

The Supporting Information for

Temporal and spatial variations of atmospheric unintentional PCBs emissions in Chinese mainland from 1960 to 2019

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The authors declare no competing financial interest.

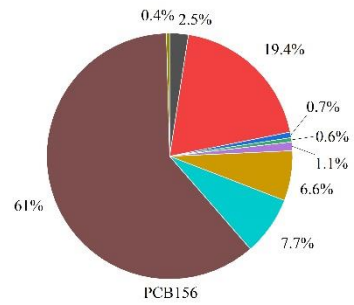
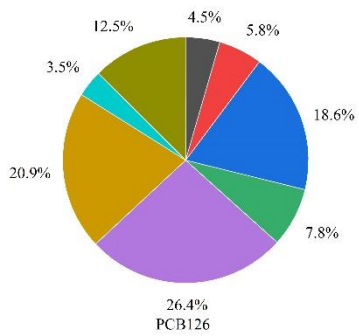
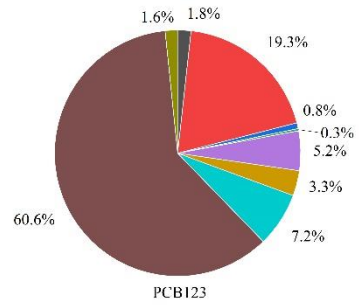
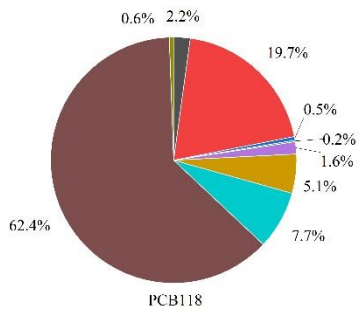
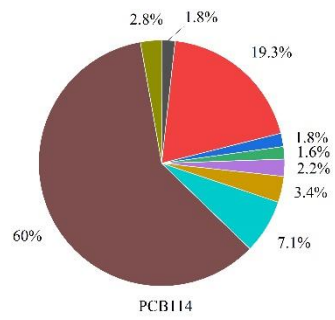
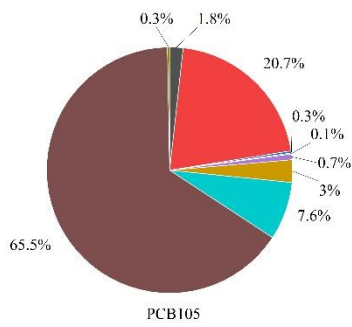
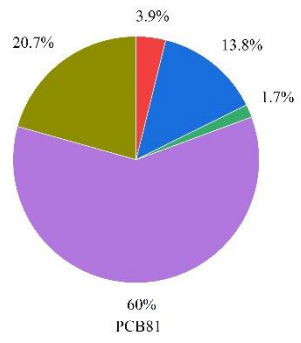
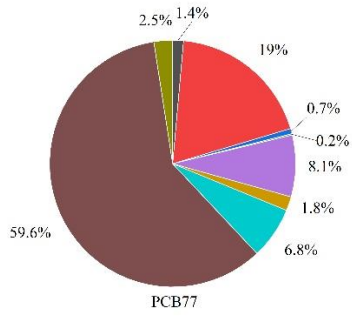
This material includes 18 pages, 5 tables, and 8 figures.

List of Tables

- S1. List of PCBs emission sources with best estimate (BE) and geometric standard deviations (SD) of EFs.
- S2. Emission profiles (percentage of mass) of 12 dioxin-like UP-PCBs used to derived congener specific EFs.
- S3. List of 12 PCBs congeners included in this study and their TEF values.
- S4. Definitions of the seven regions in China.
- S5. TEQ/u (g WHO-TEQ per unit of mass of UP-PCBs emissions) for nine source categories.

List of Figures

- S1. Emission profiles for 9 source categories of 12 UP-PCBs.
- S2. Spatial distribution of per-capita UP-PCBs emissions.
- S3. Emission densities and per capita emissions of UP-PCBs for each province in China in 2019.
- S4. Scatter plots of emission densities with population densities and GDP/Area.
- S5. Comparison of emission densities (A) and per capita emissions (B) among various source categories.
- S6. Time trends of UP-PCBs emissions from various source categories from 1960 to 2019 in seven regions of China.
- S7. Time trends of emission intensities for China and seven regions.
- S8. Relationship between per capita GDP and emission intensities (EIs) with all provincial data from 1960 to 2019 and only provincial data in 2019.



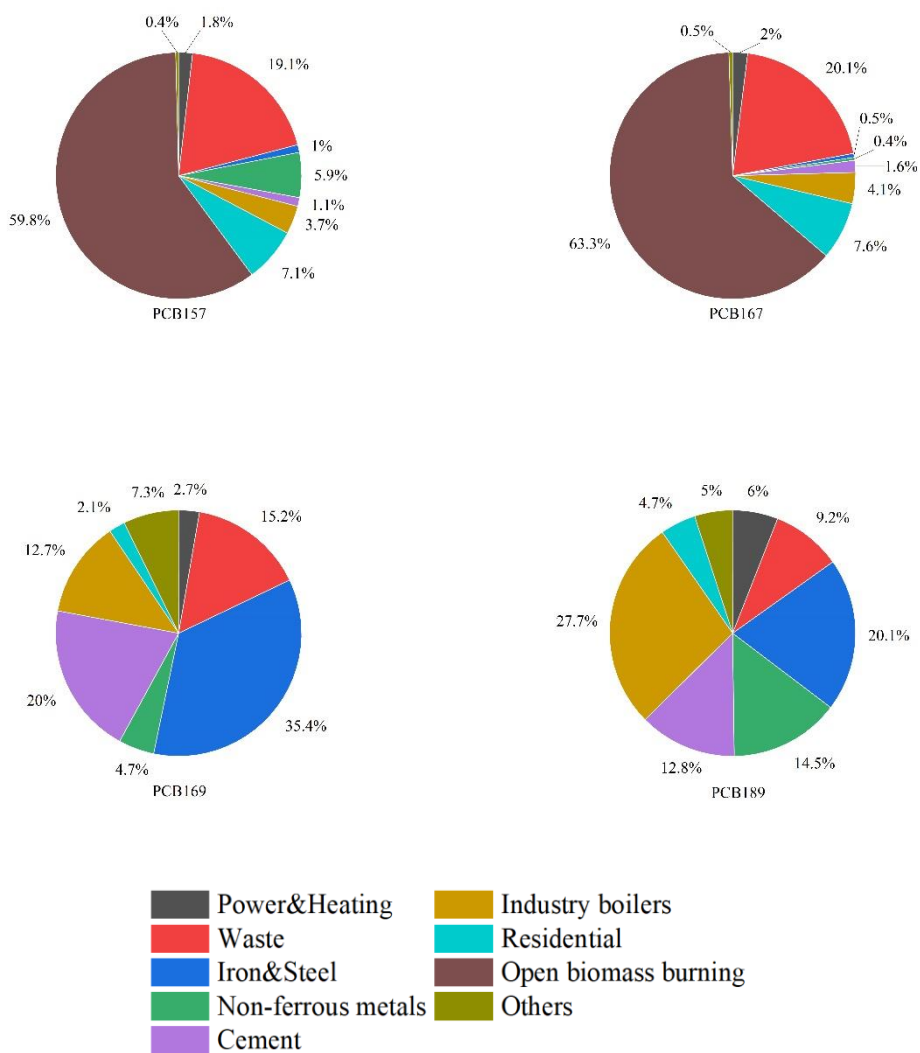


Fig. S1. Emission profiles for 9 source categories of 12 UP-PCBs.

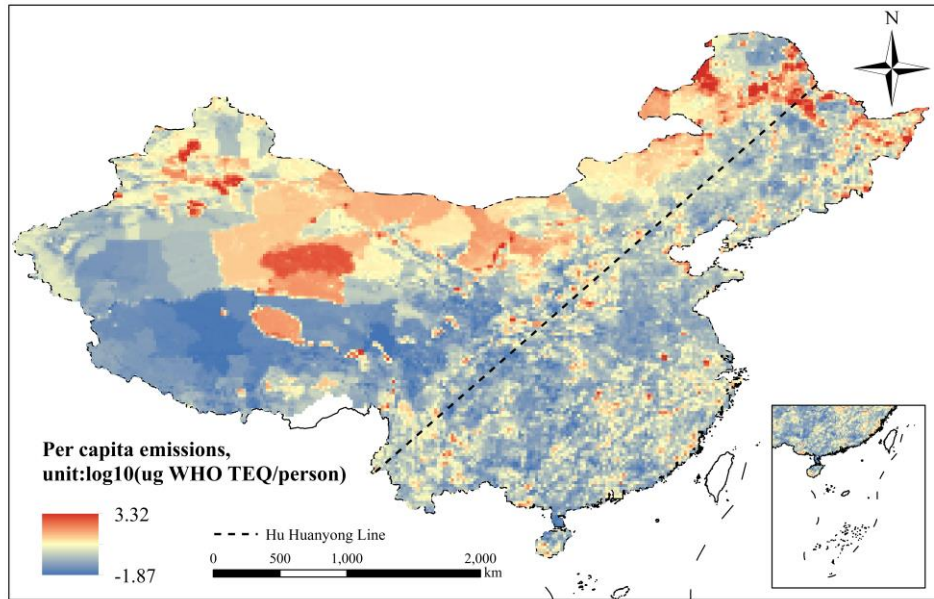


Fig. S2. Spatial distribution of per-capita UP-PCBs emissions

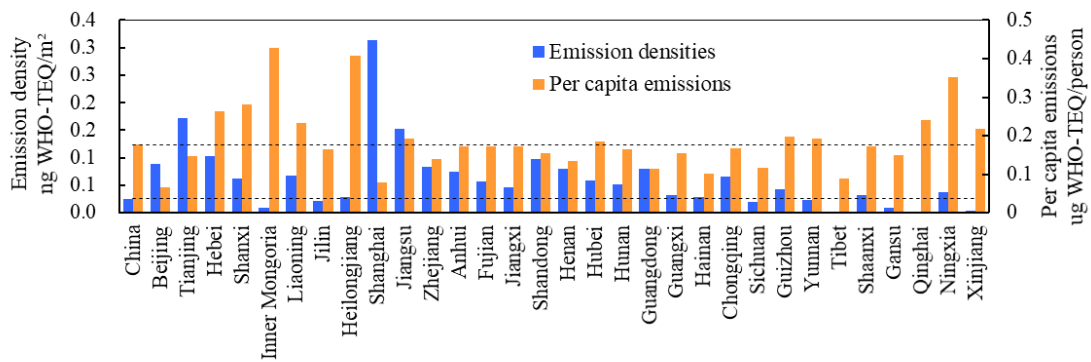


Fig. S3. Emission densities and per capita emissions of UP-PCBs for each province in China in 2019. The two dashed lines shows the national averages.

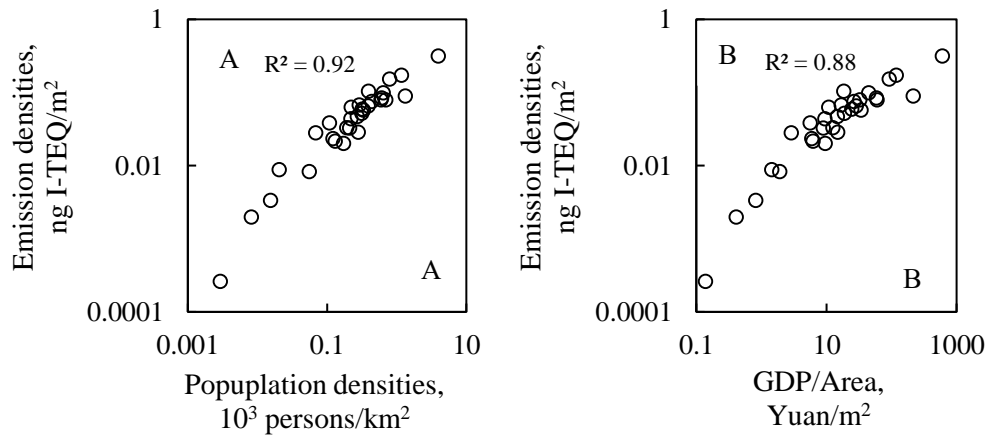


Fig. S4. Scatter plots of emission densities with population densities (A) and GDP/Area (B).

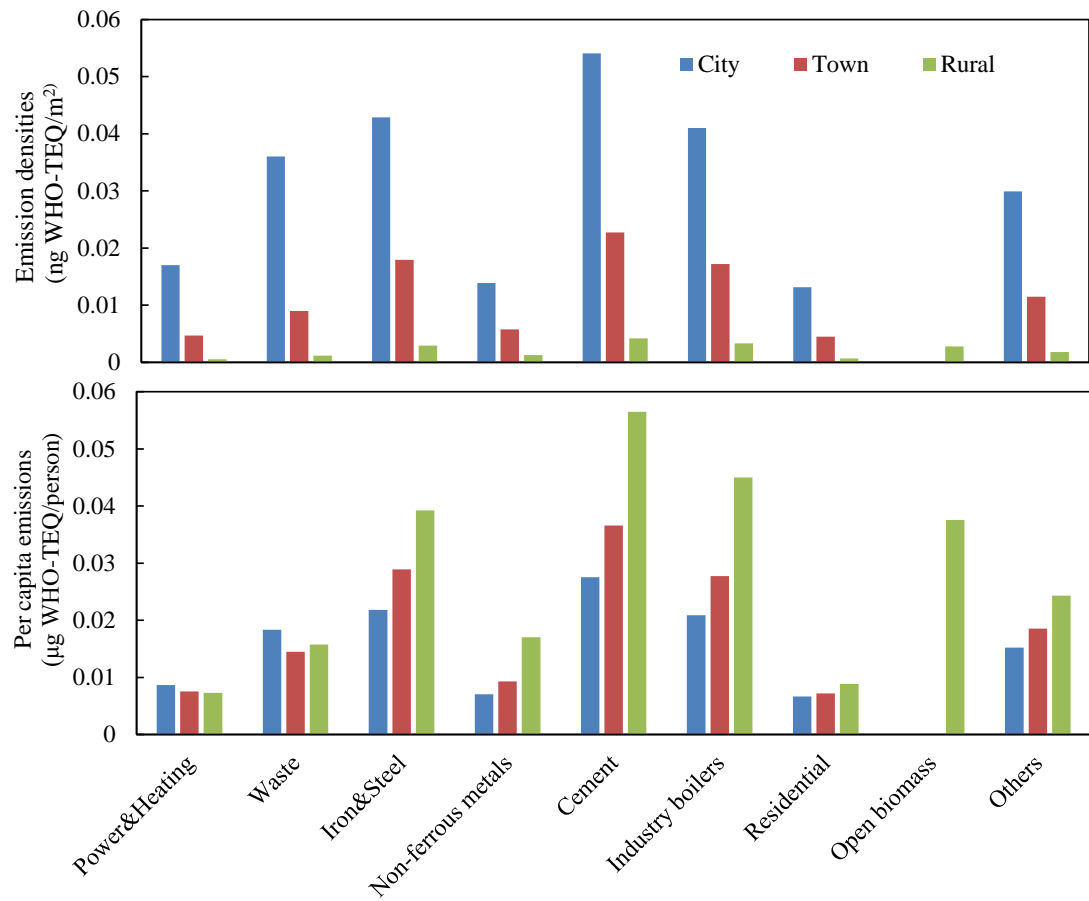
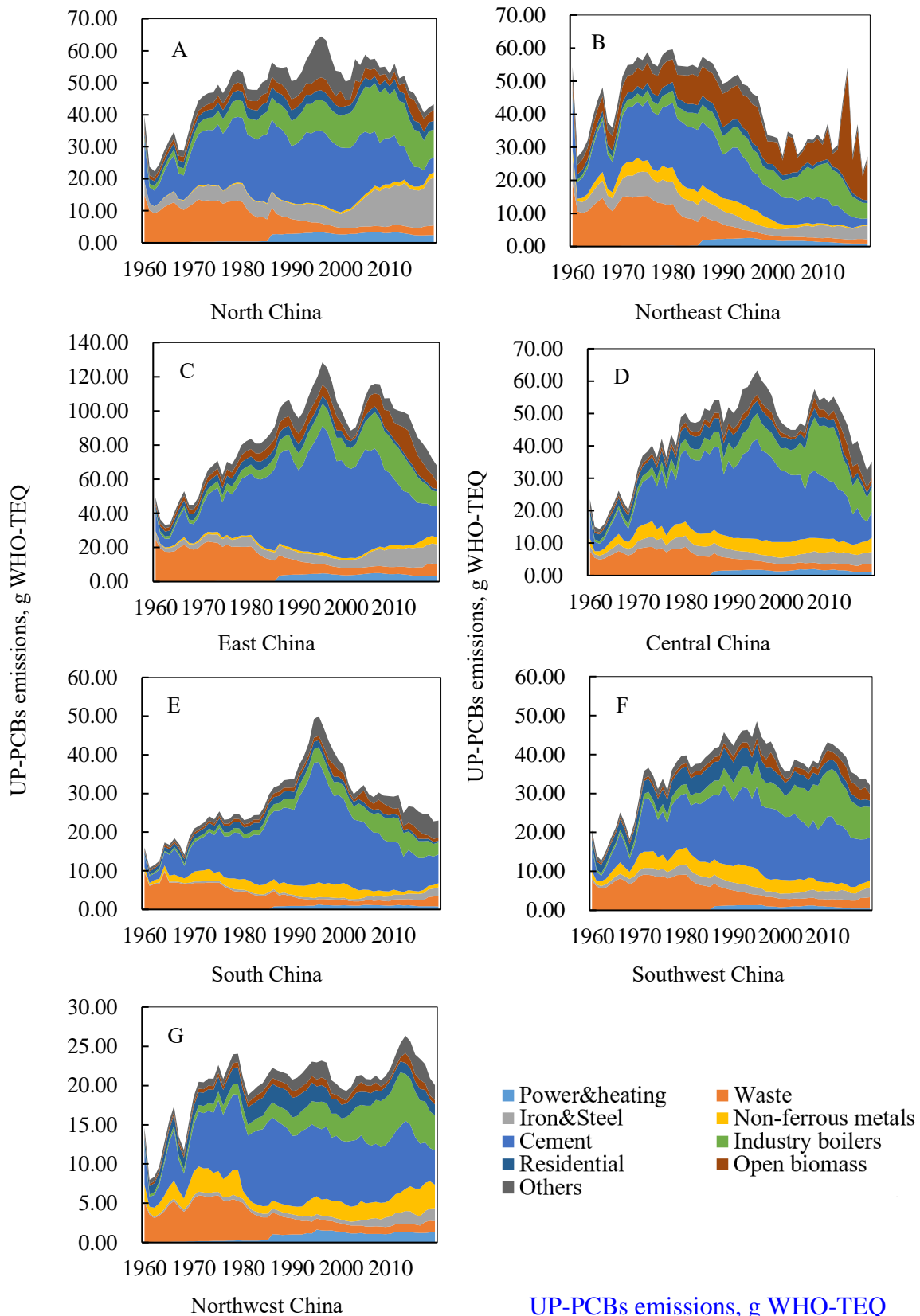


Fig. S5. Comparison of emission densities (A) and per capita emissions (B) among various source categories.



UP-PCBs emissions, g WHO-TEQ

Fig. S6. Time trends of UP-PCBs emissions from various source categories from 1960 to 2019 in seven regions of China including North China (A), Northeast China (B), East China (C), Central China (D), South China (E), Southwest China (F), and Northwest China (G).

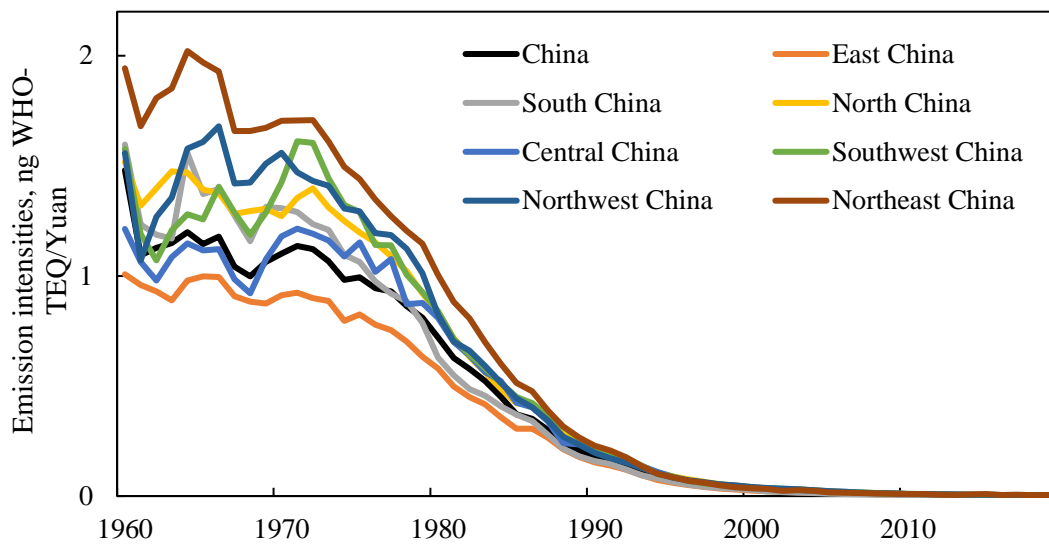


Fig. S7. Time trends of emission intensities for China and seven regions.

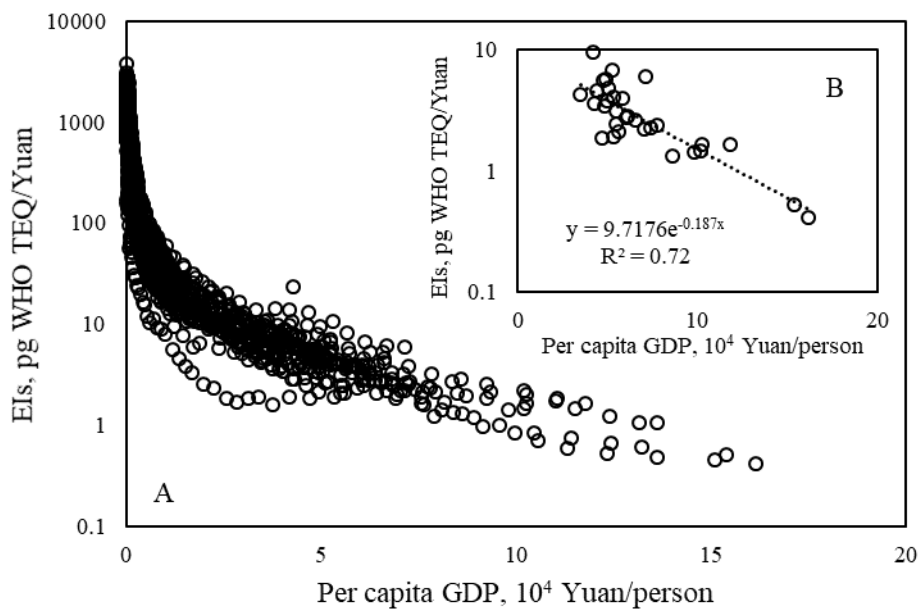


Fig. S8. Relationship between per capita GDP and emission intensities (EIs) with all provincial data from 1960 to 2019 (A) and only provincial data in 2019 (B).

Table S1. List of PCBs emission sources with best estimates (BE) and geometric standard deviations (SD) of EFs. The units of EF are ng WHO-TEQ/cigarette for cigarette smoking, ng WHO-TEQ/body for cremation of corpses, and ng WHO-TEQ/kg fuel consumed or product produced for all other sources.

Categories	Fuel/Product	No.	Sub-source	BE	SD	Ref ^a
Power & Heating	Anthracite/ Coke/ Bituminous coal/ Lignite/ Peat	1-5	Uncontrolled/ Controlled	0.105/0.0018	0.07/0.38	1
	Gas/Diesel	6		0.000823	0.1	2
	Residue fuel oil	7		0.000823	0.1	2
	Solid biomass	8	Uncontrolled/ Controlled	0.662/0.066	0.1	2
	Natural gas liquids	9		0.000823	0.1	2
Waste	Municipal waste	10	Uncontrolled/ Controlled	15.31/0.036	1.32	3,4,5,6
	Industry waste	11	Uncontrolled/ Controlled	15.31/0.036	1.32	3,4,5
	Non-organized waste burning	12		15.31	1.32	3,4,5
	Medical waste	13	Uncontrolled/ Controlled	13.29/1.77	0.09/1.04	1,3,5
Iron & Steel	Sintering iron	14	Uncontrolled/ Controlled	0.379/0.016	0.60	2,3,5,7,8
	Electric arc furnace	15	Uncontrolled/ Controlled	0.628/0.084	0.59	2,3,7,8,9
	Open Hearth Furnace	16	Uncontrolled/ Controlled	0.01/0.001	0.59	6
	Oxygen Blown Converter	17	Uncontrolled/ Controlled	0.01/0.001	0.59	6
	Hot rolling	18	Uncontrolled/ Controlled	0.62/0.025	1.01	1,2,3,10
Non-ferrous metals	Aluminum	19	Uncontrolled/ Controlled	1.37/0.14	0.48	2,6,7,8,9,11
	Lead	20	Uncontrolled/ Controlled	40/0.36	1.39	2,3,8
	Magnesium	21	Uncontrolled/ Controlled	2.462/0.2462	0.74	6,12
	Zinc	22	Uncontrolled/ Controlled	100/0.324	1.47	1,2,6,7,9
	Copper	23	Uncontrolled/ Controlled	0.92/0.092	0.88	1,2,6,7,8,13,14
	Secondary lead	24	Uncontrolled/ Controlled	1.1/0.002	1.67	2,3,8
	Secondary zinc	25	Uncontrolled/ Controlled	100/1.33	1.25	1,6,15

	Secondary copper	26	Uncontrolled/ Controlled	9.77/0.83	1.55	1,6,9,11,13
	Secondary aluminum	27	Uncontrolled/ Controlled	1.616/0.162	0.19	2
Cement	Cement	28	Uncontrolled/ Controlled	3.844/0.216	0.58	2,3,7
	Machine diesel	29		0.000823	0.1	2
Industry	Anthracite/ Coke/ Bituminous coal/ Lignite/ Peat	30-34	Uncontrolled/ Controlled	0.105/0.0018	0.07/0.38	1
boilers	Natural gas liquid	35		0.000823	0.1	2
	Residue fuel	36		0.000823	0.1	2
	Solid biomass	37		0.066	0.1	2
	Anthracite	38		0.084	0.65	16,17,18
	Coke/ Bituminous coal/ Lignite/ Peat	39-42		0.084	0.65	16,17,18
Residential	Natural gas liquids/Kerosene	43-44		0.000823	0.1	2
	Firewood	45		0.016	0.17	16
	Straw/Dung cake	46-47		0.016	0.17	16
	Agricultural waste	48		1.525	0.89	6
Open biomass	Savanna	49		0.03	0.89	16,9
	Forest fire/Deforestation/Peat/Woodland	50-53		0.1	0.89	16,19
			Beehive			
	Coke production	54	coke/Uncontrolled/ Controlled	0.2/0.2/0.002	0.96	2,7
Others	Brick production	55	Uncontrolled/ Controlled	0.20/0.02	0.50	2
	Gas Flaring/ Machine diesel in agriculture	56-57		0.000823	0.1	2
	Pulp and paper mill	58	Uncontrolled/ Controlled	0.696/0.070	0.5	2
	Cremation of corpses	59	Uncontrolled/ Controlled	0.006/0.0006	0.5	2

Vinyl chloride monomer manufacturing facilities	60	Uncontrolled/ Controlled	0.122/0.012	0.5	2
Motor gasoline/ Aviation gasoline/ Biodiesel/ Jet Kerosene/ Gas& Diesel	61-65		0.000823	0.1	2
Lime	66	Uncontrolled/ Controlled	0.108/0.011	0.1	2

Notes: ^a1.Pham et al., 2019; 2.Cui et al., 2013; 3.Liu et al., 2013b; 4.Zhang et al., 2011; 5.Li et al., 2017; 6. UNEP; 7.Cui et al., 2015; 8.Yu et al., 2006; 9.Antunes et al., 2012; 10.Lv et al., 2011; 11.Ba et al., 2009b; 12.Nie et al., 2011; 13.Nie et al., 2012a; 14.Hu et al., 2013; 15.Ba et al., 2009a; 16.Lee et al., 2005; 17.Hopan et al., 2009; 18. Šyc et al., 2010; 19.Lohmann et al., 2006

Table S2. Emission profiles (percentage of mass) of 12 dioxin-like UP-PCBs used to derived congener specific EFs.

No.	PCB77	PCB81	PCB105	PCB114	PCB118	PCB123	PCB126	PCB156	PCB157	PCB167	PCB169	PCB189	Refs
1-5	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
6	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
7	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
8	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005
9	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
10	25.2	6.7	10.0	3.7	15.9	4.8	10.6	5.6	3.7	3.8	5.0	5.1	Wang et al., 2010a; Wang et al., 2010b; Liu et al., 2013
11	25.2	6.7	10.0	3.7	15.9	4.8	10.6	5.6	3.7	3.8	5.0	5.1	Wang et al., 2010a; Wang et al., 2010b; Liu et al., 2013a
12	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005
13	13.3	8.4	21.1	6.2	11.5	6.2	8.4	5.9	3.5	4.6	4.3	6.5	Liu et al., 2013a
14	18.1	4.4	11.6	2.8	33.3	4.5	11.9	4.7	2.0	2.3	1.8	2.6	Aries et al., 2006; Lv et al., 2011; Liu et al., 2013a
15	18.5	3.1	18.1	2.3	37.4	5.7	5.9	4.0	1.4	1.2	1.1	1.4	Liu et al., 2013a
16	18.5	3.1	18.1	2.3	37.4	5.7	5.9	4.0	1.4	1.2	1.1	1.4	Liu et al., 2013a
17	10.8	1.9	17.5	2.3	42.8	4.6	3.2	9.6	2.8	1.2	2.2	1.1	Li et al., 2014
18	11.9	6.3	13.7	5.6	34.5	6.1	4.6	6.0	3.0	3.0	2.5	2.8	Nie et al., 2012b
19	0.0	0.0	0.0	0.0	0.0	0.0	90.7	0.0	0.0	0.0	9.3	0.0	Yu et al., 2006
20	0.0	0.0	0.0	0.0	0.0	0.0	96.7	0.0	0.0	0.0	3.3	0.0	Yu et al., 2006
21	7.0	0.4	19.1	14.0	33.1	5.6	0.2	12.3	4.4	2.0	0.0	2.0	Nie et al., 2011
22	0.0	0.0	0.0	0.0	0.0	0.0	93.6	0.0	0.0	0.0	6.4	0.0	Yu et al., 2006

23	10.4	1.6	13.4	7.5	20.2	7.3	1.3	9.7	9.7	9.7	0.3	9.0	Nie et al., 2012a; Yu et al., 2006
24	89.7	0.0	0.0	0.0	0.0	0.0	9.5	0.0	0.0	0.0	0.9	0.0	Ba et al., 2009a; Yu et al., 2006
25	0.0	0.0	0.0	0.0	0.0	0.0	2.8	0.0	96.9	0.0	0.3	0.0	Ba et al., 2009a; Yu et al., 2006
26	5.0	1.7	9.8	9.8	12.2	8.6	1.9	14.7	12.3	12.3	0.7	11.0	Ba et al., 2009b; Nie et al., 2012a; Yu et al., 2006
27	7.6	5.8	16.5	0.0	66.4	0.0	3.0	0.0	0.0	0.0	0.7	0.0	Ba et al., 2009b; Yu et al., 2006
28	39.9	5.5	9.2	1.4	27.3	9.4	2.1	2.1	0.7	1.8	0.3	0.4	Juan et al., 2016; Liu et al., 2013a
29	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
30-34	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
35	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
36	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
37	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005
38	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
39-42	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
43-44	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
45	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005
46-47	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005
48	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005
49	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005
50-53	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005

54	20.5	3.8	21.0	3.2	41.2	4.3	1.3	2.1	0.6	0.8	0.4	0.7	Liu et al., 2009
55	39.9	5.5	9.2	1.4	27.3	9.4	2.1	2.1	0.7	1.8	0.3	0.4	Juan et al., 2016; Liu et al., 2013a
56-57	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
58	0.0	7.1	0.0	71.1	0.0	0.0	21.1	0.0	0.0	0.0	0.7	0.0	Yu et al., 2006
59	11.6	0.0	31.9	1.4	42.0	4.3	0.0	4.3	1.4	2.9	0.0	0.0	Lee et al., 2005
60	7.5	8.9	18.6	6.1	40.4	9.0	0.9	5.1	1.1	0.7	0.1	1.5	Liu et al., 2012
61-65	5.6	0.0	22.4	1.3	53.8	3.7	1.0	7.3	1.4	2.9	0.1	0.5	Lee et al., 2005
66	39.9	5.5	9.2	1.4	27.3	9.4	2.1	2.1	0.7	1.8	0.3	0.4	Juan et al., 2016; Liu et al., 2013a

Table S3. List of 12 PCBs congeners included in this study and their TEF values.

Congeners	Abbreviation	WHO 1998 TEF ¹	WHO 2005 TEF ²
3,3',4,4'-tetraCB	PCB77	0.0001	0.0001
3,4,4',5-tetraCB	PCB81	0.0001	0.0003
2,3,3',4,4'-pentaCB	PCB105	0.0001	0.00003
2,3,4,4',5-pentaCB	PCB114	0.0005	0.00003
2,3',4,4',5-pentaCB	PCB118	0.0001	0.00003
2',3,4,4',5-pentaCB	PCB123	0.0001	0.00003
3,3',4,4',5-pentaCB	PCB126	0.1	0.1
2,3,3',4,4',5-hexaCB	PCB156	0.0005	0.00003
2,3,3',4,4',5'-hexaCB	PCB157	0.0005	0.00003
2,3',4,4',5,5'-hexaCB	PCB167	0.00001	0.00003
3,3',4,4',5,5'-hexaCB	PCB169	0.01	0.03
2,3,3',4,4',5,5'-hexaCB	PCB189	0.0001	0.00003

Notes:¹Van den Berg et al., 1998; ²Van den Berg et al., 2006.

Table S4. Definitions of the seven regions in China.

Regions	Provinces
North China	Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia
Northeast China	Liaoning, Jilin, Heilongjiang
East China	Shanghai, Jiangsu, Zhejiang, Anhui, Fujian, Jiangxi, Shandong
Central China	Henan, Hubei, Hunan
South China	Guangdong, Guangxi, Hainan
Southwest China	Chongqing, Sichuan, Guizhou, Yunnan, Tibet
Northwest China	Shaanxi, Gansu, Qinghai, Ningxia, Xinjiang

Table S5. TEQ/u (g WHO-TEQ per unit of mass of UP-PCBs emissions) for nine source categories.

Sources	Values, g WHO-TEQ/g.
Power & heating	5×10^{-4}
Waste burning	1×10^{-4}
Iron & Steel production	6.8×10^{-3}
Non-ferrous metals	4.6×10^{-3}
Cement production	2.2×10^{-3}
Industry boilers	1.1×10^{-3}
Residential	1×10^{-4}
Open biomass	4×10^{-5}
Others	3×10^{-3}

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