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Integrating point sources to map anthropogenic atmospheric mercury emissions in China, 1978–2021

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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, Epro 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

Equation S5: Normalized Mean Bias (NMB)

$$\text{NMB} = \frac{\sum_i (Sim_i - Obs_i)}{\sum_i Obs_i}$$

Equation S6: Normalized Mean Error (NME)

$$\mathrm{NME} = \frac{\sum_i |Sim_i - Obs_i|}{\sum_i Obs_i}$$

Equation S7: Root Mean Square Error (RMSE)

$$\mathrm{RMSE} = \sqrt{\frac{\sum_i (Sim_i - Obs_i)^2}{n}}$$

Equation S8: Pearson Correlation Coefficient (R)

$$\mathrm{R} = \frac{\sum_i (Sim_i - \overline{Sim})(Obs_i - \overline{Obs})}{\sqrt{\sum_i (Sim_i - \overline{Sim})^2 \sum_i (Obs - \overline{Obs})^2}}$$

Where *Sim* is simulated Hg^0 concentration, ng/m^3. *Obs* is observed Hg^0 concentration, ng/m^3. i is an index representing each data point in the dataset, n is the total number of data points.

Text S3 Equations for decline rate

Equation S9: An example calculation for decline rate of Hg^0 concentration at Changbai

$$Decline\ rate = \frac{obs_{2013} - obs_{2021}}{obs_{2013}}$$

Where obs_{2013} is observed Hg^0 concentration at 2013, ng/m^3. obs_{2021} is observed Hg^0 concentration at 2021, ng/m^3.

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

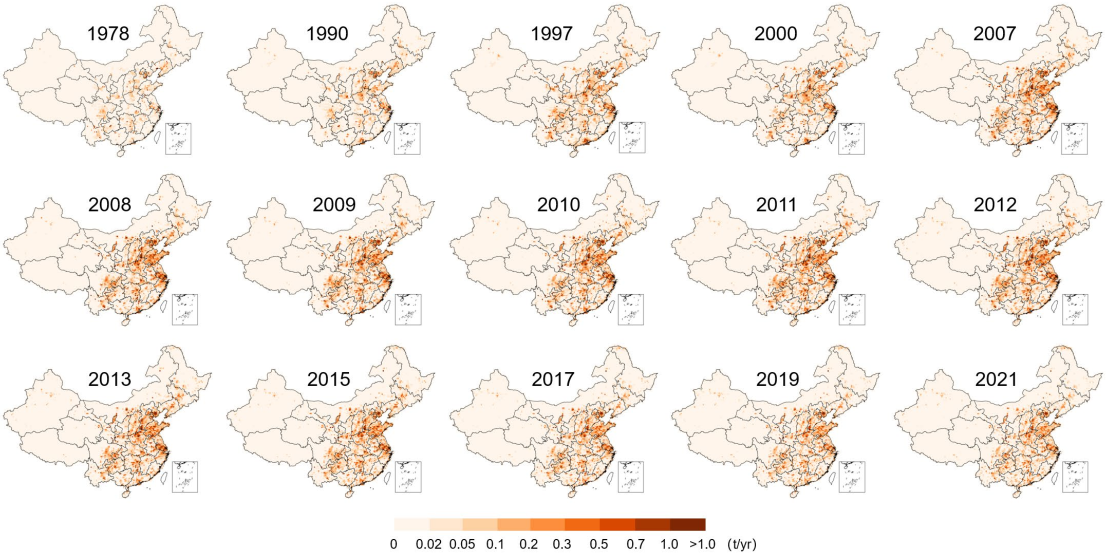
Category	Type of activity level	References
Coal-fired power plant	Coal consumption in power plants	China energy statistical yearbooks (NESA, 1986-2022); Chinese statistics of electric power industry (CEPYEC, 1993-2021); Chinese environmental statistics (CEMS, 2013-2017)
Coal-fired industrial boilers	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	
Cement production	(SK/RK)CYC	1.0±0.5	Normal	38.0	38.0	24.0	
	(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.

72 

73 **Figure S1 Temporal and spatial distribution of anthropogenic mercury emissions.**

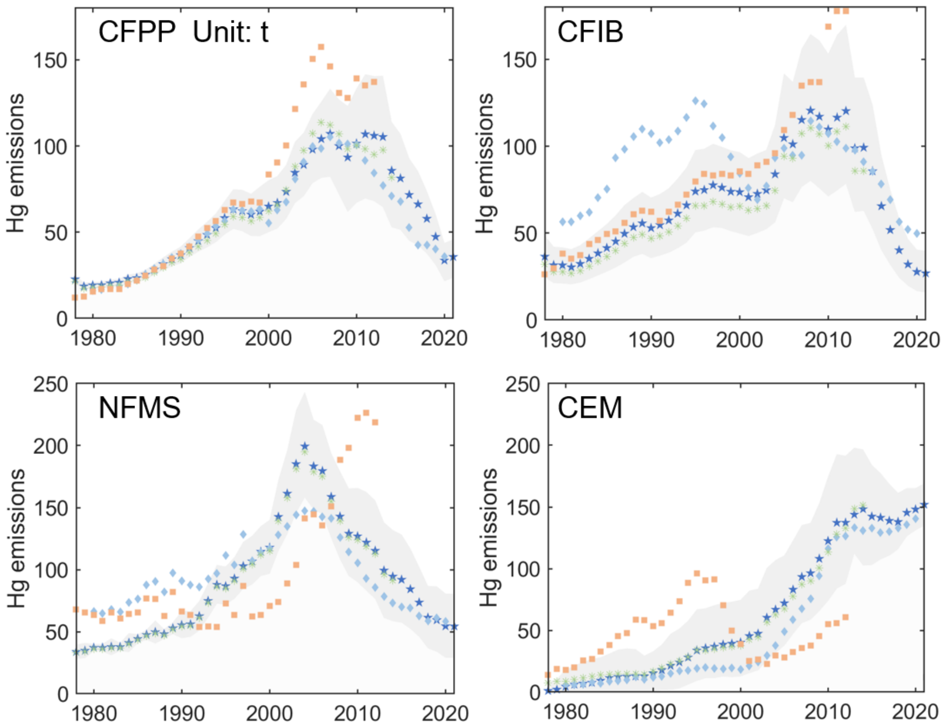


Figure S2 Comparison in key sectors with previous emission inventories.

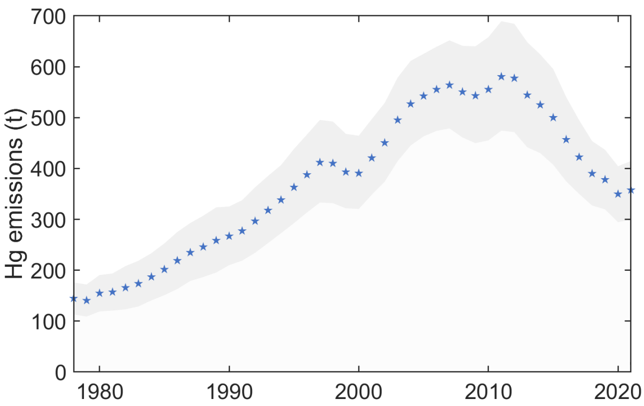


Figure S3 Uncertainty range of anthropogenic mercury emissions during 1978-2021.

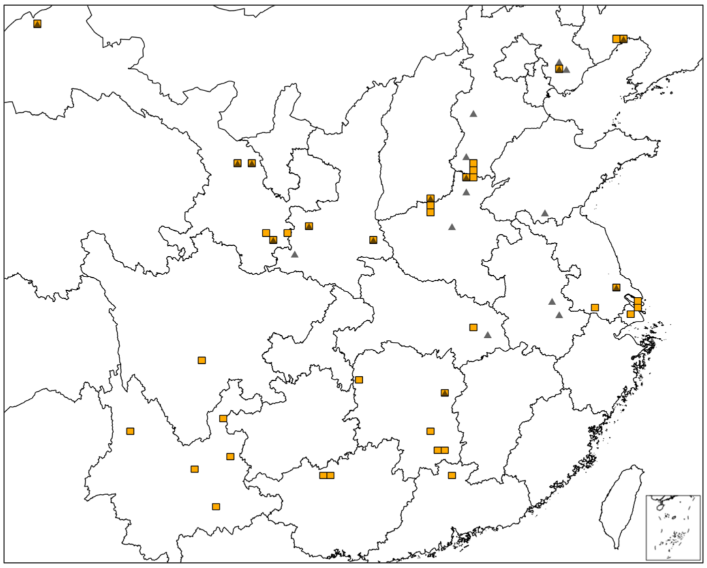


Figure S4 Comparation of hotspots between cumulative emissions and emissions at 2021. The triangle represents the 2021 atmospheric mercury emission hotspots, and the square represents the historical cumulative emission hotspots.

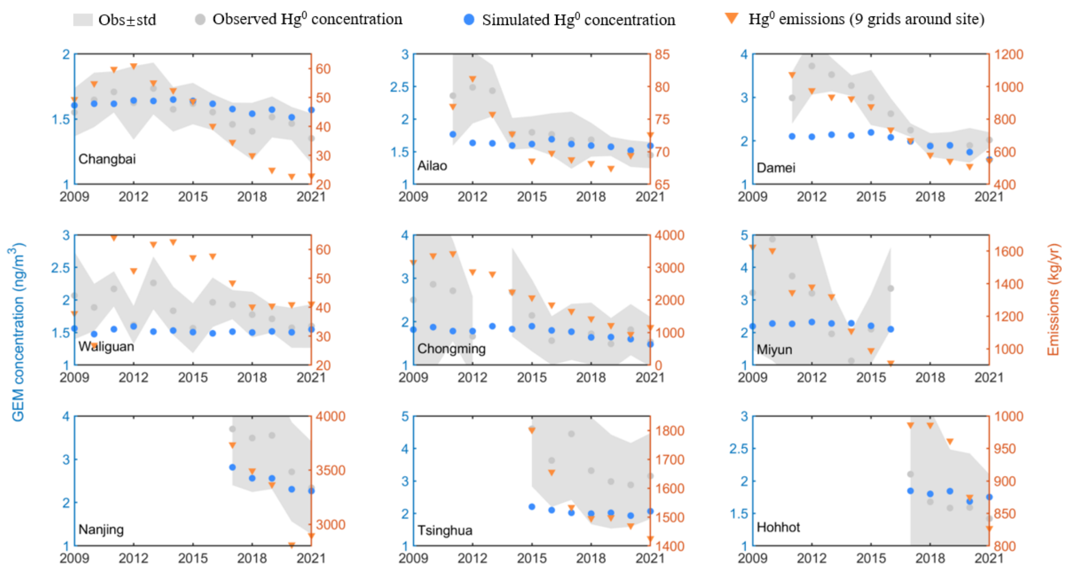
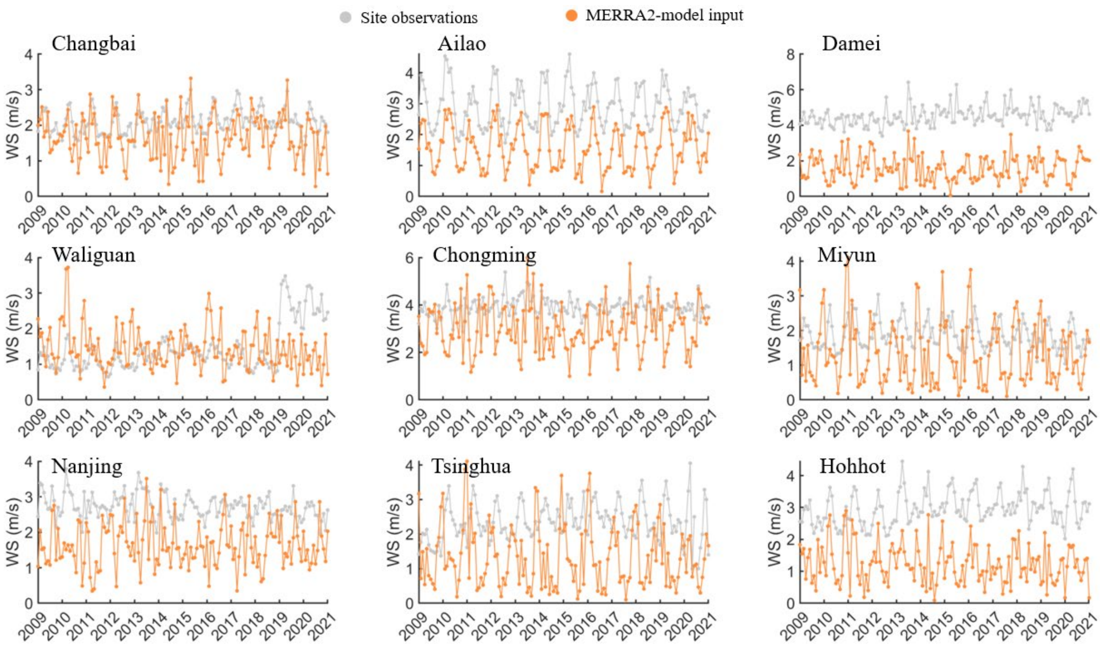


Figure S5 Long-term comparison of trends in annual observed and simulated Hg^0 concentration and emissions

89 

90 **Figure S6 Long-term comparison of trends in monthly observed and model input wind speed**

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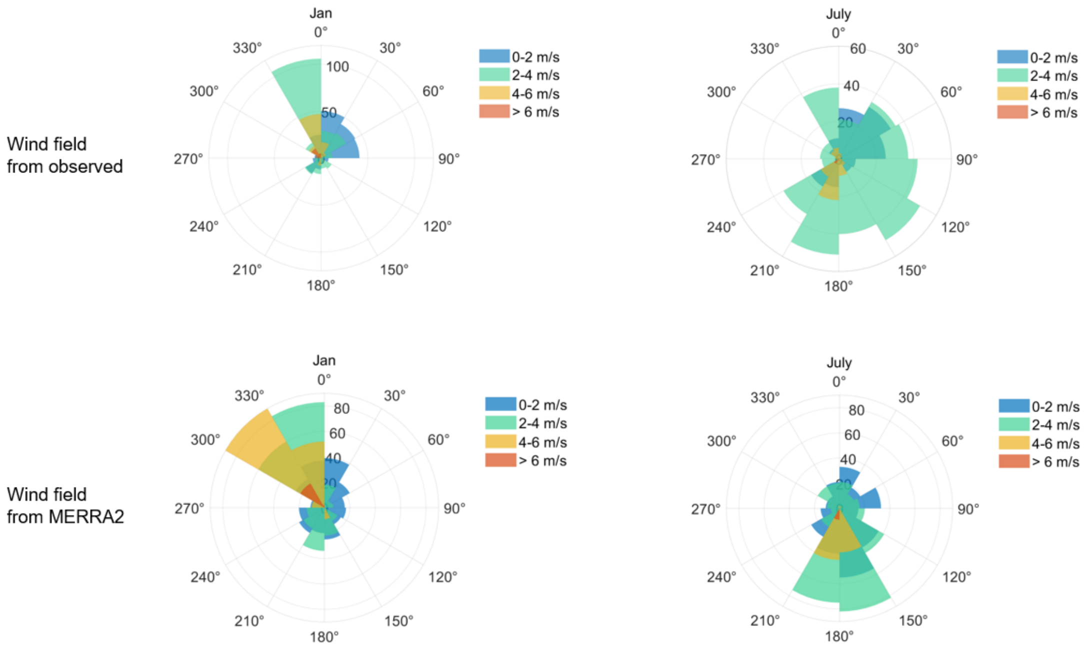
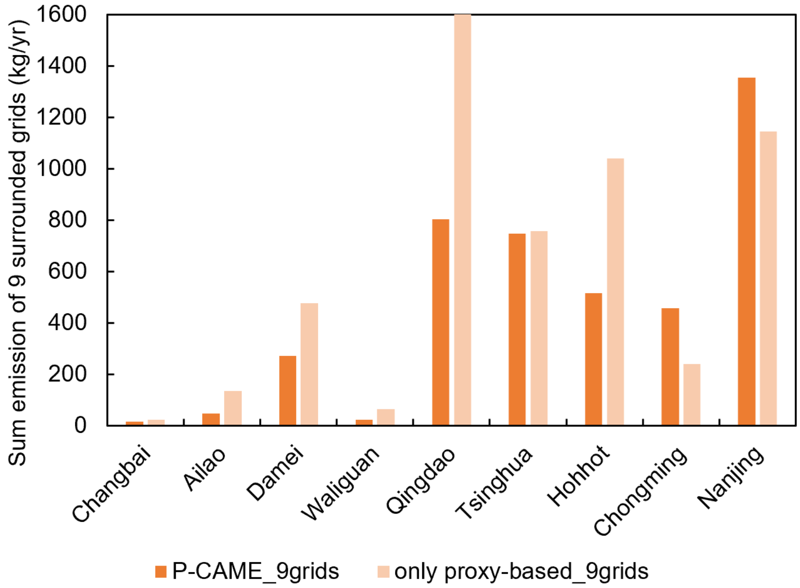


Figure S7 **Comparison of wind fields between observations and MERRA2 in Beijing**



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96 **Figure S8 Comparison of emissions from surrounding 9 grids between P-CAME and only proxy-**

97 **based inventory at 2020**

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