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Evaluating the effects of China's pollution controls on inter-annual trends and uncertainties of atmospheric mercury emissions

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SUPPLEMENT

Number of tables: 7

Number of figures: 4

Table S1. Database of Hg release ratios by combustion type.

Combustion Type	Release Ratios (%)	References
PC	99.6	Zhou et al. (2008)
	98.2 98.2 98.1 98.1	Luo (2009)
	99.6 100 92.5	X Yang et al. (2007)
	99.1 99.8 99.9	Zhu et al. (2001)
	99.5 99.8	Wang et al. (2008a)
	95.9 94.2 96.3 98.4 99.7 99.8	Chen et al. (2006)
	99.96 99.04 99.98 99.93 100	Wang et al. (2010a)
	95.4 99.9	Wang et al. (2008b)
	99.92 99.91	Xu et al. (2010)
	99.99 100 99.96 97.5	Wang et al. (2011)
Grate	93.75	Wang et al. (2003)
	98	Streets et al. (2005)
CFB	96	L Yang et al. (2007)
	100	Wang et al. (2010a)
	99	Tian et al. (2012)

Table S2. Database of Hg removal efficiencies by air pollutant control device (APCD).

Control Device	Removal Efficiencies (%)	References
ESP[a]	10.78 1.62	Wang et al. (2008b)
	13.5	Zhou et al. (2008)
	24.4 36.16 43.33 15.41	Luo (2009)
	38.7	Wang et al. (2008c)
	0.4 32.78 23.46	L Yang et al. (2007)
	10.8 4.7 33.1	Xu et al. (2010)
	31.1 31.2 24.5	Zhu et al. (2001)
	6.0 20.0 4.0	Wang et al. (2008a)
	24.8	Duan et al. (2005)
	35.7	X Yang et al. (2007)
	2.17 8.75 23.4	Wang et al. (2011)
	36.5 46.4 10.3 6.1 23.8	Wang et al. (2010a)
	25.2	Li (2011)
	2.61 5.83 7.91 15.1	C Wu et al. (2010)
	23.1 23.8 29.4 17.8 30.3 32.2	Chen et al. (2006)
	12.8 17.4	Li et al. (2013)
	29.4	Shi et al. (2013)
ESP[b]	50.6	L Yang et al. (2007)
	59.6	Gao et al. (2007)
	18.5	Wang et al. (2010a)
ESP+FGD	67.8 70.0 81.36 27.5 35.5	Wang et al. (2010a)
	20.9 33.9 69.4 70.6 62.1 74.2	Hu (2009)
	19.1 2.1	Wang et al. (2008c)
	9.6	L Yang et al. (2007)
	73.5 84.9 54.4	Xu et al. (2010)
	62.3 4.4 95.9 44.6	Wang et al. (2011)
	67.3 78.3	Li (2011)
	62.2 55.3 69.5 65.2	C Wu et al. (2010)
	68.7	Shi et al. (2013)
	25.4 38.4	Li et al. (2013)

Table S2 (continued)

Control Device	Removal Efficiencies (%)	References
FF	75.8	L Yang et al. (2007)
	84.0 20.7	Zhang et al. (2008)
	76.9 23.0	Wang et al. (2008a)
WET	8.7	Kilgroe et al. (2001)
	4.3	Chu and Porcella (1995)
	26	Afonso and Senior (2001)
CYC	0.1	Kilgroe et al. (2002)
	12	Huang et al. (2004)
	0	US EPA (1997)
SCR+FGD / SCR+ESP+FGD	82.2	Li (2011)
	36.5	Wang et al. (2010a)
	85	Wang et al. (2010b)
	82.2	Zhi et al. (2013)
	95	Cheng et al. (2009)
	74.1	Chen et al. (2008)
	81	Unpublished data from Jiangsu Environmental Monitoring Center

[a] ESP applied with PC (pulverized combustion); [b] ESP applied with CFB (circulating fluidized bed).

Table S3. Database of Hg emission factors of non-ferrous smelting by technology.

Metal	Smelting Process	Mercury emission factor (g/t)			
		Li et al. (2010)	Wang et al. (2010c)	Zhang et al. (2012)	Wu et al. (2012)
Zn	EP	5.70	0.50	0.57	0.59
	ISP	122.00		2.98	6.02
	RZSP	34.00			6.16
	EZF				13.80
	AZ	75.00			45.75
Pb	RPSP			1.00	1.19
	ISP				6.07
	SMP			0.49	10.16
	SPP				29.35
Cu	FFSP			0.23	0.26
	RPSP			0.09	0.28
	RLEP				0.38
	IFSP				1.07
	EF				14.96

Table S4. Database of mass fractions of Hg speciation by source.

Source category		Mercury speciation (%) (Hg^{2+} Hg^{p} Hg^{0})	References
Coal combustion	ESP	(11.4 0.0 88.6) (17.1 0.0 82.9)	Wang et al. (2008b)
		(53.5 0.1 46.4)	Zhou et al. (2008)
		(39.5 0.1 60.4)	Wang et al. (2008c)
		(11.4 0.0 88.6) (45.8 0.0 54.2) (8.1 0.0 91.9)	L Yang et al. (2007)
		(30.5 0.9 68.6) (30.2 0.0 69.8) (66.1 2.4 31.5)	Xu et al. (2010)
		(8.1 0.0 91.9) (44.7 0.0 55.3) (17.1 0.0 82.9)	Wang et al. (2008a)
		(74.0 0.0 26.0)	Duan et al. (2005)
		(7.5 0.0 92.5)	X Yang et al. (2007)
		(67.5 0.1 32.4) (49.4 0.0 50.6) (64.0 0.0 36.0) (14.3 0.7 85.0) (30.6 0.0 69.4)	Wang et al. (2010a)
	ESP+FGD	(24.8 0.0 75.2) (18.3 0.0 81.7) (8.9 0.0 91.1) (6.2 0.0 93.8) (11.5 0.0 88.5)	Wang et al. (2010a)
		(5.4 0.0 94.6) (3.9 0 96.1)	Wang et al. (2008b)
		(4.0 0.0 96.0)	L Yang et al. (2007)
		(27.7 0.6 71.7) (14.9 0.0 85.1) (53.7 2.5 43.8)	Xu et al. (2010)
	FF	(65.4 24.5 10.1)	Zhang et al. (2008)
		(74.5 8.8 16.7) (75.6 0.0 24.4)	Wang et al. (2008a)
		(78 10 12)	L Yang et al. (2007)
	SCR+ESP / SCR+ESP+FGD	(10.7 0.0 89.5)	Wang et al. (2010a)
		(20.9 0.0 79.1) (15.6 0.0 84.4) (16.6 0.0 83.4) (17.7 0.0 82.3)	Zhong et al. (2010)
		(84.1 0.0 15.9)	Chen et al. (2008)
	CYC/WET	(78.0 2.0 20.0)	Streets et al. (2005)
		(36.0 25.0 39.0)	Y Wu et al. (2010)
Non-ferrous metal smelting	Zinc	(90.0 4.0 6.0)	Wang et al. (2010c)
		(58.0 0.0 42.0) (61.0 0.0 39.0)	Zhang et al. (2012)
		(70.0 1.0 19.0)	Wu et al. (2012)
	Pb	(39.0 0.0 61.0) (40.0 0.0 60.0)	Zhang et al. (2012)
	Cu	(32.0 0.0 68.0) (68.0 0.0 32.0)	Zhang et al. (2012)
Solid wastes burning		(95.5 0.4 4.1) (63.8 2.6 33.6) (57.7 1.3 41.0) (69.9 4.0 26.1) (98.2 0.8 1.0) (94.9 0.4 4.7) (98.4 0.2 1.4) (88.3 0.1 11.6) (97.3 0.4 2.3) (95.8 0.2 4.0)	Chen et al. (2013)
Biomass burning		(13 40 47) (5 3 92) (0 34 66) (1 1 98) (0 1 99) (4 11 85) (15 21 64) (0 0 100) (10 14 76) (1 11 88) (7 6 87) (3 12 85) (12 4 84) (3 11 86) (10 25 65) (5 27 68) (9 37 54) (10 15 75) (0 46 54) (14 15 71) (10 11 79) (0 25 75) (1 28 71) (3 32 65) (0 28 72)	Zhang et al. (2013)

Table S5. National annual emissions of Hg^0 by source category from 2005 to 2012.

Source category	2005	2006	2007	2008	2009	2010	2011	2012
Coal-fired power plants	**85.8**	**94.3**	**100.1**	**98.7**	**101.5**	**103.2**	**115.9**	**107.7**
Industry	**255.9**	**264.9**	**279.1**	**282.6**	**287.8**	**286.9**	**285.3**	**276.0**
Cement production	28.2	28.4	28.5	26.2	26.0	21.5	19.2	18.6
Coal use	7.4	7.7	7.8	7.5	7.8	7.5	7.4	7.3
Iron & steel plants	11.4	12.7	13.1	13.7	15.1	16.3	17.6	18.1
Heating boilers	5.3	6.0	7.1	7.6	7.7	8.9	9.6	10.2
Other industrial boilers	18.0	19.6	22.3	24.0	24.6	23.8	25.9	25.8
Nonferrous metal smelting	38.9	43.5	50.3	46.4	47.8	48.0	41.4	33.7
Zinc	12.5	14.2	16.9	17.2	18.4	20.9	20.3	19.7
Lead	24.1	27.2	31.7	27.6	27.7	25.3	20.6	13.3
Copper	2.3	2.1	1.7	1.7	1.7	1.7	0.6	0.7
Gold mining	129.3	128.8	128.9	127.8	128.5	125.9	126.9	128.0
Large scale	12.4	11.9	12.0	10.9	11.6	9.0	10.0	11.1
Artisanal and small scale	116.9	116.9	116.9	116.9	116.9	116.9	116.9	116.9
Other miscellaneous processes	24.9	25.9	28.9	36.9	38.0	42.6	44.7	41.5
Mercury mining	13.0	10.8	11.4	19.0	20.3	22.6	22.6	19.2
Battery/fluorescent lamp production	6.0	6.9	7.8	8.7	8.0	8.0	8.0	8.0
PVC production	5.6	7.1	8.6	8.0	8.5	10.8	12.8	13.1
Oil and gas combustion	0.2	1.1	1.1	1.2	1.2	1.2	1.3	1.3
Residential & commercial sector	**22.0**	**21.6**	**22.2**	**22.3**	**23.5**	**24.1**	**25.2**	**26.6**
Coal burning	7.5	7.0	6.8	7.7	8.3	8.7	9.1	9.6
Biofuel use/biomass open burning	7.8	7.9	7.4	7.0	7.1	6.2	6.3	6.3
Solid waste incineration	1.3	1.4	1.5	1.5	1.6	1.7	1.7	2.0
Municipal	0.2	0.3	0.4	0.4	0.6	0.7	0.7	1.0
Rural	1.1	1.1	1.1	1.1	1.0	1.0	1.0	1.0
Oil and gas combustion	5.3	5.2	6.5	6.0	6.5	7.4	8.1	8.7
Total	**363.7**	**380.8**	**401.4**	**403.6**	**412.7**	**414.2**	**426.5**	**410.3**
Total coal combustion	**135.3**	**147.3**	**157.2**	**159.3**	**165.0**	**168.4**	**185.6**	**178.7**

Table S6. National annual emissions of Hg^{2+} by source category from 2005 to 2012.

Source category	2005	2006	2007	2008	2009	2010	2011	2012
Coal-fired power plants	**55.0**	**52.0**	**49.4**	**42.9**	**37.4**	**35.3**	**38.4**	**34.7**
Industry	**194.2**	**207.3**	**225.3**	**224.6**	**231.2**	**230.3**	**229.5**	**224.9**
Cement production	31.1	32.1	32.8	28.6	29.1	21.8	19.2	19.6
Coal use	7.7	8.8	9.6	9.3	10.8	11.5	12.6	13.3
Iron & steel plants	10.6	11.7	12.0	12.3	13.5	14.6	15.8	16.1
Heating boilers	10.2	11.5	13.6	14.7	14.8	17.1	18.6	19.7
Other industrial boilers	34.5	37.5	42.8	46.1	47.2	45.9	50.1	49.8
Nonferrous metal smelting	47.7	53.7	62.7	60.5	63.6	68.1	62.3	56.3
Zinc	29.6	33.8	40.2	40.9	43.7	49.8	48.2	47.0
Lead	15.8	17.8	20.7	18.0	18.1	16.5	13.4	8.7
Copper	2.3	2.1	1.7	1.7	1.7	1.7	0.6	0.7
Gold mining	55.4	55.2	55.2	54.8	55.1	53.9	54.4	54.9
Large scale	5.3	5.1	5.1	4.7	5.0	3.8	4.3	4.8
Artisanal and small scale	50.1	50.1	50.1	50.1	50.1	50.1	50.1	50.1
Other miscellaneous processes	4.8	5.5	6.1	7.6	7.9	8.7	9.2	8.6
Mercury mining	2.4	2.0	2.1	3.6	3.8	4.2	4.2	3.6
Battery/fluorescent lamp production	1.1	1.3	1.5	1.6	1.5	1.5	1.5	1.5
PVC production	1.0	1.3	1.6	1.5	1.6	2.0	2.4	2.5
Oil and gas combustion	0.2	0.8	0.9	0.9	1.0	0.9	1.0	1.0
Residential & commercial sector	**21.6**	**21.6**	**22.8**	**23.5**	**25.2**	**26.7**	**28.1**	**30.8**
Coal burning	7.9	7.5	7.3	8.3	8.9	9.4	9.8	10.3
Biofuel use/biomass open burning	0.5	0.5	0.5	0.5	0.5	0.4	0.4	0.4
Solid waste incineration	8.9	9.4	9.8	9.9	10.7	11.0	11.4	13.1
Municipal	1.5	2.2	2.7	3.0	3.8	4.4	4.9	6.8
Rural	7.4	7.3	7.1	7.0	6.8	6.7	6.5	6.4
Oil and gas combustion	4.3	4.2	5.2	4.8	5.2	5.9	6.5	7.0
Total	**270.8**	**280.9**	**297.5**	**291.0**	**293.8**	**292.3**	**296.0**	**290.5**
Total coal combustion	**125.8**	**129.1**	**134.7**	**133.5**	**132.7**	**133.9**	**145.3**	**143.9**

Table S7. National annual emissions of Hg^P by source category from 2005 to 2012.

Source category	2005	2006	2007	2008	2009	2010	2011	2012
Coal-fired power plants	**3.9**	**3.2**	**3.0**	**2.6**	**1.7**	**1.5**	**1.6**	**1.2**
Industry	**23.2**	**24.0**	**25.8**	**26.9**	**27.4**	**25.4**	**25.8**	**25.8**
Cement production	5.1	5.3	5.4	4.3	4.3	2.4	1.7	1.8
Coal use	1.0	1.2	1.3	1.2	1.4	1.4	1.5	1.6
Iron & steel plants	4.2	3.5	3.1	3.7	3.8	3.8	3.4	3.4
Heating boilers	2.6	2.9	3.4	3.7	3.7	4.1	4.4	4.7
Other industrial boilers	8.8	9.5	10.8	11.5	11.8	10.9	11.9	11.8
Nonferrous metal smelting	0.9	1.0	1.2	1.2	1.3	1.4	1.4	1.4
Zinc	0.9	1.0	1.2	1.2	1.3	1.4	1.4	1.4
Other miscellaneous processes	1.6	1.8	2.0	2.5	2.5	2.8	3.0	2.8
Mercury mining	0.8	0.7	0.7	1.2	1.3	1.4	1.4	1.2
Battery/fluorescent lamp production	0.4	0.4	0.5	0.5	0.5	0.5	0.5	0.5
PVC production	0.3	0.4	0.5	0.5	0.5	0.7	0.8	0.8
Oil and gas combustion	0.0	0.2	0.2	0.2	0.2	0.2	0.3	0.3
Residential & commercial sector	**17.7**	**16.6**	**16.4**	**17.9**	**19.1**	**20.0**	**20.9**	**22.0**
Coal burning	14.6	13.5	13.1	14.8	15.9	16.8	17.5	18.5
Biofuel use/biomass open burning	2.0	2.0	1.9	1.8	1.8	1.6	1.6	1.6
Solid waste incineration	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2
Municipal	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1
Rural	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Oil and gas combustion	1.1	1.0	1.3	1.2	1.3	1.5	1.6	1.7
Total	**44.8**	**43.8**	**45.2**	**47.4**	**48.2**	**46.8**	**48.2**	**48.9**
Total coal combustion	**35.1**	**33.8**	**34.7**	**37.5**	**38.2**	**38.4**	**40.3**	**41.1**

Table S8. Comparisons of emissions estimates between this work and global inventories for given sources.

		CPP [a]	HB & OIB [a]	RC & ROG [a]	CEM [a]	ISP [a]	NMS [a]	LGM [a]	ASGM [a]
2010	This work	140	111	52	46	35	118	13	167
	UNEP [b]	97	55	29	85	23	71	13	167
	IIASA [c]	148	38	28		135 [f]			187
2008	This work	144	108	45	59	30	108	16	167
	EDGAR [d]		172 [g]	31	67	12	22	7	173
2005	This work	145	87	40	64	26	87	18	167
	UNEP [e]		387 [h]		85	14	66	45	156
	EDGAR [d]		128 [g]	27	52	13	16	6	136

[a] CPP: coal-fired power plants; HB: heating boilers; OIB: other industrial boilers; RC: residential coal combustion; ROG: residential oil & gas combustion; CEM: cement production; ISP: iron & steel plants; NMS: nonferrous metal smelting, LGM: large-scale gold mining; and ASGM: artisanal and small-scale gold mining.

[b] AMAP/UNEP (2013)

[c] Rafaj et al. (2013)

[d] Muntean et al. (2014)

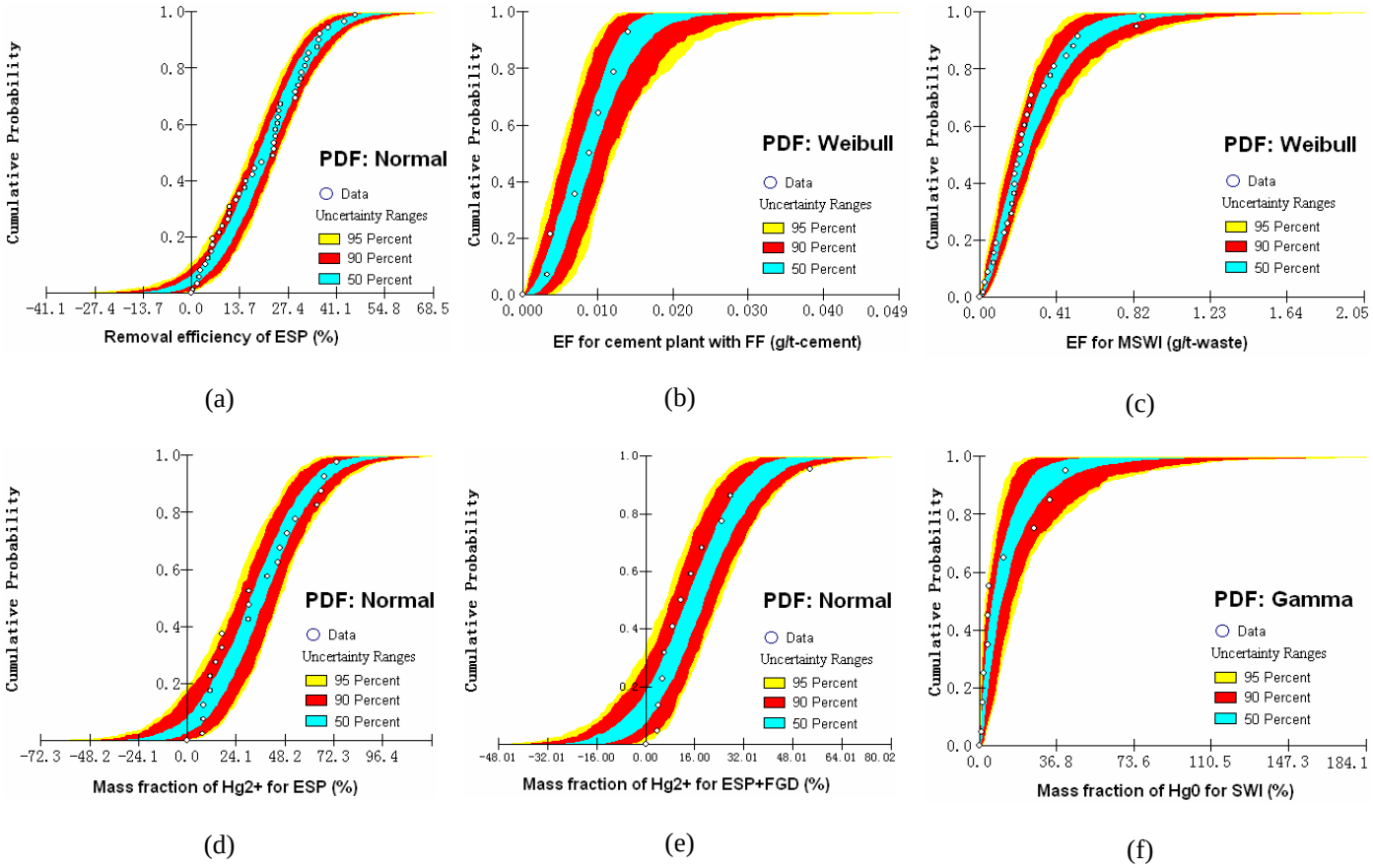
[e] AMAP/UNEP (2008) and Pacyna et al. (2010)

[f] Emissions from industrial processes including CEM, ISP and NMS

[g] Emissions from CPP and HB & OIB

[h] Emissions from CPP, HB & OIB and RC & ROG

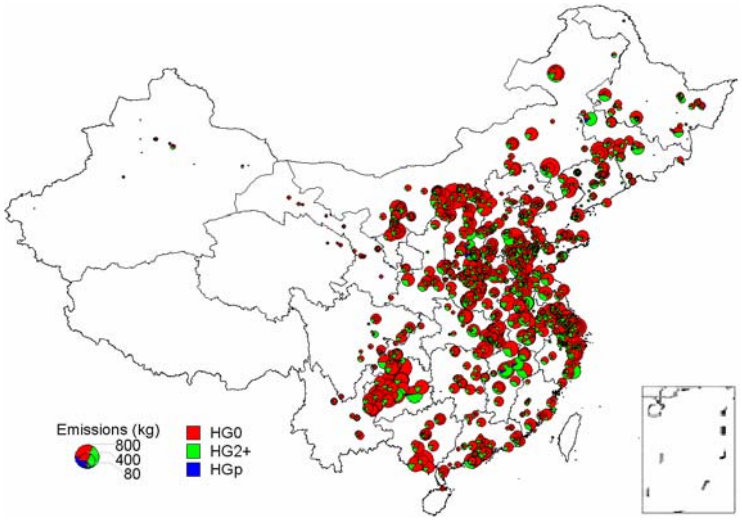
Figure S1



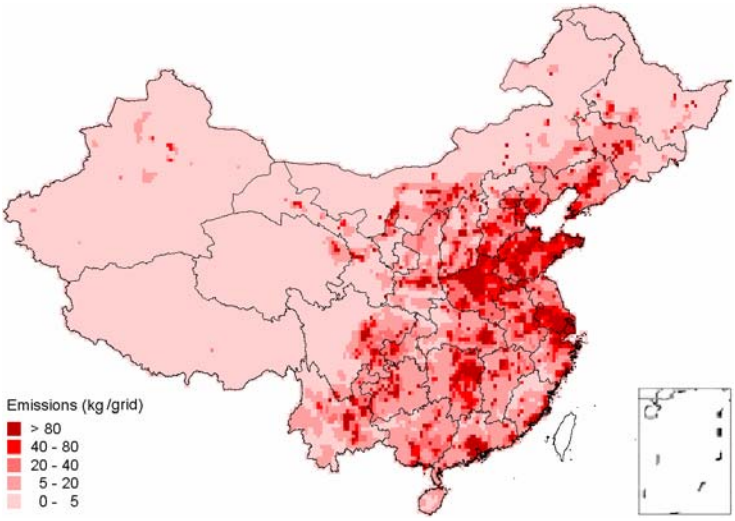
(a) (b) (c)

(d) (e) (f)

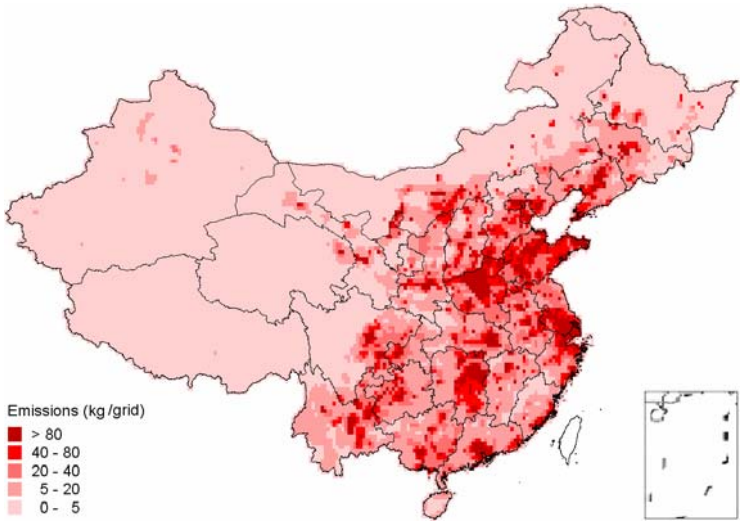
Figure S2



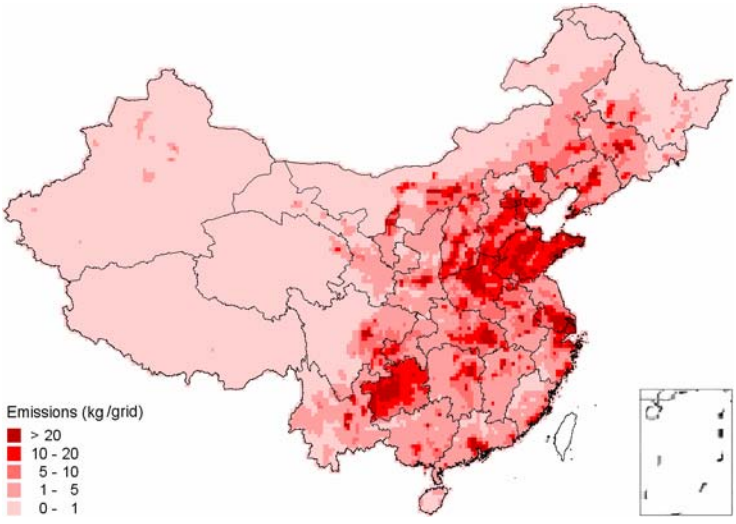
(a) Power sector



(b) Hg^0



(c) Hg^{2+}



(d) Hg^p

Figure S3

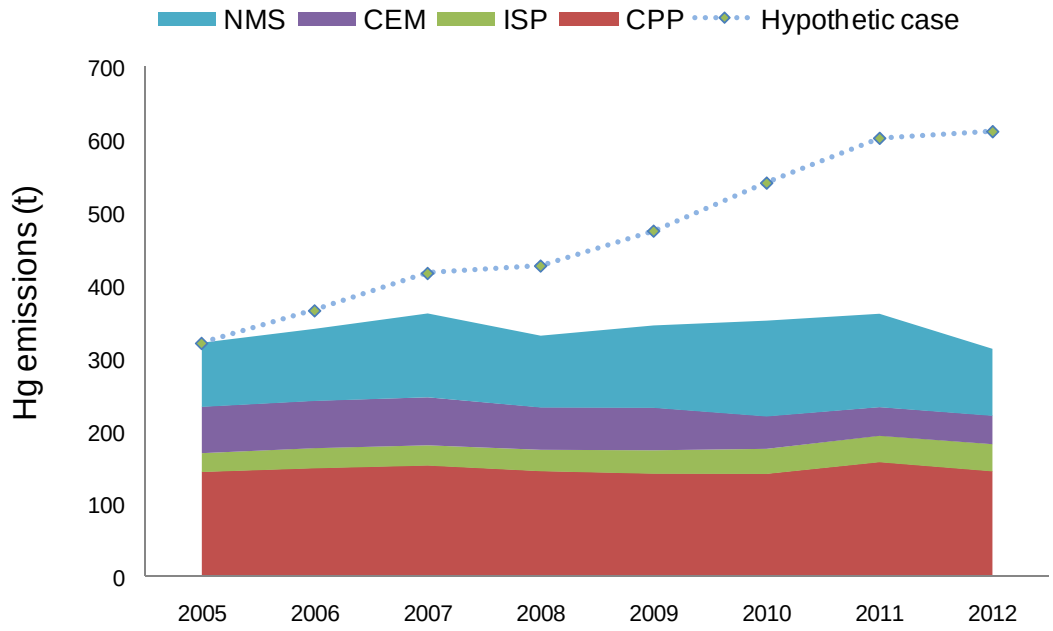
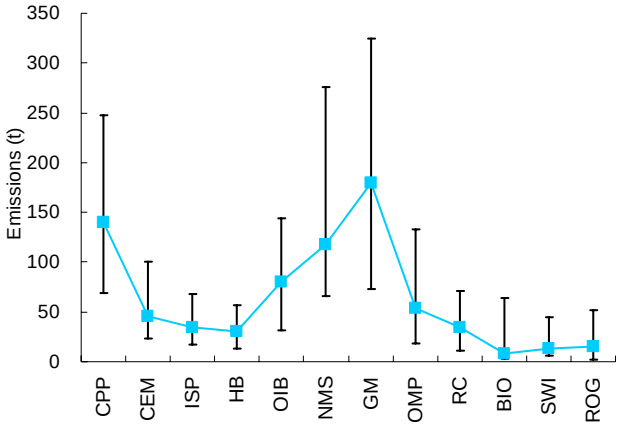
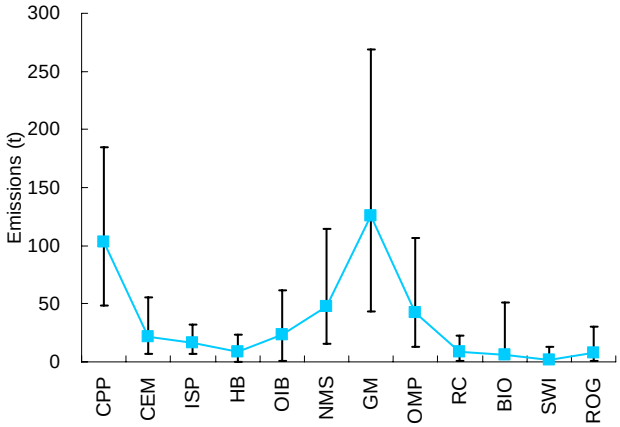

NMS
CEM
ISP
CPP
Hypothetic case
Hg emissions (t)
700
600
500
400
300
200
100
0
2005
2006
2007
2008
2009
2010
2011
2012

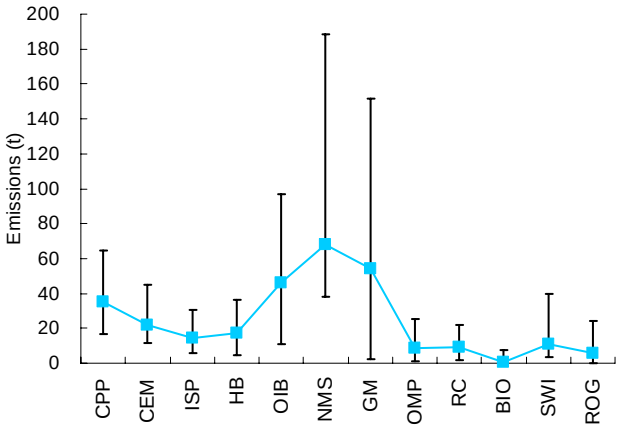
Figure S4



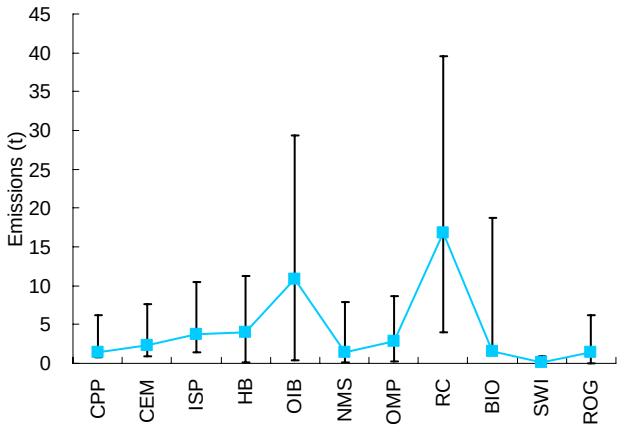
(a) Total Hg



(b) Hg^0



(c) Hg^{2+}



(d) Hg^p

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