Supplement of Atmos. Chem. Phys., 22, 11187–11202, 2022
https://doi.org/10.5194/acp-22-11187-2022-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

Measurement report: Atmospheric mercury in a coastal city of Southeast China – inter-annual variations and influencing factors

Jiayan Shi et al.

Correspondence to: Lingling Xu (linglingxu@iue.ac.cn) and Jinsheng Chen (jschen@iue.ac.cn)

CWT regions identification

The potential source regions of GEM were defined as follows:

Regions	Region description
Fujian	Fujian province.
East China (EC)	Shanghai, Jiangsu, Zhejiang, Anhui, Jiangxi and Shandong provinces.
Southwest China (SWC)	Chongqing, Sichuan, Guizhou, Yunnan, and Tibet provinces.
Central China (CC)	Henan, Hubei and Hunan provinces.
North China (NC)	Beijing, Tianjin, Hebei, Shanxi and Inner Mongolia provinces.
Fujian	Fujian province.
Philippine sea and Taiwan Strait (PhiS+TW)	Philippine Sea and Taiwan island.
Philippine sea and East China Sea (PhiS+ECS)	Philippine Sea and East China Sea.
South China Sea (SCS)	South China Sea.

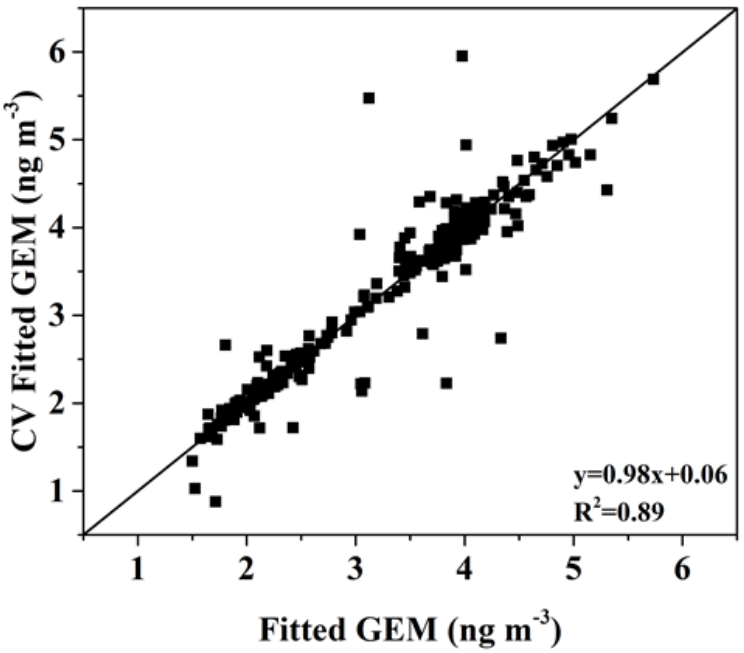

Figure S1. The results of 10-fold cross-validation test. The strong correlation, with a determination coefficient (R^2) of approximately 0.89, demonstrated the accuracy of GAMs

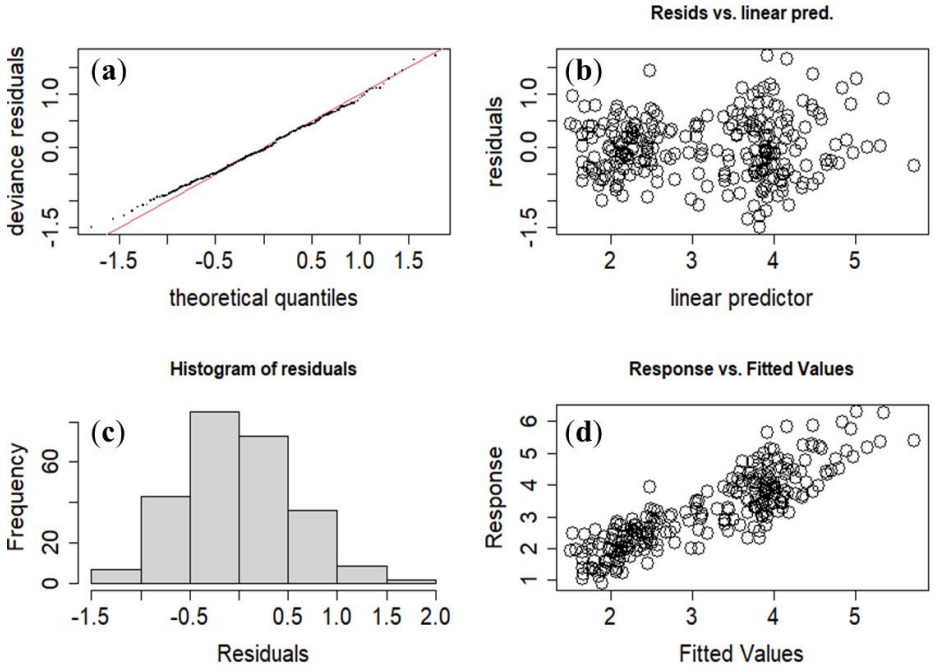

Figure S2. The assessment results of the GAMs: (a) quantile-quantile (QQ) plots, (b) The Resids vs. linear (c) Histogram of residuals (d) Response vs. Fitted Values

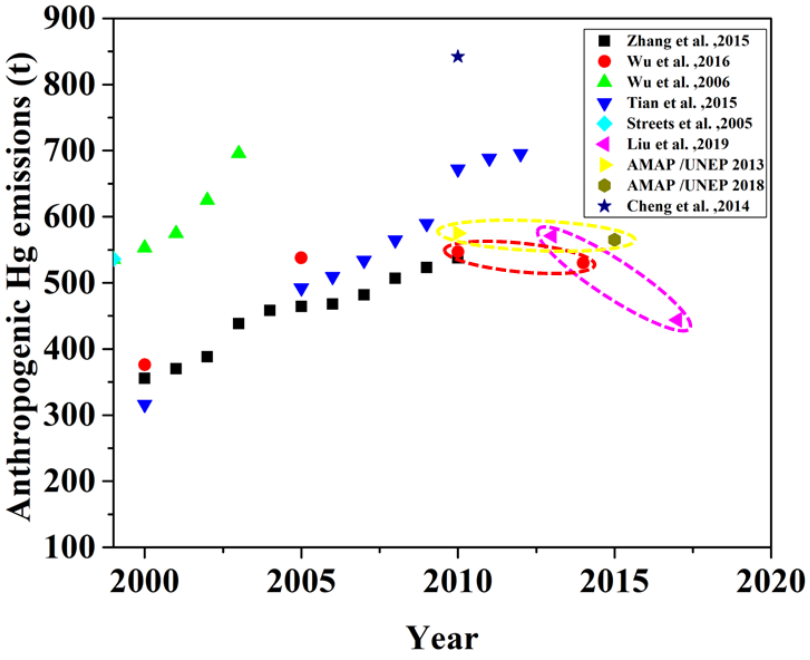

Figure S3. Anthropogenic mercury emissions from China reported in the literature (Streets et al., 2005; Wu et al., 2006; Cheng et al., 2015; Tian et al., 2015; Zhang et al., 2015; Wu et al., 2016; Liu et al., 2019).

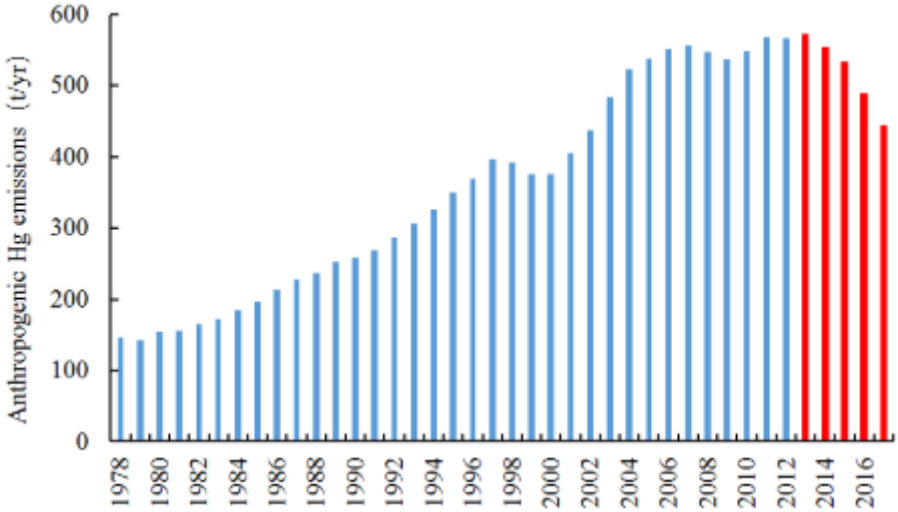

Figure S4. Anthropogenic mercury emissions during 1978-2017 in China (Liu et al., 2019).

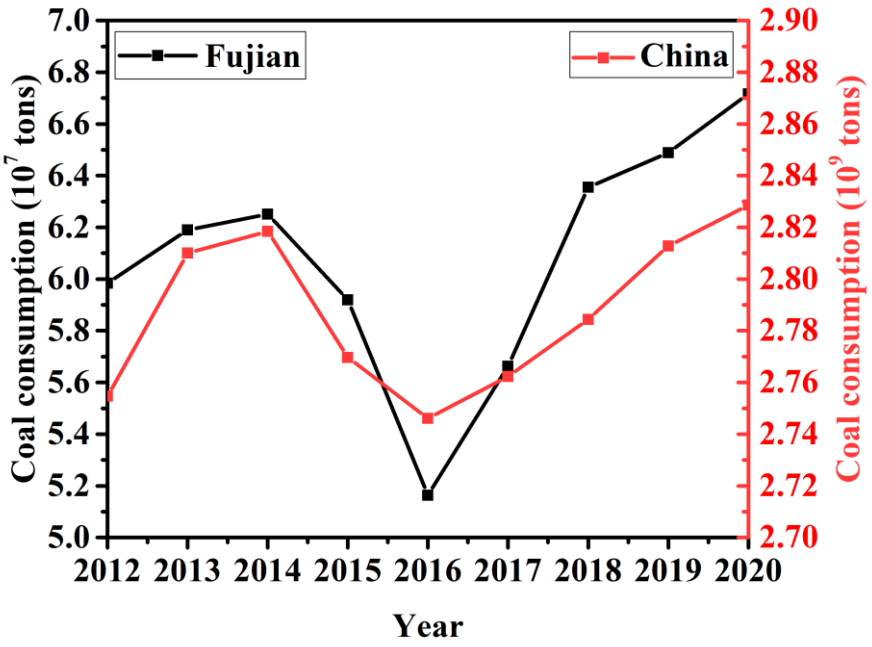

Figure S5. Statistics of coal consumption in China and Fujian Province during 2012 – 2020.

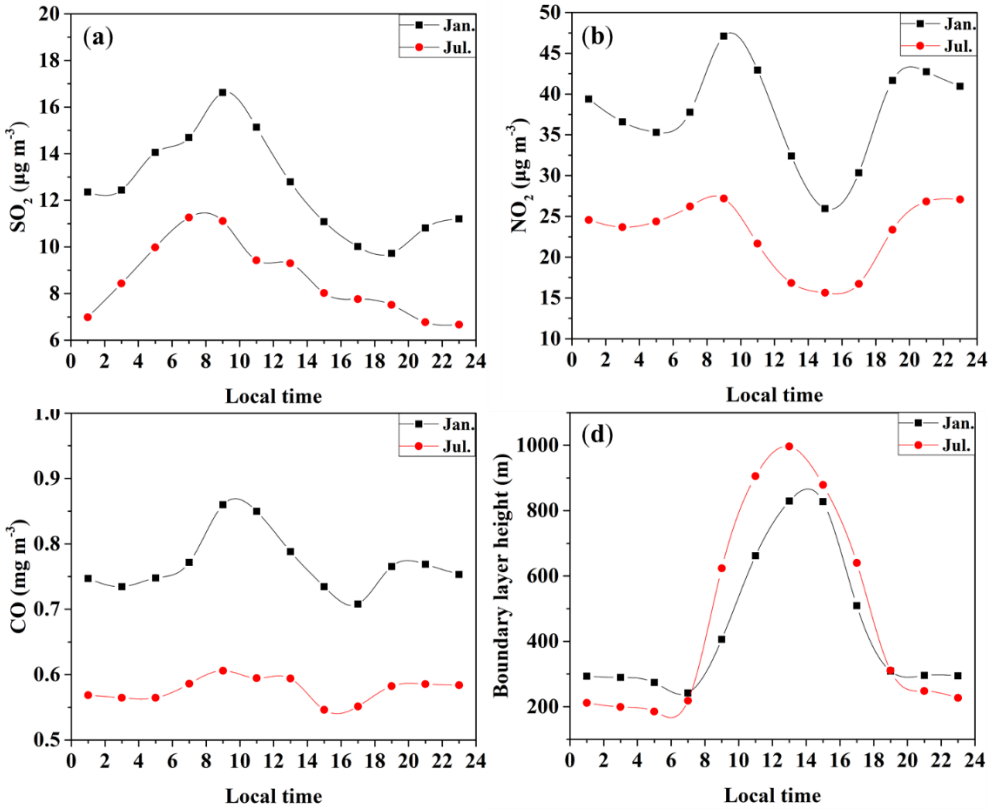

Figure S6. The diurnal trend of (a) SO_2, (b) NO_2, (c) CO and (d) boundary layer height in January and July during the whole study period.

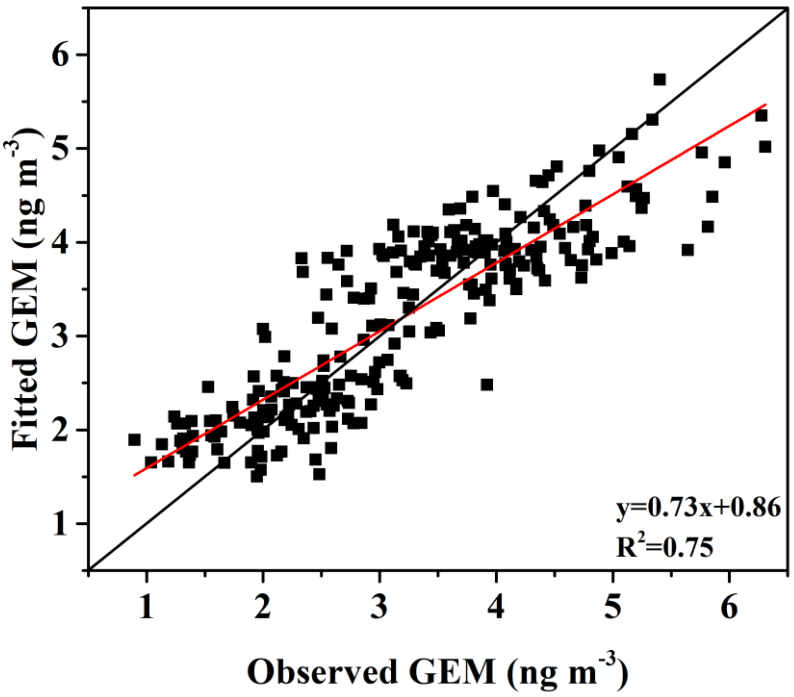

Figure S7. Relationships between the fitted and the observed GEM concentrations.

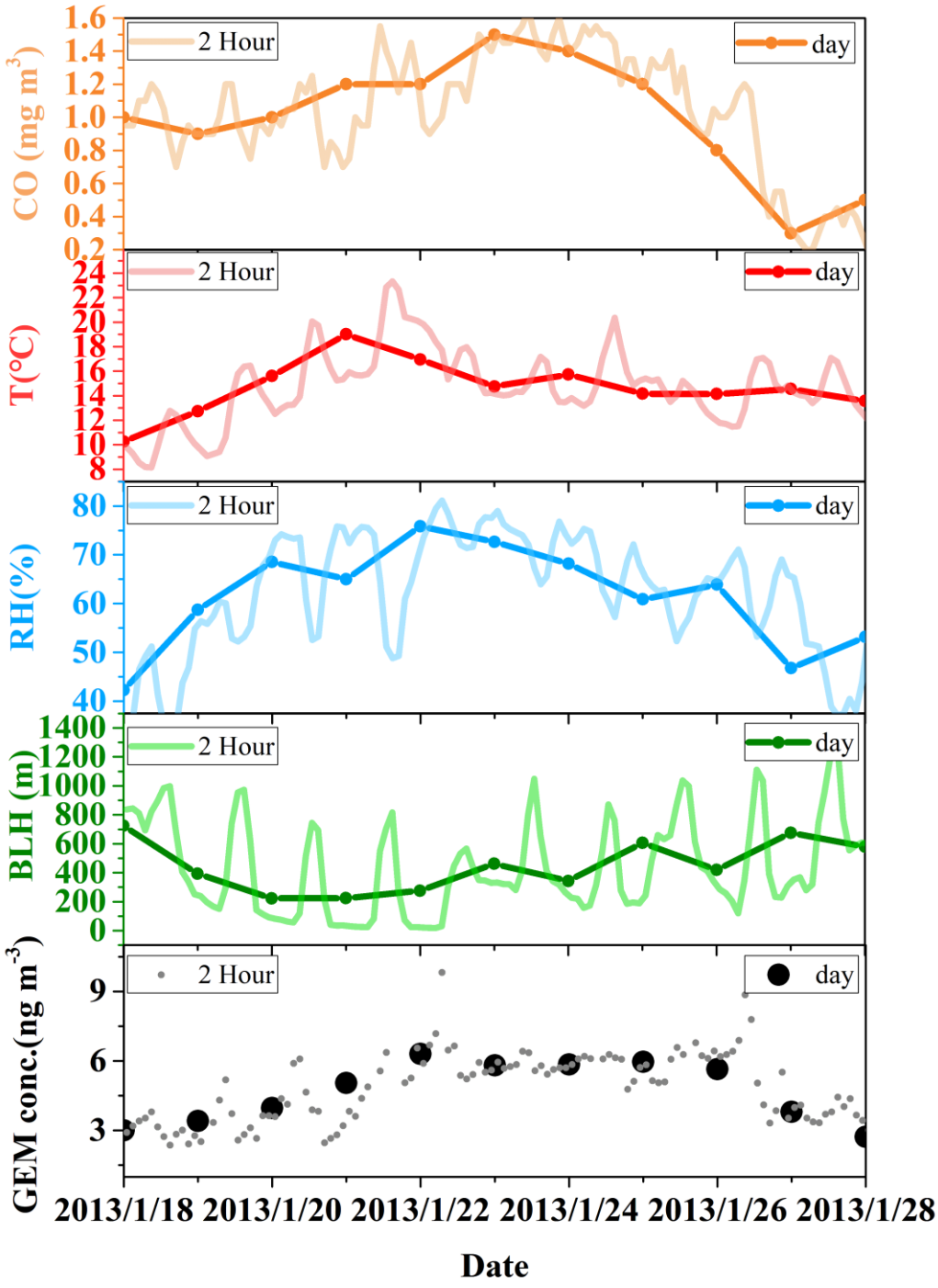

Figure S8. Time series of CO (ng m^{-3}), temperature (°C), relative humidity (%), boundary layer height (m) and GEM concentrations (ng m^{-3}) for a case study.

Table S1. Test variables in the GAMs.

Categories	Parameters	Data source
Emissions	SO_2、NO_2、O_3、CO、$PM_{2.5}$、PM_{10}	
Surface meteorology	Temperature Relative humidity Wind speed Wind direction Surface air pressure	Observation data from Xiamen air quality monitoring site
High-altitude meteorology	Boundary layer height Downward UV radiation at the surface Low cloud cover	European Centre for Medium-Range Weather Forecasts (ECMWF) reanalysis (https://www.ecmwf.int)
Air transmission	24h-Latitude 24h-Longitude	HYSPLIT model

Table S2. Final selected variables for the GAMs.

Variables	e.d.f	F	Approximate significance
s(CO)	6.68	6.65	<0.05
te (T, BLH, RH)	22.16	1.86	<0.05
te (24h-Latitude, 24h- Longitude)	10.09	2.60	<0.05

Note: e.d.f stands for estimated degree of freedom, F is an indicator for importance assessment, s () represents the smooth function, te () represents the interaction functions of tensors.

Table S3. Statistics of coal consumption and SO_2, NO*x* emissions in Fujian Province during 2012 – 2020

Year	Coal (10000 tons of SCE)	Coal for Households (10000 tons of SCE)	SO_2 (10000 tons)	NO*x* (10000 tons)
2012	5983.76	83.00	37.13	46.72
2013	6190.35	54.95	36.10	43.83
2014	6251.02	31.30	35.60	41.17
2015	5919.53	30.10	33.79	37.90
2016	5163.44	26.86	18.93	26.18
2017	5662.19	24.00	13.39	27.72
2018	6355.41	23.00	12.19	28.41
2019	6488.76	21.35	12.10	30.15
2020	6716.21	10.50	/	/

Table S4. Pearson correlations results of GEM and tracer pollutants in January and July in each study year.

Month (Season)	Year	SO_2	NO_2	O_3	CO	PM_{10}	$PM_{2.5}$
January (Winter)	2012	0.15^{*}	0.32^{**}	-0.33^{**}	0.14^{*}	0.11^{*}	/
	2013	0.38^{**}	0.35^{**}	-0.12^{*}	$\mathbf{0.53^{**}}$	0.45^{**}	$\mathbf{0.53^{**}}$
	2015	0.16^{**}	0.41^{**}	-0.28^{**}	$\mathbf{0.50^{**}}$	0.10	0.39^{**}
	2017	0.15^{**}	0.17^{**}	-0.25^{**}	/	0.19^{**}	0.26^{**}
	2020	0.32^{**}	$\mathbf{0.60^{**}}$	$\mathbf{-0.54^{**}}$	0.48^{**}	0.34^{**}	$\mathbf{0.53^{**}}$
July (Summer)	2012	0.44^{**}	0.34^{**}	-0.08	0.22^{**}	0.39^{**}	-0.04
	2013	0.23^{**}	0.18^{**}	-0.09	0.26^{**}	0.24^{**}	0.29^{**}
	2015	0.21^{**}	0.18^{**}	-0.18^{**}	0.25^{**}	0.20^{**}	0.12
	2017	0.18^{**}	0.38^{**}	-0.18^{*}	0.31^{**}	0.14	-0.01
	2020	/	0.19^{**}	-0.09	/	0.25^{**}	0.21^{**}

$P < 0.01$, **; $P < 0.05$, *; $r > 0.5$ are show in bold phase.

References

Cheng, K., Wang, Y., Tian, H. Z., Gao, X., Zhang, Y. X., Wu, X. C., Zhu, C. Y., and Gao, J. J.: Atmospheric Emission Characteristics and Control Policies of Five Precedent-Controlled Toxic Heavy Metals from Anthropogenic Sources in China, Environmental Science & Technology, 49, 1206-1214, https://doi.org/10.1021/es5037332, 2015.

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, Environ Sci Technol, 53, 8938-8946, https://doi.org/10.1021/acs.est.9b02428, 2019.

Streets, D., Hao, J., Wu, Y., Jiang, J., Chan, M., Tian, H., and Feng, X.: Anthropogenic mercury emissions in China, Atmospheric Environment, 39, 7789-7806, https://doi.org/10.1016/j.atmosenv.2005.08.029, 2005.

Tian, H. Z., Zhu, C. Y., Gao, J. J., Cheng, K., Hao, J. M., Wang, K., Hua, S. B., Wang, Y., and Zhou, J. R.: Quantitative assessment of atmospheric emissions of toxic heavy metals from anthropogenic sources in China: historical trend, spatial distribution, uncertainties, and control policies, Atmospheric Chemistry and Physics, 15, 10127-10147, https://doi.org/10.5194/acp-15-10127-2015, 2015.

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

Wu, Y., Wang, S., Streets, D. G., Hao, J., Chan, M., and Jiang, J.: Trends in anthropogenic mercury emissions in China from 1995 to 2003, Environ Sci Technol, 40, 5312-5318, https://doi.org/10.1021/es060406x, 2006.

Zhang, L., Wang, S., Wang, L., Wu, Y., Duan, L., Wu, Q., Wang, F., Yang, M., Yang, H., Hao, J., and Liu, X.: Updated Emission Inventories for Speciated Atmospheric Mercury from Anthropogenic Sources in China, Environmental Science & Technology, 49, 3185-3194, https://doi.org/10.1021/es504840m, 2015.