Mercury Export from Mainland China to Adjacent Seas and Its Influence on the Marine Mercury Balance

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Supporting Information

34 pages, 8 tables, 6 figures

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Mercury Discharges from Rivers

Details of the estimation methods are provided in the Methods section. The areas of the four adjacent seas and major rivers in China are presented in Figure S1. The Hg concentrations in China's river mouths are provided in Table S1. The Hg concentrations of each river are assumed to have remained unchanged every five years. The Hg fluxes of the eight rivers were used to estimate the contributions from all the rivers according to their proportions in the total riverine water discharge. Among the eight major rivers, the Yangtze River, Pearl River and Yellow River were the three largest rivers in China, comprising 76% of the total riverine water discharge. Therefore, the annual variations in the Hg that was discharged from these three rivers from 1984 to 2013 are presented.

Mercury Discharges from Industrial Wastewater and Domestic Sewage

The ratio of Hg(D) to Hg(P) was set to 3:7 based on the literature because of a lack of sediment concentration data from industrial wastewater and domestic sewage.[1-7] The method that was developed by Ye et al. was applied to estimate the historical trends of Hg discharge from industrial wastewater and domestic sewage.[8] The trends of Hg concentrations in the total wastewater discharge from 1980 to 2010 according to a previous study were used to estimate the variations in the trends from 1984 to 2013 because annual Hg concentration data were limited for each sector of wastewater discharge.[9] Industrial wastewater data for coastal cities and domestic sewage data for coastal provinces from the China Environmental Statistics Yearbook were used to estimate the Hg flux that was exported into the seas from these two sources.[10]

Mercury Discharges from Groundwater

A ratio of 3:7 was set as a proportion of Hg(D) and Hg(P), as reported in a previous study, because of a lack of sediment concentration data for groundwater in China.[11] The Hg fluxes that were exported into the four seas were estimated based on the lengths of the coastlines. When calculating the annual change in the Hg flux from groundwater, the groundwater discharge was estimated based on the riverine water discharge (see Methods), and the Hg concentration was assumed to be invariant from 1984 to 1995, from 1996 to 2005, and from 2006 to 2013, respectively.

Mercury Export from Non-point Sources

The weighted means of the erosion moduli were calculated based on the results of various previous studies in the study area. The SEs of the mean erosion moduli were estimated for each erosion modulus and their study area. All the Hg concentration data for the coastal soil were the average concentrations of certain administrative regions. The Hg concentrations of the coastal soil in China did not change obviously in recent years. For example, the average Hg concentrations of Hebei Province were 38±25 μg/kg in 2010[12] and 36±39 μg/kg in 1990.[13] The average Hg concentrations in the topsoil of Tianjin City were 73±35 μg/kg in 2009[14] and 84±127 μg/kg in 1990.[13] Luo et al. investigated the Hg concentrations of soil in South China and found that the concentrations ranged from 31 to 150 μg/kg from 2011 to 2013, while the Hg concentrations of soil in South China ranged from 78 to 150 μg/kg in 1990.[15]

Mercury Exportation from Coastal Erosion

The weighted means of the erosion rates along the coastlines of the four seas were obtained based on the lengths of the coastlines from different studies. The SEs of the mean erosion rates were estimated for each erosion rate and study length.

Mercury Mass Balance of China's Seas

The box model was applied to simulate the Hg mass balance of China's four adjacent seas. Some of the Hg(P) was deposited in the sea margin, and additional proportions were transported to the open Pacific Ocean. The ratios of suspended sediment that was deposited in China's seas and the sea margin sediments that were transported to the open ocean were adopted according to Oguri et al.[16]

Table S1. Hg Concentrations in Mouths of Chinese Rivers

time	[Hg(T)] (ng/L)	[Hg(D)] (ng/L)	[Hg(P)] (ng/L)	reference
Songhua River [a]				
2005.12	50 ± 29	13 ± 5.9[b]	37 ± 26	17
2009.9	3.2 ± 1.1	2.1 ± 0.5	1.1 ± 1.1	18
Liao River				
2006.5	30			19
2006.7	80			19
2006.9	40			19
2006.5	27			19
2006.7	92			19
2006.9	11			19
Luanhe River				
2008.4	240			20
2008.4	220			20
Haihe River				
2010.Summer	10 ± 7.8			21
2001	710			22
2005	140 (120-240)			22
2004.8	110 ± 100			23
Yellow River				
1984.5	75			24
1984.8	100			24
2005.5	110			25
2005.9	160			25
2005.5	110	34	73	26
2005.9	160	78	80	26
2005.11	65	27	38	26
2006-2007.Spring	48 (22-89)			27
2006-2007.Summer	67 (38-140)			27
2006-2007.Autumn	43 (34-67)			27
2006-2007.Winter	41 (17-56)			27
2008.12	190	110	78	28
2009.1	320	120	200	28
2009.2	210	120	86	28
2009.3	240	140	98	28
2009.4	280	140	140	28
2009.5	240	130	110	28
2009.6	130	120	12	28
2009.7	150	130	23	28
2009.8	230	110	120	28
2009.9	180	110	65	28

2009. 10	150	110	42	28
2009.11	160	110	53	28
2011.5		98		29
2011.8		47		29
Yangtze River				
1985.8	18	2	16	30
	24	6	18	30
	20	1	19	30
	42	20	22	30
	36	18	18	30
1986.3	48	2	46	30
	44	1	43	30
	54	1	53	30
	50	1	49	30
	81	27	54	30
1984	23			31
1985	23			31
1986	24			31
1987	28			31
1988	30			31
1989	85			31
1990	36			31
1991	40			31
1992	64			31
1993	72			31
1994	45			31
1995	40			31
1996	32			31
1997	76			31
1998	28			31
1999	36			31
2000	66			31
2001	55			31
2002	24			31
2003	30			31
2004	40			31
2005	34			31
2006	70	46±11		31
2006.1		40		31
2006.2		40		31
2006.3		38		31
2006.4		56		31
2006.5		72		31
2006.6		61		31

2006.7		52		31
2006.8		43		31
2006.9		41		31
2006. 10		42		31
2006.11		37		31
2006.12		35		31
2006-2007.Spring	130 (40-350)			27
2006-2007.Summer	140 (39-240)			27
2006-2007.Autumn	150 (30-260)			27
2006-2007.Winter	130 (31-230)			27
2008.8		(37–610) [c]	(51–640)	32
2009.5		(13-53) [d]		33
2009.8		(5-180)		33
2010.4		52		29
2011.6		69		29
2011.8		68		29
2013.1	50			34
2013.2	80			34
2013.3	160			34
2013.4	80			34
2013.5	50			34
2013.6	80			34
2013.7	120			34
2013.8	150			34
2013.9	50			34
2013. 10	120			34
2013.11	50			34
2013.12	120			34
Qiantang River				
2001-2004	100 ± 70			35
2011.8		21		36
		22		36
		63		36
		58		36
		48		36
		61		36
		55		36
2012		8 ± 3		37
Minjiang River				
2008	140			38
2012	170 (13-730)			39
Pearl River				
1980.8	34	3	30	40
1980.8	41	5	35	40

1980.8	33	2	30	[40]
1980.8	38	2	35	[40]
1982	30 ± 19			[41]
1981	24			[42]
1986	14			[42]
1990	60			[42]
1995	42			[42]
2002	18			[42]
2006-2007.Spring	36 (21-47)			[27]
2006-2007.Summer	16 (4-35)			[27]
2006-2007.Autumn	26 (12-43)			[27]
2006-2007.Winter	23 (15-34)			[27]
2009	17 ± 3.4	12 ± 2.2	5.9 ± 4.4	[43]
2009	24 ± 4.4	15 ± 6.1	8.2 ± 2.6	[43]
2009	24 ± 10	16 ± 3.7	8 ± 7.3	[43]
2009	18 ± 3.6	13 ± 3.9	5.5 ± 5.2	[43]
2009	21 ± 6.7	14 ± 4.5	6.9 ± 5.1	[43]
2009	17 ± 3.8	9 ± 1.7	8 ± 4.8	[43]
2009	17 ± 5.7	6.3 ± 2.2	11 ± 5.5	[43]
2009	19 ± 5.0	7.9 ± 5.9	11 ± 10	[43]

a. Hg concentrations of Songhua River are used for calculating K_D value.

b. While a literature just provided the concentrations of Hg(T) (ng/L) and Hg(D) (ng/L), [Hg(T)] – [Hg(D)] is used to estimate the concentration of Hg(P) (ng/L).[43, 44]

c. Outliers were removed when estimating the annual changes of Hg fluxes of Yangtze River (Z = 5.15).

d. If an article just provided the range of Hg concentration, the midrange value is used for estimating the mean.[45] Same method was adopted for other pathways.

Table S2. Hg Concentrations in Industrial Wastewater in Major Industry Sectors in China

	[Hg(T)] (μg/L)	reference	[Hg(T)] (μg/L)	reference
Agricultural Services				
	0.39 ± 0.72	[46]	0.013 ± 0.007	[47]
	0.14 ± 220	[9]	0.82	[48]
Mining and Washing of Coal				
	52	a		
Mining and Processing of Ferrous Metal Ores				
	0.22	[49]		
Mining and Processing of Non-ferrous Metal Ores				
	0.68	[50]	11	[3]
	50	[51]	6.5	[4]
	0.62	[1]	2.1	[5]
	(0.058-7.0)	[2]	0.34	[52]
	(0.025-7.0)	[2]	17 (6.7-30)	[53]
	(0.02-11)	[2]	0.12	[54]
Mining and Processing of Non-metal Ores				
	7.4	[55]		
Manufacture of Textile, Manufacture of Textile Wearing				
	1.3	[56]	24	[56]
	5.9	[56]	67	[56]
	22	[56]	0.083	[49]
Printing, Reproduction of Recording Media				
	1.6	[57]	0.19	[57]
Manufacture of Articles for Culture, Education and Sport Activity				
	0.26	[57]		
Manufacture of Raw Chemical Materials and Chemical Products				
	0.15	[57]	0.15 ± 0.010	[58]
	0.21 ± 0.029	[58]	0.45 ± 0.023	[58]
	0.096 ± 0.003	[58]	24	[59]
	0.13 ± 0.009	[58]	49	[60]
Smelting and Pressing of Ferrous Metals				
	0.22	[49]		
Smelting and Pressing of Non-ferrous Metals				
	5.8	[51]	0.442 ± 0.121	[61]
	43	[51]		
Manufacture of Electrical Machinery, Instrument and General Equipment				
	73	[62]	790	[62]
	46	[62]	510	[62]
	55	[62]	93	[62]
Waste Recycle				

	25	[6]		
Production and Supply of Electric Power and Heat				
	57 ± 0.64	[63]	12	[64]
	32	[64]	390	[65]

a. Estimation of the Hg concentration in wastewater of coal mining and washing is made by multiplying the coal washing rate (51%),[66] the removal rate of Hg (30%),[67] the average Hg concentration of coal in China (0.19 mg/kg),[68] coal output (2.5×10^9 t in *China Energy Statistical Yearbook* in 2012) and wastewater generated by coal mining and washing (1.4×10^9 t in *China Environment Statistical Yearbook* in 2012).[10, 69]

Table S3. Hg Concentrations in Domestic Sewage from Urban Areas

	[Hg(T)] (μg/L)	reference	[Hg(T)] (μg/L)	reference
non-treated discharge				
	1.2 ± 0.60	7	1.1	70
	(0.41-0.77)	71	3.0	72
	0.55 ± 0.10	73	17	72
	0.53	70	2.8	72
	0.54	70	1.7	72
	0.61	70	3.2	72
	1.0	70	0.88	72
	0.21	70	2.6	72
	1.3	70	2.0	72
	2.2	70	3.2	72
	0.33	70	3.3	72
	0.21	70	4.5	72
	0.63	70	2.1	72
	1.8	70	0.99	72
	1.0	70	3.1	72
	1.3	70	2.0	72
	1.4	70	0.97	72
	0.87	70	0.37	72
	1.2	70	0.55	72
	1.9	70	1.1	57
	0.89	70	5.2	57
	1.5	70	0.41	74
	1.5	70	0.31	75
	1.5	70	0.20	49
treated discharge				
	0.022 ± 0.0064	7	0.03	74
	(0.39-0.57)	71	0.04	75
	0.13 ± 0.10	73	0.11	49
	0.17	72		

Table S4. Hg Concentrations in Groundwater

place	time	[Hg(D)] (ng/L)	reference
Bohai Sea			
Tianjin	2008	16 (10-47)	[48]
southern Jilin province	1999	10	[76]
Nenjiang basin	1999	49	[76]
Second Songhua river basin	1999	24	[76]
Tumen river basin	1999	10	[76]
Mudanjiang river basin	1999	10	[76]
Hebei plain	2008	17	[77]
Yellow Sea			
Shaying river basin	2007-2009	40	[78]
the Beijing-Hangzhou Grand Canal	2000	220	[79]
Yalu river basin	1999	10	[76]
East China Sea			
southern Yangtze river delta	1998	15 ± 12	[80]
	1998	18 ± 12	[80]
	1998	8.2 ± 5.2	[80]
	1998	20 ± 12	[80]
	1998	6.7 ± 2.0	[80]
	1998	12 ± 9.3	[80]
eastern Jianghan plain	1999	11	[54]
Poyang Lake	1999	9.6	[54]
Southern Yangtze river delta	1994	11	[81]
South China Sea			
Foshan	2008	50	[82]
Pearl river delta	2011	160(10-950) [Hg(T)]	[83]

Table S5. Erosion Modulus of Different Regions in China, Non-point Source

place	time	study area (km^2)	erosion modulus ($ton/km^2 \cdot a$)	reference
Bohai Sea				
region surrounding the Bohai Sea	2012	500000	1500	84
Hebei province	2010	190000	550	85
Fuxin city	2015	10000	1900	86
Luanping county	2011	3200	3800	87
Beijing city	2004	4000	2400	88
western Pingquan county	2011	19	2400	89
Xihe basin	2011	29	710	90
Yellow Sea				
Taizhou city	2012	4	150	91
	2012	5.4	3000	91
	2012	16	500	91
Yimeng mountain area	2011		4200	92
Yimeng mountain area	2010		4700	93
downtown of Qingdao city	2005	190	280	94
Chengyang district	2005	550	350	94
Laoshan district of Qingdao	2005	390	1100	94
Huangdao district of Qingdao	2005	270	510	94
Laixi city of Qingdao	2005	1500	440	94
Jimo city of Qingdao	2005	1800	480	94
Pingdu city of Qingdao	2005	3200	580	94
Jiaozhou city of Qingdao	2005	1300	580	94
Jiaonan city of Qingdao	2005	1800	980	94
East China Sea				
Changting county, Fujian	2007	980	980	95
Xiazhuang village of Zhangzhou city	2005		2500	96
Taihu Basin	2008	23000	2000	97
Zhejiang province	2014	98000	400	98
Fujian province	2014	120000	300	98
Jurong city	2011		2600	99
South China Sea				
Guangdong province	2009	180000	2200	100
Guangdong province	2014	180000	720	101
Dongping town of Yangjiang city	2004	16	5300	102
Conghua distrct	2011	2000	1900	103
Raoping county	2013	1700	1600	104
Dongjiang river basin	2010	35000	1900	105
Songtao reservoir basin	2009	14000	370	106

Note: Erosion modulus means the mass of soil eroded by waterpower in a given time and area, with the unit of $t/km^2 \cdot yr$.

Table S6. Erosion Rates of Different Regions in China, Coastal Erosion

place	time	study length (km)	erosion rate (m/yr)	reference
Bohai sea				
beach of Beidaihe	2012	12	2.5	[107]
Tianjin Binhai New Area	2009	270	0.26	[108]
east coast of Laizhou bay	2008	150	2.5	[109]
Qinhuangdao city	2009	140	2.5	[110]
Qinhuangdao city	2012	160	2.8	[111]
Qinhuangdao city	2008	130	1.9	[112]
Jinmeng bay of Qingdao	2014	3.4	1.6	[113]
Hebei province	2008	37	0.7	[114]
Beach surrounding Bohai sea	2013	2700	1.5	[115]
Laizhou bay	2006	190	2	[116]
Feiyan beach	2005	37	0.21	[117]
east coast of Laizhou bay	2006	210	3.5	[118]
south coast of Laizhou bay	2006	110	27	[119]
Beihai beach of Gaizhou	2014	1.9	0.7	[120]
Tengfangshen beach of yingkou	2014	1.5	0.9	[120]
Yingchengzi beach of Dalian	2014	1.7	0.8	[120]
Zhujiatun beach of Jinzhou	2014	14	0.8	[120]
Daizi beach of Lvshun	2014	1.5	3	[120]
Liaodong bay	2005	140	2.5	[121]
Shandong province	2013	1600	1.5	[122]
Yellow river delta	2005		400	[123]
Yellow sea				
Old Yellow river estuary	2002		43	[124]
Old Yellow river estuary	2013	320	6.3	[125]
Lv si shore	2002		20	[124]
Lv si shore	2009	14	20	[126]
Xiuzhen-Xinzhuang shore	2002		15	[124]
south coast of Shandong	2000	33	1	[127]
Lingshan bay	2012		7	[128]
Weihai city	2012	120	1.5	[129]
Xuejia island	2002	18	0.021	[130]
Shandong province	2013	1600	1.5	[122]
Liangshui bay of Dalian city	2014	15	1.4	[120]

Bofengzi bay of Lvshun city	2014	5.4	4.7	120
Chengtou mountain	2014	2	2	120
Longkou-Chefoo shore	2014	170	2.2	131
East China sea				
Zhejiang province and Shanghai city	2011		4	132
Luchao-Zhong harbor	1993		50	133
Dongshan island of Fujian	2015	22	2.7	134
Fujian province	2011	1	3	132
Fujian province	2009	4600	1	135
South coast of Fujian	2006	1300	1.6	136
Southeast coast of Xiamen	2001	7.6	2	137
Longfengtou bay	2013	0.22	11	138
Banyue bay	2013	2.5	6.2	139
South China sea				
Silver beach	2011	5	10	140
west coast of Haikou bay	2013	8	23	141
Boao beach of Hainan	2011	2.5	4.7	142
Bangtang bay of Hainan	2003	4	12	143
Hainan province	2003	240	5.7	144
Lanqing bay	2014	82	2.3	145
Leizhou Peninsula	2008	1100	2	146
east coast of Moyang river estuary shore	2012		15	147
Nanhai district	2007	2	20	148
Guangdong province	2011		9	132
Sanya bay	2007	3.4	0.4	149
Shantou city	2008	15	1.4	150
Xiangshan-Changweijiao beach of Xiamen	2013	1.5	7.9	138

Note: erosion rate means the speed of coastline erosion by sea wave, with the unit of m/yr.

Table S7. Hg Concentrations in Coastal Soil

place	sample size	mean ± SD [Hg(T)] (μg/kg)
Liaoning province	116	37 ± 24 [a]
Hebei province	148	36 ± 39
Tianjin city	41	84 ± 130
Shandong province	117	19 ± 12
Jiangsu province	83	290 ± 720
Shanghai city	20	95 ± 52
Zhejiang province	76	86 ± 67
Fujian province	87	93 ± 81
Guangdong province	167	78 ± 85
Guangxi province	150	150 ± 170

a. The data is acquired from *Chinese Soil Element Background Value 1990.*[13]

Table S8. Parameters of Chinese Adjacent Seas Used to Develop the Marine Hg Mass Balance Model

parameter (unit)	value	reference
atmosphere		
air/sea Hg(0) exchange（ng/m^2h）	2.4(2.3-2.5)	151, 152
dry deposition（ng/m^2h）	1.3	153
wet deposition（ng/m^2h）	2.1	153
seawater		
area of Bohai Sea (km^2)	7.7×10^4	154
area of Yellow Sea (km^2)	3.8×10^5	155
area of East China Sea (km^2)	7.7×10^5	155
area of South China Sea (km^2)	3.5×10^6	156
average depth of Bohai Sea (m)	20	154
average depth of Yellow Sea (m)	44	155
average depth of East China Sea (m)	370	155
average depth of South China Sea (m)	1200	157
oceanic current of Bohai Sea to Yellow Sea (Sv)	4.8×10^{-3}	153
oceanic current of Yellow Sea to East China Sea (Sv)	0.32	153
oceanic current of East China Sea to North Pacific Ocean (Sv)	1.2	158
oceanic current of North Pacific Ocean to South China Sea (Sv)	6.4	159
oceanic current of South China Sea to East China Sea (Sv)	1.2	159
oceanic current of South China Sea to other oceans (Sv)	1.7	159
Hg in Bohai Sea water (ng/L)	41	160-163
Hg in Yellow Sea water (ng/L)	1.7	164
Hg in East China Sea water (ng/L)	2.2	165
Hg in South China Sea water (ng/L)	1.2	166
Hg in North Pacific Ocean water (ng/L)	0.35	167
sediment		
Hg in Bohai Sea sediment (ng/g)	33(24-39)	168-170
Hg in Yellow Sea sediment (ng/g)	24(15-30)	168, 169, 171
Hg in East China Sea sediment (ng/g)	29(18-37)	168, 172
Hg in South China Sea sediment (ng/g)	37(2.0-88)	173, 174
Hg evasion from sediment ($ng/m^2\cdot h$)	0.83[a]	175
sediment transport rate ($g/m^2\cdot h$)	0.10	16
suspended sediment deposition in continental shelf (%)	40[b]	16
suspended sediment deposition in open ocean (%)	60[b]	16
fishing		
Hg in fish of Bohai Sea (μg/g)	0.54	176
Hg in fish of Yellow Sea (μg/g)	0.19	177
Hg in fish of East China Sea (μg/g)	3.0×10^{-2}	178
Hg in fish of South China Sea (μg/g)	0.15	179
fishing in Bohai Sea (t/yr)	2.0×10^6	180

fishing in Yellow Sea (t/yr)	2.3×10^{6}	180
fishing in East China Sea (t/yr)	5.1×10^{6}	180
fishing in South China Sea (t/yr)	3.3×10^{6}	180

a. Due to the lack of study of Hg evasions from oceanic sediment in Chinese adjacent seas, the study of Boszke is referenced and the average Hg volatilization rate (0.83 $ng/m^2\cdot h$.) is adopted. [175]

b. According to previous study,[16] 40% of the sediment would deposit in continental shelf and 60% would deposit in the open ocean near China.

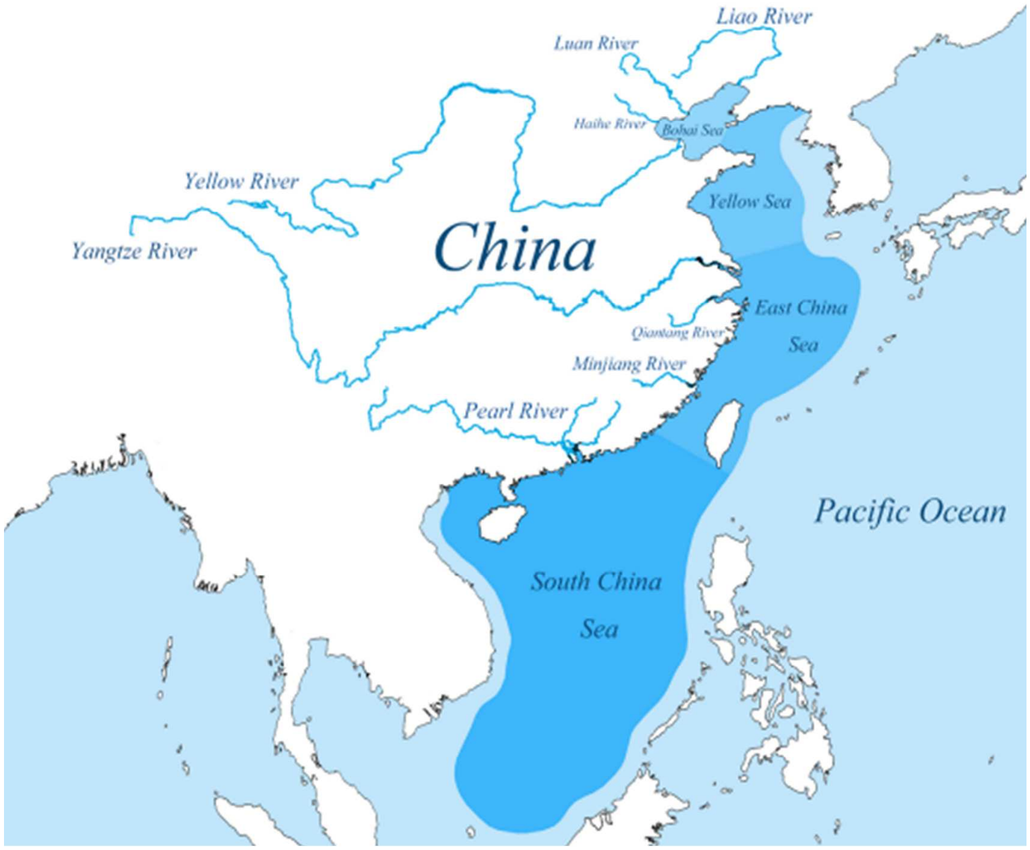

Figure S1. Study area of the four adjacent seas (including Bohai Sea, Yellow Sea, East China Sea and South China Sea)

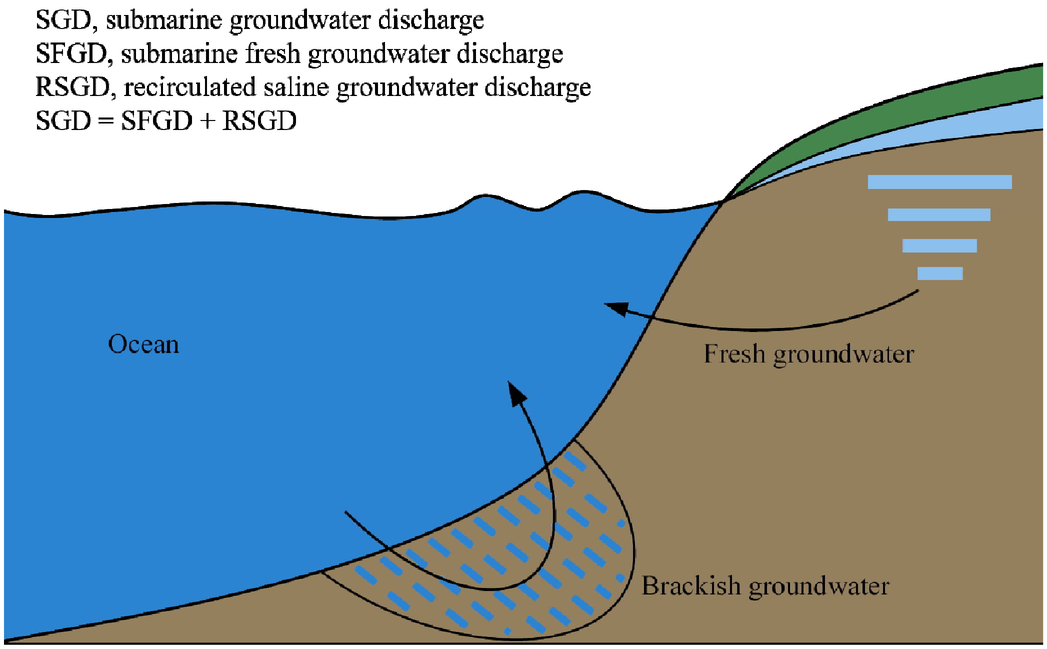

Figure S2. Schematic depiction (no scale) of SGD, SFGD and RSGD (built on previous study).[181] SFGD is used to calculate the Hg flux from groundwater discharged into the seas

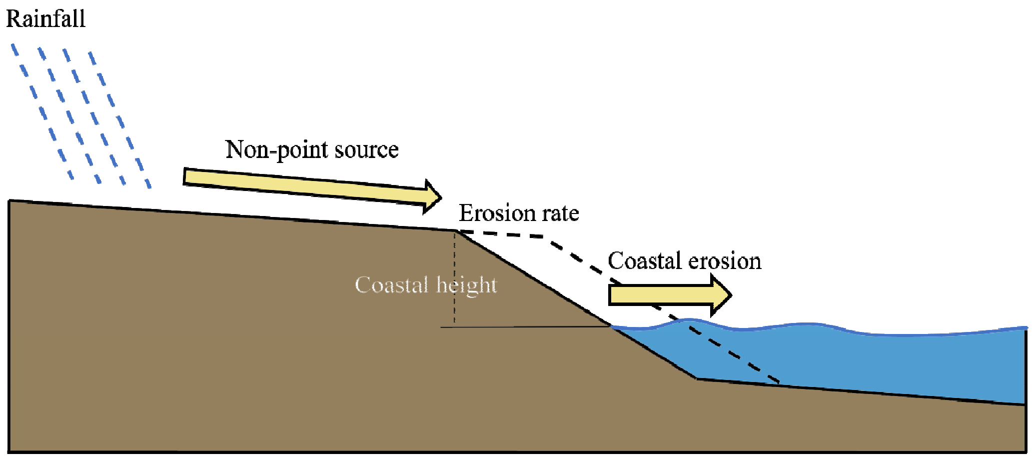

Figure S3. Schematic depiction (no scale) of non-point source and coastal erosion contributions

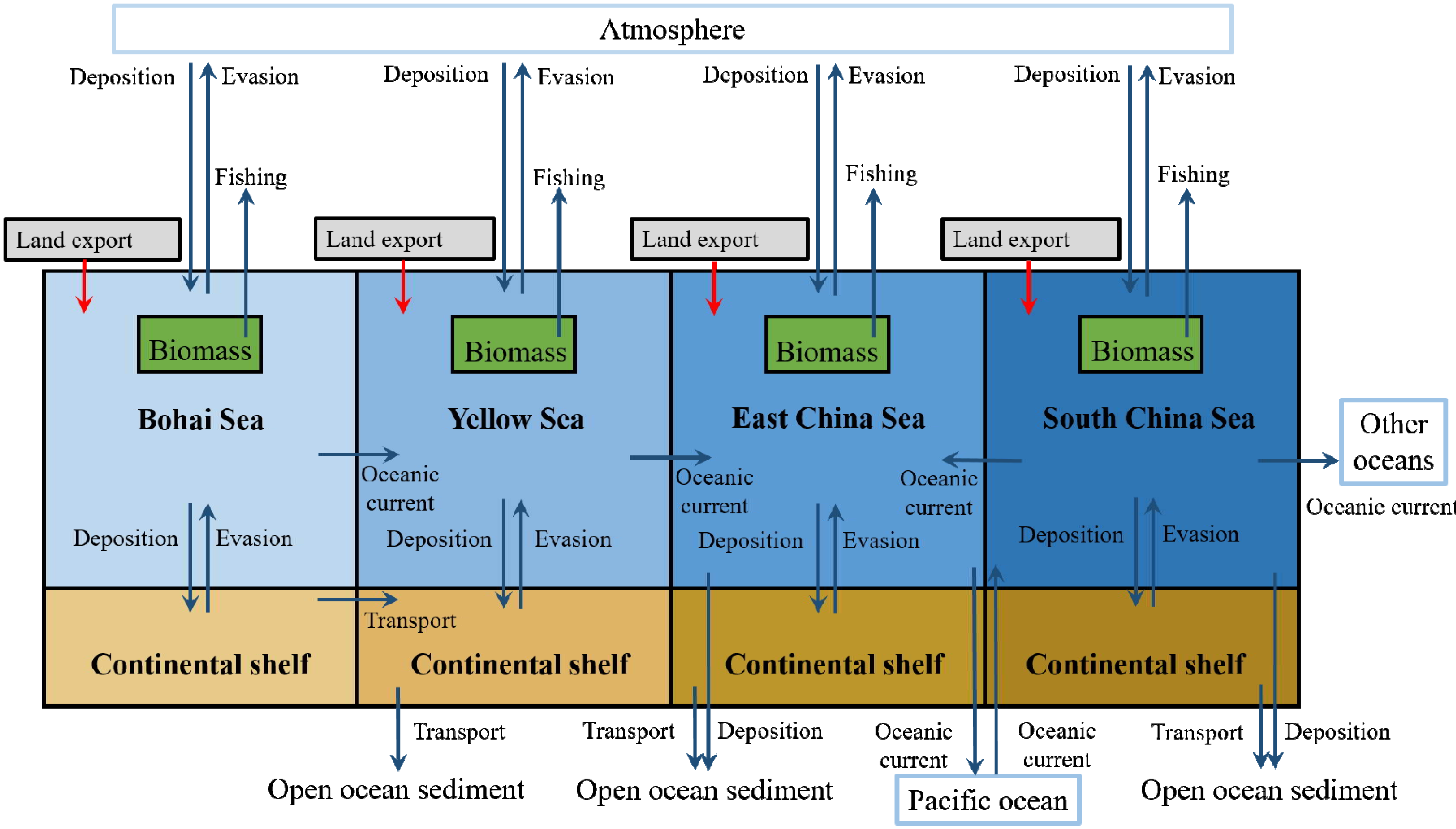

Figure S4. Schematic model of Hg mass balance in the four adjacent seas

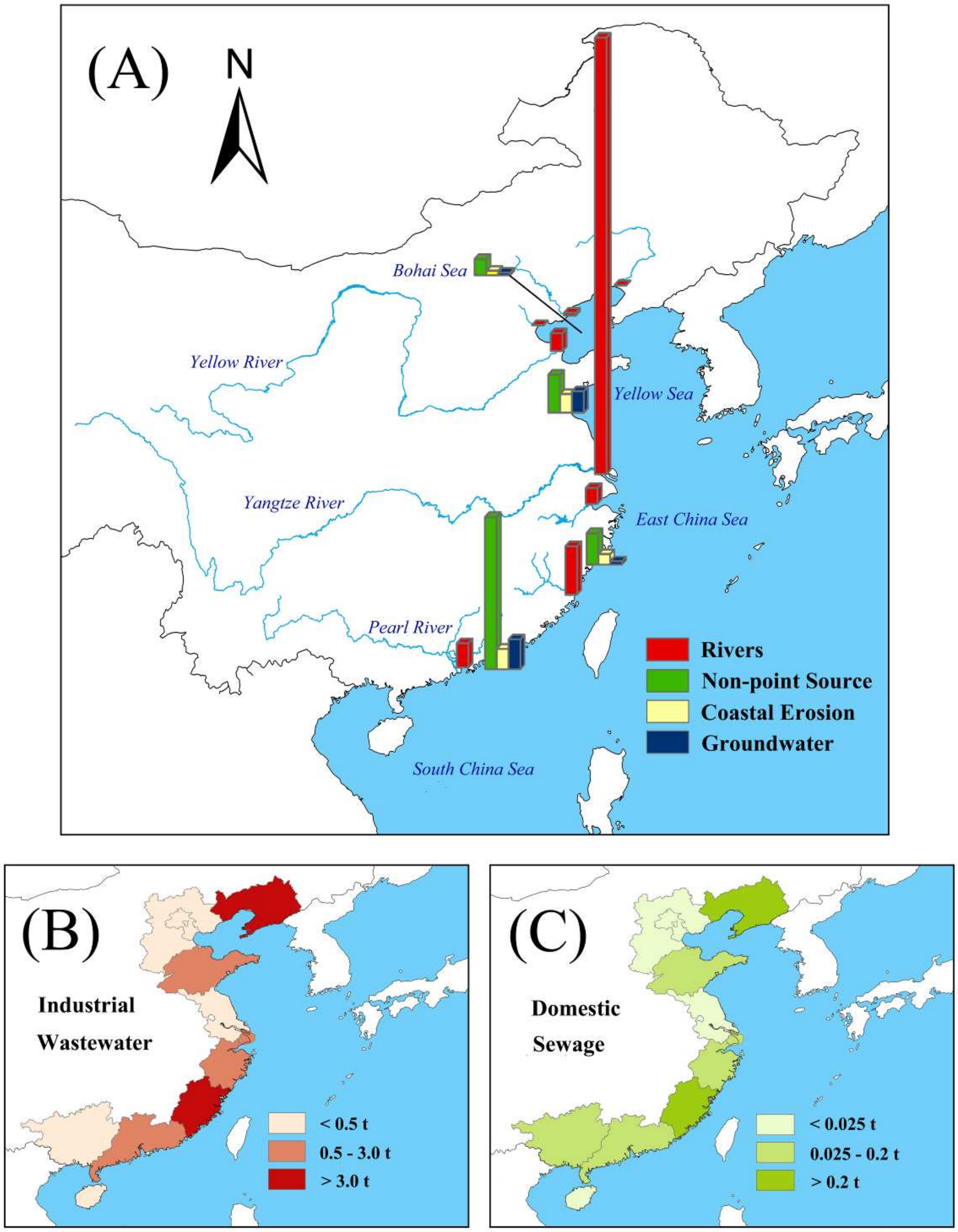

Figure S5. Total Hg exported from mainland China in 2012

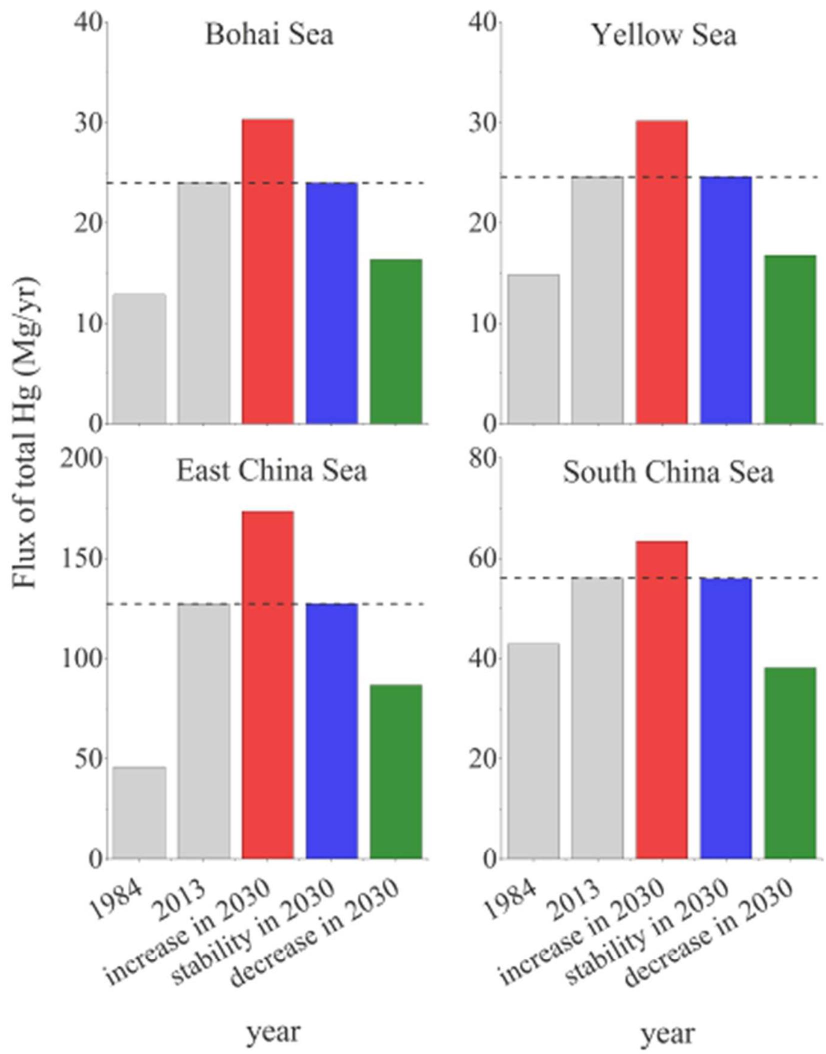

Figure S6. Total Hg exported from mainland China into adjacent seas in 1984 and 2013, and prediction in 2030

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