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Development of a unit-based industrial emission inventory in the Beijing–Tianjin–Hebei region and resulting improvement in air quality modeling

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There are several studies estimating emissions in BTH region. We compare emissions reported after 2014 (the base year is 2010 or later) with our study, which are shown in **Fig.S3**.

For SO_2, NOx and $PM_{2.5}$, in Beijing, the total emissions in our study (for 2014) are significantly lower than those in previous studies (for 2010-2013), probably because stringent control measures have been implemented during 2010-2014. In Tianjin and Hebei, the emission estimates from different studies are much closer to each other. For NMVOCs, Wang et al. (2014), Wu et al. (2017) and Cai et al. (2017) generally have similar results to this study. The NMVOCs emissions by Zheng et al. (2017) are higher in Beijing and Tianjin, and lower in Hebei compared with other studies. For NH_3 emissions, the estimates from various studies are generally similar, except that the estimate of Zhou et al. (2015) is much higher.

We compare the meteorological parameters simulated by WRFv3.7 with observational data obtained from the National Climatic Data Center (NCDC), where hourly or 3-h observations are available for about 28 sites in the innermost domain. The variables of interest include the temperature at 2 m, wind speed at 10 m, and humidity at 2 m. The statistical indices used include the mean observation (Mean OBS), mean simulation (Mean SIM), mean bias, gross error, root mean square error (RMSE), and index of agreement (IOA), the definitions of which are provided in Emery et al. (2001). Table S1 summarizes the model performance statistics as well as the benchmarks suggested by Emery et al. (2001). Obviously almost all indices fall within the benchmark range except that the MB of humidity slightly exceeds the range in July. In summary, the statistics indicate an overall decent performance of meteorological predictions.

We summarize the model performance for major air pollutants at each individual site in Beijing (12 sites out of a total of 80 sites in the BTH region) in Table S8-S11. The time series of $PM_{2.5}$ concentration at representative urban sites (Wanshouxigong and Dongsi) and suburban sites (Huairou and Shunyi) is shown in Fig.S7-S8. For the urban sites, the concentrations of $PM_{2.5}$, SO_2 and NO_2 are much lower with the unit-based inventory than with the proxy-based inventory. For the suburban sites, however, the concentrations are either slightly higher or slightly lower with the unit-based inventory than with the proxy-based inventory. The situation for ozone is quite the opposite. The ozone concentration at urban sites is higher with the unit-based inventory than with the proxy-based inventory.

In suburban sites, it is lower with the unit-based inventory than with the proxy-based inventory. In addition, for the simulations with the unit-based inventory, the NME and MFE are usually lower than those with the proxy-based inventory while the correlation efficient is usually higher, which means that the error is generally smaller and the trend is more similar to the observation when the unit-based inventory is used.

The figures of time series of $PM_{2.5}$ concentration corroborates the preceding conclusion. At urban sites, the concentration with the unit-based inventory is substantially lower than that with the proxy-based inventory throughout the simulation periods. For suburban sites, the concentration is slightly lower with the unit-based inventory than that with the proxy-based inventory in most of the simulation period.

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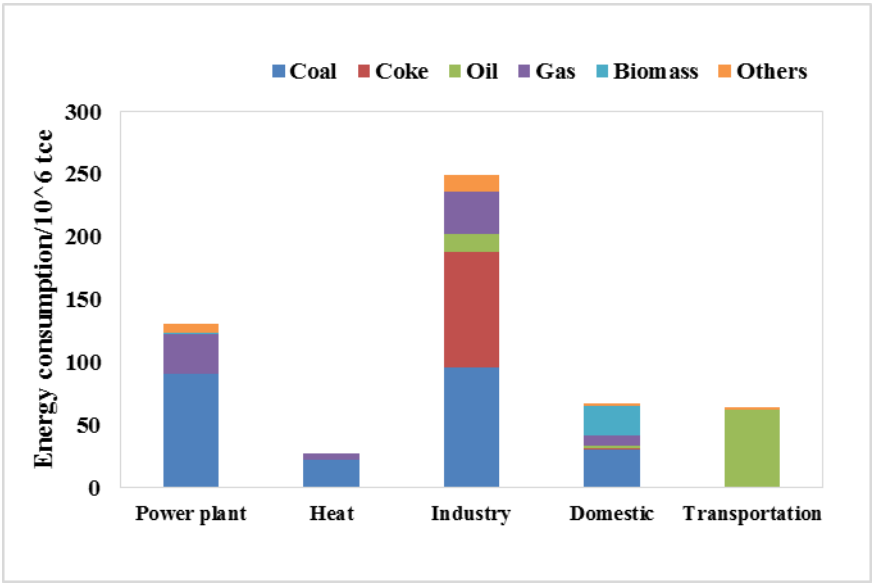


Fig.S1. Energy consumption in Jing-Jin-Ji region in 2014.

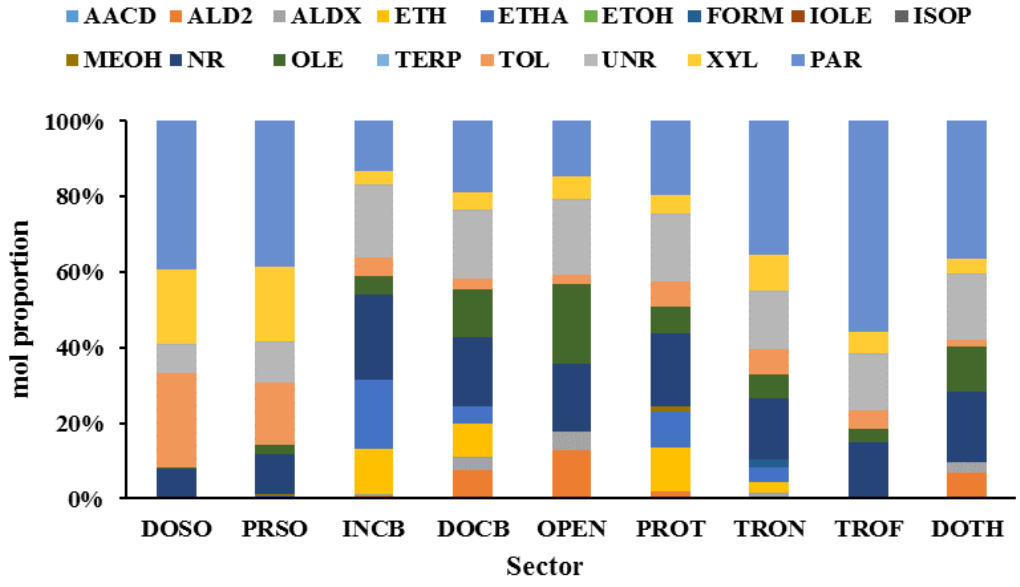
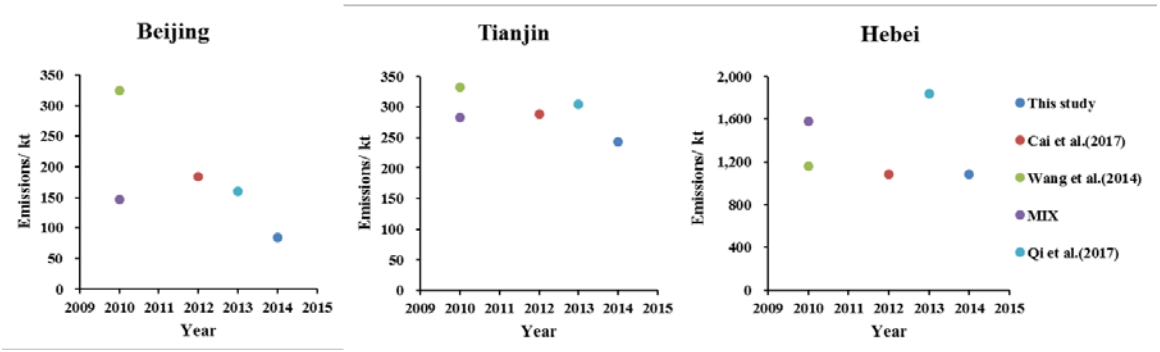
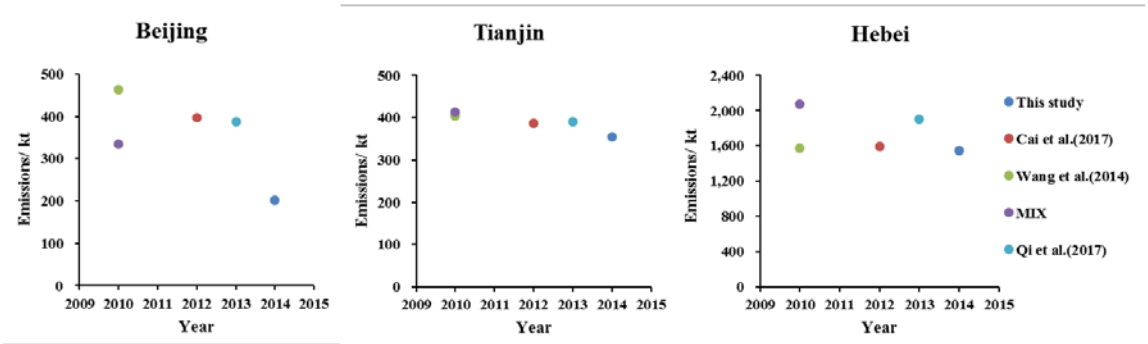


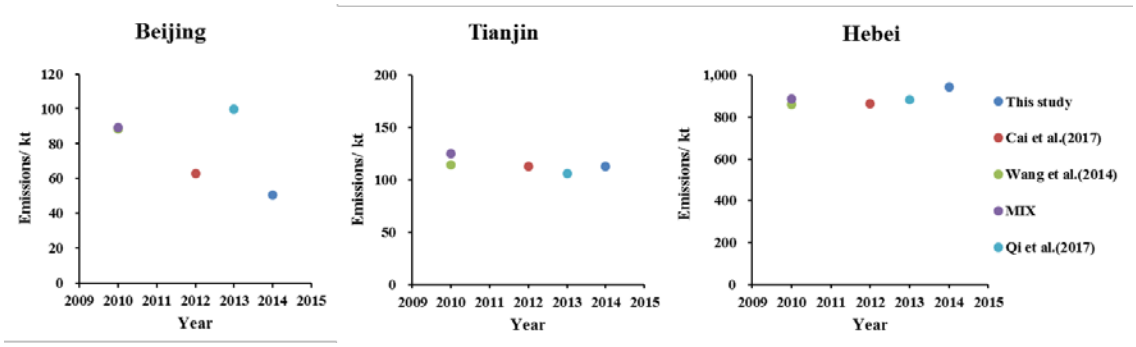
Fig.S2. NMVOCs speciation by sector according to the Carbon Bond 05 (CB05) gas chemistry scheme. DOSO is domestic solvent use; PRSO is industrial solvent use; INCB is industrial combustion; DOCB is domestic combustion; OPEN is open burning; PROT is industrial process; TRON is on-road transport; TROF is off-road transport; DOTH is other domestic use.



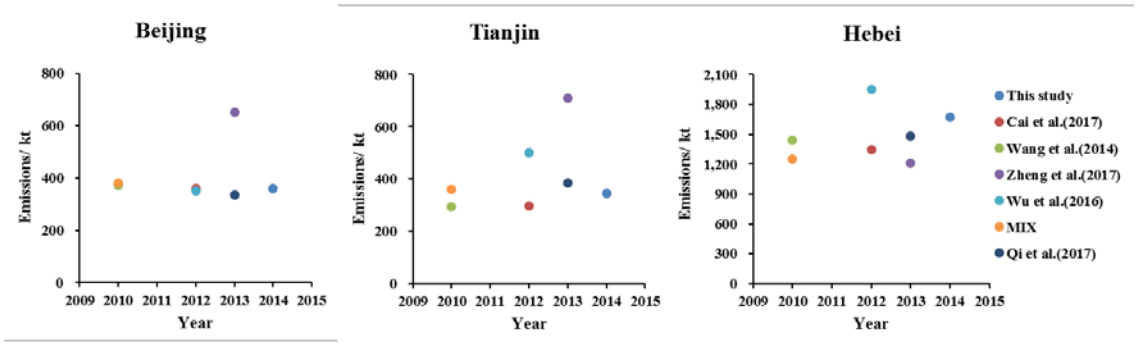
(a)SO_2



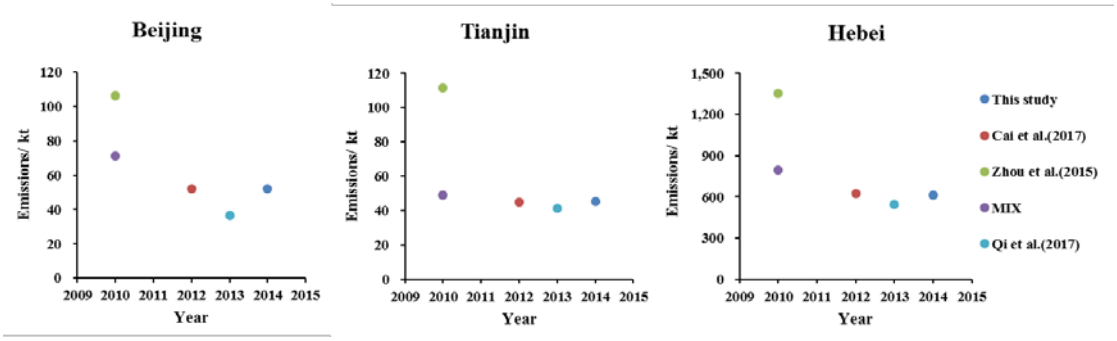
(b)NOx



(c)$PM_{2.5}$



(d)NMVOCs



(e)NH_3

Fig.S3. Emissions compared with other studies.

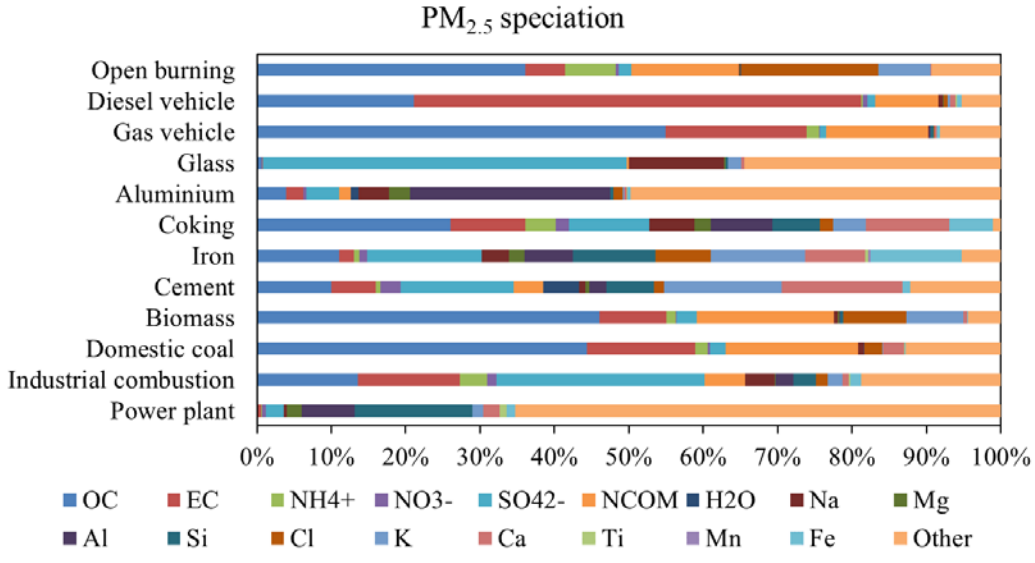


Fig.S4. $PM_{2.5}$ speciation profile of major sectors.

Fig.S5. The observational sites in Beijing.

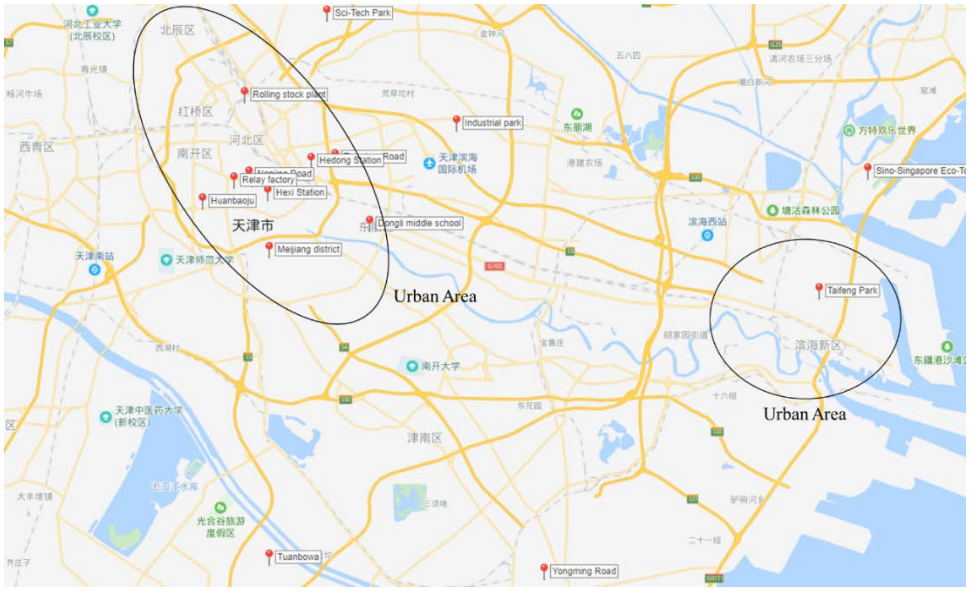


Fig.S6 The observational sites in Tianjin

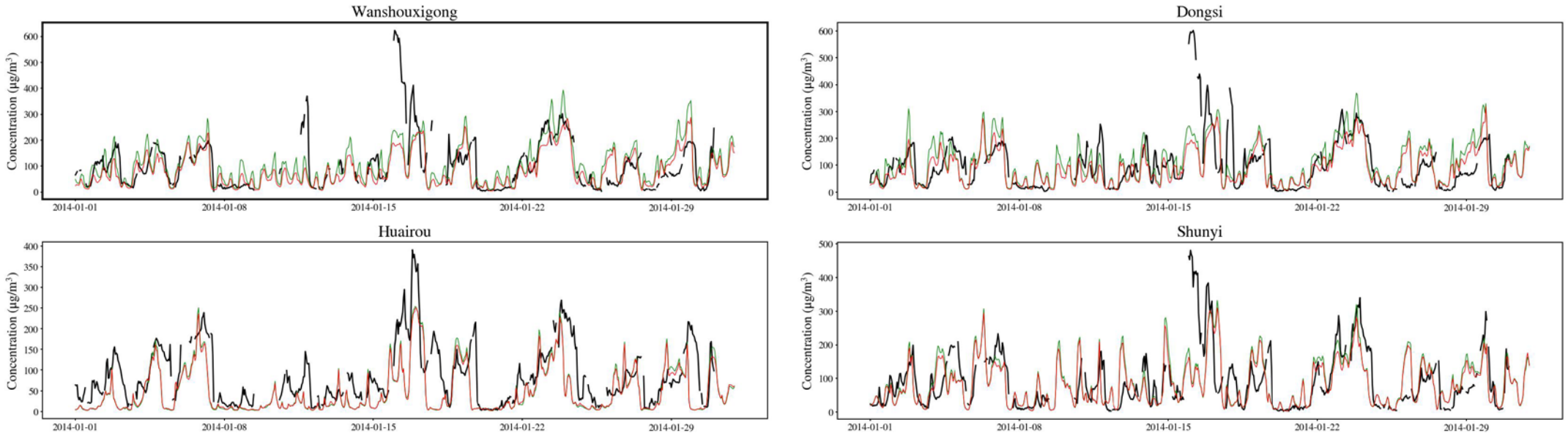


Fig.S7 The $PM_{2.5}$ concentration in January in Beijing (The black, green and red lines represent observation, results with proxy-based and unit-based inventories)

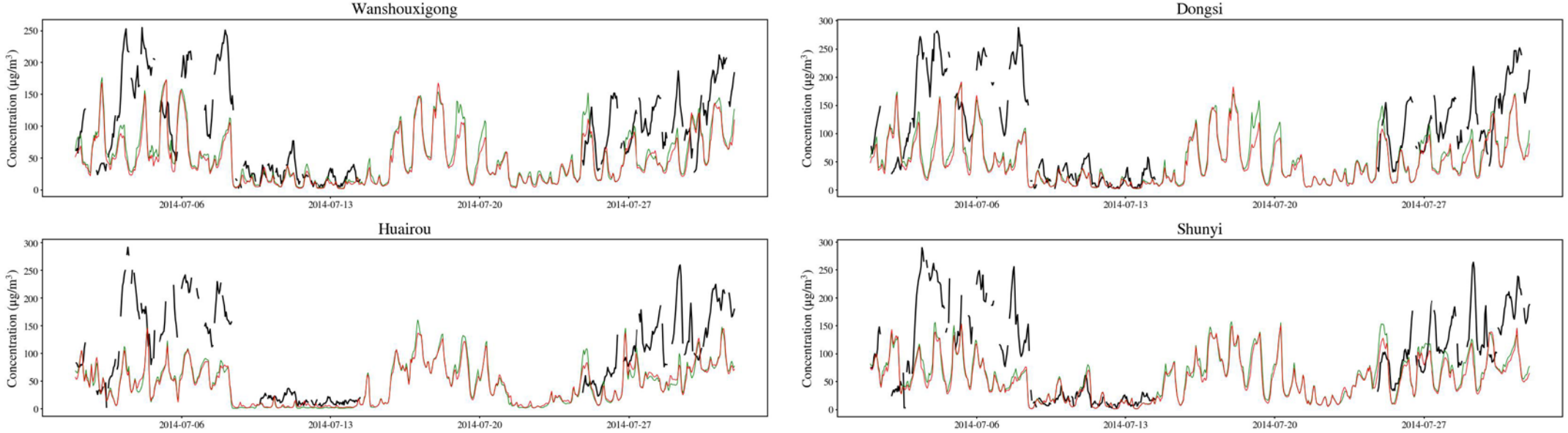


Fig.S8 The $PM_{2.5}$ concentration in July in Beijing

Table S1 Statistics for validation results of WRF model.

Variable	statistics	Unit	January	July	Benchmark
Wind Speed	Mean OBS	m/s	2.68	2.53	
	Mean SIM	m/s	2.65	2.48	
	Mean Bias	m/s	-0.03	-0.05	≤±0.5
	Gross Error	m/s	1.2	1.14	≤2
	RMSE	m/s	1.77	1.56	≤2
	IOA		0.71	0.67	≥ 0.6
Wind Direction	Mean OBS	deg	230.79	205.37	
	Mean SIM	deg	244.79	202.55	
	Mean Bias	deg	3.33	-1.18	≤±10
	Gross Error	deg	41.6	45.93	
Temperature	Mean OBS	K	270.30	298.54	
	Mean SIM	K	269.87	298.77	
	Mean Bias	K	-0.43	0.23	≤±0.5
	Gross Error	K	1.78	2.04	≤2
	RMSE	K	2.28	2.6	
	IOA		0.96	0.94	≥ 0.8
Humidity	Mean OBS	g/kg	1.5	14.0	
	Mean SIM	g/kg	1.6	12.9	
	Mean Bias	g/kg	0.07	-1.11	≤±1
	Gross Error	g/kg	0.31	2.1	≤2
	RMSE	g/kg	0.44	2.7	
	IOA		0.87	0.8	≥0.6

Table S2 Proxies used in the proxy-based inventory for each sector.

Sector	Allocate to county	Allocate to grid
Power plant, steel, cement	GDP of secondary industry	Population density

Industrial combustion, other industrial process	GDP of secondary industry	Population density
Domestic fuel	Total GDP	Population density
Domestic biomass	GDP of first industry	Population density
Transportation	GDP of tertiary industry	Road
Open burning	GDP of first industry	Population density
Livestock	GDP of first industry	Population density
Fertilizer application	GDP of first industry	Population density
Domestic solvent use	Total GDP	Population density
Industrial solvent use	GDP of secondary industry	Population density

Table S3 Comparison of industrial sectors covered in previous studies and this study (The underlined sectors are newly added to this study).

Study	Sector	Region
Zhao et al. (2008), Chen et al. (2014), Liu et al. (2015), Li et al. (2017)	Power plants	China
Wang et al. (2016), Wu et al. (2015)	Iron plants	China
Lei et al. (2011), Chen et al. (2015)	Cement plants	China
Qi et al. (2017)	Power plants, iron plants, cement factories, coking factories, heating plants, other industries	BTH
This study	Power plants, iron plants, cement factories, coking factories, nonferrous metals, glass factories, brick factories, lime factories, ceramics factories, refinery factories, chemical plants, industrial	BTH

	boilers

Table S4 Types of products used for each sector.

Industrial sector	Product
Power plant	Use energy consumption to estimate
Industrial boiler	Use energy consumption to estimate
Iron and steel production	Pig iron, crude steel, rolled steel
Non-ferrous metal smelter	Alumina, aluminum, copper
Coking	Coke
Cement	Cement, clinker
Glass	Glass
Brick	Brick
Lime	Lime
Ceramics	Ceramics
Refinery	Crude oil, ethylene
Chemical industries	Ammonia, caustic soda, soda ash, sulfuric acid, nitric acid

Table S5 Data source of stack information

Sector	Stack height	Flue gas temperature	Chimney diameter	Flue gas velocity
Power plant	Compilation of power industry statistics	National information platform of pollutant discharge permit		
Cement	Emission standard of air pollutants for cement industry			
Glass, brick, lime and ceramics industries	Emission standard of air pollutants for industrial kiln and furnace			

Non-ferrous metal smelter, coking, refinery and chemical industries	National information platform of pollutant discharge permit

Table S6 Vertical distribution of emissions for each industrial sector in the unit-based inventory with plume rise considered

Layer	Sigma value	Level height (m)	Power plants		Iron plants		Cement plants		Industrial boilers		Industrial process	
			Jan	Jul	Jan	Jul	Jan	Jul	Jan	Jul	Jan	Jul
1	0.995	35	0%	0%	0%	0%	3%	3%	3%	4%	3%	6%
2	0.99	85	0%	0%	0%	0%	9%	11%	21%	21%	28%	31%
3	0.98	140	0%	0%	0%	6%	26%	32%	32%	41%	45%	36%
4	0.96	210	6%	9%	60%	85%	49%	49%	39%	31%	20%	24%
5	0.94	310	15%	17%	38%	9%	13%	5%	5%	2%	3%	3%
6	0.91	440	47%	45%	2%	0%	1%	0%	0%	0%	0%	0%
7	0.86	610	31%	29%	0%	0%	0%	0%	0%	0%	0%	0%

Table S7 Vertical distribution of emissions for each industrial sector in the proxy-based inventory

Layer	Sigma value	Level height (m)	Power plants	Iron plants	Cement plants	Industrial boilers	Industrial process
1	0.995	35	0%	6%	6%	50%	6%
2	0.99	85	10%	26%	26%	30%	26%
3	0.98	140	10%	68%	68%	20%	68%
4	0.96	210	30%	0%	0%	0%	0%
5	0.94	310	20%	0%	0%	0%	0%
6	0.91	440	20%	0%	0%	0%	0%
7	0.86	610	10%	0%	0%	0%	0%

5

Table S8 The statistics for model performance of $PM_{2.5}$ with proxy-based and unit-based inventories

Months	Sites	Concentration			NMB		NME		MFB		MFE		R	
		proxy-based	unit-based	OBS	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based
Jan	Wanshouxigong	111.2	88.0	108.4	3%	-19%	54%	49%	20%	-3%	62%	60%	0.58	0.60
	Dingling Tomb	36.8	36.9	69.3	-47%	-47%	54%	53%	-59%	-55%	72%	68%	0.67	0.68
	Dongsi	112.2	91.6	104.1	8%	-12%	58%	51%	24%	6%	64%	61%	0.55	0.56
	Heaven Temple	110.7	90.3	97.6	13%	-8%	58%	51%	31%	10%	64%	61%	0.59	0.60
	Nongzhanguan	92.6	77.7	101.9	-9%	-24%	52%	50%	4%	-10%	59%	58%	0.57	0.59
	Guanyuan	110.5	86.4	100.6	10%	-14%	60%	51%	23%	1%	65%	61%	0.55	0.56
	Haidian	86.8	70.0	109.3	-21%	-36%	55%	53%	-17%	-35%	66%	67%	0.53	0.54
	Shunyi	89.4	83.3	92.3	-3%	-10%	56%	54%	8%	3%	62%	61%	0.55	0.55
	Huairou	49.8	48.5	86.9	-43%	-44%	57%	55%	-71%	-69%	86%	82%	0.61	0.62
	Changping	74.7	70.3	85.6	-13%	-18%	54%	51%	-4%	-8%	58%	56%	0.57	0.58
	Olympic center	93.6	82.5	94.8	-1%	-13%	56%	50%	9%	1%	62%	59%	0.57	0.59
	Gucheng	77.0	63.2	102.0	-25%	-38%	50%	51%	-19%	-37%	60%	64%	0.59	0.60
Jul	Wanshouxigong	55.7	50.0	96.4	-42%	-48%	55%	58%	-51%	-61%	69%	75%	0.53	0.52
	Dingling Tomb	24.6	26.1	83.7	-71%	-69%	74%	72%	-106%	-102%	112%	109%	0.55	0.57
	Dongsi	57.3	52.2	110.0	-48%	-53%	57%	59%	-54%	-61%	72%	75%	0.56	0.55
	Heaven Temple	58.0	52.5	103.3	-44%	-49%	56%	58%	-52%	-61%	70%	74%	0.51	0.50
	Nongzhanguan	54.4	50.2	91.7	-41%	-45%	54%	55%	-54%	-59%	70%	72%	0.50	0.50
	Guanyuan	54.8	49.8	99.6	-45%	-50%	55%	57%	-59%	-67%	73%	78%	0.56	0.56
	Haidian	42.8	39.9	99.8	-57%	-60%	61%	63%	-88%	-91%	93%	95%	0.60	0.60
	Shunyi	60.2	55.8	101.7	-41%	-45%	54%	55%	-41%	-47%	67%	70%	0.58	0.57
	Huairou	44.8	45.0	101.2	-56%	-56%	60%	59%	-89%	-78%	96%	85%	0.68	0.68
	Changping	37.2	39.0	91.9	-60%	-58%	65%	63%	-77%	-74%	86%	84%	0.65	0.67
	Olympic center	50.5	47.1	104.8	-52%	-55%	57%	59%	-73%	-77%	81%	83%	0.60	0.59
	Gucheng	38.2	36.9	97.2	-61%	-62%	63%	64%	-96%	-98%	99%	100%	0.64	0.63

Table S9 The statistics for model performance of NO_2 with proxy-based and unit-based inventories

Months	Sites	Concentration			NMB		NME		MFB		MFE		R	
		proxy-based	unit-based	OBS	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based
Jan	Wanshouxigong	135.3	96.0	81.1	67%	18%	81%	47%	43%	11%	59%	46%	0.60	0.64
	Dingling Tomb	39.9	41.4	37.2	7%	11%	60%	58%	4%	11%	60%	60%	0.64	0.66
	Dongsi	135.1	101.4	66.3	104%	53%	116%	75%	59%	37%	72%	60%	0.43	0.42
	Heaven Temple	134.9	101.8	73.4	84%	39%	94%	61%	48%	21%	60%	50%	0.57	0.58
	Nongzhanguan	108.0	93.9	68.0	59%	38%	79%	59%	33%	26%	58%	51%	0.60	0.63
	Guanyuan	141.7	99.9	76.1	86%	31%	103%	61%	42%	12%	63%	52%	0.59	0.59
	Haidian	106.1	80.0	93.5	13%	-14%	66%	50%	-12%	-32%	62%	60%	0.47	0.48
	Shunyi	100.7	92.7	56.8	77%	63%	93%	80%	42%	37%	62%	58%	0.61	0.61
	Huairou	54.8	54.0	56.7	-3%	-5%	72%	68%	-45%	-41%	85%	81%	0.55	0.57
	Changping	102.3	95.3	57.0	80%	67%	98%	87%	39%	34%	60%	57%	0.54	0.56
	Olympic center	113.9	100.0	67.7	68%	48%	86%	66%	43%	37%	63%	58%	0.60	0.62
	Gucheng	95.0	73.6	75.9	25%	-3%	61%	46%	12%	-9%	55%	52%	0.61	0.64
Jul	Wanshouxigong	22.1	18.4	41.3	-46%	-56%	59%	62%	-72%	-86%	83%	92%	0.17	0.15
	Dingling Tomb	5.7	7.2	17.1	-67%	-58%	70%	63%	-116%	-102%	118%	106%	0.34	0.37
	Dongsi	25.6	22.6	43.5	-41%	-48%	57%	58%	-65%	-72%	78%	81%	0.22	0.19
	Heaven Temple	24.5	21.0	36.4	-33%	-42%	56%	56%	-53%	-63%	73%	77%	0.24	0.21
	Nongzhanguan	22.8	21.7	44.9	-49%	-52%	60%	60%	-78%	-77%	87%	84%	0.27	0.27
	Guanyuan	21.4	18.1	42.2	-49%	-57%	61%	62%	-75%	-87%	86%	93%	0.11	0.12
	Haidian	14.9	13.7	54.3	-73%	-75%	74%	76%	-119%	-123%	121%	124%	0.07	0.09
	Shunyi	21.6	19.7	28.1	-23%	-30%	39%	41%	-36%	-43%	54%	57%	0.66	0.64
	Huairou	11.9	12.8	25.0	-52%	-49%	62%	60%	-96%	-86%	105%	95%	0.39	0.40
	Changping	15.1	17.1	32.5	-53%	-47%	58%	54%	-85%	-75%	90%	81%	0.27	0.28
	Olympic center	20.1	18.5	48.6	-59%	-62%	64%	64%	-93%	-96%	97%	99%	0.23	0.25
	Gucheng	12.4	12.2	45.6	-73%	-73%	74%	74%	-118%	-118%	119%	118%	0.23	0.24

Table S10 The statistics for model performance of SO_2 with proxy-based and unit-based inventories in January

Months	Sites	Concentration			NMB		NME		MFB		MFE		R	
		proxy-based	unit-based	OBS	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based
Jan	Wanshouxigong	102.2	93.4	62.0	65%	51%	77%	67%	60%	51%	69%	65%	0.51	0.51
	Dingling Tomb	36.0	36.7	35.4	2%	3%	73%	71%	-9%	-4%	70%	69%	0.43	0.44
	Dongsi	99.8	91.8	56.7	76%	62%	86%	75%	64%	57%	72%	67%	0.52	0.53
	Heaven Temple	99.7	92.0	47.1	112%	95%	124%	112%	81%	74%	88%	84%	0.30	0.30
	Nongzhanguan	88.0	82.7	58.9	50%	40%	69%	62%	49%	45%	64%	61%	0.50	0.52
	Guanyuan	101.2	91.6	54.8	85%	67%	97%	84%	67%	58%	77%	72%	0.51	0.51
	Haidian	89.7	81.7	58.2	54%	40%	81%	73%	48%	40%	72%	70%	0.47	0.46
	Shunyi	68.9	66.1	44.0	57%	50%	79%	75%	56%	53%	73%	71%	0.48	0.47
	Huairou	46.3	46.5	45.6	2%	2%	66%	63%	-8%	-4%	74%	71%	0.40	0.40
	Changping	66.3	64.0	57.6	15%	11%	58%	56%	12%	8%	56%	56%	0.46	0.46
	Olympic center	87.2	82.6	58.3	50%	42%	72%	66%	45%	42%	65%	62%	0.50	0.50
	Gucheng	80.6	72.4	52.9	52%	37%	79%	69%	47%	38%	71%	66%	0.52	0.52
Jul	Wanshouxigong	69.9	66.5	5.6	1144%	1083%	1168%	1110%	149%	145%	156%	153%	-0.31	-0.31
	Dingling Tomb	9.7	10.5	4.6	112%	128%	168%	178%	52%	58%	97%	98%	0.12	0.11
	Dongsi	74.6	71.8	9.6	680%	650%	696%	669%	135%	133%	141%	141%	-0.05	-0.06
	Heaven Temple	67.3	64.0	7.4	805%	762%	831%	790%	136%	133%	146%	144%	-0.27	-0.28
	Nongzhanguan	62.9	61.0	8.7	622%	600%	649%	627%	127%	128%	137%	138%	-0.07	-0.08
	Guanyuan	77.0	73.5	8.5	802%	761%	842%	802%	149%	147%	155%	153%	0.04	0.04
	Haidian	62.5	60.2	12.2	413%	394%	444%	424%	96%	95%	122%	120%	-0.26	-0.26
	Shunyi	36.8	33.5	6.5	463%	412%	498%	454%	112%	106%	130%	128%	-0.13	-0.15
	Huairou	22.9	23.7	4.5	405%	422%	435%	451%	112%	119%	126%	129%	-0.01	-0.02
	Changping	28.2	30.6	5.4	421%	466%	457%	493%	114%	122%	127%	133%	-0.06	-0.02
	Olympic center	64.2	61.8	5.0	1174%	1127%	1193%	1147%	141%	140%	149%	149%	-0.09	-0.08
	Gucheng	44.1	43.0	5.2	741%	720%	767%	744%	128%	128%	140%	139%	-0.19	-0.17

Table S11 The statistics for model performance of ozone with proxy-based and unit-based inventories

Months	Sites	Concentration			NMB		NME		MFB		MFE		R	
		proxy-based	unit-based	OBS	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based	proxy-based	unit-based
Jan	Wanshouxigong	11.0	14.0	12.0	-8%	17%	100%	115%	-86%	-71%	153%	149%	0.43	0.40
	Dingling Tomb	47.2	46.4	35.4	33%	31%	56%	54%	20%	21%	80%	78%	0.60	0.61
	Dongsi	12.0	14.7	23.3	-49%	-37%	71%	75%	-116%	-106%	139%	137%	0.50	0.44
	Heaven Temple	11.5	14.2	20.8	-45%	-32%	76%	82%	-108%	-97%	144%	143%	0.46	0.41
	Nongzhanguan	15.9	17.5	20.9	-24%	-16%	67%	69%	-87%	-80%	128%	127%	0.61	0.59
	Guanyuan	12.6	16.1	16.0	-21%	1%	82%	94%	-78%	-62%	144%	142%	0.50	0.43
	Haidian	18.3	22.8	14.9	23%	54%	99%	122%	-48%	-34%	139%	138%	0.53	0.42
	Shunyi	20.4	21.3	22.9	-11%	-7%	57%	57%	-66%	-59%	116%	113%	0.71	0.70
	Huairou	40.8	40.2	26.2	56%	53%	87%	86%	13%	13%	101%	100%	0.53	0.52
	Changping	26.9	28.1	25.9	4%	9%	55%	55%	-34%	-28%	90%	89%	0.65	0.65
	Olympic center	17.2	18.5	15.6	10%	18%	84%	92%	-50%	-47%	136%	136%	0.58	0.52
	Gucheng	20.6	25.1	31.5	-35%	-20%	64%	65%	-84%	-64%	112%	105%	0.51	0.47
Jul	Wanshouxigong	57.8	58.5	104.8	-45%	-44%	55%	55%	-89%	-86%	104%	102%	0.63	0.62
	Dingling Tomb	106.6	107.4	115.2	-7%	-7%	40%	38%	1%	1%	40%	39%	0.49	0.56
	Dongsi	55.1	55.5	92.8	-41%	-40%	57%	57%	-82%	-80%	104%	102%	0.55	0.54
	Heaven Temple	58.6	59.0	101.3	-42%	-42%	55%	55%	-82%	-78%	102%	99%	0.63	0.61
	Nongzhanguan	63.4	63.1	107.0	-41%	-41%	57%	57%	-58%	-57%	99%	98%	0.56	0.56
	Guanyuan	56.5	57.3	98.7	-43%	-42%	61%	61%	-79%	-77%	107%	105%	0.52	0.52
	Haidian	68.5	69.3	83.4	-18%	-17%	58%	57%	-28%	-26%	93%	92%	0.54	0.55
	Shunyi	70.2	71.2	101.9	-31%	-30%	44%	44%	-37%	-33%	63%	61%	0.70	0.69
	Huairou	92.8	91.1	112.0	-17%	-19%	41%	41%	-12%	-14%	46%	46%	0.54	0.54
	Changping	91.4	90.1	115.3	-21%	-22%	44%	42%	-17%	-18%	52%	51%	0.51	0.55
	Olympic center	64.3	64.7	90.3	-29%	-28%	59%	59%	-45%	-43%	99%	97%	0.52	0.52
	Gucheng	80.5	80.4	100.4	-20%	-20%	51%	50%	-15%	-15%	73%	72%	0.58	0.59