Supporting Information

Increases of Total Mercury and Methylmercury Releases from Municipal Sewage into Environment in China and Implications

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Supporting Figures

Figure S1. Eight regions in China. A is North China, B is Northeast China, C is East China, D is Central China, E is South China, F is Southwest China, G is Northwest China, and H is Tibetan region. South China Sea isn't included in the map.

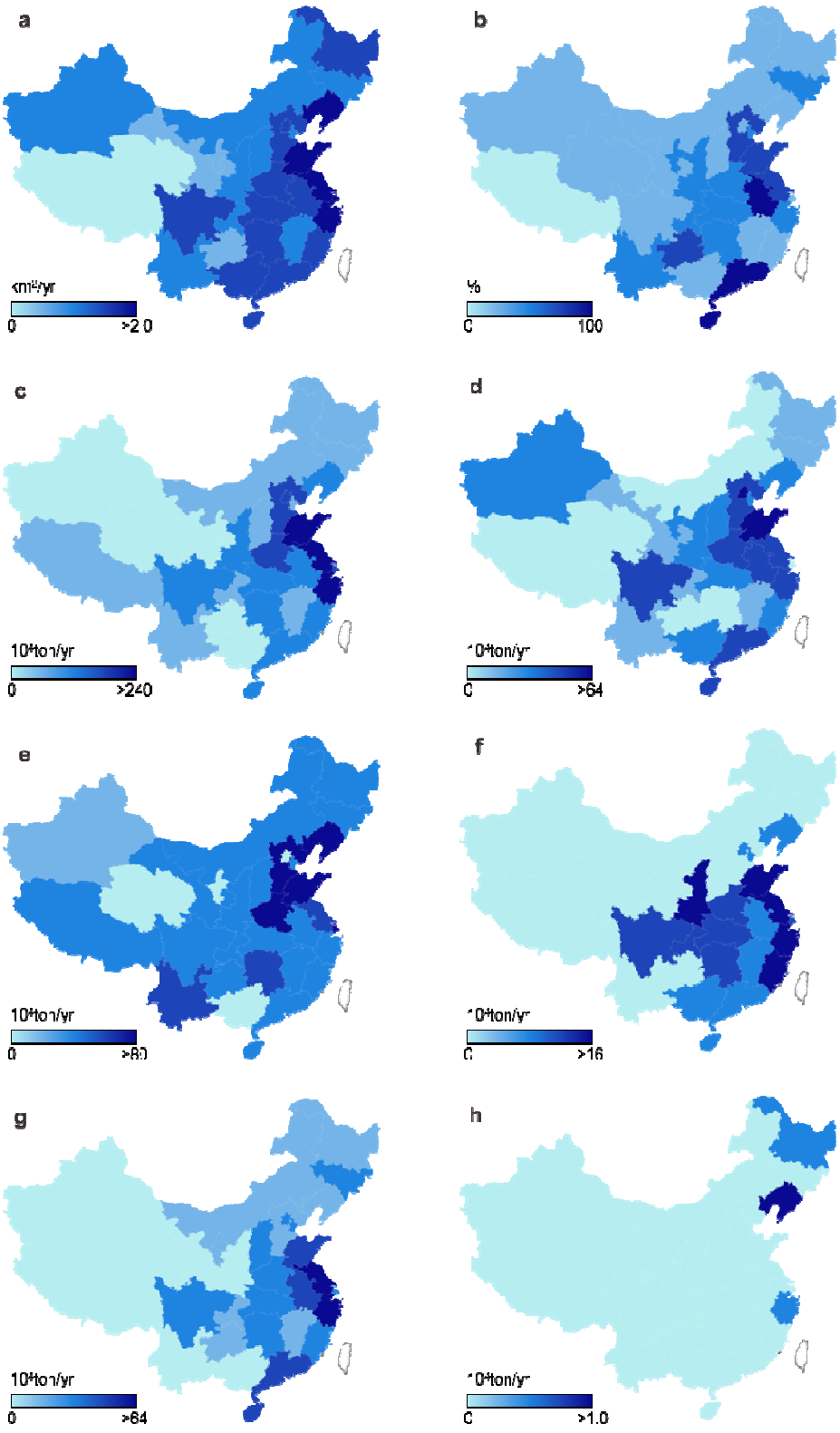

Figure S2. Basic data associated with the life cycle of municipal sewage in China in

2015 used in the material flow analysis. **a** is generation of municipal sewage in each province of China (*China Environmental Statistical Yearbook*);[1] **b** is treatment ratio of municipal sewage (*China Environmental Statistical Yearbook*); **c** is generation of sewage sludge (*China Environment Yearbook*);[2] **d** is sewage sludge discharged into landfill (*China Environment Yearbook*); **e** is sewage sludge discharged into cropland (*China Environment Yearbook*); **f** is sewage sludge used for building materials (*China Environment Yearbook*); **g** is incineration of sewage sludge (*China Environment Yearbook*); **h** is sewage sludge irregularly dumped into natural land (*China Environment Yearbook*).

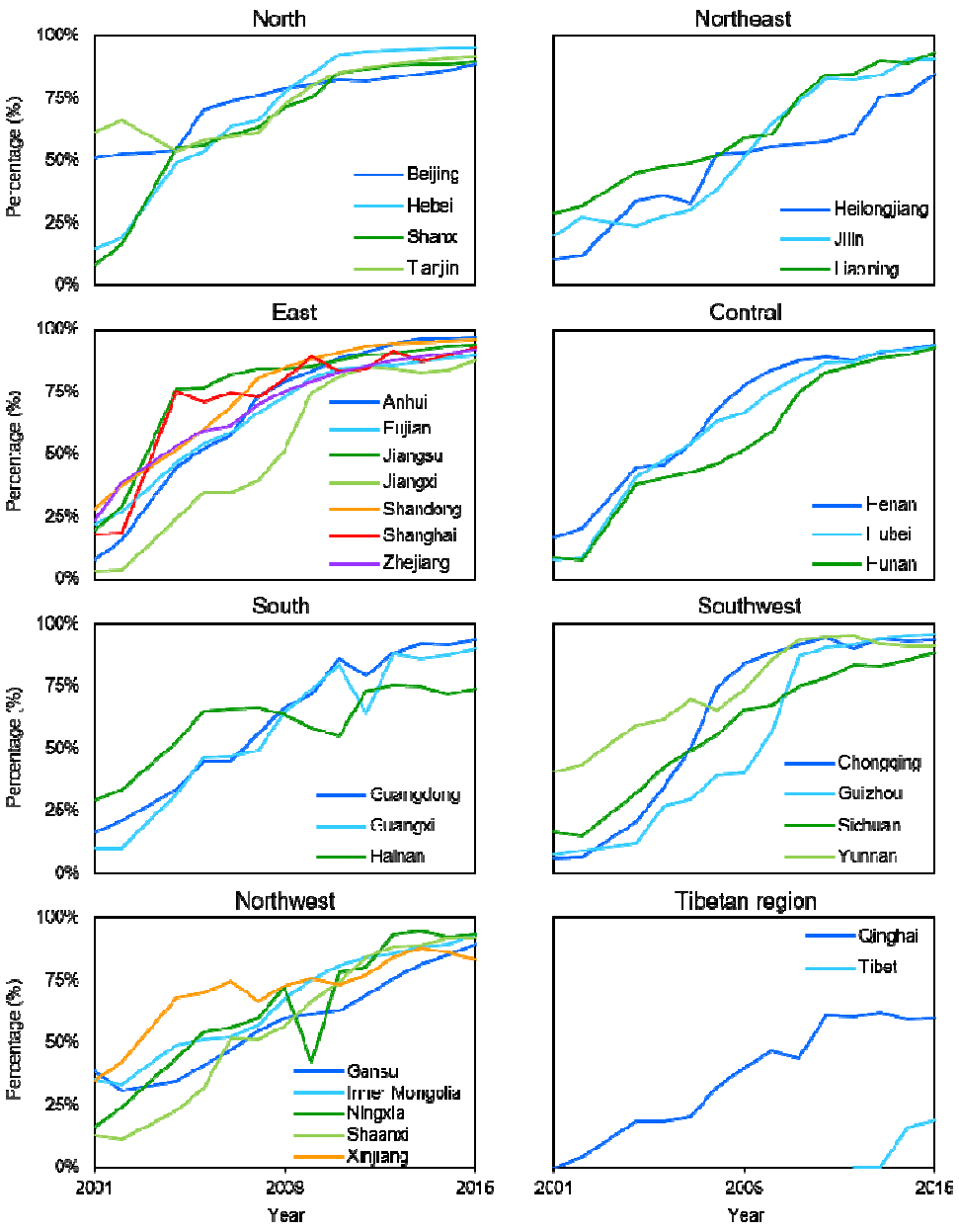

Figure S3. Trend of treatment ratio of municipal sewage of each province from 2001 to 2015. Data are referenced from the *China Environmental Statistical Yearbook*.[1]

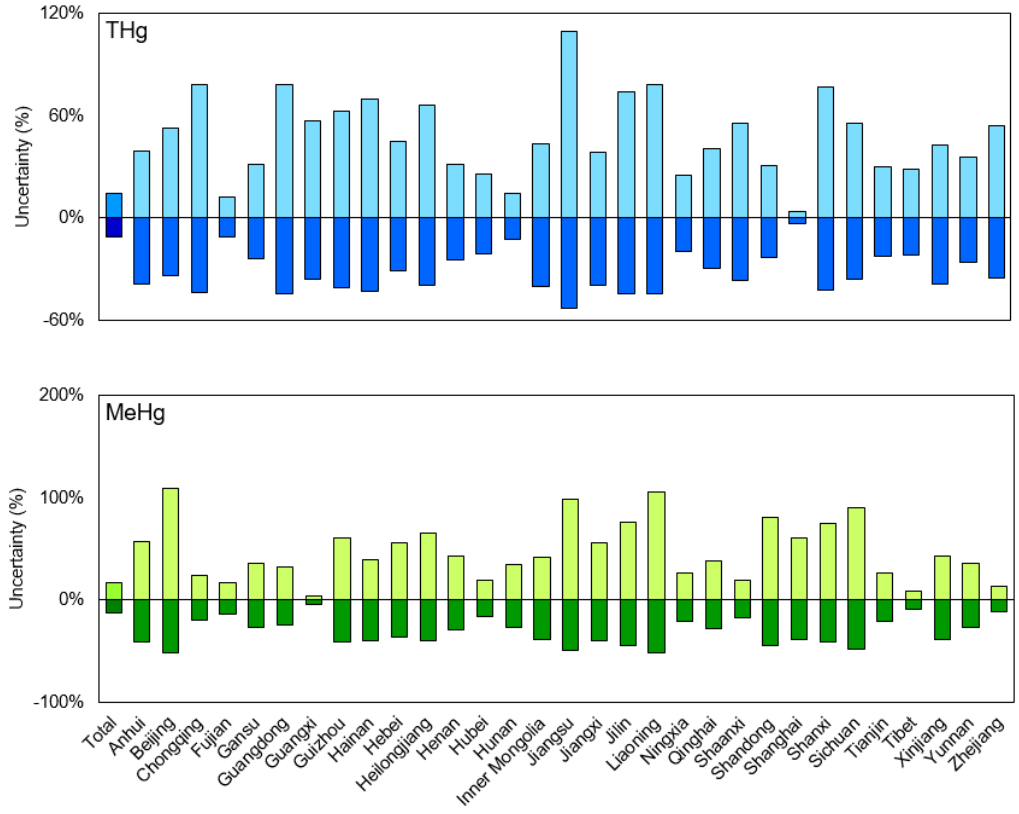

Figure S4. Uncertainties (P20-80 confidence interval) of THg and MeHg released from municipal sewage of each province in China in 2015.

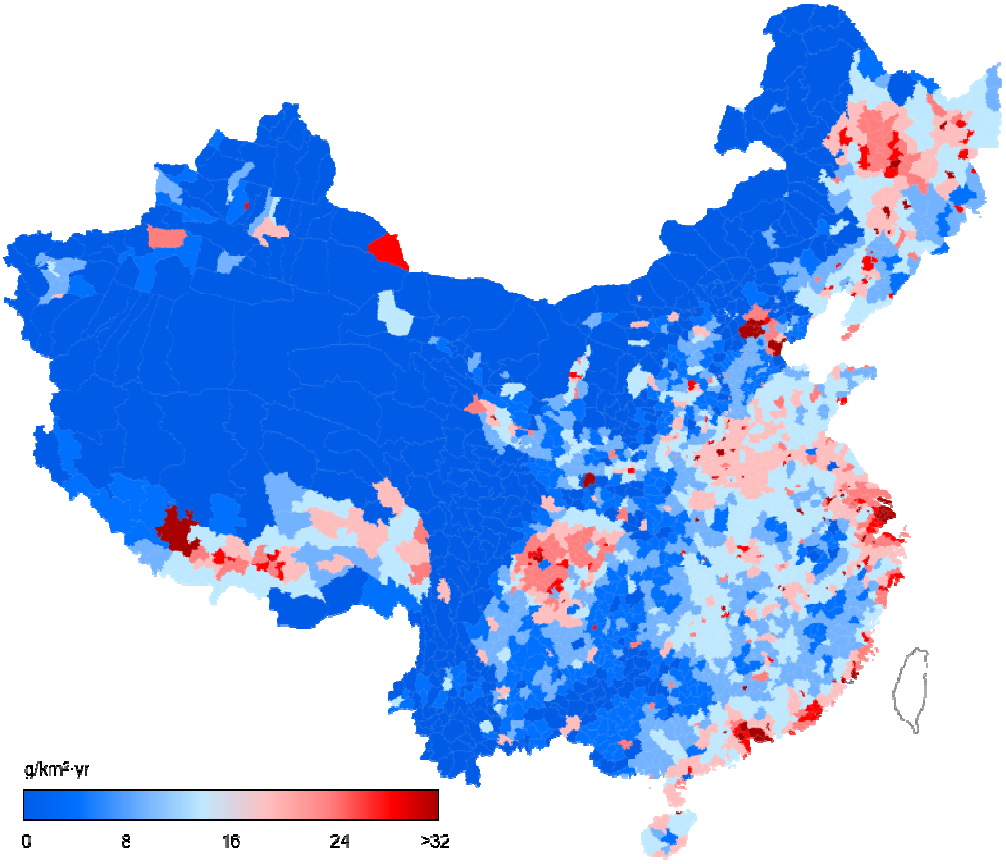

Figure S5. Distribution of THg released from municipal sewage into aquatic environment in China in 2015.

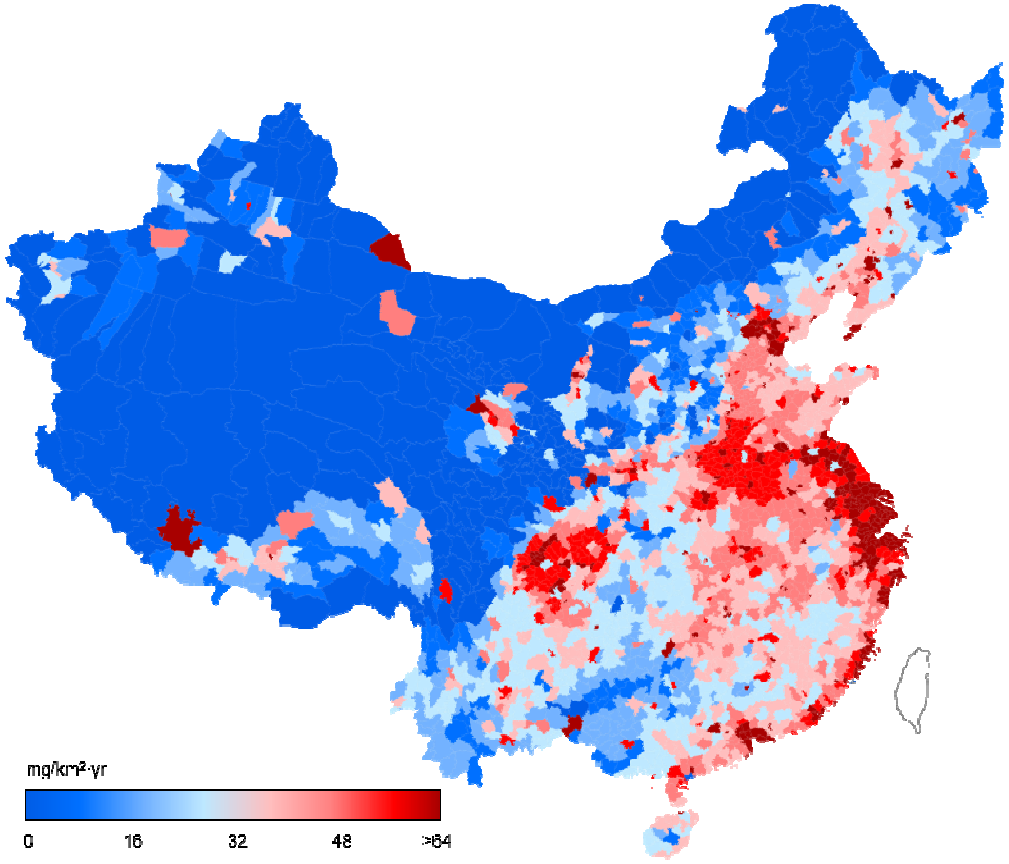

Figure S6. Distribution of MeHg released from municipal sewage into aquatic environment in China in 2015.

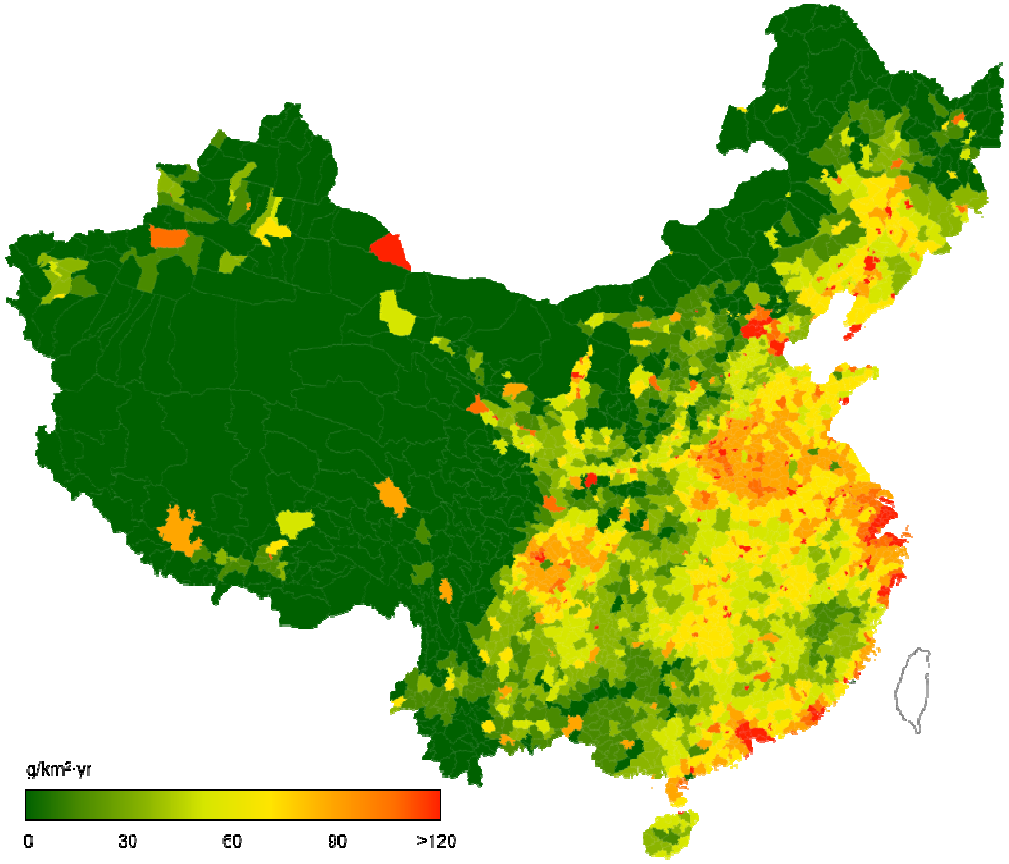

Figure S7. Distribution of THg released from municipal sewage into land in China in 2015.

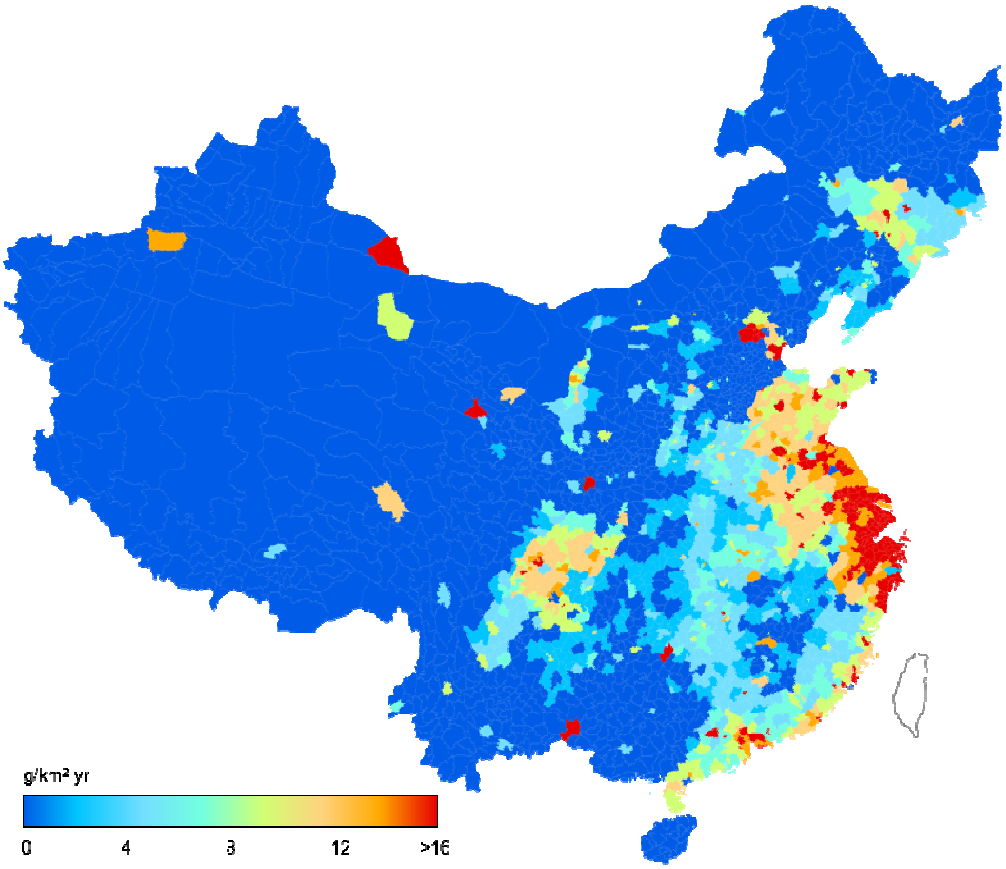

Figure S8. Distribution of THg emitted from municipal sewage into atmosphere in China in 2015.

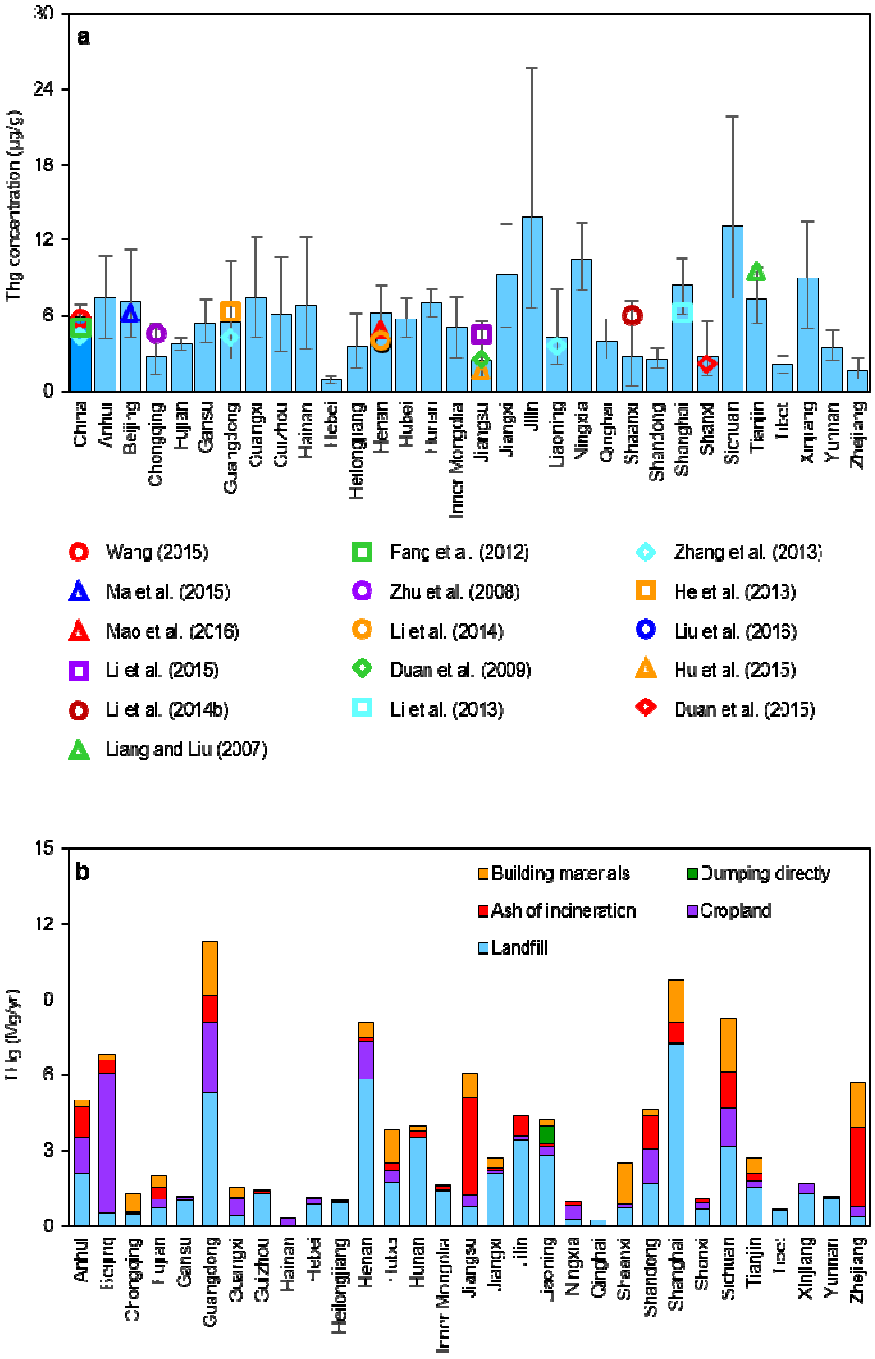

Figure S9. Modeling results of THg concentration in sewage sludge and compared with measurement data from published literatures (**a**), and THg released from municipal sewage into different sinks of land in each province in China in 2015 (**b**). Column and bar in figure **a** are median value and P20-80 confidence interval,

respectively. Measurement data are referenced from Wang (2015),[3] Fang et al. (2012),[4] Zhang et al. (2013),[5] Ma et al. (2015),[6] Zhu et al. (2008),[7] He et al. (2013),[8] Mao et al. (2016),[9] Li et al. (2014a),[10] Liu et al. (2016),[11] Li et al. (2015),[12] Duan et al. (2009),[13] Hu et al. (2015),[14] Li et al. (2014b),[15] Li et. (2013),[16] Duan et al. (2015),[17] Liang and Liu (2007).[18]

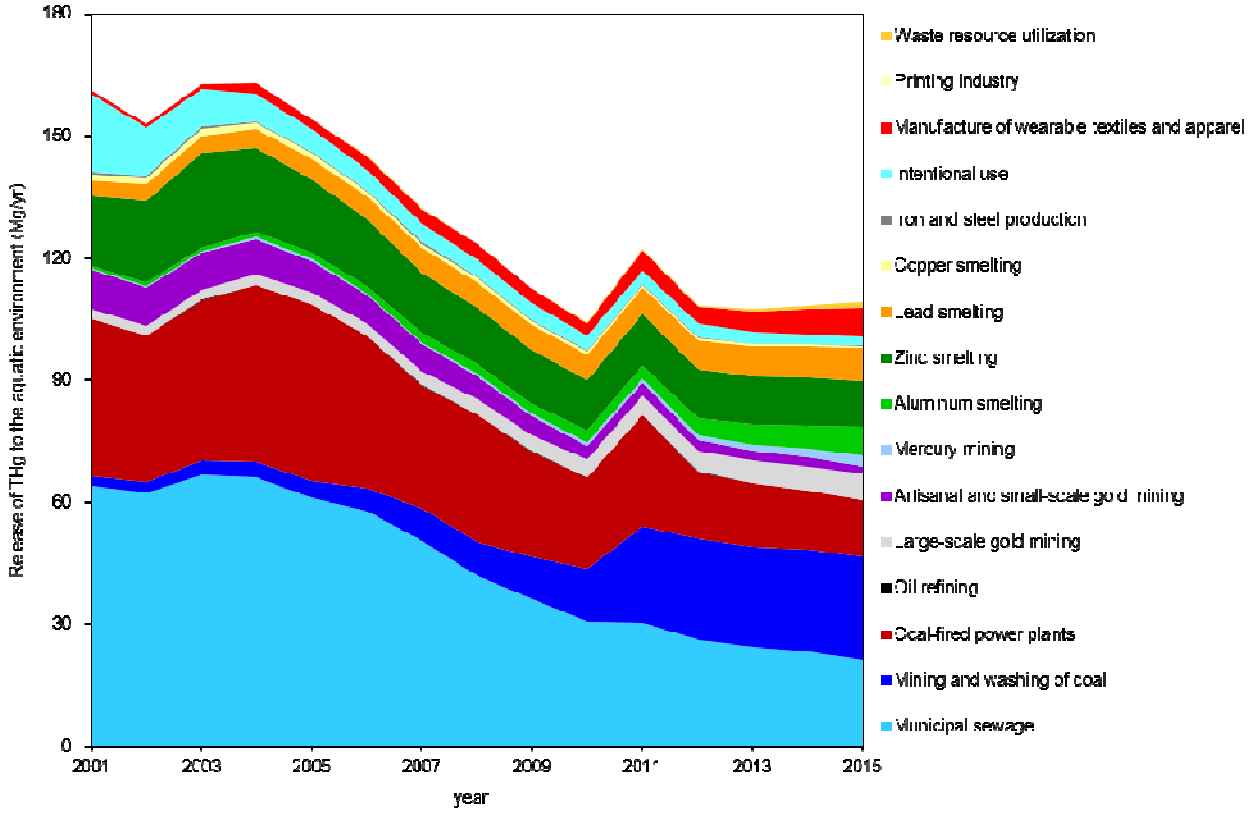

Figure S10. Updates of THg released from direct anthropogenic sources in China from 2001 to 2015.

Supporting Tables

Table S1. Municipal sewage treatment plants and sampling information

Region	Province	MSTP[a] ID	Influent (untreated sewage)	Effluent (treated sewage)	Treatment technology[b]	Sewage flow (m^3/d)	THg removal ratio (%)	MeHg removal ratio (%)
North China								
	Beijing	CP-1	√ [c]	√	A^2/O	20,000	98%	91%
	Beijing	CP-2	√	√	CAST	20,000	94%	90%
	Beijing	CY-1	√	√	MBR	100,000	100%	97%
	Beijing	CY-2	√	√	OD	40,000	98%	96%
	Beijing	CY-3	√	√	A^2/O	600,000	90%	93%
	Beijing	CY-4	√	√	OD	350,000	88%	73%
	Beijing	FT-1	√	√	A^2/O	80,000	93%	89%
	Beijing	FT-2	√	√	SBR	80,000	99%	91%
	Beijing	FT-3	√	√	A^2/O	43,000	93%	95%
	Beijing	HD-1	√	√	A^2/O	550,000	100%	90%
	Beijing	HD-2	√	√	A^2/O	21,000	96%	99%
	Beijing	TZ-1	√	√	ASP	950,000	98%	54%
	Tianjin	TG-1	√	× [d]	A/O	64,000	n/a[e]	n/a
	Hebei	SJZ-1	√	×	A^2/O	490,000	n/a	n/a
	Hebei	SJZ-2	√	×	ASP	160,000	n/a	n/a
	Shanxi	CZ-1	√	√	OD	160,000	85%	90%

	Shanxi	TY-1	√	×	A^2/O	150,000	n/a	n/a
	Shanxi	TY-2	√	×	A^2/O	70,000	n/a	n/a
Northeast China								
	Liaoning	SY-1	√	√	BAF	400,000	95%	89%
	Liaoning	SY-2	√	×	BP	400,000	n/a	n/a
	Liaoning	DL-1	√	×	CAST	80,000	n/a	n/a
	Liaoning	DL-2	√	×	BAF	100,000	n/a	n/a
	Heilongjiang	HEB-1	√	√	A/O	140,000	99%	97%
	Heilongjiang	HEB-2	√	√	A/O	290,000	96%	91%
	Heilongjiang	HEB-3	√	√	CAST	27,000	96%	85%
East China								
	Fujian	XM-1	√	√	BAF	260,000	87%	86%
	Fujian	XM-2	√	√	OD	190,000	96%	54%
	Jiangsu	NJ-1	√	×	A^2/O	640,000	n/a	n/a
	Jiangsu	NJ-2	√	×	SBR	58,000	n/a	n/a
	Jiangsu	SZ-1	√	×	ASP	140,000	n/a	n/a
	Jiangsu	SZ-2	√	×	ASP	110,000	n/a	n/a
	Jiangsu	SZ-3	√	×	ASP	42,000	n/a	n/a
	Shandong	JN-1	√	×	A^2/O	340,000	n/a	n/a
	Shanghai	CN-1	√	√	SO	75,000	99%	96%
	Shanghai	HK-1	√	√	A/O	52,000	96%	66%
	Zhejiang	HZ-1	√	√	A^2/O	380,000	94%	78%
Central China								
	Henan	LY-1	√	√	A^2/O	190,000	96%	78%
	Henan	LY-2	√	×	A^2/O	160,000	n/a	n/a
	Hubei	WH-1	√	√	A^2/O	300,000	95%	86%

	Hunan	CS-1	√	√	A^2/O	n/a	97%	70%
South China								
	Guangdong	GZ-1	√	√	A^2/O	1,200,000	96%	88%
	Guangdong	SZ-1	√	×	A^2/O	750,000	n/a	n/a
	Guangdong	SZ-2	√	×	A^2/O	290,000	n/a	n/a
	Guangxi	NN-1	√	×	ASP	310,000	n/a	n/a
	Guangxi	NN-2	√	×	ASP	290,000	n/a	n/a
Southwest China								
	Chongqing	SPB-1	√	√	BP	18,000	95%	86%
	Sichuan	CD-1	√	√	BAF	210,000	93%	87%
	Sichuan	XC-1	√	√	OD	42,000	94%	91%
	Yunnan	KM-1	√	√	A^2/O	300,000	94%	95%
Northwest China								
	Gansu	LZ-1	√	×	A^2/O	50,000	n/a	n/a
	Gansu	LZ-2	√	×	ASP	170,000	n/a	n/a
	Gansu	LZ-3	√	×	SO	130,000	n/a	n/a
	Gansu	LZ-4	√	×	A^2/O	40,000	n/a	n/a
	Ningxia	YC-1	√	√	SBR	100,000	97%	93%
	Ningxia	YC-2	√	√	CAST	70,000	95%	89%
	Ningxia	YC-3	√	√	A^2/O	200,000	93%	81%
	Shannxi	XA-1	√	√	OOD	170,000	95%	89%
	Shannxi	XA-2	√	√	A^2/O	380,000	95%	89%
	Shannxi	XA-3	√	√	A^2/O	n/a	97%	92%
Tibetan region								
	Qinghai	XN-1	√	√	A^2/O	74,000	97%	93%
	Qinghai	XN-2	√	√	A^2/O	56,000	98%	96%

Tibet	LS-1	√	√	n/a	50,000	99%	96%

Note: a) municipal sewage treatment plants; b) A/O: anoxic/oxic process, A^2/O: anaerobic-anoxic-oxic process, ASP: activated sludge process, BAF: biological aerated filter, BP: biomembrance process, CAST: cyclic activated sludge technology, MBR: membrane Bio-Reactor, OD: oxidation ditch, SO: secondary oxidation, SBR: sequencing batch reactor, OOD: orbal oxidation ditch; c) have sample; d) no sample; e) not available.

Table S2. Comparison of THg and MeHg concentrations with previous studies

Region	Influent (untreated sewage)			Effluent (treated sewage)			Reference
	n[a]	THg[b] (ng/L)	DHg[c] (ng/L)	n	THg (ng/L)	DHg (ng/L)	
Beijing, North China		560-14,000	n/d[d]		n/d	n/d	Shi et al. (2007)[19]
Guangdong, South China		n/d	10-310		n/d	n/d	Zhao et al. (2014)[20]
Henan, Central China		260-2,800	n/d		2.7-410	n/d	Mao et al. (2016)[9]
Henan, Central China		3,100	n/d		170	n/d	Chen et al. (2006)[21]
Henan, Central China		530-2,300	11-61		11-29	3.1-6.9	Li et al. (2014a)[10]
Minnesota, USA		170	n/d		3.5	n/d	Balogh and Nollet (2008)[22]
Onondaga, USA		310	n/d		25	n/d	Gbondo-Tugbawa et al. (2010)[23]
Winnipeg, Canada		61	n/d		7.0	n/d	Bodaly et al. (1998)[24]
Sao Paulo, Brazil		130	n/d		50	n/d	Da et al. (2007)[25]
North China	18	4,500±3,200[e]	33±39	13	210±220	23±20	This study
Northeast China	7	5,000±4,500	20±5.0	4	170±180	33±3.3	This study
East China	11	3,300±3,000	24±5.7	5	150±120	40±18	This study
Central China	4	3,200±1,300	26±6.1	2	130±55	31±2.0	This study
South China	5	2,100±1,600	43±47	1	93±74	17±1.5	This study
Southwest China	4	3,600±3,000	45±22	3	230±230	33±7.9	This study
Northwest China	10	3,200±1,400	140±90	3	140±62	47±33	This study
Tibetan region	3	6,900±6,200	50±33	3	85±43	28±5.5	This study
China	62	3,400±2,600	39±24	34	160±130	33±12	This study
		TMeHg[f] (ng/L)	DMeHg[g] (ng/L)		TMeHg (ng/L)	DMeHg (ng/L)	

Henan, Central China		1.5-9.6	n/d		0.10-1.0	n/d	Mao et al. (2016)[9]
Henan, Central China		6.7-50	0.55-8.1		0.42-1.2	0.13-0.62	Li et al. (2014a)[10]
Onondaga, USA		5.1	n/d		1.5	n/d	Gbondo-Tugbawa et al. (2010)[23]
Winnipeg, Canada		2.2	n/d		0.20	n/d	Bodaly et al. (1998)[24]
North China	18	8.5±9.4	4.3±6.4	13	0.74±0.89	0.45±0.61	This study
Northeast China	7	5.3±4.6	2.7±1.6	4	0.58±0.64	0.98±0.80	This study
East China	11	6.9±6.3	4.1±2.8	5	1.4±1.2	1.1±0.65	This study
Central China	4	5.7±3.2	2.2±1.1	2	1.2±0.70	0.41±0.35	This study
South China	5	2.5±0.65	2.0±0.47	1	0.31±0.28	0.25±0.10	This study
Southwest China	4	11±10	5.7±4.6	3	1.1±1.3	1.4±1.4	This study
Northwest China	10	4.6±3.7	1.8±1.5	3	0.50±0.37	0.60±0.26	This study
Tibetan region	3	14±4.6	1.6±1.1	3	0.69±0.35	0.35±0.22	This study
China	62	6.5±5.5	3.4±2.6	34	1.0±0.82	0.80±0.59	This study

Note; a) number of MSTPs; b) total Hg, calculated by dissolved Hg (ng/L) + particulate Hg (ng/L); c) dissolved Hg; d) no data; e) mean value ± standard deviation; f) calculated by dissolved MeHg (ng/L) + particulate MeHg (ng/L); g) dissolved MeHg.

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