±10.7	8	10,156,300	154	(Iverfeldt, 1991;Munthe et al., 1995;Lee et al., 2000;Schwesig and Matzner, 2000;Lindberg et al., 2007;Larssen et al., 2008)
South America	72	72	1	9,436,410	679	(Fostier et al., 2000)
Africa				6,164,310	160*	Lack of observational data
Oceania				1,951,370	51*	Lack of observational data
Global total					**1338**	

(Estimated throughfall deposition: * indicates the values were calculated using the global median throughfall Hg deposition flux and the forest area in these regions)

Table S5. A statistical summary of reported forest soil emission fluxes and estimated annual forest soil emission fluxes over the world

Region	Forest soil Hg emission flux ($\mu g\ m^{-2}\ yr^{-1}$)			Forest area (km^2)	Estimated forest soil emission ($Mg\ yr^{-1}$)	Reference
	Range	Median	N			
Asia	3.3-81.2	30.4±23.5	11	5,775,220	176	(Wang et al., 2006;Fu et al., 2008;Fu et al., 2012;Ma et al., 2013)
North America	-1.3-45.9	9.2±11.4	15	6,847,010	63	(Carpi and Lindberg, 1998;Poissant and Casimir, 1998;Zhang et al., 2001;Nacht and Gustin, 2004;Schroeder et al., 2005;Kuiken et al., 2008a;Kuiken et al., 2008b;Choi and Holsen, 2009)
Europe (including Russia)	-0.1-9.6	2.4±4.3	4	10,156,300	24	(Schroeder et al., 1989;Xiao et al., 1991;Ferrara et al., 1997;Lindberg et al., 1998)
South America	6.0	6.0	1	9,436,410	57	(Carpi et al., 2014)
Africa				6,164,310	47*	Lack of observational data
Oceania				1,951,370	15*	Lack of observational data
Global total					**381**	

(Estimated forest soil emission: * indicates the values were calculated using the global median forest soil Hg emission flux and the forest area in these regions)

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