Supporting Information for:

Integrating Point Sources to Map Anthropogenic Atmospheric Mercury Emissions in China, 1978–2021

Yuying Cui[1,2], Qingru Wu[1,2*], Shuxiao Wang[1,2], Kaiyun Liu[3], Shengyue Li[1,2], Zhezhe Shi[1,2], Daiwei Ouyang[1,2], Zhongyan Li[4], Qinqin Chen[1,2], Changwei Lü[5,6], Fei Xie[5,6], Yi Tang[7], Yan Wang[8], Jiming Hao[1,2]

[1]State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China

[2]State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex, Beijing 100084, China

[3]College of Environmental Science and Engineering, North China Electric Power University, Beijing, 102206, PR China

[4]Weiyang College, Tsinghua University, Beijing 100084, China

[5]School of Ecology and Environment, Inner Mongolia University, 010021, Hohhot, China

[6]Institute of Environmental Geology, Inner Mongolia University, 010021, Hohhot, China

[7]State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, 100012, Beijing, China

[8]College of Electromechanical Engineering, Qingdao University of Science and Technology, Qingdao 266061, China

Correspondence to: Qingru Wu (qrwu@tsinghua.edu.cn)

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Text S1 Equations for P-CAME emission inventory

Equation S1: Emissions calculation for Tier 1

$$E_{i,k,t}=\sum_{j}(A_{i,j,t}\times ef_{i,j,k,t})=\sum_{j}(A_{i,j,t}\times c_{i,p,t}(x)\times R_{i,j}(y)\times(1-\eta_{i,j}(y,z))\times\theta_{i,j,k}(x,y,z))$$

Equation S2: Emissions calculation for Tier 2

$$E_{i,k,t}=\sum_{p}(A_{i,p,t}\times ef_{i,p,k,t})=\sum_{p}(A_{i,p,t}\times c_{i,p,t}(x)\times R_{i,j}(y)\times(1-\eta_{i,p}(y,z))\times\theta_{i,p,k}(x,y,z))$$

Where *E* is the emission, kg. i is the sector. j is the enterprise. k is mercury species. t is year. p is the province. *A* is the activity level. *ef* is the emission factor. x is the type of fuel or raw materials. y is the type of combustion or production process. z is the type of APCD. *c* is Hg concentration of fuel or raw materials, g/t. *R* is the release rate of combustion or production process, %. η is the probabilistic distribution of Hg removal efficiency of a certain type of APCD combination, %. θ is the proportion of mercury species, %.

Equation S3: Emissions calculation for Tier 3

$$E_{i,k,t}=\sum_{p}(A_{i,p,t}\times ef_{i,p,k,t})=\sum_{p}(A_{i,p,t}\times\theta_{i,p,k}\times[(ef_{a_i}-ef_{b_i})\times\exp\left(-\frac{(t-t_0)^2}{2\times S_i^2}\right)+ef_{b_i}])$$

Where ef_a is the emission factor pre-1990, g/t. ef_b is the best emission factor that could be achieved, g/t. t_0 is the time when the technology transition begins (pre-1990), yr. *S* is the shape parameter of the curve. The largest emission factor for one sector from the literature was set as ef_a while the most recent localized emission factor was used as ef_b.

Equation S4: Spatial distribution of Tier2 and Tier3

$$E_{i,j}=[E_{pro}]_i\times\frac{GDP_k}{\sum_{pro}GDP_k}\times\frac{POP/Road_j}{\sum_{city}POP/Road_j}$$

Where *E* is emission of the grid, i is the sectors, *E*pro is the emissions of the province where the grid is located, *GDP* is the gross domestic product, k is the city which belongs to the province, *POP* is the population, *Road* is the area of the driveway in every grid, j is the grid which belongs to the city.

Text S2 Equations for bias calculation

Calculation method of normalized mean bias (NMB)

$$\mathrm{NMB} = \frac{\sum_i (Simulation_i - Observation_i)}{\sum_i Observation_i} \qquad \text{(Equation S5)}$$

Calculation method of normalized mean error (NME)

$$\mathrm{NME} = \frac{\sum_i |Simulation_i - Observation_i|}{\sum_i Observation_i} \qquad \text{(Equation S6)}$$

Table S1 Categories, calculation and spatial distribution of Hg emission sectors

Tier	Abbreviation	Sector	Calculation method	Spatial distribution method
1	CFPP	Coal-fired power plant	Technology-based emission factors	Latitude and longitude
1	Zn	Zinc smelting		
1	Pb	Leading smelting		
1	Cu	Copper smelting		
1	CEM	Cement production		
1	ISP	Iron and steel production		
1	CFIB	Coal-fired industrial boiler		
1	MSWI	Municipal solid waste incineration		
1	LSGP	Large-scale gold production		
2	RBL	Residential coal combustion	Dynamic technology-based emission factors	Population
2	OCC	Other coal combustion		GDP2 and Population
2	CSP	Caustic soda production		
2	PVC	Cholor-alhali production		
2	BAP	Battery production		
2	FLU	Fluorescent lamp production		
2	THP	Thermometer production		
2	SMP	Sphygmomanometer production		
3	BIO	Biomass fuel combustion	Time-varying emission factors	GDP1 and Population
3	Hg	Mercury production		GDP2 and Population
3	Al	Aluminum production		
3	ASGM	Artisanal and small-scale gold mining		
3	SOC	Stationary oil combustion		GDP3 and Road map
3	MOC	Mobile oil combustion		
3	CRE	Cremation		Population

Notes: GDP1 represents GDP of primary industry, GDP2 represents GDP of the secondary industry, GDP3 represents GDP of the tertiary industry.

Table S2 Activity level and references

Category	Type of activity level	References
Coal-fired power plant	Coal consumption in power plants	China energy statistical yearbooks (NESA, 1986-2022); Chinese statistics of electric power industry (CEPYEC, 1993-2021); Chinese environmental statistics (CEMS, 2013-2017)
Coal-fired industrial boilers	Coal consumption in industrial boilers	China energy statistical yearbooks (NESA, 1986-2022); Chinese environmental statistics (CEMS, 2013-2017)
Residential coal combustion	Coal consumption in residents	China energy statistical yearbooks (NESA, 1986-2022); Chinese environmental statistics (CEMS, 2013-2017)
Other coal combustion	Coal consumption in others	China energy statistical yearbooks (NESA, 1986-2022); Chinese environmental statistics (CEMS, 2013-2017)
Stationary oil combustion	Oil consumption in stations	China statistical yearbooks (NBS, 1981-2022)
Mobile oil combustion	Oil consumption in mobile vehicles	China statistical yearbooks (NBS, 1981-2022)
Biomass fuel combustion	Biomass consumption	China rural energy statistical yearbook (MA, 1997-2021)
Municipal solid waste incineration	Waste incineration amount	China energy statistical yearbook (NESA, 1986-2022)
Cremation	Corpse numbers	China civil affairs statistical yearbook (MCA, 1980-2017)
Copper smelting	Copper yield	Yearbooks of nonferrous metals industry of China (CMRA, 1991-2020); Chinese environmental statistics (CEMS, 2013-2017)
Leading smelting	Leading yield	Yearbooks of nonferrous metals industry of China (CMRA, 1991-2020); Chinese environmental statistics (CEMS, 2013-2017)
Zinc smelting	Zinc yield	Yearbooks of nonferrous metals industry of China (CMRA, 1991-2020); Chinese environmental statistics (CEMS, 2013-2017)
Large-scale gold production	Gold yield	Chinese environmental statistics (CEMS, 2013-2017)
Artisanal and small-scale gold mining	Gold yield	(Wu et al., 2016)
Aluminum smelting	Aluminum yield	Chinese environmental statistics (CEMS, 2013-2017)
Mercury production	Mercury yield	Yearbooks of nonferrous metals industry of China (CMRA, 1991-2020); Chinese environmental statistics (CEMS, 2013-2017)
Cement production	Cement yield	Chinese environmental statistics (CEMS, 2013-2017)
Iron and steel production	Pig steel yield	China steel yearbooks (CISI, 1985-2022); Chinese environmental statistics (CEMS, 2013-2017)
Chlor-alkali production	Vinyl chloride yield	China statistical yearbooks (NBS, 1981-2022) Chinese environmental statistics (CEMS, 2013-2017) Report for national mercury investigation of China (MEE, 2012)
Caustic soda production	Caustic soda yield	
Battery production	Battery yield	
Fluorescent lamp production	Fluorescent lamp yield	
Thermometer production	Thermometer yield	
Sphygmomanometer production	Sphygmomanometer yield	

Table S3 Hg removal efficiencies and speciation profiles for APCDs combination

Sectors	APCDs	Hg removal efficiency (%)	Probabilistic distribution	Speciation profile (%)			Ref.
				Hg^0	Hg^{2+}	Hg^P	
Coal-fired power plant	NOC/CYC	1.0±0.5	Weibull	38.0	38.0	24.0	(Liu et al., 2019; Zhang et al., 2023; Wu et al., 2016)
	WET	23±8	Normal	65.0	33.0	1.0	
	ESP	32±23	Weibull	58.0	41.0	1.3	
	FF	67±30	Normal	50.0	49.0	0.5	
	ESP+WFGD	60±22	Weibull	84.0	16.0	0.6	
	FF+WFGD	86±10	Normal	78.0	21.0	1.0	
	ESP-FF+WFGD	88±16	Normal	84.0	16.0	0.6	
	SCR+ESP+WFGD	70±21	Normal	72.0	27.0	1.0	
	SCR+FF+WFGD	88±7	Normal	37.0	61.0	2.0	
	SCR+ESP+WFGD+WESP	94±3	Normal	69.0	30.0	1.0	
	SCR+ESP-FF/LTESP+WFGD	97±3	Normal	64.0	35.0	2.0	
	SNCR+ESP+WFGD	98	Normal	51.0	48.0	1.0	
	(CFB)ESP	73±8	Weibull	71.8	27.6	0.6	
	(CFB)FF	76±12	Normal	81.5	18.0	0.5	
	(CFB)SNCR+ESP+WFGD	98	Normal	51.0	48.0	1.0	
Coal-fired industrial boiler	NOC/CYC	1.0±0.5	Weibull	38.0	38.0	24.0	
	WET	23±8	Normal	65.0	33.0	1.0	
	IDRD	38±21	Normal	49.0	48.0	3.0	
	FF+WFGD	86±1	Normal	78.0	21.0	1.0	
	ESP-FF+WFGD	88±16	Normal	84.0	16.0	0.6	
	SCR+FF+WFGD	88±7	Normal	37.0	61.0	2.0	
	ESP	32±23	Normal	57.7	41.0	1.3	
	FF	67±20	Normal	50.5	48.5	1.0	
Pb/Zn/Cu smelting	None	0	Normal	34.0	56.0	10.0	
	DC	10±8	Normal	33.0	62.0	5.0	
	FGS	24±25	Normal	65.0	33.0	2.0	
	DC+FGS	41±20	Normal	41.0	54.0	5.0	
	DC+FGS+ESD+SCSA	87±3	Normal	57.0	38.0	5.0	
	DC+FGS+ESD+DCDA	97±2	Normal	46.0	49.0	5.0	
	DC+FGS+ESD+DCDA+DFGD	99.0±0.1	Normal	94.0	6.0	0.0	
	DC+FGS+ESD+DCDA+WFGD	99	Normal	65.0	35.0	0.0	
	DC+FGS+ESD+SMR+DCDA	99.0±0.1	Normal	6.0	90.0	4.0	
	DC+FGS+ESD+SMR+DCDA+DFGD	99	/	65.0	35.0	0.0	
	DC+FGS+ESD+SMR+DCDA+WFGD	99	/	56.0	34.0	10.0	
	(SK/RK)CYC	1.0±0.5	Normal	38.0	38.0	24.0	

Cement production	(SK/RK)WET	25±1	Normal	33.0	65.0	2.0	
	(SK/RK)ESP	44±29	Normal	41.0	58.0	1.0	
	(SK/RK)FF	62±28	Normal	49.0	50.0	1.0	
	(DPPT)CYC	1.0±0.5	Normal	38.0	38.0	24.0	
	(DPPT)WET	25±1	Normal	33.0	65.0	2.0	
	(DPPT)ESP	6±7	Normal	23.5	76.0	0.5	
	(DPPT)FF	6±7	Normal	23.5	76.0	0.5	
	SNCR+ESP/FF	15	Normal	48.0	51.0	1.0	
	SNCR+ESP/FF+DFGD	15	/	48.0	51.0	1.0	
	SNCR+ESP/FF+WFGD	42	/	79.5	20.1	0.3	
	SCR+ESP/FF	20	/	11.8	87.8	0.4	
	SCR+ESP/FF+WFGD	69	/	36.0	63.8	0.3	
Iron and steel production	Non/CYC	1	Normal	38.0	38.0	24.0	
	WS	23	Normal	65.0	34.0	1.0	
	ESP	29	Normal	18.0	82.0	0.0	
	FF	67	Normal	18.0	82.0	0.0	
	ESP+WFGD	57	Normal	41.0	59.0	0.0	
	ESP+DFGD+FF	72	/	0.5	99.0	0.5	
	SCR+ESP+WFGD	70	/	56.7	43.1	0.2	
	SCR+ESP+DFGD+FF	81	/	37.0	61.0	2.0	
	SCR+FF+WFGD	88	/	37.0	61.0	2.0	

Note, CYC: cyclone, WET: wet scrubber, ESP: electrostatic precipitator, FF: fabric filter, WFGD: wet flue gas desulfurization, SCR: selective catalytic reduction, WESP: wet electrostatic precipitator, LTESP: low temperature electrostatic precipitator, SNCR: selective non-catalytic reduction, CFB: circulating fluidized bed, IDRD: In-duct reaction device, DC: dust collector, FGS: flue gas scrubber, ESD: electrostatic demister, SCSA: single conversion single absorption, DCDA: double conversion double absorption, DFGD: dry flue gas desulfurization, SMR: selective multi-component reduction, SK/RK: shaft kiln or rotary kiln, DPPT: dry-process precalciner technology, WS: wet scrubbing.

Table S4 GEM observations and validation of simulation at 2021 (*GEM_obs: GEM observations, ng/m^3; GEM_std: standard error of GEM observations; GEM_sim_proxy: GEM simulation based on proxy; GEM_sim_P-CAME: GEM simulation based on P-CAME; NMB: normalized mean bias; NME: normalized mean error)

Lon	Lat	Provinces	Sites	Type	Month	GEM_obs	GEM_std	References	GEM_sim_proxy	GEM_sim_P-CAME	NMB/NME
118.77	32.08	Jiangsu	Nanjing	urban	1	1.94	/	(Sun et al., 2024)	3.31	2.91	NMB_Proxy=59% NME_Proxy=59% NMB_P-CAME=39% NME_ P-CAME =44%
					2	0.98			2.85	2.52	
					3	1.31			2.74	2.41	
					4	1.46			2.49	2.18	
					5	1.48			2.36	2.06	
					6	2.08			2.30	1.93	
					7	1.46			1.44	1.34	
					8	1.90			2.00	1.78	
					9	1.37			2.19	1.91	
					10	1.15			2.35	1.97	
111.76	40.83	Inner Mongolia	Hohhot	urban	1	1.32	0.69	This study	2.69	2.09	NMB_Proxy=57% NME_Proxy=57% NMB_P-CAME=24% NME_ P-CAME =24%
					2	1.42	0.52		2.59	2.03	
					3	1.69	0.78		2.62	2.07	
					4	1.52	0.60		2.36	1.88	
					5	1.45	0.68		1.70	1.45	
					6	1.41	0.61		1.80	1.48	
					7	1.38	0.34		1.70	1.43	
					8	1.37	0.35		1.92	1.53	
					9	1.13	0.27		2.09	1.65	
					10	1.27	0.78		2.32	1.73	
					11	1.66	1.25		2.32	1.78	
					12	1.36	0.81		2.54	1.88	
120.507	36.163	Shandong	Qingdao	urban	1	3.0	/	(Shao et al., 2022)	3.45	2.56	NMB_Proxy=23% NME_Proxy=23% NMB_P-CAME=-7% NME_ P-CAME =10%
					2	2.0			2.71	2.09	
128.113	42.40	Jilin	Mt. Changbai	rural	1	1.59	/	(Wu et al., 2023)	1.43	1.43	NMB_Proxy=-6%
					2	1.64			1.52	1.52	
					3	1.59			1.50	1.49	

					4	1.34			1.32	1.32	NME_Proxy=7% NMB_P-CAME=-6% NME_ P-CAME =7%
					5	1.23			1.26	1.26	
					6	1.12			1.20	1.20	
					7	1.22			1.12	1.12	
					8	1.13			1.10	1.10	
					9	1.16			1.16	1.16	
					10	1.35			1.19	1.19	
					11	1.52166			1.25	1.25	
101.02	24.533	Yunnan	Mt. Ailao	rural	1	1.77	/	(Wu et al., 2023)	1.96	1.89	NMB_Proxy=-2% NME_Proxy=12% NMB_P-CAME=-4% NME_ P-CAME =12%
					2	1.42			1.74	1.67	
					3	1.56			1.56	1.51	
					4	1.62			1.50	1.45	
					5	1.45			1.21	1.19	
					6	1.38			1.21	1.20	
					7	1.20			1.20	1.18	
					8	1.23			1.22	1.20	
					9	1.48			1.16	1.13	
					10	1.72			1.51	1.47	
					11	1.15			1.47	1.41	
121.9826	31.46799	Shanghai	Chongming	rural	2	1.35	/	This study	1.82	1.80	NMB_Proxy=-2% NME_Proxy=19% NMB_P-CAME=-1% NME_ P-CAME =18%
					3	1.45			1.64	1.67	
					4	1.49			1.39	1.42	
					5	1.71			1.49	1.52	
					6	1.58			1.26	1.29	
					7	1.82			1.09	1.09	
					10	0.87			1.01	1.03	
					11	1.34			1.46	1.47	
					12	1.48			1.63	1.62	

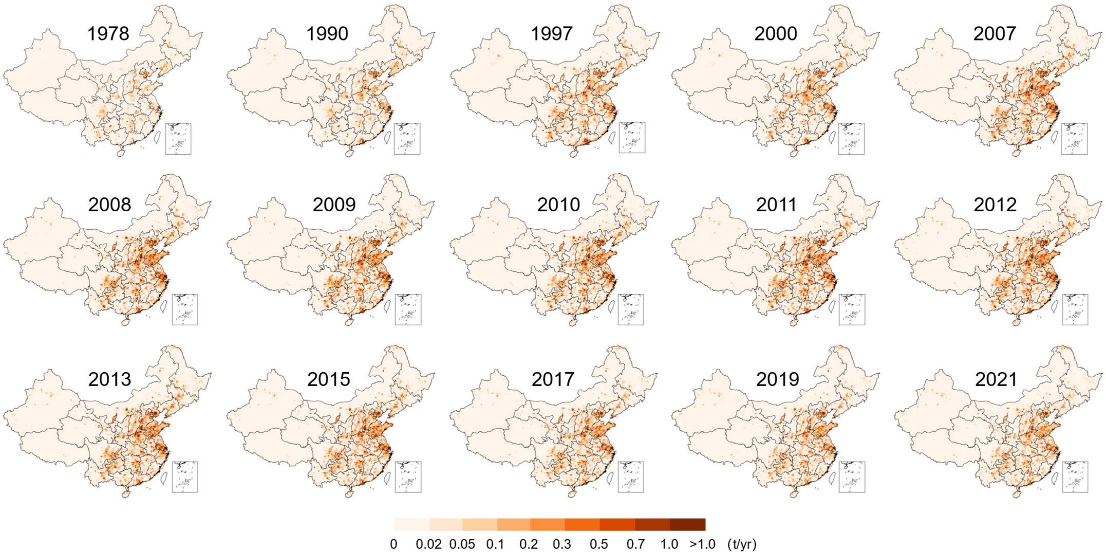

Figure S1 Spatial distribution of anthropogenic mercury emissions.

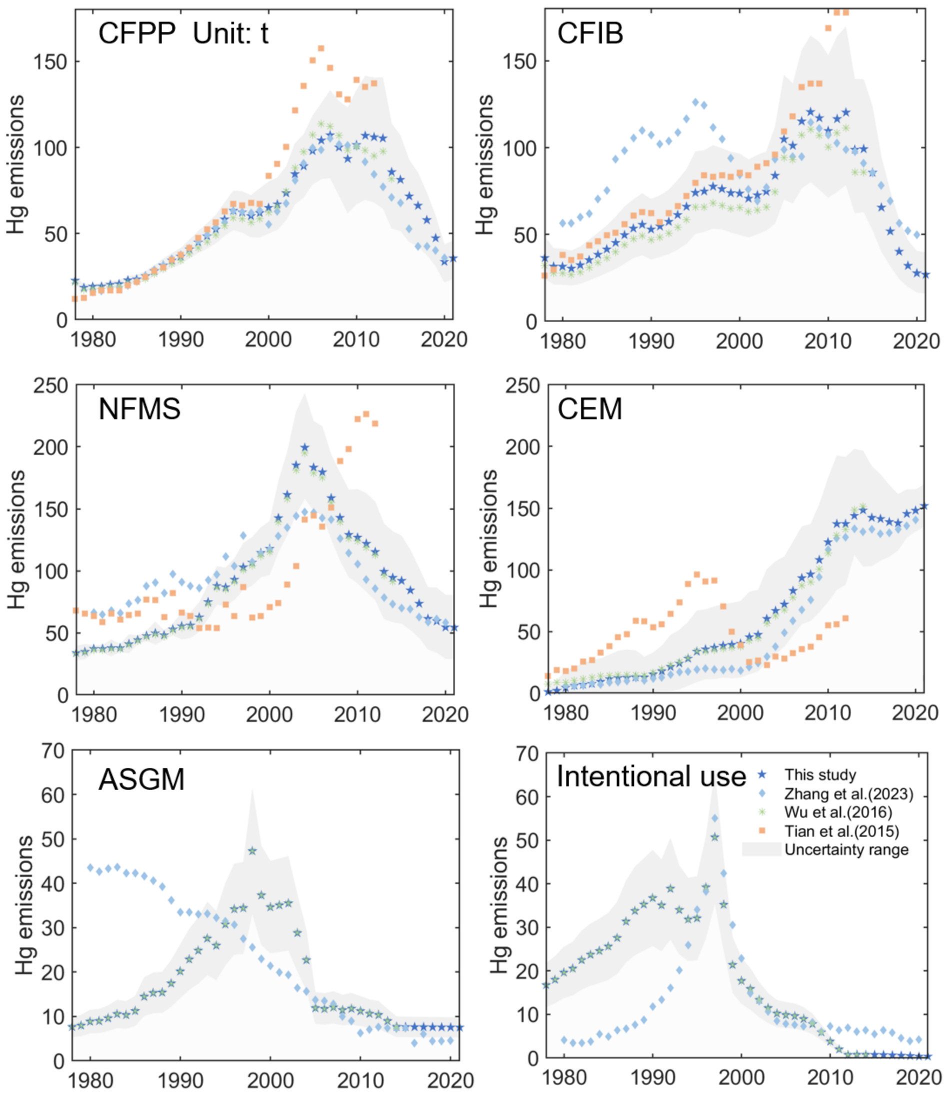

Figure S2 Comparison in key sectors with previous emission inventories.

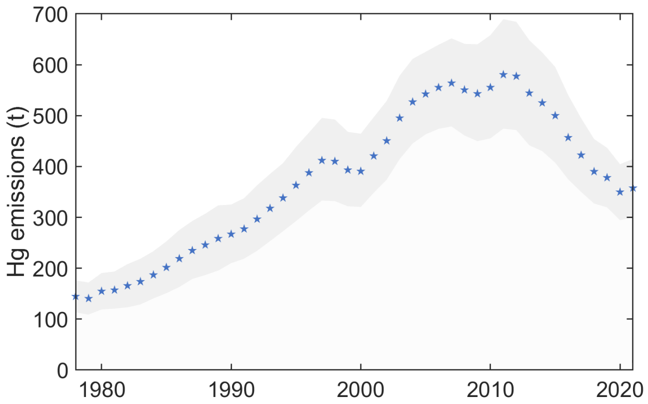

Figure S3 Uncertainty range of anthropogenic mercury emissions during 1978-2021.

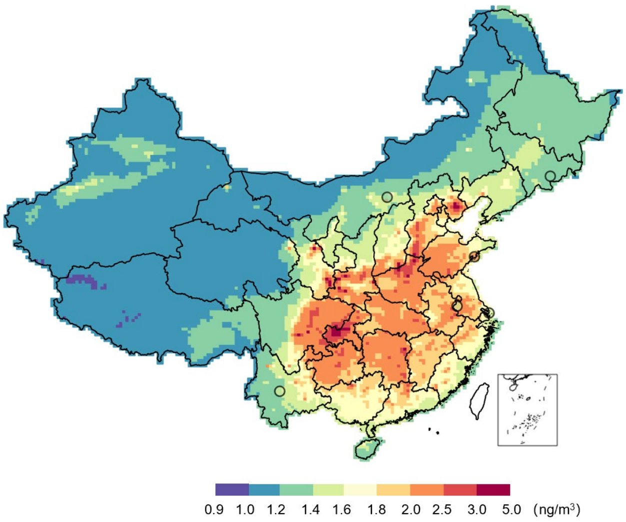

Figure S4 GEM concentrations simulated by GEOS-Chem using P-CAME and comparison with observations (Circles represent observed GEM concentrations).

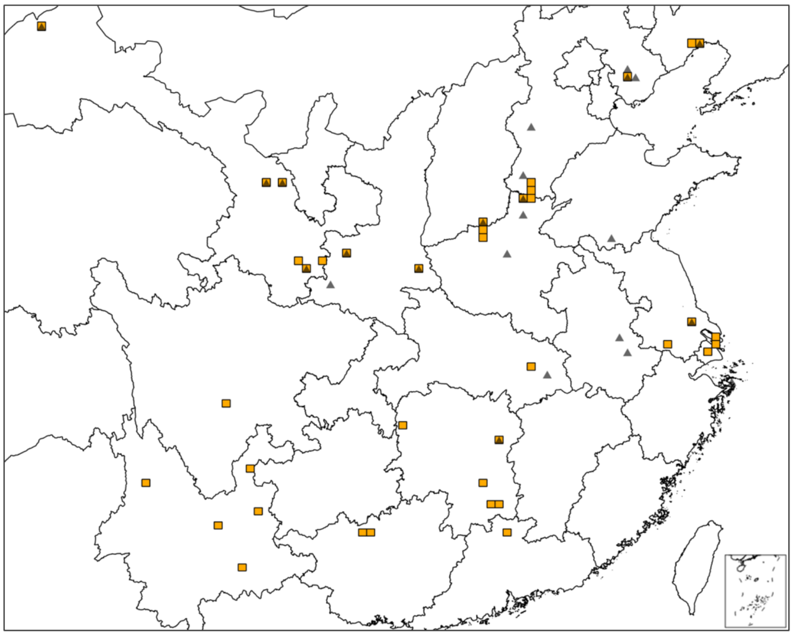

Figure S5 Comparation of hotspots between cumulative emissions and emissions at 2021. The triangle represents the 2021 atmospheric mercury emission hotspots, and the square represents the historical cumulative emission hotspots.

References

CEMS (China Environmental Monitoring Station): Chinese environmental statistics, China Environmental Monitoring Station, Beijing, 2013-2017.

CEPYEC (China Electric Power Yearbook Editorial Committee): Chinese statistics of electric power industry, China Electric Power Press, Beijing, 1993-2021.

CISI (Editorial Committee of China Iron and Steel Industry Annual Review, Ministry of Metallurgical Industry of China): China steel yearbooks, China Metallurgical Press, Beijing, 1985-2022.

CMRA (China Nonferrous Metals Industry Association): Yearbooks of nonferrous metals industry of China, China Printing Corporation, Beijing, 1991-2020.

Liu, K., Wu, Q., Wang, L., Wang, S., Liu, T., Ding, D., Tang, Y., Li, G., Tian, H., Duan, L., Wang, X., Fu, X., Feng, X., and Hao, J.: 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.

MA (Department of Science, Technology and Education, Ministry of Agriculture): China Rural energy statistical Yearbook, China Agriculture Press, Beijing, 1997-2021.

MCA (Ministry of Civil Affairs, People's Republic of China): China Civil Affairs Statistical Yearbook, China Statistics Press, Beijing, 1980-2017.

MEE (Ministry of Ecology and Environment of the People's Republic of China): Report for National Mercury Investigation of China, Beijing, 2012.

NBS (National Bureau of Statistics of the People's Republic of China): China Statistical Yearbooks, China Statistics Press, Beijing, 1981-2022.

NESA (National Energy Statistical Agency of China): China Energy Statistical Yearbooks, China Statistics Press, Beijing, 1986-2022.

Shao, L., Wang, Y., Liu, X., Liu, R., Han, K., and Zhang, Y.: Temporal variation of gaseous elemental mercury in a northern coastal city in China: Monsoon and COVID-19 lockdown effects, Atmospheric Pollution Research, 13, 101436, https://doi.org/10.1016/j.apr.2022.101436, 2022.

Sun, P., Song, Z., Qin, Y., Xu, Z., Zhang, Y., Zhong, S., and Yu, J.: Declines of gaseous element mercury concentrations at an urban site in eastern China caused by reductions of anthropogenic emission, Atmospheric Environment, 317, 10.1016/j.atmosenv.2023.120199, 2024.

Wu, Q., Wang, S., Li, G., Liang, S., Lin, C.-J., Wang, Y., Cai, S., Liu, K., and Hao, J.: Temporal Trend and Spatial Distribution of Speciated Atmospheric Mercury Emissions in China During 1978–2014, Environmental Science & Technology, 50, 13428-13435, 10.1021/acs.est.6b04308, 2016.

Wu, X., Fu, X., Zhang, H., Tang, K., Wang, X., Zhang, H., Deng, Q., Zhang, L., Liu, K., Wu, Q., Wang, S., and Feng, X.: Changes in Atmospheric Gaseous Elemental Mercury Concentrations and Isotopic Compositions at Mt. Changbai During 2015–2021 and Mt. Ailao During 2017–2021 in China, Journal of Geophysical Research: Atmospheres, 128, e2022JD037749, https://doi.org/10.1029/2022JD037749, 2023.

Zhang, Y., Zhang, L., Cao, S., Liu, X., Jin, J., and Zhao, Y.: Improved Anthropogenic Mercury Emission Inventories for China from 1980 to 2020: Toward More Accurate Effectiveness Evaluation for the Minamata Convention, Environmental Science & Technology, 57, 8660-8670, 10.1021/acs.est.3c01065, 2023.