

Supporting information

Isotopic compositions of atmospheric total gaseous mercury in ten Chinese cities and implications for land surface emissions

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Figure S1 Locations of the ten urban and one rural sites (black dot) in China investigated in the present study and the gridded surface Hg^0 emission inventory in China in 2013 simulated by Wang et al.(2016). Black square surrounding each sampling site represent the domain of $1^\circ \times 1^\circ$ in size that was used to extract the regional soil emission fluxes for the sampling site in the present study.

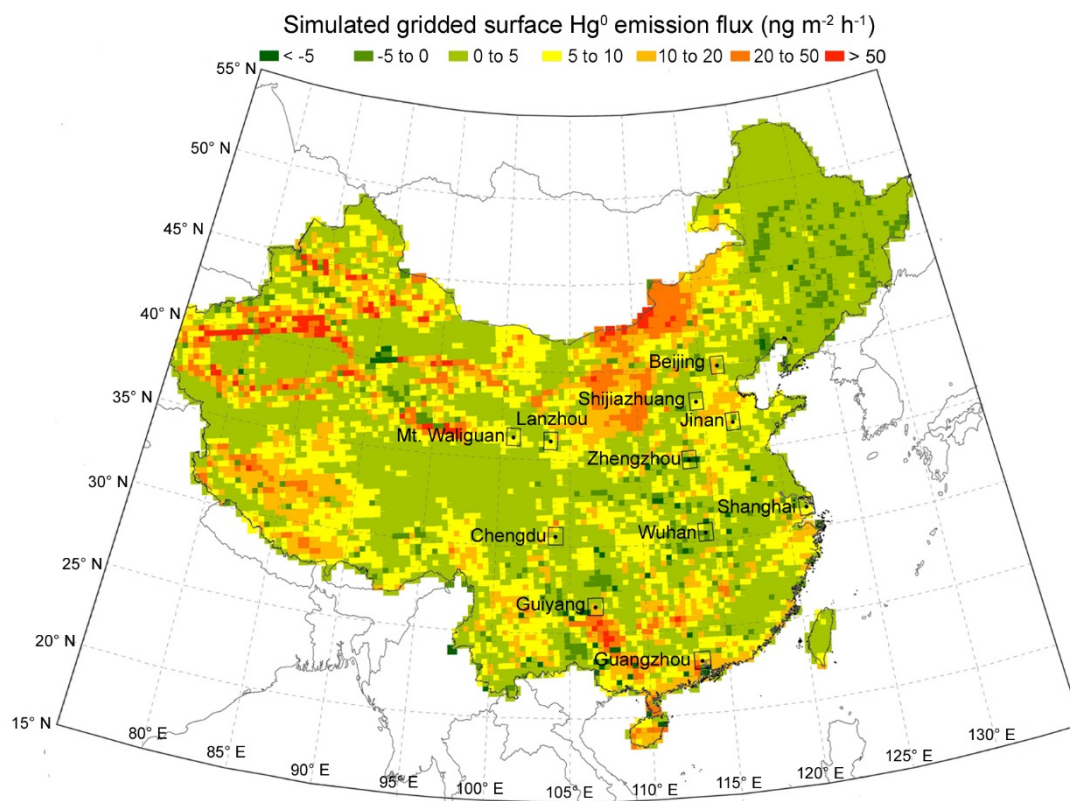


Figure S2 Distribution of the land cover types in the ten Chinese cities in 2018. Land cover type data are from Data Center for Resources and Environmental Science, Chinese Academy of Science (RESDC, <http://www.resdc.cn>) and in a spatial resolution of 1 km.

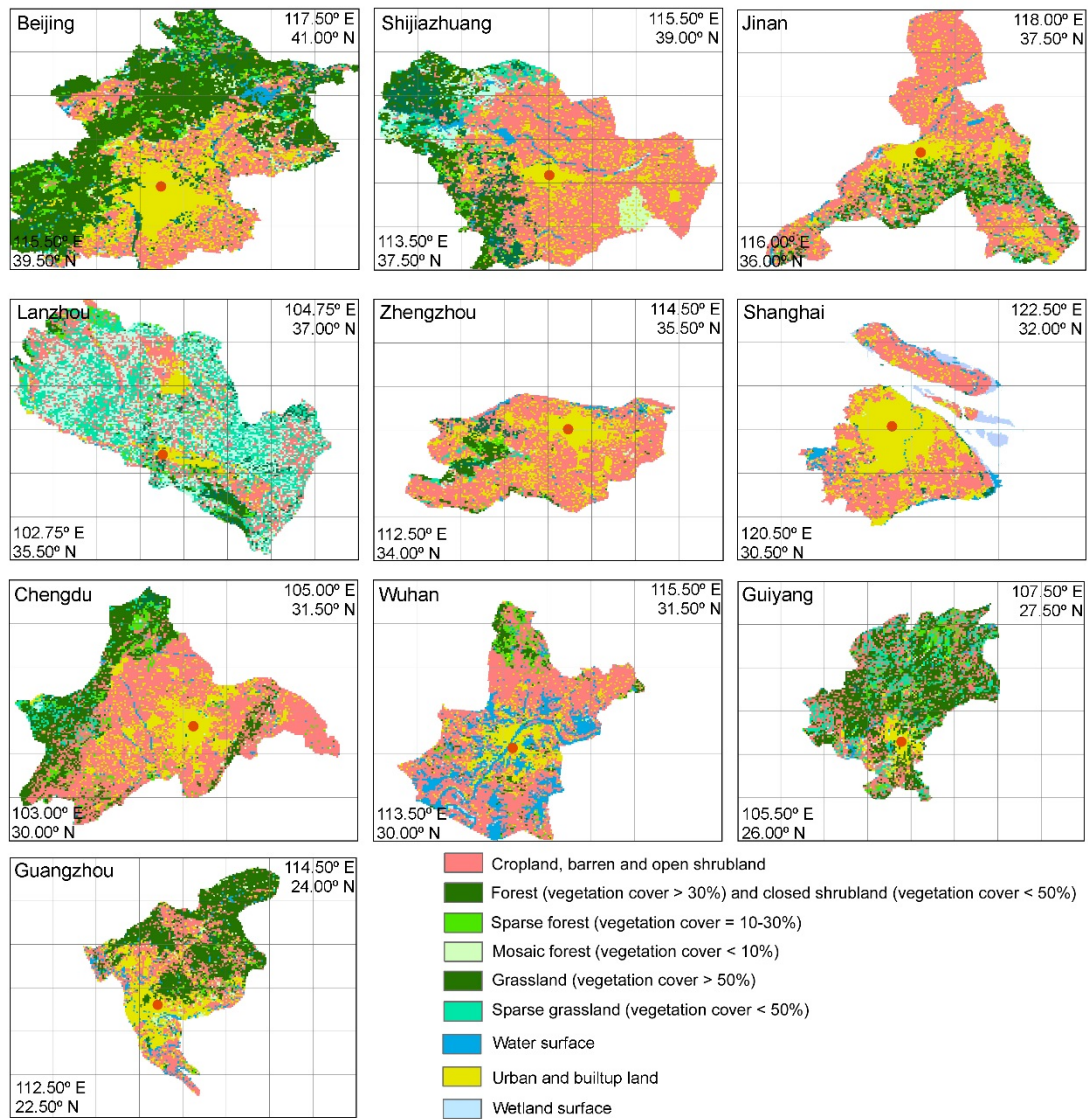


Figure S3 A diagram showing the field sampling of atmospheric total gaseous mercury (TGM)

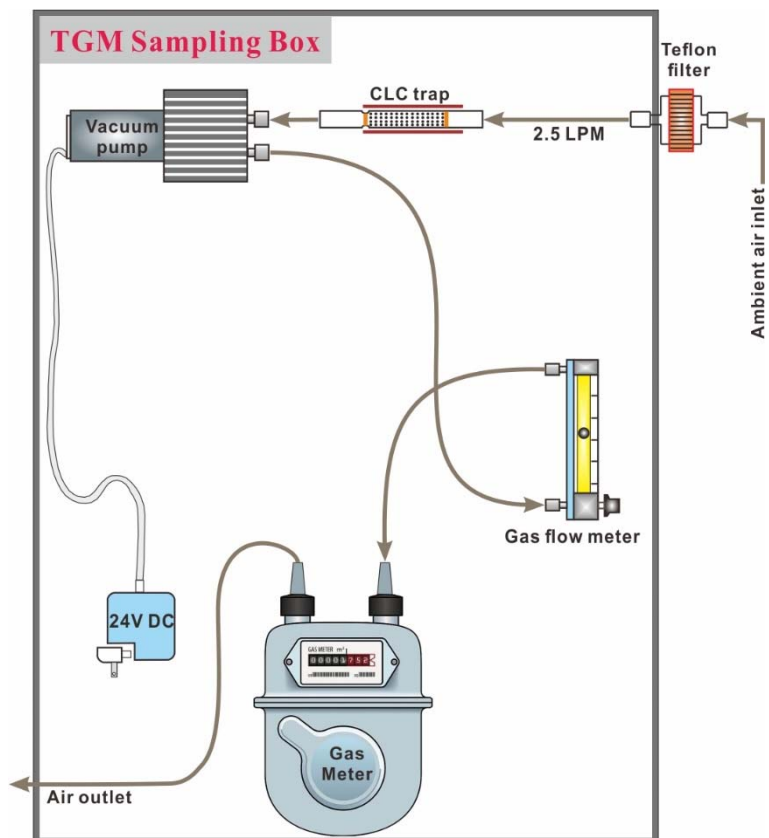


Figure S4 Daily GEM $\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ at the inlet and outlet of flux chamber, and estimated $\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ signatures of GEM emitted from soils in Guiyang and Wuhan based on binary mixing models. Lines are used to connect the daily isotopic compositions of GEM at the inlet and outlet of flux chamber and GEM emitted from soils.

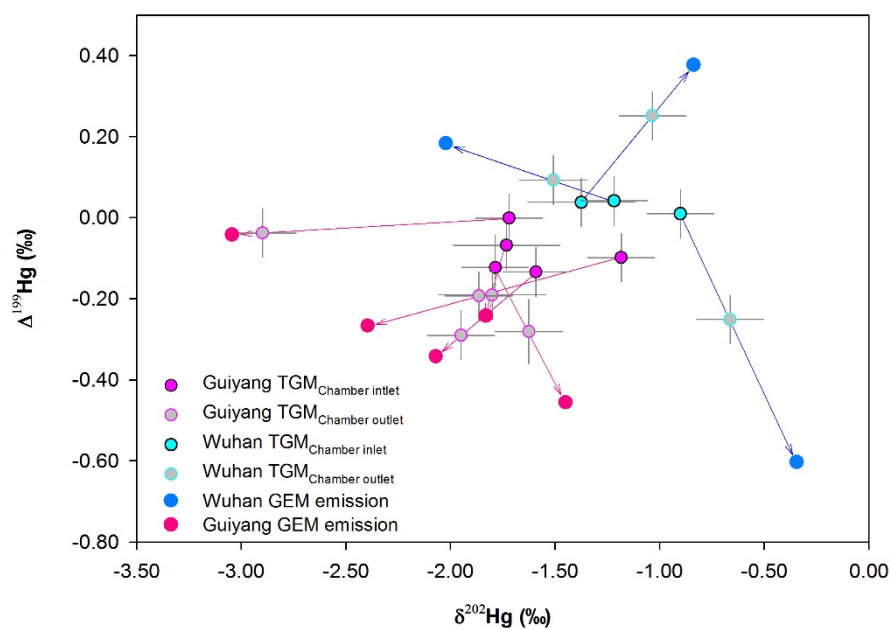


Figure S5 Summertime/wintertime concentration ratios of O₃, normalized difference vegetation index (NDVI), and CO. Dotted lines represent the summertime/wintertime ratio of 1.

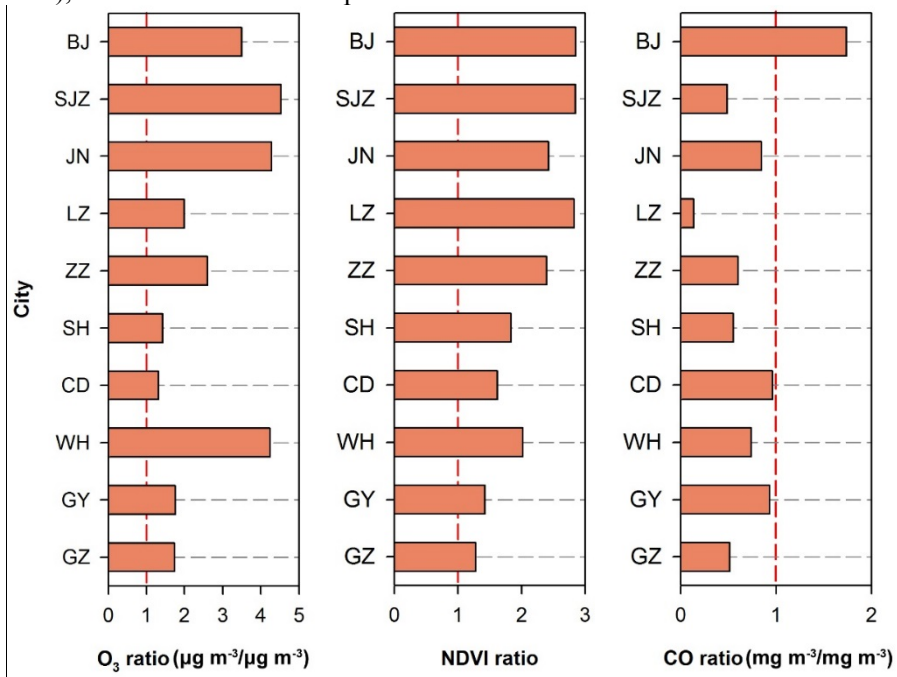


Figure S6 Wind rose diagrams of the wintertime and summertime sampling period in the ten cities

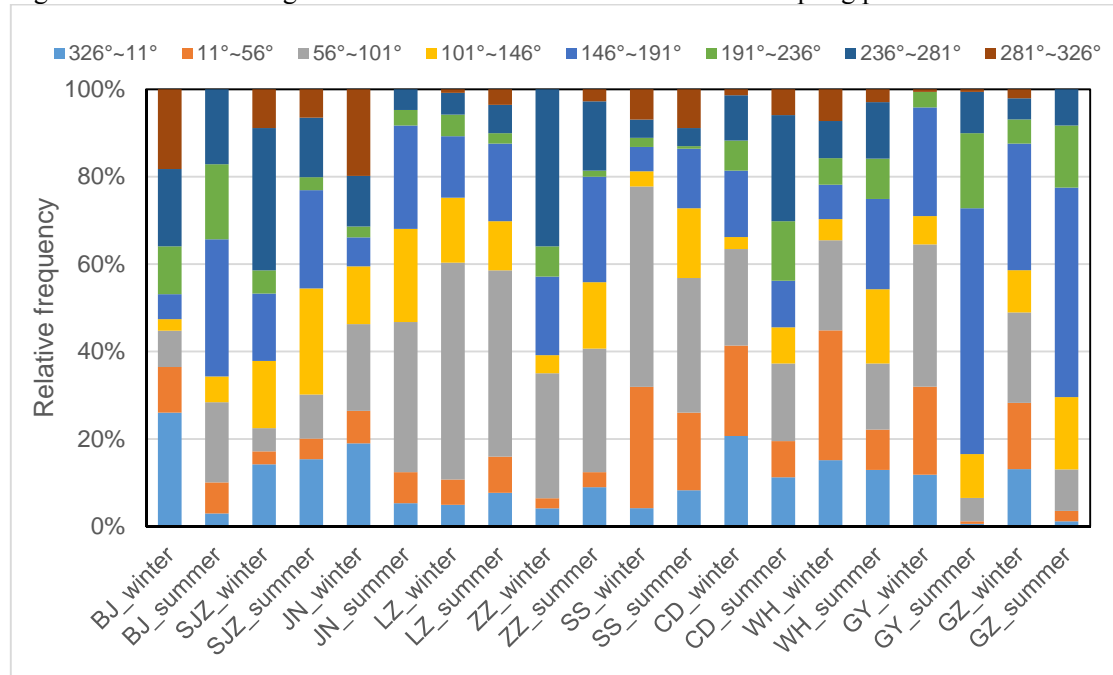


Figure S7 Effect of the soil Hg^0 (GEM) emissions on the seasonal variations in TGM $\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ in the ten Chinese cities. (A) Seasonal amplitude of $\delta^{202}\text{Hg}$ values ($\delta^{202}\text{Hg}_{\text{summer}} - \delta^{202}\text{Hg}_{\text{winter}}$) versus seasonal amplitude of simulated soil Hg^0 emissions flux $((\text{flux}_{\text{July}} - \text{flux}_{\text{January}}) / \text{flux}_{\text{July}})$. (B) Seasonal amplitude of $\Delta^{199}\text{Hg}$ values ($\Delta^{199}\text{Hg}_{\text{summer}} - \Delta^{199}\text{Hg}_{\text{winter}}$) versus seasonal amplitude of simulated soil Hg^0 emissions flux $((\text{flux}_{\text{July}} - \text{flux}_{\text{January}}) / \text{flux}_{\text{July}})$. The data in Zhengzhou (unfilled red circle) was excluded from Figure S6A because an unique deposition pattern of GEM was simulated in winter of this city (Table S5), which might confound the effect of soil GEM emissions on the seasonal variations in TGM $\delta^{202}\text{Hg}$.

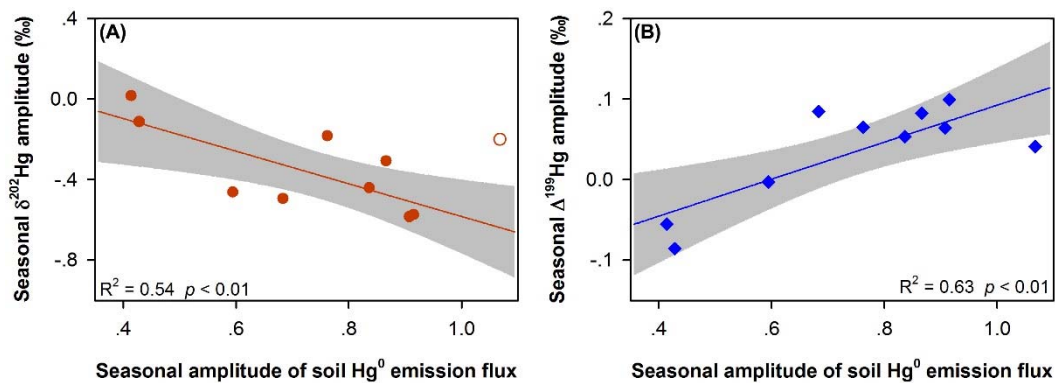


Figure S8 A York bivariate linear regression analysis between (A) daily atmospheric TGM $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$ values in the ten Chinese cities and (B) daily GEM $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$ emitted from cropland soils in Guiyang and Wuhan, China. Error bars are 2σ analytical uncertainties of measured atmospheric TGM $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$ and estimated soil emitted GEM $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$.

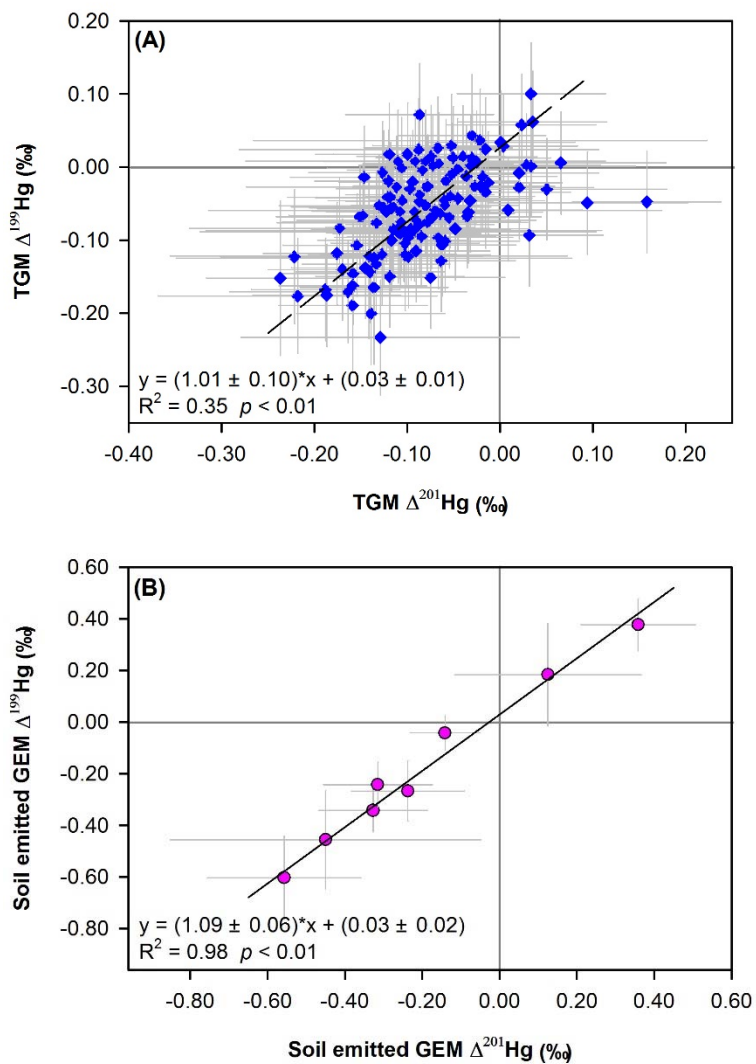


Table S1 Site location and sampling period in the present study as well as the site information and fraction of land cover types in the ten Chinese cities and Mt. Waliguan

Site	Long (°E)	Lat (°N)	Altitude (m a.s.l.)	Site category	Site information		Fraction of land cover types (%)			Season	Study period
					Geographical Location	Population (million)	Croplands, open forests, open shrublands, open grassland, and barren lands ^a	Urban and build-up lands ^b	Vegetated lands ^c		
Beijing	116.370	39.975	10	Urban site	Northern China	21.54	36.4	35.2	26.3	Winter	5 to 13 Jan 2018
										Summer	27 Jul to 3 Aug 2018
Shijiazhuang	114.505	38.043	80	Urban site	Northern China	10.95	64.2	21.3	11.1	Winter	5 to 12 Jan 2018
										Summer	29 Jun to 6 Jul 2018
Jinan	117.055	36.673	50	Urban site	Eastern China	7.46	65.6	20.8	11.4	Winter	7 to 12 Jan 2018
										Summer	29 Jun to 6 Jul 2018
Lanzhou	103.631	36.103	1483	Urban site	Northwestern China	3.75	82.3	6.8	8.9	Winter	7 to 12 Jan 2018
										Summer	27 Jul to 3 Aug 2018
Zhengzhou	113.618	34.752	110	Urban site	Central China	10.14	67.6	25.5	4.2	Winter	5 to 14 Jan 2018
										Summer	29 Jun to 5 Jul 2018
Shanghai	121.390	31.266	25	Urban site	Eastern China	23.24	49.7	41.1	1.1	Winter	20 to 26 Jan 2018
										Summer	27 Jul to 3 Aug 2018
Chengdu	104.059	30.657	570	Urban site	Southwestern China	16.33	68.3	22	8.9	Winter	19 to 25 Jan 2018
										Summer	27 Jul to 3 Aug 2018
Wuhan	114.302	30.533	80	Urban site	Central China	9.08	57.9	15.9	3.5	Winter	19 to 26 Jan 2018
										Summer	27 Jul to 9 Aug 2018
Guiyang	106.698	26.570	1055	Urban site	Southwestern China	4.88	47°	4.9	47.1	Winter	18 to 25 Jan 2018
										Summer	29 Jun to 6 Jul 2018
Guangzhou	113.358	23.149	70	Urban site	Southern China	14.90	35.6	32	21.1	Winter	19 to 25 Jan 2018
										Summer	29 Jun to 6 Jul 2018
Mt. Waliguan	100.90	36.28	3816	Rural site	Northwestern China	----	70.7	7.3	17.0	Winter	19 Nov 2014 to 19 Feb 2015

Table S2 Mean gaseous elemental mercury (GEM) and gaseous oxidized mercury (GOM) concentrations reported in Guiyang, Beijing, and Shanghai China as well as in Reno, USA.

Site	Long (°E)	Lat (°N)	Altitude (m a.s.l.)	Study period	Mean GEM (± 1 sd) (ng m ⁻³)	Mean GOM (± 1 sd) (pg m ⁻³)	Mean GOM fraction in TGM	Daily or weekly GOM fraction in TGM	Reference
Guiyang, China ^a	106.72	26.57	1041	Aug to Dec 2009	9.72 \pm 10.2	35.7 \pm 43.9	0.37%	0.04 to 1.58% (daily)	(Fu et al., 2011)
Beijing, China ^a	116.338	40.007		Sep 2015 to Jul 2016	4.70 \pm 3.53	18.5 \pm 22.3	0.39%	Data are not available	(Zhang et al., 2019)
Shanghai, China ^a	120.998	31.098	17	Jun to Dec 2014	4.19 \pm 9.13	21 \pm 100	0.50%	Data are not available	(Duan et al., 2017)
Reno, USA ^b	-119.805	39.537	1377	Feb to Jun 2019	1.24 \pm 0.11	33.3 \pm 12.6	2.7%	1.4 to 5.2% (weekly)	(Gustin et al., 2019)

a) GOM concentrations were measured using the Tekran 2537/1130/1135 system.

b) GOM concentrations were measured using the cation exchange membrane method on weekly basis.

Table S3 Seal blanks and breakthrough of CLC traps as well as recovery and Hg isotopic compositions of lichen CRM (BCR 482), standard addition of NIST 3133 to CLC traps, and international standard NIST SRM 3177 over analytical sessions measured in this study. The 2 σ represent the 2 times of the standard error (2 sd) of the mean isotopic compositions measured over analytical sessions.

Test category	Data source	n	Concentration (ng), recovery (%), breakthrough (%)			Isotopic composition (‰)																	
			Range	Mean	1 σ	$\delta^{199}\text{Hg}$	2 σ	$\delta^{200}\text{Hg}$	2 σ	$\delta^{201}\text{Hg}$	2 σ	$\delta^{202}\text{Hg}$	2 σ	$\delta^{204}\text{Hg}$	2 σ	$\Delta^{199}\text{Hg}$	2 σ	$\Delta^{200}\text{Hg}$	2 σ	$\Delta^{201}\text{Hg}$	2 σ	$\Delta^{204}\text{Hg}$	2 σ
Seal blank test ^a	This study	27	0.06 to 0.43 ng	0.20 ng	0.09 ng	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Breakthrough test ^b	This study	10	0.4 to 3.3%	1.1%	0.9%	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
BCR 482 ^c	This study	6	86.0 to 97.2%	92.5%	3.9%	-0.99	0.11	-0.75	0.18	-1.86	0.19	-1.68	0.21	-2.52	0.48	-0.57	0.11	0.09	0.11	-0.59	0.13	-0.03	0.17
	Literature (Estrade et al., 2010; Blum and Johnson, 2017)					-1.05 to -0.97		-0.68 to -0.74		-1.85 to -1.71		-1.67 to -1.48				-0.67 to -0.56		0.06 to 0.08		-0.68 to -0.60		-0.10	
Standard addition test ^d	This study	11	84.1 to 104.8%	93.2%	11.8%	0.01	0.05	-0.03	0.11	-0.04	0.14	-0.06	0.16	-0.19	0.29	0.02	0.07	0.00	0.07	0.01	0.08	-0.05	0.08
	Expected			100%		0.00		0.00		0.00		0.00		0.00		0.00		0.00		0.00		0.00	
NIST 3177 test ^e	This study	30	--	--	--	-0.13	0.06	-0.27	0.06	-0.43	0.09	-0.54	0.09	-0.79	0.27	0.01	0.07	0.00	0.06	-0.02	0.08	0.03	0.25
	Literature (Sun et al., 2016)					-0.14		-0.26		-0.42		-0.52				0.00		0.00		-0.02			

a): Tested by combustion of sealed field CLC traps (containing 0.5 g CLC) prepared before field sampling. The isotopic composition of these blanks were not measured due to their extremely Hg concentrations in trap solutions ($\leq 0.043 \text{ ng mL}^{-1}$).

b): Tested by in the laboratory at the flow rate of 2.5 L min^{-1} over 24 hours following the method by Fu et al.(2014).

c): Tested by combustion of Lichen CRM (BCR 482) using the double-stage combustion method.

d): Tested by extraction of Hg from diluted NIST SRM 3133 solutions (a volume of 2.5 L and Hg concentrations of 2-10 ng L^{-1}) onto CLC traps using Hg-free air at a purging flow rate of 2.5 L min^{-1} within 1.5 h, followed by preconcentration of Hg on CLC traps to trap solutions using the combustion method.

e): Tested by direct measurements of the Hg isotopic compositions in the international standard NIST SRM 3133 solutions (concentration = 0.5 or 1.0 ng mL^{-1}). Recoveries of these test were not available.

Table S4 Daily atmospheric TGM concentrations and isotopic compositions measured in this study. 2σ represents the analytical uncertainties of the isotopic compositions. δ²⁰⁴Hg and Δ¹⁹⁹Hg were not measured for some samples (--) because of the limitations in instrumental collector designs of Neptune.

Sample ID	Site	Season	Sampling period	Volume sampled (m ³)	TGM conc. (ng m ⁻³)	δ ¹⁹⁹ Hg (‰)		δ ²⁰⁰ Hg (‰)		δ ²⁰¹ Hg (‰)		δ ²⁰² Hg (‰)		δ ²⁰⁴ Hg (‰)		Δ ¹⁹⁹ Hg (‰)		Δ ²⁰⁰ Hg (‰)		Δ ²⁰¹ Hg (‰)		Δ ²⁰⁴ Hg (‰)	
						Value	2σ	Value	2σ	Value	2σ	Value	2σ	Value	2σ	Value	2σ	Value	2σ	Value	2σ	Value	2σ
BJ-01	Beijing	Winter	2018/01/05 8:00 to 2018/01/06 8:00	2.69	2.04	-0.12	0.08	-0.14	0.11	-0.23	0.14	-0.17	0.18	-0.26	0.29	-0.07	0.07	-0.06	0.07	-0.11	0.13	-0.01	0.12
BJ-02			2018/01/06 8:00 to 2018/01/07 8:00	3.13	2.46	-0.05	0.05	-0.12	0.11	-0.25	0.14	-0.23	0.16	-0.25	0.29	0.01	0.07	0.00	0.07	-0.08	0.19	0.09	0.08
BJ-03			2018/01/08 8:00 to 2018/01/09 8:00	3.13	1.06	-0.17	0.14	-0.10	0.11	-0.10	0.14	0.07	0.20	0.03	0.56	-0.19	0.09	-0.14	0.07	-0.16	0.08	-0.07	0.26
BJ-04			2018/01/09 8:00 to 2018/01/10 8:00	2.99	1.54	-0.13	0.08	0.07	0.11	-0.04	0.14	0.16	0.18	0.36	0.29	-0.17	0.07	-0.01	0.07	-0.16	0.13	0.13	0.12
BJ-05			2018/01/10 8:00 to 2018/01/11 8:00	2.54	1.54	-0.04	0.12	0.04	0.28	-0.06	0.14	0.11	0.18	0.31	0.30	-0.07	0.07	-0.02	0.19	-0.15	0.08	0.14	0.08
BJ-06			2018/01/11 8:00 to 2018/01/12 8:00	3.24	1.68	-0.10	0.12	-0.10	0.28	-0.12	0.14	-0.03	0.18	0.12	0.30	-0.10	0.07	-0.08	0.19	-0.10	0.08	0.18	0.08
BJ-07			2018/01/12 8:00 to 2018/01/13 8:00	3.33	2.87	-0.11	0.05	-0.29	0.11	-0.49	0.14	-0.53	0.16	-0.80	0.29	0.02	0.07	-0.02	0.07	-0.09	0.19	0.00	0.08
BJ-08		Summer	2018/07/27 8:00 to 2018/07/28 8:00	2.67	2.68	-0.12	0.09	-0.20	0.11	-0.43	0.14	-0.49	0.16	-0.64	0.29	0.00	0.07	0.04	0.07	-0.07	0.08	0.09	0.09
BJ-09			2018/07/28 8:00 to 2018/07/29 8:00	2.39	2.58	-0.19	0.09	-0.26	0.11	-0.56	0.14	-0.59	0.16	-0.88	0.29	-0.04	0.07	0.04	0.07	-0.12	0.08	0.00	0.09
BJ-10			2018/07/29 8:00 to 2018/07/30 8:00	2.57	2.96	-0.11	0.05	-0.13	0.11	-0.30	0.14	-0.28	0.16	-0.45	0.29	-0.04	0.07	0.01	0.07	-0.09	0.08	-0.03	0.08
BJ-11			2018/07/30 8:00 to 2018/07/31 8:00	2.93	2.68	-0.03	0.05	-0.15	0.11	-0.40	0.14	-0.41	0.16	-0.62	0.29	0.07	0.07	0.06	0.07	-0.09	0.08	-0.01	0.08
BJ-12			2018/07/31 8:00 to 2018/08/01 8:00	3.31	3.13	-0.05	0.07	-0.12	0.11	-0.28	0.14	-0.24	0.16	-0.43	0.29	0.01	0.07	0.01	0.07	-0.09	0.08	-0.06	0.08
BJ-13			2018/08/01 8:00 to 2018/08/02 8:00	2.83	3.24	-0.10	0.07	-0.21	0.11	-0.41	0.14	-0.40	0.16	-0.63	0.29	0.00	0.07	0.00	0.07	-0.11	0.08	-0.04	0.08
BJ-14			2018/08/02 8:00 to 2018/08/03 8:00	3.02	2.67	-0.08	0.05	-0.18	0.11	-0.39	0.14	-0.37	0.16	-0.54	0.29	0.01	0.07	0.00	0.07	-0.11	0.08	0.01	0.08
SJZ-01	Shijiazhuang	Winter	2018/01/05 8:00 to 2018/01/06 8:00	3.70	2.67	-0.36	0.06	-0.35	0.11	-0.78	0.14	-0.75	0.16	--	--	-0.18	0.08	0.02	0.08	-0.22	0.15	--	--
SJZ-02			2018/01/06 8:00 to 2018/01/07 8:00	3.94	2.69	-0.39	0.06	-0.27	0.11	-0.60	0.14	-0.63	0.16	--	--	-0.23	0.08	0.05	0.08	-0.13	0.15	--	--
SJZ-03			2018/01/08 8:00 to 2018/01/09 8:00	4.05	1.88	-0.17	0.06	-0.15	0.11	-0.26	0.14	-0.17	0.16	--	--	-0.13	0.07	-0.06	0.09	-0.13	0.08	--	--
SJZ-04			2018/01/09 8:00 to 2018/01/10 8:00	3.97	1.21	-0.03	0.06	0.18	0.11	0.17	0.17	0.45	0.16	--	--	-0.14	0.07	-0.04	0.07	-0.17	0.08	--	--
SJZ-05			2018/01/10 8:00 to 2018/01/11 8:00	3.85	1.18	-0.23	0.06	-0.14	0.11	-0.22	0.17	-0.11	0.16	--	--	-0.20	0.07	-0.08	0.07	-0.14	0.08	--	--
SJZ-06			2018/01/11 8:00 to 2018/01/12 8:00	4.28	2.02	-0.20	0.05	-0.15	0.11	-0.36	0.14	-0.31	0.16	--	--	-0.12	0.07	0.01	0.07	-0.13	0.08	--	--
SJZ-07		Summer	2018/06/29 8:00 to 2018/06/30 8:00	3.74	2.46	-0.34	0.05	-0.42	0.11	-0.62	0.14	-0.73	0.16	--	--	-0.15	0.07	-0.05	0.07	-0.07	0.08	--	--
SJZ-08			2018/06/30 8:00 to 2018/07/01 8:00	3.33	2.74	-0.31	0.15	-0.42	0.19	-0.70	0.31	-0.74	0.16	--	--	-0.12	0.11	-0.05	0.13	-0.14	0.21	--	--
SJZ-09			2018/07/01 8:00 to 2018/07/02 8:00	3.26	2.21	-0.26	0.10	-0.39	0.13	-0.71	0.26	-0.71	0.16	--	--	-0.08	0.07	-0.03	0.07	-0.17	0.16	--	--
SJZ-10			2018/07/02 8:00 to 2018/07/03 8:00	3.66	4.33	-0.35	0.10	-0.65	0.13	-1.08	0.26	-1.29	0.16	--	--	-0.03	0.07	0.00	0.07	-0.11	0.16	--	--
SJZ-11			2018/07/03 8:00 to 2018/07/04 8:00	3.02	3.74	-0.26	0.07	-0.47	0.15	-0.76	0.24	-0.90	0.28	--	--	-0.03	0.07	-0.02	0.07	-0.08	0.08	--	--
SJZ-12			2018/07/04 8:00 to 2018/07/05 8:00	2.55	4.27	-0.21	0.07	-0.39	0.11	-0.70	0.14	-0.82	0.16	--	--	0.00	0.07	0.03	0.07	-0.08	0.08	--	--
SJZ-13			2018/07/05 8:00 to 2018/07/06 8:00	3.16	3.64	-0.22	0.07	-0.30	0.11	-0.55	0.14	-0.61	0.16	--	--	-0.06	0.07	0.01	0.07	-0.09	0.08	--	--

ZZ-08	Summer	2018/01/12 8:00 to 2018/01/13 8:00	3.38	3.92	-0.23	0.06	-0.34	0.11	-0.56	0.14	-0.81	0.19	--	--	-0.03	0.07	0.07	0.07	0.05	0.13	--	--
ZZ-09		2018/06/29 8:00 to 2018/06/30 8:00	2.27	3.59	-0.15	0.05	-0.29	0.11	-0.55	0.14	-0.64	0.16	--	--	0.01	0.07	0.03	0.07	-0.07	0.08	--	--
ZZ-10		2018/06/30 8:00 to 2018/07/01 8:00	2.82	5.05	-0.14	0.05	-0.24	0.11	-0.46	0.14	-0.57	0.16	--	--	0.01	0.07	0.05	0.07	-0.03	0.08	--	--
ZZ-11		2018/07/01 8:00 to 2018/07/02 8:00	2.34	3.70	-0.19	0.05	-0.29	0.11	-0.53	0.14	-0.57	0.16	--	--	-0.05	0.07	-0.01	0.07	-0.11	0.08	--	--
ZZ-12		2018/07/02 8:00 to 2018/07/03 8:00	2.62	4.08	-0.19	0.06	-0.21	0.11	-0.39	0.14	-0.47	0.16	--	--	-0.07	0.07	0.03	0.07	-0.04	0.08	--	--
ZZ-13		2018/07/03 8:00 to 2018/07/04 8:00	2.96	3.27	-0.25	0.06	-0.26	0.11	-0.44	0.14	-0.80	0.16	--	--	-0.05	0.07	0.13	0.07	0.16	0.08	--	--
ZZ-14		2018/07/04 8:00 to 2018/07/05 8:00	3.60	5.13	-0.26	0.05	-0.30	0.11	-0.52	0.14	-0.61	0.16	--	--	-0.10	0.07	0.01	0.07	-0.06	0.08	--	--
SH-01	Winter	2018/01/20 8:00 to 2018/01/21 8:00	2.64	2.96	-0.19	0.05	-0.22	0.11	-0.41	0.14	-0.44	0.16	--	--	-0.08	0.07	0.00	0.07	-0.08	0.11	--	--
SH-02		2018/01/21 8:00 to 2018/01/22 8:00	2.67	1.66	-0.21	0.05	-0.13	0.11	-0.30	0.14	-0.15	0.16	-0.28	0.29	-0.17	0.07	-0.06	0.07	-0.19	0.08	-0.05	0.08
SH-03		2018/01/22 8:00 to 2018/01/23 8:00	2.65	3.03	-0.16	0.05	-0.23	0.11	-0.44	0.14	-0.41	0.16	--	--	-0.05	0.07	-0.02	0.07	-0.13	0.11	--	--
SH-04		2018/01/23 8:00 to 2018/01/24 8:00	2.69	1.75	-0.02	0.14	-0.01	0.16	-0.01	0.14	0.15	0.16	0.23	0.29	-0.06	0.11	-0.09	0.08	-0.13	0.08	0.01	0.51
SH-05		2018/01/24 8:00 to 2018/01/25 8:00	2.61	1.80	-0.10	0.14	0.01	0.16	0.05	0.14	0.25	0.16	0.55	0.29	-0.16	0.11	-0.12	0.08	-0.14	0.08	0.18	0.51
SH-06		2018/01/25 8:00 to 2018/01/26 8:00	2.62	1.52	-0.11	0.14	0.02	0.16	-0.11	0.14	0.17	0.16	0.35	0.29	-0.15	0.11	-0.07	0.08	-0.24	0.08	0.10	0.51
SH-07	Summer	2018/07/27 8:00 to 2018/07/28 8:00	2.40	3.13	-0.24	0.05	-0.43	0.11	-0.83	0.14	-0.91	0.17	-1.27	0.29	-0.01	0.07	0.03	0.07	-0.15	0.08	0.10	0.30
SH-08		2018/07/28 8:00 to 2018/07/29 8:00	2.36	3.28	-0.23	0.10	-0.35	0.11	-0.64	0.23	-0.70	0.16	-0.98	0.53	-0.05	0.07	0.00	0.07	-0.12	0.11	0.06	0.29
SH-09		2018/07/29 8:00 to 2018/07/30 8:00	2.46	2.47	-0.10	0.05	-0.10	0.11	-0.39	0.14	-0.35	0.16	-0.55	0.38	-0.01	0.07	0.07	0.07	-0.13	0.08	-0.03	0.40
SH-10		2018/07/30 8:00 to 2018/07/31 8:00	2.73	2.63	-0.21	0.10	-0.26	0.11	-0.53	0.23	-0.53	0.16	-0.73	0.53	-0.08	0.07	0.01	0.07	-0.13	0.11	0.06	0.29
SH-11		2018/07/31 8:00 to 2018/08/01 8:00	2.39	2.35	-0.09	0.05	-0.17	0.11	-0.41	0.14	-0.46	0.16	-0.72	0.38	0.03	0.07	0.07	0.07	-0.07	0.08	-0.03	0.40
SH-12		2018/08/01 8:00 to 2018/08/02 8:00	2.77	2.02	-0.18	0.05	-0.22	0.11	-0.41	0.15	-0.46	0.16	-0.77	0.29	-0.06	0.07	0.01	0.07	-0.06	0.08	-0.08	0.28
SH-13		2018/08/02 8:00 to 2018/08/03 8:00	3.08	2.01	-0.14	0.05	-0.25	0.11	-0.48	0.15	-0.55	0.16	-0.79	0.29	0.00	0.07	0.03	0.07	-0.07	0.08	0.02	0.28
CD-01	Winter	2018/01/18 8:00 to 2018/01/19 8:00	1.08	4.76	-0.11	0.07	-0.16	0.11	-0.29	0.17	-0.37	0.16	--	--	-0.01	0.07	0.02	0.07	-0.02	0.08	--	--
CD-02		2018/01/19 8:00 to 2018/01/20 8:00	2.01	2.29	0.10	0.07	0.26	0.11	0.48	0.17	0.63	0.16	--	--	-0.06	0.07	-0.06	0.07	0.01	0.08	--	--
CD-03		2018/01/20 8:00 to 2018/01/21 8:00	2.80	4.23	-0.10	0.05	-0.22	0.11	-0.37	0.14	-0.45	0.16	--	--	0.01	0.07	0.01	0.07	-0.03	0.08	--	--
CD-04		2018/01/21 8:00 to 2018/01/22 8:00	2.89	4.70	-0.16	0.05	-0.27	0.11	-0.42	0.14	-0.52	0.16	--	--	-0.03	0.07	-0.01	0.07	-0.03	0.08	--	--
CD-05		2018/01/22 8:00 to 2018/01/23 8:00	2.79	5.37	-0.12	0.05	-0.28	0.11	-0.50	0.14	-0.59	0.16	--	--	0.03	0.07	0.02	0.07	-0.05	0.08	--	--
CD-06		2018/01/23 8:00 to 2018/01/24 8:00	3.14	6.59	-0.19	0.05	-0.40	0.11	-0.65	0.14	-0.87	0.16	--	--	0.03	0.07	0.03	0.07	0.00	0.08	--	--
CD-07		2018/01/24 8:00 to 2018/01/25 8:00	3.06	3.20	-0.32	0.05	-0.49	0.11	-0.79	0.14	-0.98	0.16	--	--	-0.07	0.07	0.00	0.07	-0.05	0.08	--	--
CD-08	Summer	2018/07/27 8:00 to 2018/07/28 8:00	2.78	3.77	-0.26	0.08	-0.53	0.11	-0.74	0.14	-0.96	0.16	-1.35	0.29	-0.02	0.07	-0.04	0.07	-0.01	0.08	0.09	0.10
CD-09		2018/07/28 8:00 to 2018/07/29 8:00	3.02	5.89	-0.29	0.09	-0.52	0.12	-0.76	0.14	-1.04	0.16	-1.56	0.29	-0.03	0.07	0.00	0.08	0.02	0.08	-0.01	0.12
CD-10		2018/07/29 8:00 to 2018/07/30 8:00	3.15	4.44	-0.21	0.09	-0.43	0.12	-0.67	0.14	-0.85	0.16	-1.24	0.29	0.00	0.07	0.00	0.08	-0.03	0.08	0.03	0.12
CD-11		2018/07/30 8:00 to 2018/07/31 8:00	3.22	3.01	-0.24	0.05	-0.47	0.11	-0.68	0.14	-0.99	0.16	-1.46	0.29	0.01	0.07	0.03	0.07	0.07	0.11	0.02	0.08

CD-12			2018/07/31 8:00 to 2018/08/01 8:00	2.52	5.44	-0.19	0.05	-0.33	0.11	-0.53	0.14	-0.75	0.16	-1.19	0.29	0.00	0.07	0.05	0.07	0.03	0.11	-0.06	0.08
CD-13			2018/08/01 8:00 to 2018/08/02 8:00	2.40	5.72	-0.31	0.05	-0.50	0.11	-0.81	0.27	-0.98	0.16	-1.45	0.29	-0.06	0.07	0.00	0.07	-0.07	0.24	0.01	0.11
CD-14			2018/08/02 8:00 to 2018/08/03 8:00	2.92	4.35	-0.17	0.05	-0.38	0.11	-0.63	0.27	-0.81	0.16	-1.18	0.29	0.04	0.07	0.03	0.07	-0.02	0.24	0.03	0.11
WH-01	Wuhan	Winter	2018/01/19 8:00 to 2018/01/20 8:00	3.07	6.54	-0.28	0.09	-0.42	0.18	-0.76	0.16	-0.91	0.31	-1.37	0.31	-0.05	0.07	0.03	0.07	-0.08	0.08	-0.02	0.15
WH-02			2018/01/20 8:00 to 2018/01/21 8:00	3.62	3.44	-0.24	0.09	-0.28	0.18	-0.52	0.16	-0.59	0.31	-0.93	0.31	-0.10	0.07	0.02	0.07	-0.08	0.08	-0.06	0.15
WH-03			2018/01/21 8:00 to 2018/01/22 8:00	2.59	3.89	-0.23	0.05	-0.36	0.11	-0.60	0.16	-0.78	0.16	-1.18	0.29	-0.03	0.08	0.03	0.08	-0.02	0.09	-0.02	0.08
WH-04			2018/01/22 8:00 to 2018/01/23 8:00	2.94	4.80	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
WH-05			2018/01/23 8:00 to 2018/01/24 8:00	3.32	2.61	-0.26	0.05	-0.31	0.11	-0.51	0.16	-0.54	0.16	-0.90	0.29	-0.12	0.08	-0.04	0.08	-0.10	0.09	-0.09	0.08
WH-06			2018/01/24 8:00 to 2018/01/25 8:00	3.31	1.71	-0.19	0.05	-0.31	0.12	-0.40	0.19	-0.38	0.16	-0.52	0.29	-0.09	0.07	-0.12	0.14	-0.11	0.21	0.05	0.15
WH-07			2018/01/25 8:00 to 2018/01/26 8:00	3.35	1.69	-0.21	0.05	-0.29	0.12	-0.38	0.19	-0.33	0.16	-0.31	0.29	-0.12	0.07	-0.12	0.14	-0.14	0.21	0.18	0.15
WH-08		Summer	2018/07/27 8:00 to 2018/07/28 8:00	2.90	3.16	-0.26	0.07	-0.52	0.11	-0.81	0.14	-1.01	0.16	--	--	-0.01	0.07	-0.01	0.07	-0.05	0.08	--	--
WH-09			2018/07/28 8:00 to 2018/07/29 8:00	2.30	3.59	-0.26	0.07	-0.50	0.11	-0.79	0.14	-0.95	0.16	--	--	-0.03	0.07	-0.03	0.07	-0.08	0.08	--	--
WH-10			2018/07/29 8:00 to 2018/07/30 8:00	2.84	4.05	-0.28	0.07	-0.54	0.11	-0.87	0.14	-1.09	0.16	--	--	0.00	0.07	0.01	0.07	-0.05	0.08	--	--
WH-11			2018/07/31 8:00 to 2018/08/01 8:00	2.53	4.41	-0.32	0.05	-0.50	0.11	-0.78	0.14	-0.92	0.16	--	--	-0.09	0.07	-0.03	0.07	-0.09	0.08	--	--
WH-12			2018/08/01 8:00 to 2018/08/02 8:00	2.55	4.21	-0.16	0.05	-0.26	0.11	-0.47	0.14	-0.52	0.16	--	--	-0.03	0.07	0.00	0.07	-0.08	0.08	--	--
WH-13			2018/08/02 8:00 to 2018/08/03 8:00	2.18	4.20	-0.16	0.05	-0.29	0.11	-0.56	0.14	-0.68	0.16	--	--	0.01	0.07	0.05	0.07	-0.05	0.08	--	--
WH-14			2018/08/03 8:00 to 2018/08/04 8:00	2.72	2.98	-0.24	0.05	-0.31	0.11	-0.58	0.14	-0.62	0.16	--	--	-0.09	0.07	0.01	0.07	-0.12	0.08	--	--
WH-15			2018/08/07 8:00 to 2018/08/08 8:00	1.45	4.83	-0.12	0.05	-0.27	0.11	-0.50	0.14	-0.53	0.16	--	--	0.02	0.07	0.00	0.07	-0.10	0.08	--	--
WH-16			2018/08/08 8:00 to 2018/08/09 8:00	3.09	4.53	-0.14	0.05	-0.31	0.11	-0.59	0.14	-0.62	0.16	--	--	0.02	0.07	0.00	0.07	-0.12	0.08	--	--
GY-01	Guiyang	Winter	2018/01/18 8:00 to 2018/01/24 8:00	2.84	9.39	-0.32	0.05	-0.83	0.11	-1.23	0.14	-1.68	0.16	-2.38	0.29	0.10	0.07	0.01	0.09	0.03	0.08	0.12	0.08
GY-02			2018/01/19 8:00 to 2018/01/20 8:00	3.22	5.81	-0.27	0.05	-0.63	0.11	-0.94	0.14	-1.29	0.16	-1.76	0.29	0.06	0.07	0.02	0.09	0.02	0.08	0.16	0.08
GY-03			2018/01/20 8:00 to 2018/01/21 8:00	3.22	5.21	-0.30	0.05	-0.58	0.11	-0.89	0.16	-1.13	0.16	-1.68	0.29	-0.01	0.07	-0.01	0.07	-0.04	0.08	0.01	0.15
GY-04			2018/01/21 8:00 to 2018/01/22 8:00	2.70	5.66	-0.24	0.05	-0.59	0.11	-0.87	0.16	-1.20	0.16	-1.90	0.29	0.06	0.07	0.01	0.07	0.04	0.08	-0.11	0.15
GY-05			2018/01/22 8:00 to 2018/01/23 8:00	2.64	5.70	-0.12	0.08	-0.24	0.13	-0.39	0.23	-0.49	0.23	-0.78	0.29	0.01	0.07	0.00	0.07	-0.03	0.08	-0.05	0.08
GY-06			2018/01/23 8:00 to 2018/01/24 8:00	3.14	4.40	-0.01	0.08	-0.09	0.13	-0.14	0.23	-0.19	0.23	-0.23	0.29	0.03	0.07	0.01	0.07	0.00	0.08	0.05	0.08
GY-07		2018/01/24 8:00 to 2018/01/25 8:00	2.39	2.91	-0.10	0.06	-0.16	0.11	-0.25	0.14	-0.36	0.16	-0.38	0.51	-0.01	0.07	0.01	0.08	0.02	0.09	0.16	0.54	
GY-08		Summer	2018/06/29 8:00 to 2018/06/30 8:00	2.51	2.06	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	
GY-09			2018/06/30 8:00 to 2018/07/01 8:00	2.72	2.16	-0.34	0.05	-0.51	0.11	-0.76	0.14	-1.14	0.16	--	--	-0.05	0.07	0.06	0.07	0.09	0.11	--	--
GY-10			2018/07/01 8:00 to 2018/07/02 8:00	3.04	2.14	-0.27	0.05	-0.37	0.11	-0.64	0.14	-0.73	0.16	--	--	-0.08	0.07	-0.01	0.07	-0.10	0.11	--	--
GY-11			2018/07/02 8:00 to 2018/07/03 8:00	3.23	2.56	-0.28	0.05	-0.50	0.11	-0.83	0.14	-0.94	0.16	--	--	-0.04	0.07	-0.03	0.07	-0.12	0.11	--	--
GY-12			2018/07/03 8:00 to 2018/07/04 8:00	2.60	2.39	-0.31	0.05	-0.47	0.11	-0.79	0.14	-0.95	0.16	--	--	-0.07	0.07	0.01	0.07	-0.07	0.08	--	--

GY-13			2018/07/04 8:00 to 2018/07/05 8:00	3.09	2.35	-0.36	0.05	-0.55	0.11	-0.90	0.14	-1.05	0.16	--	--	-0.09	0.07	-0.02	0.07	-0.11	0.08	--	--	
GY-14			2018/07/05 8:00 to 2018/07/06 8:00	3.24	2.98	-0.30	0.05	-0.65	0.11	-1.00	0.14	-1.31	0.16	--	--	0.02	0.07	0.01	0.07	-0.02	0.08	--	--	
GZ-01	Guangzhou	Winter	2018/01/18 8:00 to 2018/01/24 8:00	3.92	4.50	-0.17	0.05	-0.31	0.11	-0.55	0.14	-0.61	0.16	-0.82	0.29	-0.02	0.07	0.00	0.07	-0.09	0.08	0.08	0.08	
GZ-02				2018/01/19 8:00 to 2018/01/20 8:00	3.68	2.30	-0.15	0.05	-0.23	0.11	-0.38	0.14	-0.39	0.16	-0.70	0.29	-0.05	0.07	-0.03	0.07	-0.09	0.08	-0.11	0.08
GZ-03				2018/01/20 8:00 to 2018/01/21 8:00	3.51	2.23	-0.19	0.05	-0.35	0.11	-0.51	0.14	-0.65	0.16	-0.92	0.29	-0.03	0.07	-0.03	0.07	-0.02	0.08	0.04	0.08
GZ-04				2018/01/21 8:00 to 2018/01/22 8:00	4.33	3.20	-0.24	0.05	-0.37	0.11	-0.57	0.14	-0.72	0.16	-1.05	0.29	-0.06	0.07	-0.01	0.07	-0.03	0.08	0.01	0.08
GZ-05				2018/01/22 8:00 to 2018/01/23 8:00	3.64	5.01	-0.12	0.05	-0.14	0.11	-0.27	0.14	-0.29	0.16	-0.46	0.29	-0.04	0.07	0.01	0.07	-0.05	0.08	-0.02	0.08
GZ-06				2018/01/23 8:00 to 2018/01/24 8:00	3.41	3.03	-0.07	0.05	-0.07	0.11	-0.12	0.14	-0.11	0.16	-0.29	0.29	-0.05	0.07	-0.01	0.07	-0.03	0.08	-0.12	0.08
GZ-07				2018/01/24 8:00 to 2018/01/25 8:00	3.67	1.79	-0.18	0.08	-0.25	0.11	-0.33	0.14	-0.37	0.16	-0.64	0.29	-0.08	0.07	-0.06	0.07	-0.05	0.08	-0.08	0.10
GZ-08		Summer		2018/06/29 8:00 to 2018/06/30 8:00	3.16	1.41	-0.27	0.05	-0.31	0.11	-0.53	0.15	-0.52	0.18	-0.79	0.29	-0.14	0.07	-0.05	0.10	-0.14	0.08	-0.01	0.29
GZ-09				2018/06/30 8:00 to 2018/07/01 8:00	3.24	1.48	-0.28	0.05	-0.26	0.11	-0.48	0.15	-0.40	0.18	-0.55	0.29	-0.18	0.07	-0.06	0.10	-0.19	0.08	0.04	0.29
GZ-10				2018/07/01 8:00 to 2018/07/02 8:00	3.05	1.54	-0.20	0.05	-0.20	0.11	-0.33	0.14	-0.36	0.16	-0.59	0.29	-0.11	0.07	-0.02	0.07	-0.06	0.08	-0.06	0.24
GZ-11				2018/07/02 8:00 to 2018/07/03 8:00	3.12	2.42	-0.20	0.06	-0.24	0.11	-0.42	0.14	-0.40	0.16	-0.38	0.29	-0.10	0.07	-0.03	0.07	-0.12	0.08	0.22	0.08
GZ-12				2018/07/03 8:00 to 2018/07/04 8:00	3.01	2.39	-0.18	0.06	-0.29	0.11	-0.59	0.14	-0.63	0.16	-0.84	0.29	-0.02	0.07	0.02	0.07	-0.12	0.08	0.09	0.08
GZ-13				2018/07/04 8:00 to 2018/07/05 8:00	3.35	1.52	-0.24	0.05	-0.34	0.11	-0.47	0.14	-0.51	0.16	-0.81	0.29	-0.11	0.07	-0.08	0.07	-0.09	0.08	-0.05	0.24
GZ-14				2018/07/05 8:00 to 2018/07/06 8:00	3.04	1.62	-0.12	0.05	-0.12	0.11	-0.29	0.14	-0.23	0.16	-0.27	0.29	-0.06	0.07	-0.01	0.07	-0.12	0.08	0.07	0.08
MWLG-01	Mt. Waliguan	Winter	2014/11/19 to 2014/11/29	22.6	1.40	-0.12	0.12	-0.08	0.15	-0.23	0.14	-0.16	0.16	--	--	-0.08	0.11	0.00	0.13	-0.10	0.11	--	--	
MWLG-02				2014/11/29 to 2014/12/09	20.4	1.46	-0.12	0.05	-0.13	0.11	-0.38	0.14	-0.25	0.16	--	--	-0.06	0.07	0.00	0.09	-0.19	0.10	--	--
MWLG-03				2014/12/09 to 2014/12/19	23.4	2.12	-0.21	0.05	-0.19	0.13	-0.35	0.16	-0.26	0.16	--	--	-0.14	0.07	-0.06	0.10	-0.15	0.12	--	--
MWLG-04				2014/12/19 to 2014/12/29	21.3	1.95	-0.29	0.05	-0.33	0.11	-0.55	0.14	-0.49	0.16	--	--	-0.17	0.07	-0.08	0.07	-0.19	0.08	--	--
MWLG-05				2014/12/29 to 2015/01/09	22.4	2.00	-0.13	0.05	-0.10	0.11	-0.24	0.14	-0.11	0.16	--	--	-0.10	0.07	-0.05	0.07	-0.16	0.12	--	--
MWLG-06				2015/01/09 to 2015/01/19	23.7	1.80	-0.11	0.05	-0.04	0.11	-0.10	0.14	-0.04	0.16	--	--	-0.10	0.07	-0.02	0.07	-0.07	0.12	--	--
MWLG-07				2015/01/19 to 2015/01/29	22.4	1.95	-0.04	0.10	0.02	0.11	0.00	0.26	0.07	0.16	--	--	-0.06	0.07	-0.02	0.07	-0.06	0.19	--	--
MWLG-08				2015/01/29 to 2015/02/09	20.2	2.21	-0.15	0.06	0.01	0.11	-0.21	0.14	-0.08	0.16	--	--	-0.13	0.07	0.05	0.07	-0.15	0.08	--	--
MWLG-09				2015/02/09 to 2015/02/19	21.3	1.14	-0.13	0.15	-0.09	0.11	-0.17	0.23	-0.13	0.16	--	--	-0.10	0.12	-0.02	0.07	-0.08	0.16	--	--

Table S5 Statistical summary of mean TGM concentrations, $\delta^{202}\text{Hg}$, $\Delta^{199}\text{Hg}$, $\Delta^{200}\text{Hg}$, simulated soil-atmosphere Hg^0 exchange flux in summer, winter and the whole sampling period (Wang et al., 2016), and anthropogenic Hg^0 emission flux in the investigated cities (AMAP/UNEP, 2013).

Site	Season	TGM			$\delta^{202}\text{Hg}$ (‰)			$\Delta^{199}\text{Hg}$ (‰)			$\Delta^{200}\text{Hg}$ (‰)			Simulated soil-atmosphere Hg^0 exchange flux ($\text{ng m}^{-2} \text{h}^{-1}$)	Anthropogenic Hg^0 emission flux ($\text{ng m}^{-2} \text{h}^{-1}$)
		Mean	1sd	n	Mean	1sd	n	Mean	1sd	n	Mean	1sd	n		
Beijing	Summer	2.85	0.26	7	-0.40	0.12	7	0.00	0.04	7	0.02	0.03	7	10.3	
	Winter	1.88	0.62	7	-0.09	0.24	7	-0.08	0.08	7	-0.05	0.05	7	1.4	
	Mean	2.37	0.68	15	-0.24	0.24	15	-0.04	0.07	15	-0.01	0.05	15	5.9	30.1
Shijiazhuang	Summer	3.34	0.87	7	-0.83	0.22	7	-0.07	0.05	7	-0.02	0.03	7	10.0	
	Winter	1.94	0.67	6	-0.25	0.43	6	-0.17	0.04	6	-0.02	0.05	6	0.8	
	Mean	2.64	1.04	13	-0.54	0.44	13	-0.12	0.07	13	-0.02	0.04	13	5.4	12.1
Jinan	Summer	3.14	0.59	7	-0.79	0.29	7	-0.07	0.03	7	-0.01	0.02	7	11.9	
	Winter	1.96	1.00	4	-0.21	0.37	4	-0.14	0.02	4	-0.02	0.04	4	1.1	
	Mean	2.55	0.93	11	-0.50	0.42	11	-0.10	0.04	11	-0.02	0.03	11	6.5	36.2
Lanzhou	Summer	3.08	0.70	7	-0.92	0.32	7	-0.03	0.05	7	-0.04	0.06	7	8.5	
	Winter	2.41	0.46	5	-0.48	0.19	5	-0.08	0.05	5	-0.01	0.03	5	1.4	
	Mean	2.74	0.70	12	-0.70	0.35	12	-0.06	0.05	12	-0.03	0.05	12	4.9	60.4
Zhengzhou	Summer	3.71	1.33	6	-0.65	0.13	6	-0.04	0.04	6	0.04	0.05	6	9.1	
	Winter	2.99	0.62	8	-0.45	0.29	8	-0.08	0.05	8	0.01	0.03	8	-0.6	
	Mean	3.35	0.89	14	-0.55	0.24	14	-0.06	0.05	14	0.02	0.04	14	4.2	20.1
Shanghai	Summer	2.55	0.50	7	-0.57	0.19	7	-0.03	0.04	7	0.03	0.03	7	5.4	
	Winter	2.12	0.68	6	-0.07	0.30	6	-0.11	0.06	6	-0.06	0.04	6	1.7	
	Mean	2.34	0.61	13	-0.32	0.35	13	-0.07	0.06	13	-0.01	0.06	13	3.5	177
Chengdu	Summer	4.66	1.07	7	-0.91	0.11	7	-0.01	0.03	7	0.01	0.03	7	8.1	

	Winter	4.45	1.40	7	-0.45	0.52	7	-0.01	0.04	7	0.00	0.03	7	3.3	
	Mean	4.56	1.21	14	-0.68	0.44	14	-0.01	0.03	14	0.00	0.03	14	5.7	37.6
Wuhan	Summer	4.00	0.63	9	-0.77	0.22	9	-0.02	0.04	9	0.00	0.02	9	3.9	
	Winter	3.53	1.75	7	-0.59	0.22	6	-0.09	0.04	6	-0.03	0.07	6	0.9	
	Mean	3.76	1.22	16	-0.68	0.23	15	-0.05	0.05	15	-0.02	0.05	15	2.4	41.9
Guiyang	Summer	2.43	0.31	7	-1.02	0.20	6	-0.05	0.04	6	0.00	0.03	6	15.5	
	Winter	5.58	1.97	7	-0.90	0.56	7	0.03	0.04	7	0.01	0.01	7	8.9	
	Mean	4.01	2.14	14	-0.96	0.42	13	-0.01	0.06	13	0.01	0.02	13	12.2	11.7
Guangzhou	Summer	1.77	0.44	7	-0.43	0.13	7	-0.10	0.05	7	-0.03	0.04	7	12.5	
	Winter	3.15	1.21	7	-0.45	0.22	7	-0.05	0.02	7	-0.02	0.02	7	7.3	
	Mean	2.46	1.13	14	-0.44	0.17	14	-0.07	0.05	14	-0.03	0.03	14	9.9	56.7
Mt. Waliguan	Mean	1.78	0.36	9	-0.16	0.16	9	-0.10	0.04	9	-0.02	0.04	9		

Table S6 Monthly productions of cement, zinc, iron and steel, lead, and aluminum during winter, summer, and the all year of 2018 as well as the estimated monthly Hg emissions for the production of these source materials. Note that Hg emissions from the production of these source materials as well as from coal combustion accounted for 90% of the total anthropogenic Hg emissions in China (Liu et al., 2019).

Season	Source material production ^a					Hg emission (Mg/month)					
	Cement (10 ⁴ ton/month)	Zinc (10 ⁴ ton/month)	Iron and steel (10 ⁴ ton/month)	Lead (10 ⁴ ton/month)	Aluminum (10 ⁴ ton/month)	Cement	Zinc	Iron and steel	Lead	Aluminum	Total
Winter 2018	14970	55.5	8910	45.9	465	8.9	3.7	2.6	2.4	1.1	18.6
Summer 2018	19440	43.8	9390	39.9	423	11.5	2.9	2.7	2.1	1.1	20.1
Year 2018	17880	46.8	9090	42.0	432	10.6 ^b	3.1 _b	2.6 _b	2.2 ^b	1.1 ^b	19.8 _b

a): data are from National Bureau of Statistics, China

b): data are from Liu et al.(2019).

Reference:

- AMAP/UNEP: Geospatially distributed mercury emissions dataset 2010v1, in, 2013.
- Blum, J. D., and Johnson, M. W.: Recent Developments in Mercury Stable Isotope Analysis, *Reviews in Mineralogy & Geochemistry*, 82, 733-757, 2017.
- Duan, L., Wang, X. H., Wang, D. F., Duan, Y. S., Cheng, N., and Xiu, G. L.: Atmospheric mercury speciation in Shanghai, China, *Sci Total Environ*, 578, 460-468, 2017.
- Estrade, N., Carignan, J., Sonke, J. E., and Donard, O. F. X.: Measuring Hg Isotopes in Bio-Geo-Environmental Reference Materials, *Geostand Geoanal Res*, 34, 79-93, 2010.
- Fu, X. W., Feng, X. B., Qiu, G. L., Shang, L. H., and Zhang, H.: Speciated atmospheric mercury and its potential source in Guiyang, China, *Atmos Environ*, 45, 4205-4212, DOI 10.1016/j.atmosenv.2011.05.012, 2011.
- Fu, X. W., Heimburger, L. E., and Sonke, J. E.: Collection of atmospheric gaseous mercury for stable isotope analysis using iodine- and chlorine-impregnated activated carbon traps, *J Anal Atom Spectrom*, 29, 841-852, Doi 10.1039/C3ja50356a, 2014.
- Gustin, M. S., Dunham-Cheatham, S. M., and Zhang, L.: Comparison of 4 Methods for Measurement of Reactive, Gaseous Oxidized, and Particulate Bound Mercury, *Environmental Science & Technology*, 53, 14489-14495, 10.1021/acs.est.9b04648, 2019.
- Liu, K. Y., Wu, Q. R., Wang, L., Wang, S. X., Liu, T. H., Ding, D., Tang, Y., Li, G. L., Tian, H. Z., Duan, L., Wang, X., Fu, X. W., Feng, X. B., and Hao, J. M.: Measure-Specific Effectiveness of Air Pollution Control on China's Atmospheric Mercury Concentration and Deposition during 2013-2017, *Environmental Science & Technology*, 53, 8938-8946, 10.1021/acs.est.9b02428, 2019.
- Sun, G., Sommar, J., Feng, X., Lin, C.-J., Ge, M., Wang, W., Yin, R., Fu, X., and Shang, L.: Mass-dependent and -independent fractionation of mercury isotope during gas-phase oxidation of elemental mercury vapor by atomic Cl and Br, *Environmental Science & Technology*, 50, 9232-9241, 10.1021/acs.est.6b01668, 2016.
- Wang, X., Lin, C. J., Yuan, W., Sommar, J., Zhu, W., and Feng, X. B.: Emission-dominated gas exchange of elemental mercury vapor over natural surfaces in China, *Atmos Chem Phys*, 16, 11125-11143, 2016.
- Zhang, H., Wang, Z. W., Wang, C. J., and Zhang, X. S.: Concentrations and gas-particle partitioning of atmospheric reactive mercury at an urban site in Beijing, China, *Environ Pollut*, 249, 13-23, 10.1016/j.envpol.2019.02.064, 2019.