Supplementary Materials: Clearing the Air: Assessing the Effectiveness of Emission Policy in Qinhuangdao's Key Industries

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Section S1. Beijing-Tianjin-Hebei (BTH) Air Pollution Transmission Channel Cities

BTH air pollution transmission channel cities are also known as "2 + 26 cities", which are located on the transport path of air pollutants from the BTH region. "2" refers to Beijing and Tianjin, while "26" refers to the 26 cities under the jurisdiction of the four provinces around Beijing and Tianjin. They are Shijiazhuang, Tangshan, Handan, Xingtai, Baoding, Cangzhou, Langfang, and Hengshui in Hebei Province; Taiyuan, Yangquan, Changzhi, and Jincheng in Shanxi Province; Jinan, Zibo, Jining, Dezhou, Liaocheng, Binzhou, and Heze in Shandong Province; Zhengzhou, Kaifeng, Anyang, Hebi, Xinxiang, Jiaozuo, and Puyang in Henan Province (including Xiong'an New Area, Xinji City and Dingzhou City in Hebei Province, Jiyuan City in Henan Province).

Section S2. Emission Factors

1. Emissions Factors under the Baseline and BAU Scenario

The emission factors used in this study are derived from previous studies. The emission factors of this study primarily refer to the emission inventory guidelines for various pollution sources issued by the Ministry of Ecology and Environment [1–4] and the "Technical Manual for the Preparation of Urban Air Pollutant Emission Inventories" [5].

For SO_2, PM, BC, and OC produced by coal burning, the material balance algorithm was used to calculate their emission factors in Equations (1)–(4):

$$EF_{SO_2}=2 \times S \times (1\text{-}sr) \tag{1}$$

$$EF_{PM}=A\text{ar} \times (1\text{-}ar) \times f_{PM} \tag{2}$$

$$EF_{BC}=EF_{PM_{2.5}} \times f_{PM} \tag{3}$$

$$EF_{OC}=EF_{PM_{2.5}} \times f_{OC} \tag{4}$$

where S is the average base sulfur content received by coal burning; sr is the proportion of sulfur into bottom ash, %; Aar is the average base ash received from coal burning; ar is the proportion of ash entering the bottom ash, %; f_{PM} refers to the proportion of particulate matter in a certain particle size range ($PM_{2.5}$ and PM_{10}) in the total particulate matter generated by emission sources; f_{BC} is the proportion of BC to $PM_{2.5}$; f_{OC} is the proportion of OC to $PM_{2.5}$.

For other air pollutants, the emission factors referred to the "Technical Manual for the Preparation of Urban Air Pollutant Emission Inventories" [5]. For the aviation source in Qinhuangdao, the emission factors of SO_2 referred to previous research of our team [6].

2. The Ultra-Low Emission Factors of PM_{10}, SO_2, and NOx

For the iron and steel industry, the ultra-low emission factors for PM_{10}, SO_2, and NOx in Qinhuangdao were calculated by combining the concentration limits specified in the "Standards for Ultra-low Emission of Air Pollutants in the Iron and Steel Industry" (DB13/2169-2018) [7] issued by Hebei Province in 2018 and the theoretical flue gas volume (Tables S3–S6) [8,9]. For the cement and flat glass industries, the calculation method for

ultra-low emission factors was similar to that of the iron and steel industry. It involves using the concentration limits for PM_{10}, SO_2, and NOx specified in the "Standards for Ultra-low Emission of Air Pollutants in the Cement Industry" (DB 13/2167—2020) and the "Standards for Ultra-low Emission of Air Pollutants in the Flat Glass Industry" (DB 13/2168—2020) issued by Hebei Province in 2020 [10], combined with the benchmark emissions proposed by the Ministry of Ecology and Environment (Tables S7 and S8) [11,12]. These factors were then used to calculate the ultra-low emissions of PM_{10}, SO_2, and NOx in the cement and flat glass industries. The emission factors were calculated by Equation (5):

$$EF_{p,s} = C_{p,s} \times V_{p,s} \tag{5}$$

where C represents the ultra-low concentration limits emitted from three typical industries (mg/m^3) and V represents the theoretical flue gas volume or the benchmark emissions (Nm^3/t of product).

Section S3. Calculation Formula

Emissions of pollutants were calculated using the emission factor method provided in Equation (6):

$$E = EF \times A \times (1 - \eta) \tag{6}$$

where E is the emission of each source, EF is the emission factor, A is the activity data, and η represents the removal efficiency of the control device.

1. Fossil Fuel Combustion Sources

Fossil fuels have propelled the development of modern society. The sources include power plants, industrial combustion, and residential fuel combustion. Emissions from power plants were calculated using a bottom–up approach. Detailed unit-based data, including the geographical location, installed capacity, sulfur content, fuel consumption, electricity production, and emission control technologies of power plants in Qinhuangdao were gathered and checked to estimate emissions. The related calculation data were collected from the literature, reports, and city-level statistical yearbooks.

The emissions were calculated using the emission factor method as in Equation (7):

$$E_p = \sum_{i,j,k,l} A_{i,j,k} \times EF_{p,j,k} \times (1 - \eta_{p,j,k,l}) \tag{7}$$

where E represents the total emissions of pollutants; p, i, j, k, l represent the pollutant type, plant, source category, type of fuel, and emission control technology, respectively. A is the activity data; EF represents the assumed average emission factor; η is the removal efficiency.

2. On-Road Mobile Sources

The emissions of on-road mobile sources were calculated using Equation (8):

$$E_p = \sum_{i} P_i \times VKT_i \times EF_{i,p} \tag{8}$$

where E is the total on-road mobile emissions, including SO_2, NOx, CO, PM_{10}, $PM_{2.5}$, and VOCs; p and i represent the pollutant and vehicle type, respectively; P represents the annual total number of motor vehicles; VKT is the annual average vehicle-kilometers travelled; and EF is the emission factor.

3. Non-Road Mobile Sources

Non-road mobile emissions originate from airplanes, ships, agricultural machinery, construction machinery, and agricultural transport vehicles. The non-road

mobile sources are characterized by a long residence time and high fuel consumption, and hence, require significant attention. Air pollutant emissions from non-road mobile sources were estimated using the top-down method. The pollutant calculation method is based on the "Technical Guidelines for the Preparation of Air Pollutant Emission Inventory from Non-road Mobile Sources (Trial)" [1]. SO_2 emissions were calculated with a mass balance method by Equation (9) and the emissions of other pollutants by airplanes were calculated according to Equation (10):

$$E_{SO2,m} = 2 \times S_m \times Q_m \qquad (9)$$

$$E_p = \sum LTO \times EF_p \times 10^{-3} \qquad (10)$$

where p is the type of pollutant; m is the fuel type; S is the sulfur content; Q is the consumption; E is the total amount of p-type pollutants, t/a; LTO is the number of aircraft take-offs at Beidaihe Airport, LTO/a; EF is the pollutant emission factor of the aircraft, kg/LTO.

4. Industrial Processes

Twenty sub-categories of industrial process sources were considered, such as the production of cement, tire, vegetable oil, coke, flat glass, synthetic fiber, synthetic ammonia, and iron and steel. It was difficult to acquire facility-level activity data from each factory due to the presence of numerous small factories that were scattered in Qinhuangdao. Emissions from industrial processes were estimated based on the quantum of annual production and emission factors of the unit product. Detailed information was obtained from government statistical data and field investigations including data on production technology, production type, yield, geographic location, and emission control devices.

5. Solvent Use Source

Solvents were the main source of emission of VOCs and were classified as industrial and non-industrial sources. The solvent use source in Qinhuangdao City was calculated according to the combination of point sources and area sources. Among them, printing and dyeing, automobile spraying, furniture coating, and adhesive use were combined with statistical yearbook data, and other industries used statistical yearbook data or department survey data.

6. Fugitive Dust

Fugitive dust sources primarily consisted of construction dust, road dust, yard dust, and soil dust. The content of road dust investigation primarily included the type, length, frequency of sprinkler, cleaning methods, and so on. The construction dust survey content primarily included the project name, latitude and longitude, construction type, floor area, construction area, construction stage, control measures, construction active month, and so on. The yard dust investigation content primarily included the yard name, latitude and longitude, material name, moisture content, cumulative loading and unloading, material loading and unloading times, stacking surface area, stacking height, control measures, etc. Soil dust survey content primarily included the soil type, area, bare degree, plant species, etc. The calculation method of emissions referred to the "Technical Guidelines for the Preparation of Dust Source Particulate Matter Emission Inventory (Trial)" issued by the Ministry of Ecology and Environment [2].

7. Agricultural Sources

Agriculture was the main source of NH_3 emissions, including livestock, soil, nitrogen-fixing crops, nitrogen fertilizer application, and human discharge. Ammonia emissions from agricultural sources were estimated using activity data of each sub-category and specific emission factors. The crop-cultivated area provided activity data for soil

background and nitrogen-fixing plants, while the population value provided activity data for human discharge. Urea and several nitrogen fertilizers including ammonium bicarbonate, ammonium nitrate, ammonium sulfate, and other nitrogen fertilizers were considered for fertilizer application. In this study, NH_3 emissions from five livestock types, including pigs, rabbits, goats, poultry, cows, and other cattle, were estimated. The activity data of each sub-category were gathered from the local government statistics and the Rural Statistical Yearbook.

8. Waste Treatment Sources

Waste treatment sources included solid waste treatment and sewage treatment. Solid waste treatment included solid waste landfill and solid waste incineration. The activity-level data primarily came from 2016 environmental statistics and Qinhuangdao Urban Management Bureau. There are 7 waste treatment enterprises and 16 sewage treatment plants in Qinhuangdao City. The specific formula was shown in Equation (6).

9. Biomass Burning

The emissions from biomass burning were calculated using Equation (11):

$$E_{i,j} = \sum_{i,j} P_i \times C_i \times D_i \times B_i \times F_i \times EF_{i,j} \tag{11}$$

where E is the total emissions of pollutants, including SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, $VOCs$, and NH_3; i, and j represent the crop and pollutant type, respectively; P is the annual crop production; C is the grain-to-straw ratio of different crops; D is the dry matter content of different crops; B is the straw burning rates for different crops; and F is the consumption efficiency for various crops.

10. Storage and Transportation

The storage and transportation sources referred to the emission source of volatile organic compounds caused by the leakage of crude oil, gasoline, diesel oil, and natural gas during storage, transportation, and handling. Storage and transportation sources were processed according to point sources, and $VOCs$ of oil storage, oil transportation, and gas stations are estimated. The specific formula is shown in Equation (6).

11. Others

Other emission sources referred to the collection of air pollutant emission sources not covered by the above source classification, and currently only included cooking sources. According to the guidelines, the estimation of catering sources in Qinhuangdao included the emission of catering enterprises. The calculation formula is as follows:

$$E_i = \sum_i n \times V \times H \times \eta \times EF_i \times 10^{-9} \tag{12}$$

where Ei is the emission of pollutant i, t; n is the number of furnace heads, number; V is the flue gas emission rate, m^3/h; H is the total annual operating time, h; EFi is the emission coefficient of pollutant i, mg/m^3; η is the flue gas removal efficiency of kitchen gas range hood.

12. The Ultra-Low Emissions

The emissions of SO_2, NO_x, and PM_{10} under the ULE and PPC scenarios were calculated using the predicted activity data and the ultra-low emission factors calculated in Equation (5), as shown in Equation (13) [8]:

$$E_{p,s,y} = A_{s,y} \times EF_{p,s} \tag{13}$$

where p represents the emission pollutant species; s represents the emission sources; y represents the year; E represents the emissions of air pollutants from the production of different industries (kg); A is the activity data of the production (t); and EF is the emission factor that reflects the mass of emissions per emission source of production (kg/t).

Section S4. Spatial Allocation of the Emission Sources

The emission inventory was spatially allocated into 1 km × 1 km grid cells using the Geographic Information System (GIS) and relevant allocation surrogates to characterize the sources. For point sources with accurate location information, such as power plants and cement plants, emissions were allocated directly to the grid cells based on their latitude and longitude. Emissions from road mobile sources were treated as line sources, and road network traffic information based on road type was used as a surrogate to spatial emission allocation. The emissions from area sources related to land types, such as agricultural sources and non-road mobile sources, were spatially allocated by land use types. Emissions from residential burning and other sources were distributed mainly according to population density. Road network data, land use data were provided by Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (RESDC). Population data were extracted from the LandScan Global Population Data (1 km) in 2016. Detailed spatial surrogates for each emission source are listed in Table S2 of the Supplementary Material.

Table S1. Emission source categorization in Qinhuangdao City.

Category	Sub-Category	Fuel or Activity Types
Fossil fuel combustion	Power plants	Coal
	Industrial boilers	Coal, carbon, coke, natural gas, fuel oil, blast furnace gas
	Residential combustion	Liquefied petroleum gas (LPG), natural gas, coke, coal
Industrial processes	Iron and steel	Coke, pellet ore, sintering ore, pig iron, crude steel, steel billet
	Casting	machine component, architectural parts, precision casting
	Non-metallic minerals	Cement, tile, lime, clinker, glass, artificial board
	Petrochemical (products)	Crude oil processing and petroleum product manufacturing, Nitrogen fertilizer manufacturing
	Papermaking and paper products	Corrugated paper, paper board
	Other industries	food manufacturing, tea, wine and beverages, rubber and plastics, furniture manufacturing
Solvent use	Pesticide use	The consumption of pesticide
	Surface coating	Construction painting, automotive spraying, and furniture coatings
	Printing and dyeing	Paint consumption of automobile spraying
	Other solvent use	Asphalting paving, wood production, and adhesive use
Mobile sources	On-road mobile	Buses, passenger cars, taxis, trucks, low-speed trucks, three-wheeled vehicles, motorcycles
	Non-road mobile	Agricultural machinery, airplanes, ships, construction machinery, and agricultural transport vehicles

Fugitive dust	Yard dust	Cement, timber processing, mining process, other crafts
	Road dust	Road levels
	Soil dust	Soil type
	Construction dust	Real estate construction, road construction
Biomass burning	Straw burning	Cooking, heating
Agriculture sources	Livestock	Pigs, cows, fowl other cattle
	Soil background	Area of field and fertilizer consumption
	Nitrogen fixing plants	peanut, soybean
	Nitrogen fertilizer application	Nitrogenous compound fertilizer
	Human discharge	Number of people not using sanitary toilets in rural areas
Storage and transportation	Oil and gas storage and transformation	Oil transportation, oil storage depots
	Gas station	Gasoline storage
Waste disposal	Sewage treatment	Sewage treatment volume
	Waste treatment	Waste treatment volume
Other sources	Cooking	Annual operating time, flue gas emission rate

Table S2. Spatial distribution surrogates of each emission source.

Source	Spatial Surrogates	Data Source
Fossil fuel combustion	Longitude and latitude, population density	
On-road mobile	Road network	
Non-road mobile	Longitude and latitude of the airport, railway network, river, cultivated land	
Industrial processes	Longitude and latitude	Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences
Solvent use	Longitude and latitude, cultivated land, population density	
Fugitive dust	Cultivated land, longitude and latitude	
Agriculture	Cultivated land	
Storage and transportation	Longitude and latitude	
Waste disposal	Longitude and latitude	
Others	Longitude and latitude	

Table S3. Emission concentration and theoretical flue gas rate for sintering.

Process	Emission Source	Level	Scale Structure	Concentration (DB 2169-2018) (mg/Nm³)			Theoretical Flue Gas Rate (Nm³/t)
				SO₂	NOₓ	PM₁₀	
Sintering	Fuel crushing	First	≥180 m²	35	50	10	74.5
		Second	90–180 m²	35	50	10	73.67
		Third	<90 m²	35	50	10	73.8
	Raw material preparation	First	≥180 m²	35	50	10	555
		Second	90–180 m²	35	50	10	557.67
		Third	<90 m²	35	50	10	639.6
	Head	First	Desulfurization and Denitration (Dry)	35	50	10	1356.75
		First	Desulfurization and Denitration (Wet)	35	50	10	1510.5

	Second	Desulfurization and without Denitration (Dry)	35	50	10	3053.8
	Second	Desulfurization and without Denitration (Wet)	35	50	10	2743
	Third	Without the desulfurization	35	50	10	2743
Tail	First	≥180 m²	35	50	10	1329
	Second	90–180 m²	35	50	10	1330.63
	Third	<90 m²	35	50	10	1337.9
Size stabilization system	First	≥180 m²	35	50	10	439.2
	Second	90–180 m²	35	50	10	442.75
	Third	<90 m²	35	50	10	442.8

Table S4. Emission concentration and theoretical flue gas rate for pelletizing.

Process	Emission Source	Level	Scale Structure	Concentration (DB 2169-2018) (mg/Nm³)			Theoretical Flue Gas Rate (Nm³/t)
				SO₂	NOₓ	PM₁₀	
Pelletizing	Firing	First	Desulfurization (Dry)	35	50	10	2750.75
		First	Desulfurization (Wet)	35	50	10	3062.5
		Second	Without the desulfurization	35	50	10	3301
	Size stabilization system	First	≥1,200,000 t/a	35	50	10	1948
		Second	600,000–1,200,000 t/a	35	50	10	2020
		Third	<600,000 t/a	35	50	10	1947.88
	Raw material preparation	First	≥12,00,000 t/a	35	50	10	10,883.33
		Second	600,000 t/a–1,200,000 t/a	35	50	10	11,318.6
		Third	<600,000 t/a	35	50	10	11,318.75

Table S5. Emission concentration and theoretical flue gas rate for blast furnace (BF).

Process	Emission Source	Level	Scale Structure	Concentration (DB 2169-2018) (mg/Nm³)			Theoretical Flue Gas Rate (Nm³/t)
				SO₂	NOₓ	PM₁₀	
BF	Ore tank	First	≥1200 m³	50	150	10	3465
		Second	400–1200 m³	50	150	10	3457.2
		Third	<400 m³	50	150	10	3444.6
	Hot stove	First	≥1200 m³	50	150	10	1267
		Second	400–1200 m³	50	150	10	1571
		Third	<400 m³	50	150	10	2001
	Blast furnace casting	First	≥1200 m³	50	150	10	960
		Second	400–1200 m³	50	150	10	985.2
		Third	<400 m³	50	150	10	1515.75

Table S6. Emission concentration and theoretical flue gas rate for basic oxygen furnace (BOF).

Process	Emission Source	Level	Scale Structure	Concentration (DB 2169-2018) (mg/Nm³)			Theoretical Flue Gas Rate (Nm³/t)
				SO₂	NOₓ	PM₁₀	
BOF	Hot metal pretreatment	First	≥100 t	-	-	10	747
		Second	30–100 t	-	-	10	723
		Third	<30 t	-	-	10	746.8
	Primary off gas	First	≥100 t	50	150	50	633.6
		Second	30–100 t	50	150	50	660.2
		Third	<30 t	50	150	50	579.5
	Secondary off gas	First	≥100 t	50	150	10	1451
		Second	30–100 t	50	150	10	1325.67
		Third	<30 t	50	150	10	1812.6

Table S7. Emission concentration and theoretical flue gas rate for flat glass.

Process	Emission Source	Level	Scale Structure	Concentration (DB 2168-2020) (mg/Nm³)			Benchmark Displacement (Nm³/t)
				SO₂	NOₓ	PM₁₀	
Floating	Melting	First	>900 t	50	200	10	3200
		Second	600–900 t	50	200	10	4080
		Third	500–600 t	50	200	10	4220
			≤500 t	50	200	10	4410
Rolling	Melting	-	-	50	200	10	4394

Table S8. Emission concentration and benchmark displacement for cement.

Process	Emission Source	Concentration (DB 2168-2020) (mg/Nm³)			Benchmark Displacement (Nm³/t)
		SO₂	NOₓ	PM₁₀	
Cement manufacturing	Cement kiln and clinker warehouse	100	30	100	5210.78

Table S9. The product output of the three industries in the past decade (unit: 10⁴t/a; for flat glass: 10⁴ boxes/a, and 20 boxes of flat glass= 1 t flat glass).

Output	2021	2020	2019	2018	2017	2016	2015	2014	2013	2012
Pig-iron	20,202.98	22,903.76	21,774.37	21,396	17,997.27	18,398.37	17,382.3	16,941.95	17,027.6	16,358.54
Crude steel	22,496.45	24,976.95	24,157.7	23,723.37	19,121.47	19,259.97	18,832	18,530.3	18,849.6	18,048.4
Steel	29,559.38	31,320.12	28,409.63	26,916.86	24,551.08	26,150.42	25,244.3	23,995.2	22,861.6	21,026.1
Cement	11,354.63	11,859.97	10,527.39	9554.3	9125.5	9898.58	9126.17	10,721.46	12,747.38	13,131.84
Flat glass	13,486.63	13,728.37	16,357.29	12,156.03	13,780.23	13,693.57	14,615.36	15,844.52	11,836.4	14,898.03

Table S10. The fluctuating range of the three industries in the past decade.

Fluctuating Range	2021	2020	2019	2018	2017	2016	2015	2014	2013	2012
Pig-iron	9.8%	24.5%	18.3%	16.3%	−2.2%	0.0%	−5.5%	−7.9%	−7.5%	−11.1%
Crude steel	16.8%	29.7%	25.4%	23.2%	−0.7%	0.0%	−2.2%	−3.8%	−2.1%	−6.3%
Steel	13.0%	19.8%	8.6%	2.9%	−6.1%	0.0%	−3.5%	−8.2%	−12.6%	−19.6%
Cement	14.7%	19.8%	6.4%	−3.5%	−7.8%	0.0%	−7.8%	8.3%	28.8%	32.7%
Flat glass	−1.5%	0.3%	19.5%	−11.2%	0.6%	0.0%	6.7%	15.7%	−13.6%	8.8%

Table S11. The uncertainties of activity levels.

Parameter	Distribution [1]	Parameter	Distribution
Industrial fossil fuel use	Normal (CV: 10%)	Volatile raw materials use	Normal (CV: 10%)
Industrial production	Normal (CV: 10%)	Material transportation	Normal (CV: 10%)
Non-road fuel use	Normal (CV: 20%)	petrol sales volume	Normal (CV: 10%)
Vehicle mileage travelled	Normal (CV: 5%)	petrol transport volume	Normal (CV: 10%)
Livestock number	Normal (CV: 10%)	petrol storage	Normal (CV: 10%)
Pesticide application	Normal (CV: 10%)	Sewage treatment volume	Normal (CV: 10%)
Agriculture production	Normal (CV: 30%)	Hazardous waste disposal volume	Normal (CV: 10%)
Industrial coating consumption	Normal (CV: 10%)	Domestic waste disposal volume	Normal (CV: 10%)

[1] CV means Coefficient of Variation, and its value in Tables S11–S20 referred to the literature [13–15].

Table S12. The uncertainties of emission factors for SO_2. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 10%)
	Residential fuel combustion	Normal (CV: 10%)
	Industrial boilers	Normal (CV: 10%)
Industrial process		Normal (CV: 20%)
On-road mobile		Normal (CV: 10%)
Non-road mobile source		Normal (CV: 10%)

Table S13. The uncertainties of emission factors for NOx. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 5%)
	Mining and Manufacturing industry	Normal (CV: 10%)
	Residential combustion	Normal (CV: 10%)
	Civil boilers	Normal (CV: 10%)
	Petrochemical (Industrial boilers)	Normal (CV: 10%)
On-road mobile		Normal (CV: 20%)
Non-road mobile		Normal (CV: 20%)
Process source		Normal (CV: 20%)

Table S14. The uncertainties of emission factors for CO. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 5%)
	Mining and Manufacturing industry	Normal (CV: 10%)
	Residential combustion	Normal (CV: 10%)
	Petrochemical (Industrial boilers)	Normal (CV: 10%)
	Civil boilers	Normal (CV: 10%)
On-road mobile		Normal (CV: 20%)
Non-road mobile		Normal (CV: 10%)
Process source	Kiln exhaust gas	Normal (CV: 20%)
	Cement	Normal (CV: 20%)

Table S15. The uncertainties of emission factors for PM$_{2.5}$. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 20%)
	Mining and Manufacturing industry	Normal (CV: 10%)
	Petrochemical (Industrial boilers)	Normal (CV: 10%)
	Civil boilers	Normal (CV: 10%)
	Residential combustion	Normal (CV: 10%)
On-road mobile		Normal (CV: 20%)
Non-road mobile	Agricultural machinery	Normal (CV: 10%)
Process source	Cement	Normal (CV: 20%)
	Kiln exhaust gas	Normal (CV: 20%)
Dust source		Normal (CV: 20%)

Table S16. The uncertainties of emission factors for PM$_{10}$. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 10%)
	Mining and Manufacturing industry	Normal (CV: 20%)
	Petrochemical (Industrial boilers)	Normal (CV: 20%)
	Civil boilers	Normal (CV: 20%)
	Residential combustion	Normal (CV: 20%)
On-road mobile		Normal (CV: 20%)
Non-road mobile	Agricultural machinery	Normal (CV: 20%)
Process source	Cement	Normal (CV: 20%)
	Kiln exhaust gas	Normal (CV: 20%)
Fugitive dust		Normal (CV: 20%)

Table S17. The uncertainties of emission factors for VOCs. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 10%)
	Residential combustion	Normal (CV: 20%)
	Mining and Manufacturing industry	Normal (CV: 20%)
	Petrochemical (Industrial boilers)	Normal (CV: 20%)
	Civil boilers	Normal (CV: 20%)
On-road mobile		Normal (CV: 20%)
Non-road mobile	Agricultural machinery	Normal (CV: 20%)
Process source	Cement	Normal (CV: 10%)
	Kiln exhaust gas	Normal (CV: 20%)
	Pesticide application	Normal (CV: 20%)
Solvent use	Industrial painting	Normal (CV: 20%)
	printing and dyeing	Normal (CV: 20%)
Source of storage and transportation	Oil and gas emissions (oil depots)	Normal (CV: 20%)
	Gas station	
	Oil and gas emissions (oil transportation companies)	Normal (CV: 20%)
Other	Hazardous waste disposal	Normal (CV: 20%)
	Domestic waste disposal	Normal (CV: 20%)

Table S18. The uncertainties of emission factors for NH3. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 20%)
	Mining and Manufacturing industry	Normal (CV: 10%)
	Civil boilers	Normal (CV: 10%)
Industrial possesses	Cement	Normal (CV: 10%)
	Kiln exhaust gas	Normal (CV: 10%)
On-road mobile		Normal (CV: 20%)
Waste treatment	Sewage treatment	Normal (CV: 10%)
	Hazardous waste disposal	Normal (CV: 10%)
	Domestic waste disposal	Normal (CV: 10%)
	Livestock	Normal (CV: 10%)
	Soil background	Normal (CV: 10%)
Agriculture	Nitrogen fixing crops	Normal (CV: 10%)
	Nitrogen fertilizer application	Normal (CV: 5%)
	Human discharge	Normal (CV: 10%)

Table S19. The uncertainties of emission factors for BC. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 20%)
	Mining and Manufacturing industry	Normal (CV: 10%)
	Petrochemical (Industrial boilers)	Normal (CV: 10%)
	Civil boilers	Normal (CV: 10%)
	Residential combustion	Normal (CV: 10%)
On-road mobile		Normal (CV: 20%)
Non-road mobile	Agricultural machinery	Normal (CV: 10%)
Process source	Cement	Normal (CV: 20%)
	Kiln exhaust gas	Normal (CV: 20%)

Table S20. The uncertainties of emission factors for OC. 95% CIs are provided in the parentheses.

Source	Sector	Distribution
Fossil fuel combustion	Power plant	Normal (CV: 20%)
	Mining and Manufacturing industry	Normal (CV: 10%)
	Petrochemical (Industrial boilers)	Normal (CV: 10%)
	Civil boilers	Normal (CV: 10%)
	Residential combustion	Normal (CV: 10%)
On-road mobile		Normal (CV: 20%)
Non-road mobile	Agricultural machinery	Normal (CV: 10%)
Process source	Cement	Normal (CV: 20%)
	Kiln exhaust gas	Normal (CV: 20%)

Table S21. CALPUFF model parameter settings.

Pattern	Parameter	Setting
Grid	Lower-left coordinate	(630,073 m, 4,359,925 m)
	Number of grids in the X direction	24
	Number of grids in the Y direction	30
	Number of grids in the Y direction	10
	Grid spacing	5 km
	Base time zone	UTC + 08:00 Beijing
	Projection	UTM 50N
	DATUM	WGS-84

CALMET	Run Option	Use surface and overwater stations with 3D.DAT for upper air
	Temperature and relative humidity	Use 3D.DAT temperature at surface and upper levels
CALPUFF	Chemical transformation	Computed internally (MESOPUFF II Scheme)
	Species (Participate in chemical transformation)	SO_2, NO_x, SO_4, NO_3, HNO_3
	Receptor	The monitoring station in Qinhuangdao
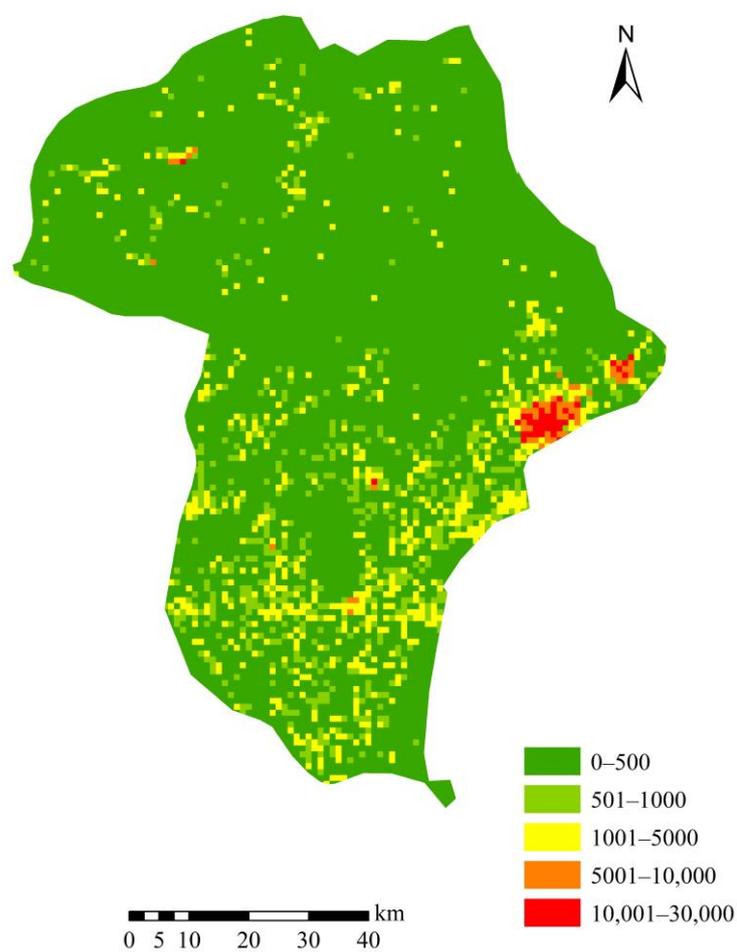
Figure S1. Distribution of population density in Qinhuangdao.
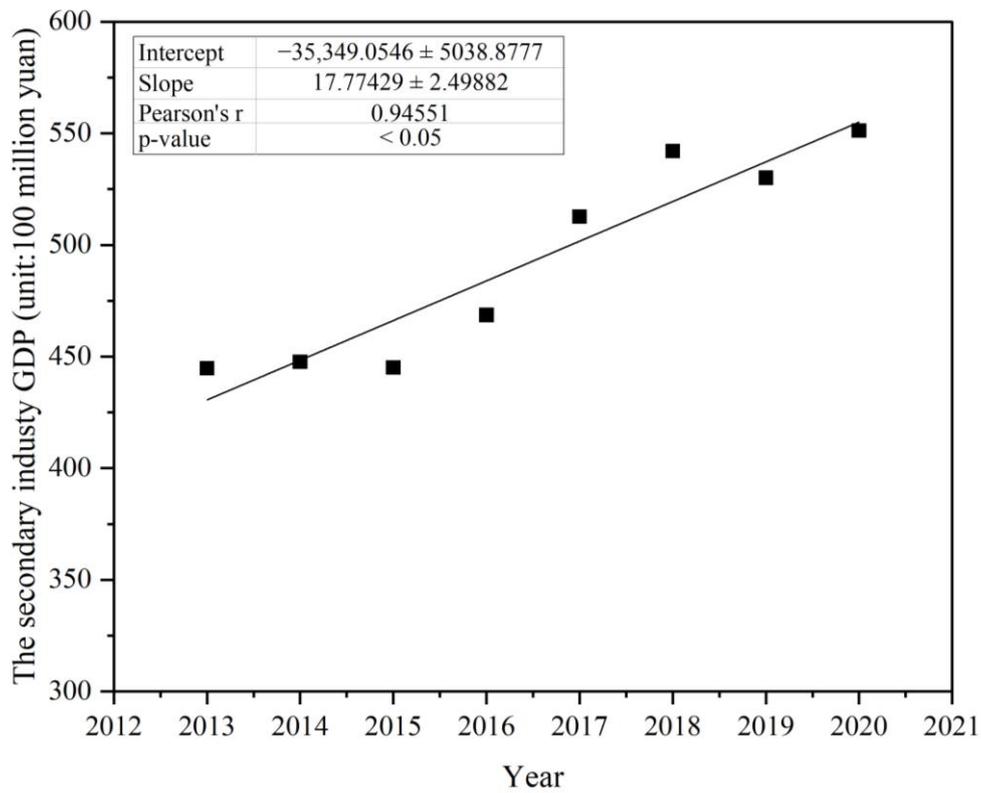
Figure S2. The prediction of the secondary industry GDP.
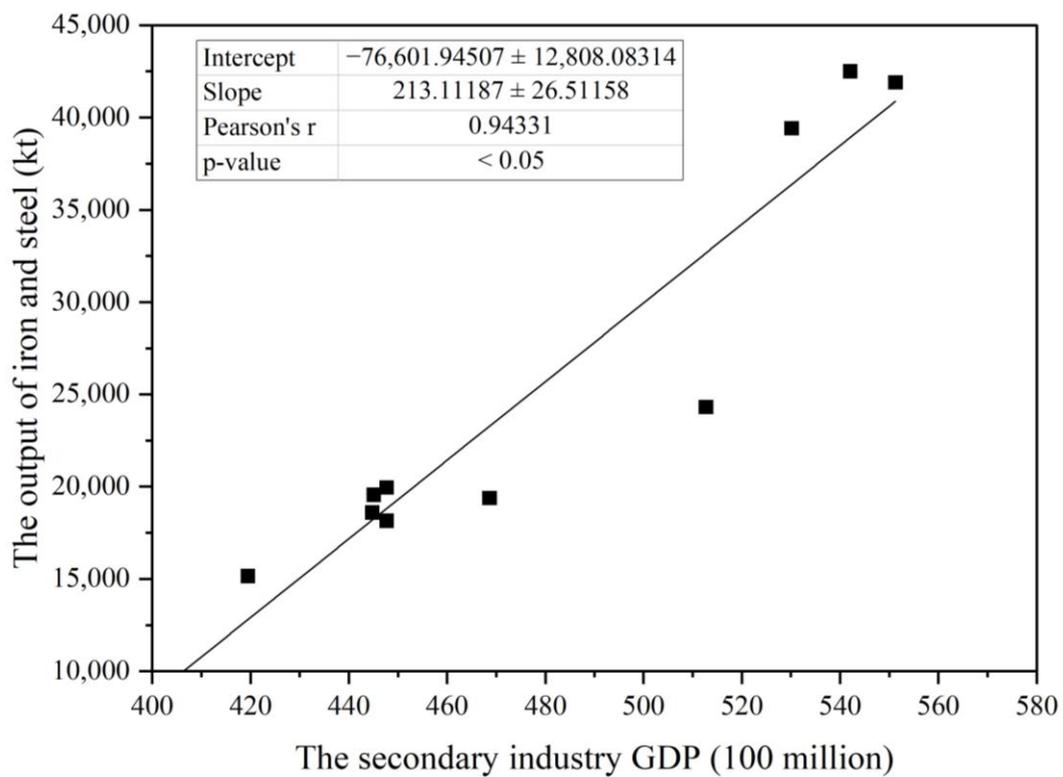
Figure S3. The relationship between the output of iron and steel and the secondary industry GDP.
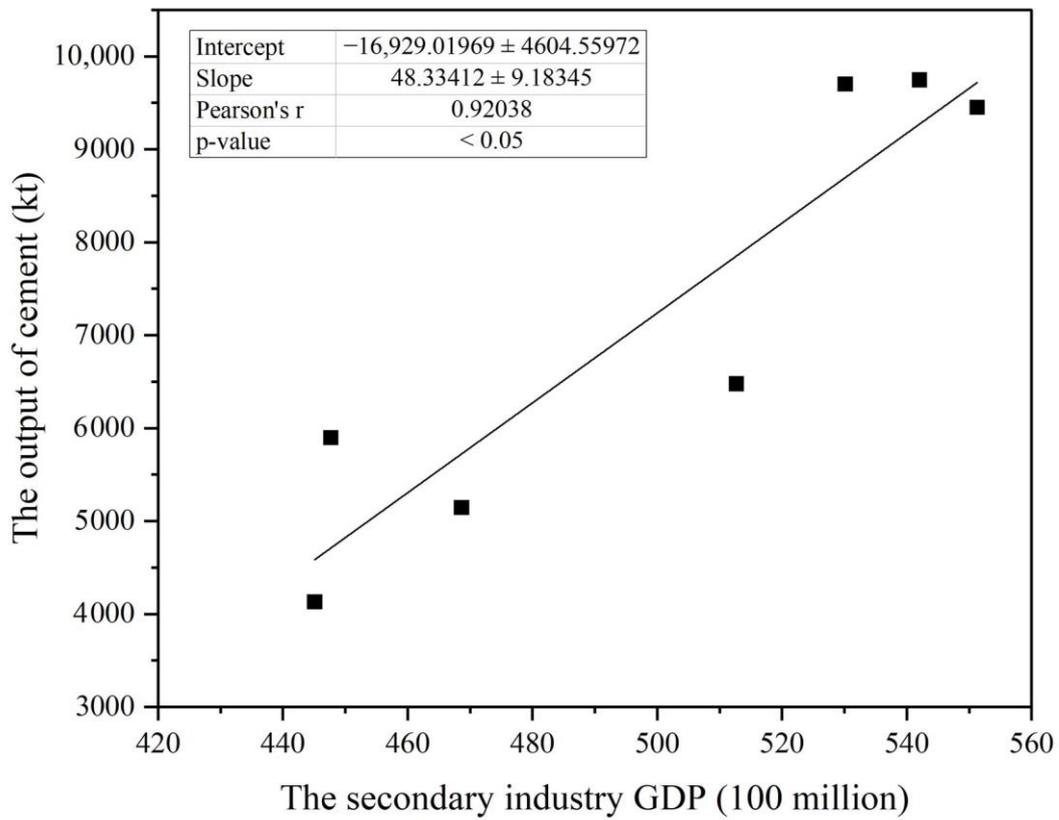
Figure S4. The relationship between the output of cement and the secondary industry GDP.
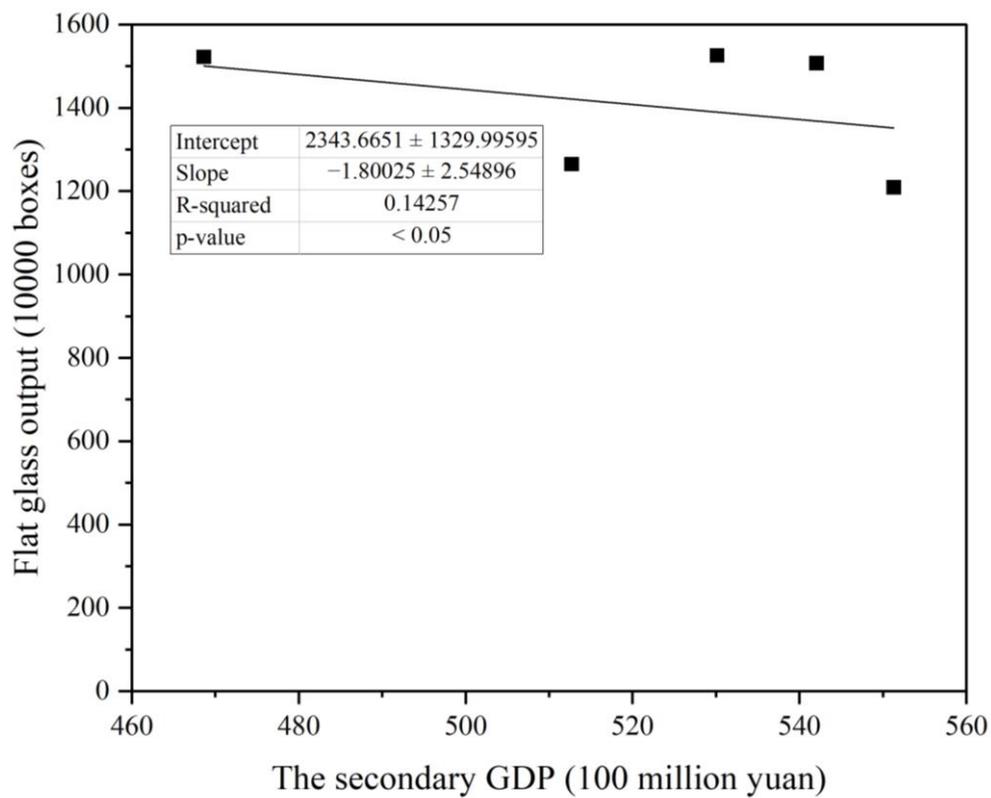
Figure S5. The relationship between the output of flat glass and the secondary industry GDP.
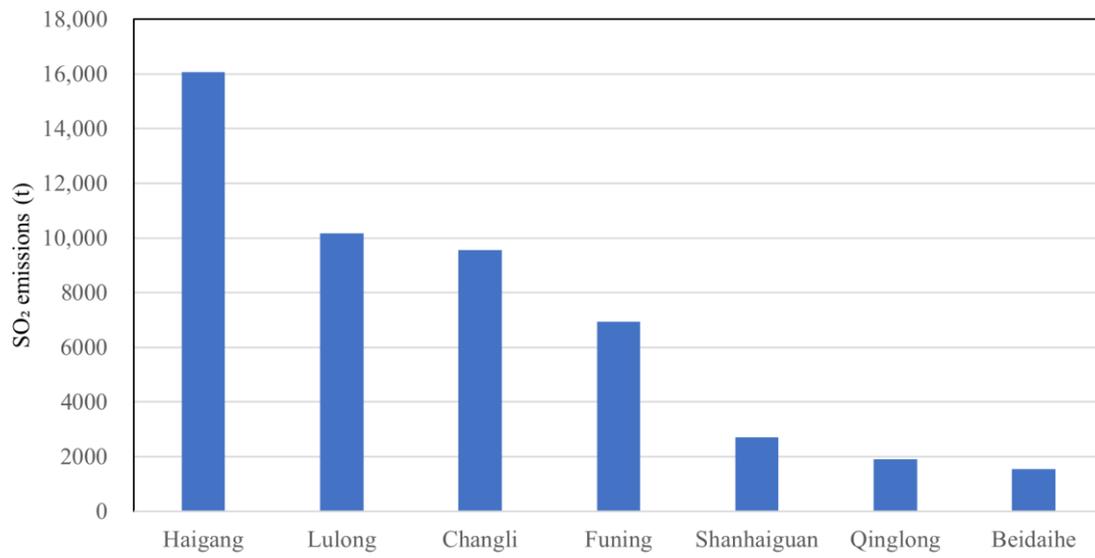
(**a**)
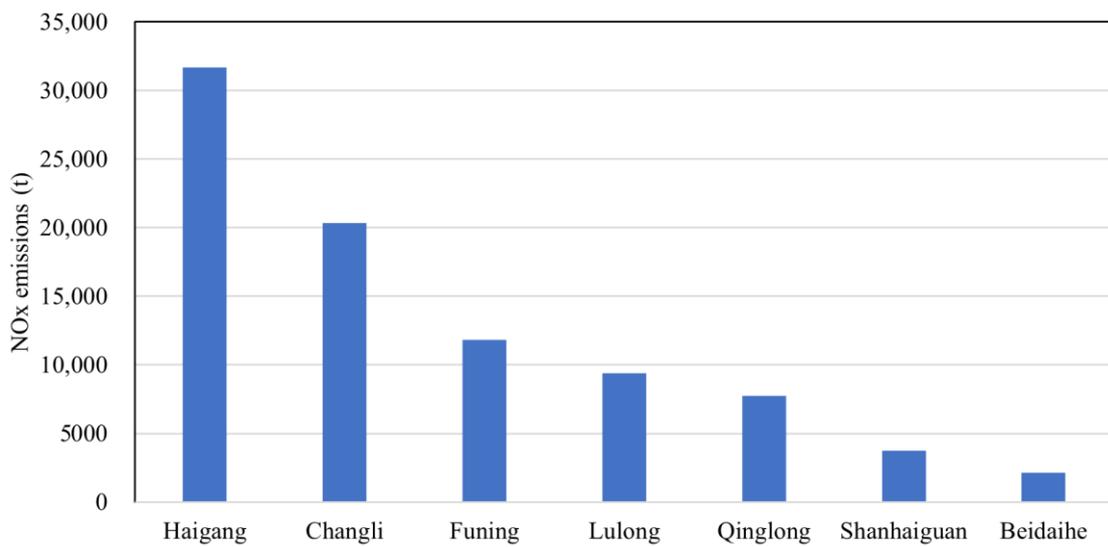
(**b**)
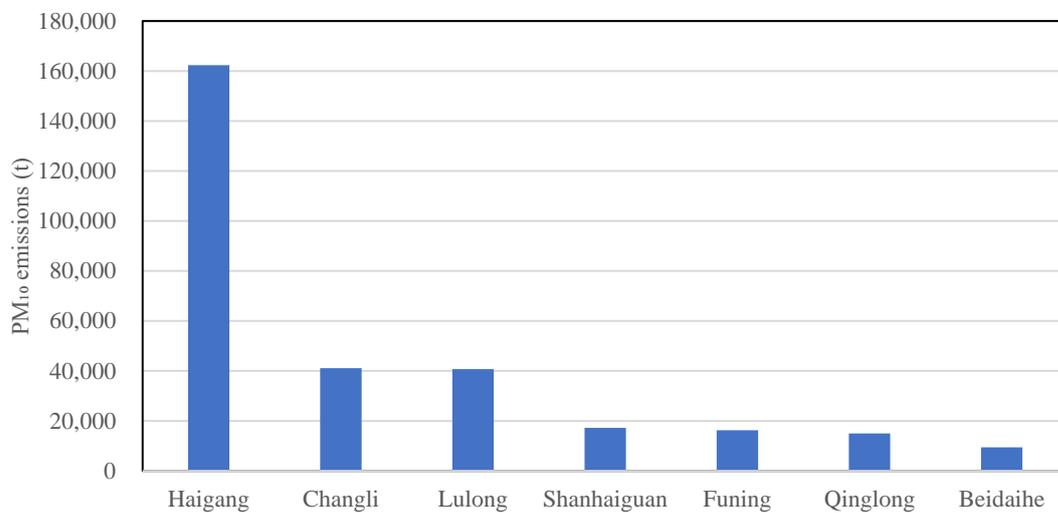
(**c**)
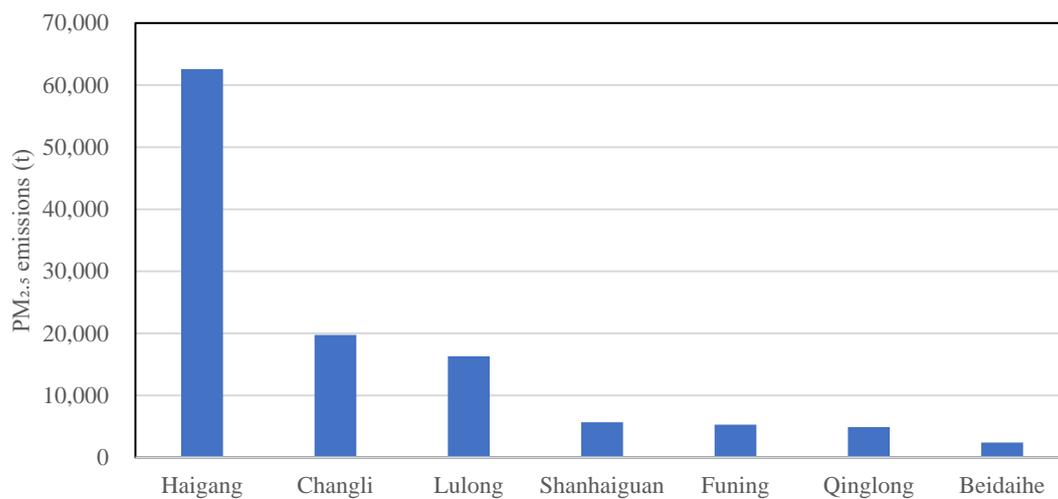
(**d**)
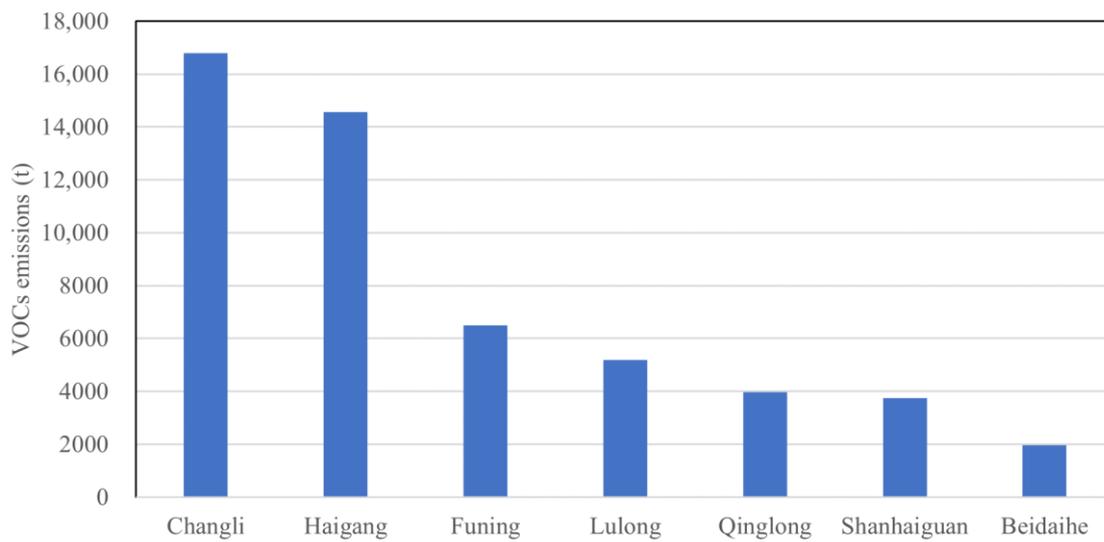
(**e**)
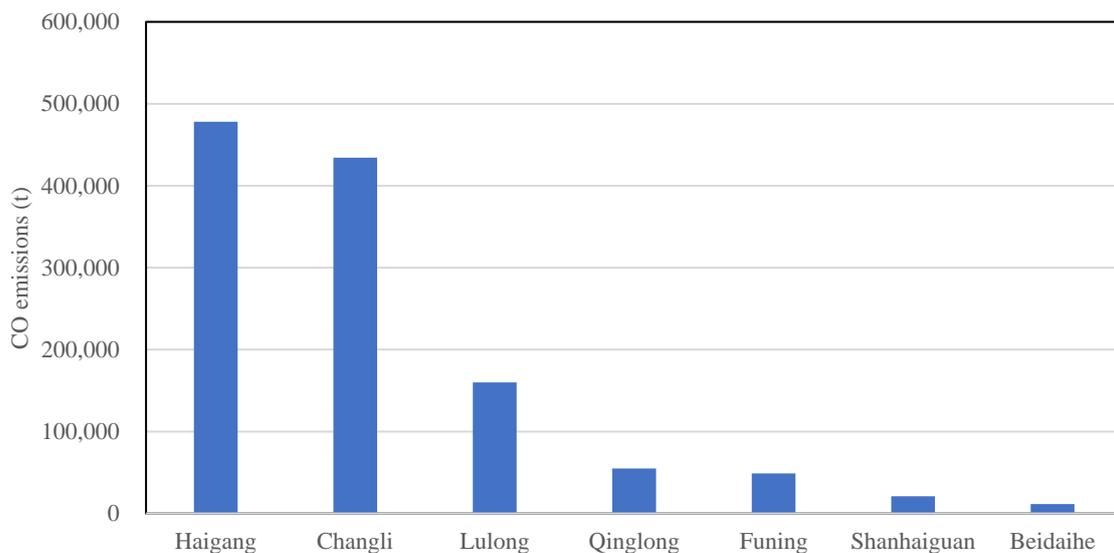
(**f**)
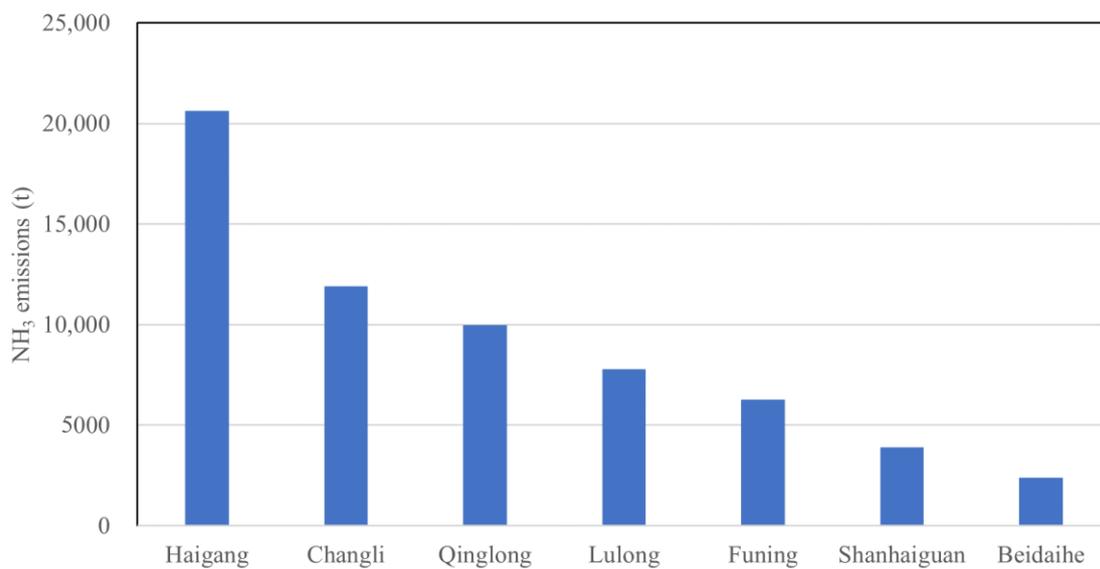
(**g**)
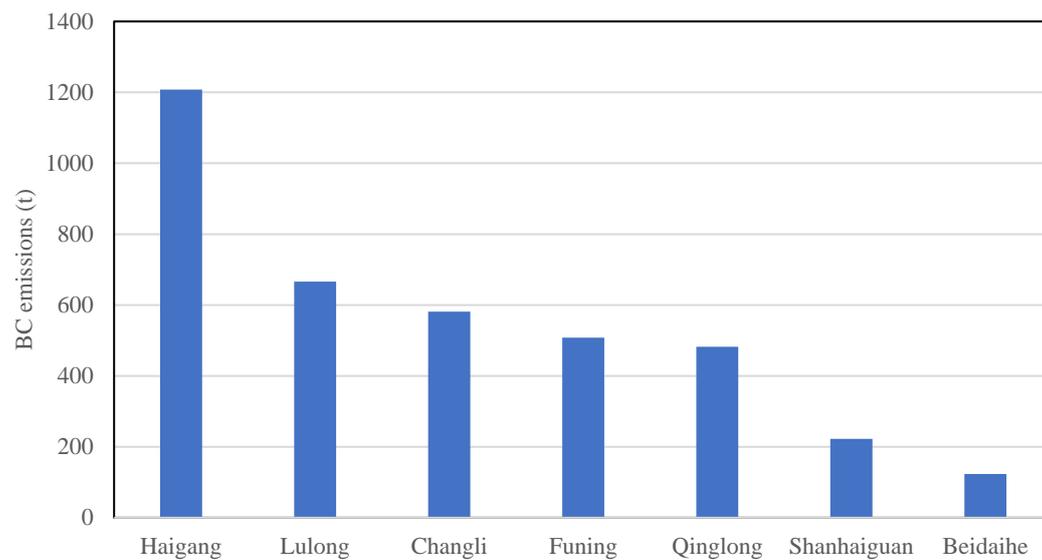
(**h**)
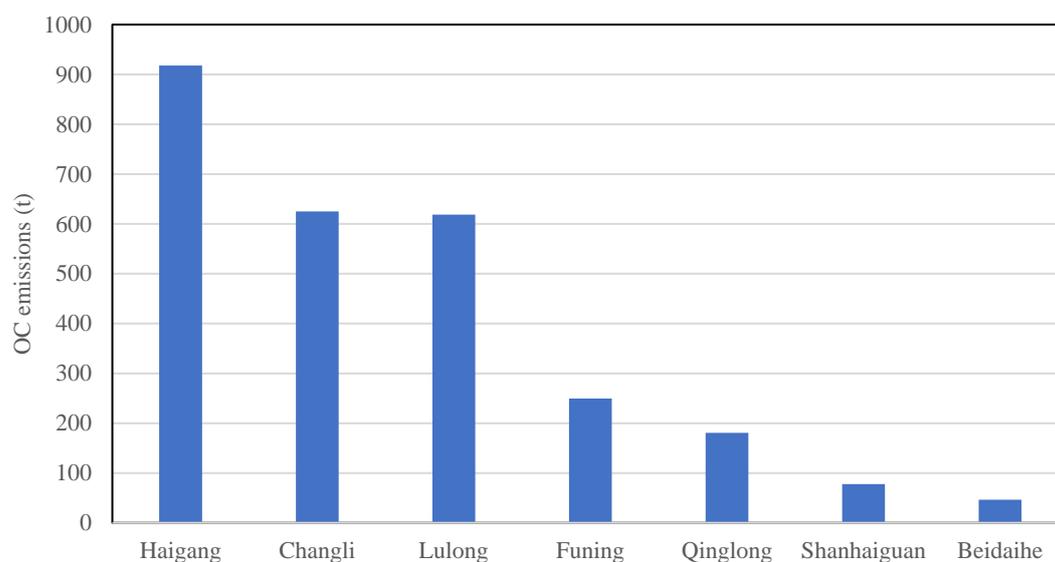
(**i**)

**Figure S6.** County-level contributions to the total emissions for nine pollutants (**a–i**) in Qinhuang-dao.
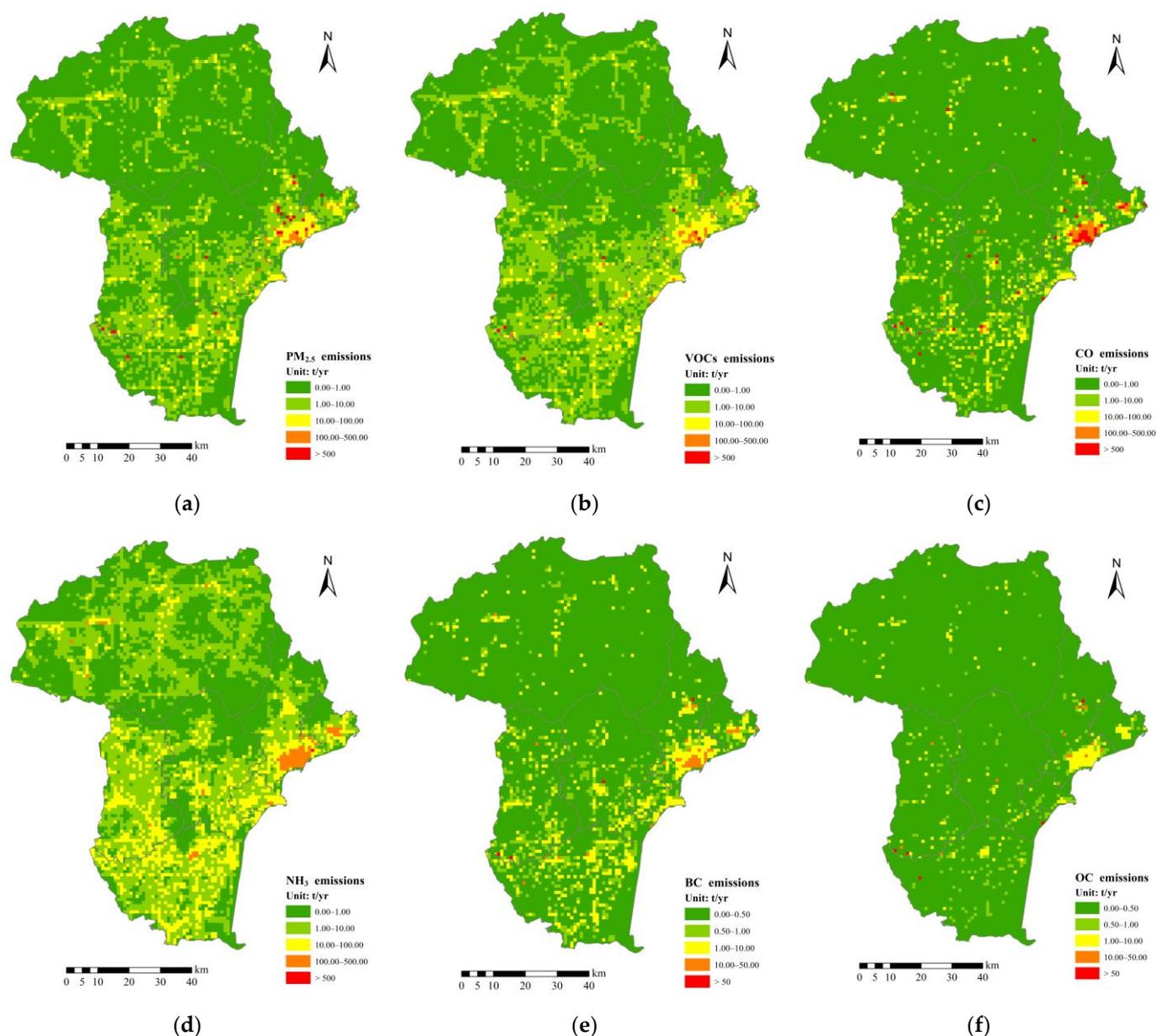
Figure S7. Spatial distribution characteristics of air pollutant emissions in Qinhuangdao for 2016. (**a**) PM2.5; (**b**) VOCs; (**c**) CO; (**d**) NH3; (**e**) BC; (**f**) OC.

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