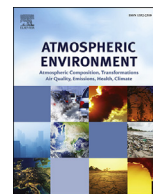




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A high-resolution air pollutants emission inventory in 2013 for the Beijing-Tianjin-Hebei region, China



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H I G H L I G H T S

- An updated high-resolution emission inventory of 2013 for the BTH region is established.
- Facility-based emissions are calculated for power and key industries sources.
- The spatial distribution of emission is investigated.
- The emission inventory is evaluated by uncertainty analysis.

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A B S T R A C T

We developed a high-resolution Beijing-Tianjin-Hebei (BTH) regional air pollutants emission inventory for the year 2013. The inventory was established using a bottom-up approach based on facility-level activity data obtained from multiple data sources. The estimates from the BTH 2013 emission inventory show that the total emissions of SO₂, NO_x, PM_{2.5}, PM₁₀, CO, NMVOC, NH₃, BC, and OC were 2,305, 2,686, 1,090, 1,494, 20,567, 2,207, 623, 160, and 254 Gg, respectively. The industry sector is the largest emissions source for SO₂, NO_x, PM_{2.5}, PM₁₀, CO, and NMVOC in the BTH region, contributing 72.6%, 43.7%, 59.6%, 64.7%, 60.3%, and 70.4% of the total emissions, respectively. Power plants contributed 11.8% and 23.3% of the total SO₂ and NO_x emissions, respectively. The transportation sector contributed 28.9% of the total NO_x emissions. Emissions from the residential sector accounted for 31.3%, 21.5%, 46.6% and 71.7% of the total PM_{2.5}, NMVOC, BC and OC emissions, respectively. In addition, more than 90% of the total NH₃ emissions originate from the agriculture sector, with 44.2% from fertilizer use and 47.7% from livestock. The spatial distribution results illustrate that air pollutant emissions are mainly distributed over the eastern and southern BTH regions. Beijing, Tianjin, Shijiazhuang, Tangshan and Handan are the major contributors of air pollutants. The major NMVOC species in the BTH region are ethylene, acetylene, ethane and toluene. Ethylene is the biggest contributor in Tianjin and Hebei. The largest contributor in Beijing is toluene. There is relatively low uncertainty in SO₂ and NO_x emission estimates, medium uncertainty in PM_{2.5}, PM₁₀ and CO emission estimates, and high uncertainties in VOC, NH₃, BC and OC emission estimates. The proposed policy recommendations, based on the BTH 2013 emission inventory, would be helpful to develop strategies for air pollution control.

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1. Introduction

Rapid economic growth in China has led to severe air pollution, which causes air pollution-related problems, such as smog and

haze. The Beijing-Tianjin-Hebei (BTH) region is one of China's most developed regions as well as one of the regions experiencing the most severe air pollution problems. Air quality monitoring data released by the Ministry of Environmental Protection show that the average PM_{2.5} concentration for the BTH region in 2013 was 106 µg/m³, which is 47% higher than the national average, and the number of heavy air pollution days was over 70 (China Environmental State

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Communique, 2013). In September 2013, the China State Council released the *Action Plan for Air Pollution Prevention and Control (APAPPC)*. The goal of the APAPPC is to improve the air quality of the entire country, particularly in key regions, including the BTH region, Yangtze River Delta (YRD) region and Pearl River Delta (PRD) region. For the BTH region, there are specific targets set in the plan, such as reducing PM_{2.5} level by 25% based on 2012 level and controlling the annual average PM_{2.5} concentration in Beijing under 60 µg/m³ (APAPPC). Policy makers and researchers are facing formidable challenges in forming effective strategies for air quality management to achieve these goals.

Air pollutants emission inventories are essential for understanding air pollution emission sources and facilitate the development of effective strategies for air pollution prevention. Researchers and policy makers have focused on emission inventories in the past decades. Several emission inventories have been developed for China. Inventories have been developed on the national scale (Streets et al., 2003; Zhang, 2005; Ohara et al., 2007; Zhang et al., 2009), for an individual source (Zhao et al., 2008; Lei et al., 2011a; Lu et al., 2011b), and for a specific pollutant (Zhang et al., 2007; Wei et al., 2008; Fu et al., 2008; Su et al., 2011b; Zhao et al., 2013). In recent years, major developed regions have been affected by air pollution. Regional joint prevention and control of air pollution has become an important air pollution control strategy for China. Many regional emission inventories have been developed. Fu et al. (2013) established an emission inventory in 2010 for 25 cities in the YRD region, and Huang et al. (2011) estimated the emissions of 16 major cities in 2007 for the YRD region. Zheng et al. (2009) developed a high-resolution emission inventory in 2006 for the PRD region. However, for the BTH region, the majority of previous studies have only focused on an individual pollutant or developed on the province scale. Zhou et al. (2015) established an ammonia emission inventory for the BTH region in 2010. Su et al. (2011a) established an emission inventory for non-methane volatile organic compounds in 2008. Wang et al. (2003) established an emission inventory for biogenic volatile organic compounds for Beijing in 2003. Zhao et al. (2012b) established an emission inventory on province level for 8 provinces in Huabei region, which contains BTH region. No previous studies have provided a comprehensive and detailed emission inventory for the BTH region. Furthermore, because factory- and facility-level activity data are difficult to access in China, the majority of the previous studies have used the emission estimation methods depended on province-level statistical data. As a result, it is difficult to allocate emissions to specific sources, making it difficult to meet the requirements of air quality simulation or to develop pollution control measures. Along with the implementation of the APAPPC, the BTH region would develop several specific air pollution emission control measures and have stricter air pollution emission standards. Therefore, an updated and high-resolution air pollutants emission inventory with knowledge of NMVOC speciation of the BTH region is critical as the basis for policy development and evaluation.

In this study, a comprehensive emission inventory for the BTH region in 2013 was developed based on facility-level activity data obtained from multiple sources. The pollutants analyzed were SO₂, NO_x, PM_{2.5}, PM₁₀, CO, NMVOC, NH₃, BC, and OC. The emissions were distributed into 3 km × 3 km resolved grids to describe the spatial characteristics of air pollutant emissions in the BTH region. Section 2 describes the methodology and data sources. The results for sectoral emissions, spatial distributions, NMVOC speciation, and uncertainties assessment are presented in Section 3. The implications and future improvements are also discussed.

2. Data and methodology

2.1. General methodology

The BTH region is located in northern China and includes Beijing, Tianjin and 11 cities in Hebei, for a total of 13 cities (see Fig. 1). The BTH region is one of the most economically developed regions in China. In 2013, it represented 2.3% of the national territory and 8.0% of the population, generated 10.9% of the total national GDP, and owned 13.0% of the nation's vehicle fleet (China National Statistics Yearbook, 2014). Moreover, it is also the region with the worst air pollution in China. In 2013, the annual average PM_{2.5} concentration in the BTH region was 106 µg/m³. In this study, the BTH region was set between latitudes 36.05N–42.62N and longitudes 113.46E–119.85E.

A bottom-up approach was adopted in this study to develop a high-resolution emission inventory for the BTH region in 2013. This method has been developed and validated by our previous study (Zheng et al., 2017). We used the same method to compile a complete emission inventory over the BTH region. The emission sources in our inventory were divided into five major sectors: power, industry, residential, transportation and agriculture. These five sectors were further subdivided by fuel type, product type and technology. These sectors were treated as point, nonpoint and mobile sources with corresponding methods for emission estimation.

Power and industry sectors were treated as point sources. The number of point sources considered in this study was 12,461, which include most of the boilers and major emission facilities used in power plants, heating plants, iron and steel factories, cement factories, coking factories and other industries. The point source emissions were estimated based on detailed facility-level investigation data using Equation (1):

$$E_{i,s} = \prod_n A_i F_{i,s} (1 - \eta_{i,n,s}), \quad (1)$$

where A is the activity data, F is the uncontrolled emission factor, η is the removal efficiency, and the subscripts i, s, and n represent the facility, air pollutant type (i.e., SO₂, NO_x, PM_{2.5}, PM₁₀, CO, NMVOC, NH₃, BC and OC) and type of air pollution control device, respectively.

The residential, non-road transportation and agriculture sectors, which are diffuse sources without identifiable stacks, were treated as nonpoint sources and estimated at the province level using Equation (2):

$$E_{j,k,m,s} = A_{j,k} X_{j,k,m} F_{j,k,m,s} \sum_n (Y_{j,k,m,n} \times (1 - \eta_{n,s})), \quad (2)$$

Where X is the fraction of activity rates contributed by a specific technology, Y is the penetration of a specific pollution control technology, and the subscripts j, k and m represent sector, fuel or product and technology, respectively. The remaining parameters are the same as in Equation (1).

On-road transportation sector was considered as mobile sources. The method established by Zheng et al. (2014) was adopted. In this study, and the on-road transportation emissions for each city were estimated using the 2013 data (e.g., vehicle numbers, fuel use and emission factors). The emissions from motorcycles were also included in this work, where provincial emissions were estimated and then allocated to city based on vehicle ownership.

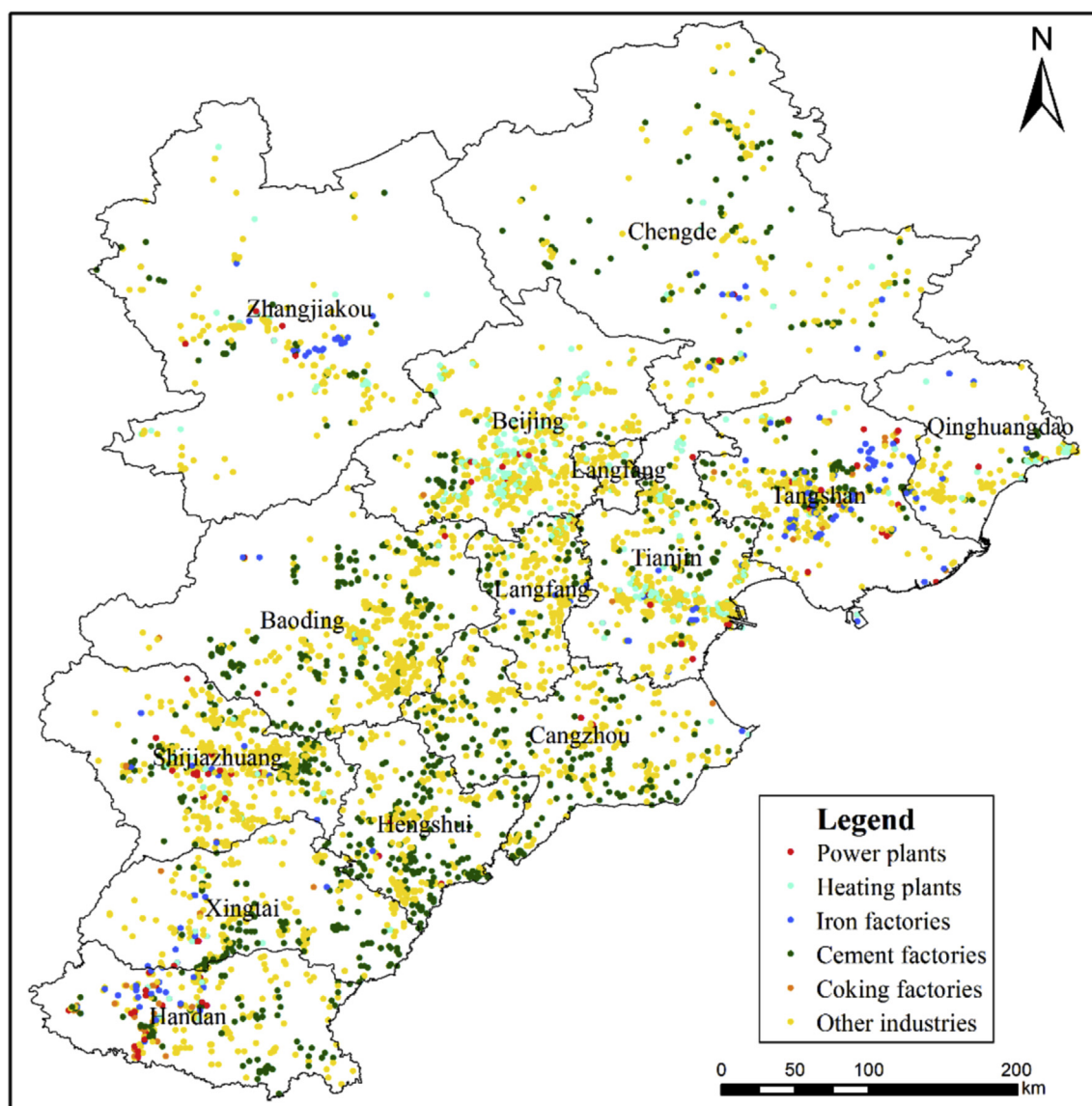


Fig. 1. Study domain and location of point sources in the BTH region.

2.2. Data sources

There are six major data sources that we draw upon in this paper: (i) Chinese Environmental Statistics, (ii) China's Pollution Source Census, (iii) Ministry of Environmental Protection Statistics, (iv) China Energy Statistical Yearbook, (v) China Industry Statistics and (vi) China Traffic Statistics. These six data sources, which were inconsistent in terms of their spatial and time scales, were assimilated to derive the parameters needed to establish the BTH inventory. The data sources used for the development of the BTH inventory are briefly summarized in Table 1.

Chinese Environmental Statistics is developed by the Ministry of Environmental Protection. It is designed to cover all industries above a designated size. It provides basic information at the factory level, including production type, product output, fuel type, and fuel consumption. However, it lacks information on production technology and control devices. In this study, we used the Chinese Environmental Statistics data for the year 2013. The China Pollution Source Census is a joint effort of multiple national ministries in China, conducted to obtain detailed information at the facility level.

It can supply detailed information at the facility level, such as capacity, process, control devices and removal efficiency. The latest records data are for the year 2010. China's Ministry of Environmental Protection Statistics contains information on the activity rate, production technology, type of control devices, and removal efficiency at the facility level. However, it only compiles information on key pollution emission industries, such as power plant, iron and steel industry, and cement industry; other industries are not covered.

These three datasets cover the majority of the point sources in the BTH region. We used the basic active factory-level data from the Chinese Environmental Statistics and then allocated them to facilities. The detailed facility information for key pollution industries was collected from China's Ministry of Environmental Protection Statistics, whereas information for other industries was obtained from the China Pollution Source Census. Certain facility-level data are only available for the year 2010. For these sources, the corresponding factory statistics in 2013 were used and split using distribution patterns from the year 2010. A factory with no facility data was treated as one facility in this study. However, these three

Table 1Data sources used to derive the parameters needed for the BTH inventory.^a

Database	Statistics level	Data Year	Data type	Description
Chinese Environmental Statistics	Factory	2013	Activity rates (Fuel use and industrial product)	Cover almost all industry plants. Updated every year.
China's Pollution Source Census	Facility	2010	Activity rates and air pollution control devices	Cover almost all industry plants with detailed statistics for each boiler and kiln. Updated every few years.
MEP ^b Statistics	Facility	2013	Activity rates, emission factors and pollution control devices	Cover all power plants and makers of coke, iron, steel, cement and flat glass. Updated every year.
China Energy Statistical Yearbook	Province	2013	Energy balance table	Cover major energy types. Updated annually but lags 2–3 years.
China Industry Statistics	Province	2013	Industrial products	Cover all industrial products. Updated every year or month.
China Traffic Statistics	City	2013	Vehicle numbers	Cover all vehicle types. Updated every year.

^a These datasets are unpublished and collected from local agencies, with the exception of those estimated at the province level.^b China's Ministry of Environmental Protection.

datasets omit certain small industries, and a proportion of rural industries are unincorporated. Therefore, macroeconomic data on fuel consumption, industrial production output and the solvent usage from the China Energy Statistical Yearbook and China Industry Statistics are needed to constrain and adjust the activity level. We scaled the small industries activity rates to match the residual after subtracting the activity rates of the major large industries from the total activity.

Owing to the lack of census data for non-point sources, parameters that are needed in the emission calculation were derived from statistical data at the province level, such as residential fuel consumption, non-road fuel use, and fertilizer use. These data were then allocated to cities according to proxies such as population and industrial GDP.

2.3. Emission factors

Emission factors were developed using technology based methodology, where emission rates were estimated for each source according to its fuel type, combustion/industrial process and control technology (Lei et al., 2011a; Liu et al., 2015; Zhang et al., 2007, 2009). For point sources, we derived facility level emission factors of SO₂, NO_x and PM based on detailed statistics. For example, we fused the industrial datasets to obtain the ash and sulfur content of coal, combustion technology and removal efficiency of air pollution control devices related to each electric generating unit. These data were translated to unit-level emission factors of SO₂, NO_x and PM for each coal fired power plant. The emission factors of other pollutants from point sources were treated at provincial level, similar as nonpoint sources, because the factors influence emissions are only available for provinces. We traced the changes of fuel contents and penetration of technologies for each province and therefore achieved net emission factors for a fuel/product in a specific sector. Development of these emission factors are documented in our previous work (Huo et al., 2012a; Lei et al., 2011a; Shen et al., 2015; Liu et al., 2015; Zhang et al., 2007, 2009; Lei et al., 2011b). The base emission factors were generally taken from measurement samples reported in literatures, and supplemented with emission factor databases (e.g. USEPA, 2002 AP-42).

For mobile sources, we used the IVE model to calculate city-level emission factors. The base emission factors in the model were assimilated against on-road measurements taken in Chinese cities (Huo et al., 2012b, 2012c; Yao et al., 2011). The IVE model adopted correction factors of driving patterns and meteorological factors on base emission factors to reflect the real emission rates. Driving patterns data were selected from surveys in literatures (Liu et al., 2007; Yao et al., 2007; Wang et al., 2008). The meteorological factors of each city were simulated using the WRF model v3.5.1 (<http://www.wrf-model.org/>).

2.4. Spatial allocation

In this study, we first allocated all of the emissions to 30'' × 30'' grid cells and then aggregated them into an approximate 3 km × 3 km (0.025° × 0.025°) resolution. The method for spatially allocating the BTH emission inventory into grid cells depends on the source type and its spatial characteristics.

For point sources, we spatially allocated the emissions to the grid cells according to their latitude and longitude coordinates and then checked and adjusted the location. In general, the location of high-emissions industries, including power plants, heating plants, iron and steel factories, coking factories, cement factories, and glass factories were inspected individually in Google Maps. For the other point sources, the coordinates were checked according to the spatial extent at the county level and modified to the county center point if they appeared to be outside of the county in which they were supposed to be located.

The non-point emissions were allocated to a 30'' × 30'' grid cell based on the population density distribution. Population densities from 2013 LandScan Asia Population data (Oak Ridge National Laboratory, 2013) was used to aid in spatial allocation. For the residential emission sector, we also considered the urban/rural extents (Schneider et al., 2009) and allocated the urban and rural residential emissions based on the population densities in urban and rural areas, respectively.

The mobile sources were downscaled to the 30'' × 30'' spatial grids. In this study, we used a Geographic Information System (GIS) road atlas that contains road network information and traffic flow statistics specific to vehicle and road types as spatial surrogates to allocate the BTH regional mobile source emissions. The spatial proxies used for emission distribution are the same as that used by Zheng et al. (2014). The detailed procedures for the allocation process are detailed in Zheng et al. (2014).

2.5. NMVOC speciation

NMVOC speciation plays a key role in simulating ozone formation and secondary organic aerosol formation in chemical transport models (CTMs). We conducted NMVOC speciation based on the framework developed by Li et al. (2014). According to Li et al. (2014), uncertainties involved in improper species mapping are diminished when following the profile-assignment approach. The development of composite profiles and oxygenated volatile organic compound (OVOC) correction helps to reduce the uncertainties involved in profile selection. In this work, we divided total NMVOC emissions into more than 700 individual chemical species for Beijing city, Tianjin city and Hebei Province using the localized SPECIATE 4.2 database (Hsu and Divita, 2009) developed by Li et al. (2014).

2.6. Uncertainty analysis

Emission inventories are subject to uncertainties due to lack of knowledge in activity data and emission factors, or to the not accuracy or imprecision in estimates of these data. To assess the uncertainties of our new inventory, a quantitative uncertainty analysis was performed by estimating the 95% confidence interval of the emissions of each pollutant. We built a Monte Carlo framework for each city, collected uncertainty information for each parameter that can be described as a probability density function, and perform 10,000 trials to estimate the bounds of emissions around a 95% confidence interval. This method as well as the data used are well documented in previous studies (Lu et al., 2011a; Zhao et al., 2011, 2012a; Liu et al., 2015). Based on the quantitative assessment, we also made a qualitative analysis to identify the sources of uncertainty in the inventory and to give suggestions on further improvements.

3. Results

3.1. Emissions by sector in the BTH region

In 2013, the total SO₂, NO_x, PM_{2.5}, PM₁₀, CO, NMVOC, NH₃, BC, and OC emissions in the BTH region were 2,305, 2,686, 1,090, 1,494, 20,567, 2,207, 623, 160, and 254 Gg, respectively. Table 2 and Fig. 2 show the contributions of each sector to the total emissions for the 9 pollutants in the BTH region.

The power sector is a significant contributor to SO₂ and NO_x emissions in the BTH region, contributing approximately 11.8% and 23.3% of the total SO₂ and NO_x emissions, respectively. By contrast, the power sector only contributes 4.8%, 1.5% and 0.2% of the total PM_{2.5}, CO and NMVOC emissions, respectively. In 2013, there were more than 600 in-use units in the power sector in the BTH region with a total capacity of 58.8 GW (8.3 GW for Beijing, 10.4 GW for Tianjin, and 40.1 GW for Hebei). Approximately 70% of the unit capacities are equal to or larger than 300 MW, which is comparable to the national average level (74%). Few new power plants were set up in the BTH region in 2013, operating at a capacity of only above 1200 MW. Pulverized coal boilers were the dominant combustion technology used, accounting for 85% of the total capacity. The FGD penetration was 86%, with an average removal efficiency of 74%. The SCR/SNCR penetration was 27%, with an average removal efficiency of 22%.

The transportation sector is an important contributor of NO_x, CO, NMVOC and BC emissions, contributing 28.9%, 9.0%, 8.0% and 16.5%, respectively, of the total emissions in the BTH region. The BTH region has increased its focus on vehicle emissions control. Beijing and Tianjin began to implement Euro V standard for light-duty gasoline passenger cars and public fleets in 2013 and 2015, respectively. The residential sector is a major source of SO₂, PM_{2.5}, PM₁₀, CO, NMVOC, BC and OC emissions in the BTH region, contributing 14.4%, 31.3%, 26.5%, 29.2%, 21.5%, 46.6% and 71.7%, respectively, of the total emissions. The residential sector

contributes 4.1% and 4.6% of the total NO_x and NH₃ emissions, respectively. 91.9% of the total NH₃ emissions originates from the agriculture sector, with 44.2% from fertilizer use and 47.7% from livestock. Currently, the BTH region has not issued any measurement for the residential or agriculture sectors to control air pollutant emissions.

The industry sector is the largest contributor of SO₂, NO_x, PM_{2.5}, PM₁₀, CO, and NMVOC emissions in the BTH region, contributing 72.6%, 43.7%, 59.6%, 64.7%, 60.3%, and 70.4% of the total emissions, respectively. Fig. 3 illustrates the contributions of different pollutants from sources within the industry sector. Industrial boilers are the major sources of SO₂ and NO_x, contributing 44.8% and 37.1% of the total SO₂ and NO_x emissions from the industry sector, respectively. Only a few of the industrial boilers have installed flue-gas desulfurization to abate SO₂ emissions discharge, leading to a greater contribution of industrial combustion sources to SO₂ emissions than that of power plants. The iron and steel production is the dominant source of SO₂, PM_{2.5} and CO emissions, contributing 25.6%, 38.8% and 75.1%, respectively, of the total emissions from the industry sector. In addition, the cement production and coke production contribute 23.1% and 14.1% of the PM_{2.5} emission from the industry sector, respectively. For NMVOC emissions, solvent use and coke production are the major sources, contributing 46.7% and 14.1% of the emissions from the industry sector, respectively. For the industrial processes and solvent use, the Chinese government only enforces certain solvent content limits that are not strict compared to the limits of developed countries. Few end treatment facilities for VOC emissions have been installed. NH₃ emissions from the industry sector mainly originate from the ammonia related chemistry.

3.2. Emissions by city in the BTH region

The BTH 2013 inventory was spatially allocated into 3 km × 3 km grid cells using the methods presented in Section 2.4. Fig. 4 shows the spatial allocation of SO₂, NO_x, PM_{2.5}, and NMVOC emissions in the BTH region. Emissions of pollutants in the BTH region are mainly distributed over the cells of the eastern and southern BTH regions, in which industrial zones and large cities are located. The major point sources of SO₂ and PM_{2.5}, including power plants, iron and steel factories, cement factories and major industrial boilers are illustrated in Fig. 5.

The BTH inventories at the city level are shown in Table 3. Beijing, Tianjin, Shijiazhuang, Tangshan and Handan, which have developed industries, heavy traffic flows and large population, are areas with the highest emissions in the BTH region. These 5 cities account for 61.5%, 65.4%, 63.3%, 65.1%, 65.2%, 64.9%, 57.2%, and 55.0% of the total SO₂, NO_x, PM_{2.5}, PM₁₀, CO, NMVOC, BC, and OC emissions in the BTH region, respectively. However, the NH₃ emissions from these 5 cities account for approximately 43.7% of the total emissions in the BTH region, which is not significantly higher than other cities. This result is reasonable because NH₃ emissions are mainly related to agricultural production, and these 5

Table 2
Emissions by sector in the BTH region for the year 2013.

	Annual air pollutant emissions/Gg								
	SO ₂	NO _x	PM _{2.5}	PM ₁₀	CO	VOC	NH ₃	BC	OC
Power	271.5	626.2	52.6	82.8	300.1	4.5	0	0.1	0
Industry	1672.6	1174.3	649.3	966.9	12400.9	1553.2	17.9	59.0	62.7
Transportation	29.5	775.6	46.3	47.6	1853.8	176.2	4.0	26.4	9.1
Residential	331.6	110.2	341.4	396.4	6012.6	473.6	28.5	74.5	182.4
Agriculture	0	0	0	0	0	0	572.1	0	0

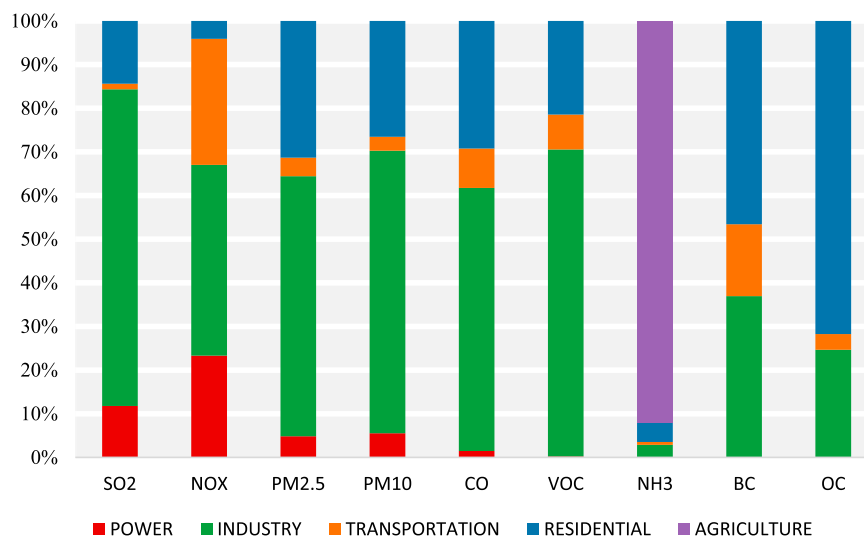


Fig. 2. Emission contributions by sector in the BTH region for the year 2013.

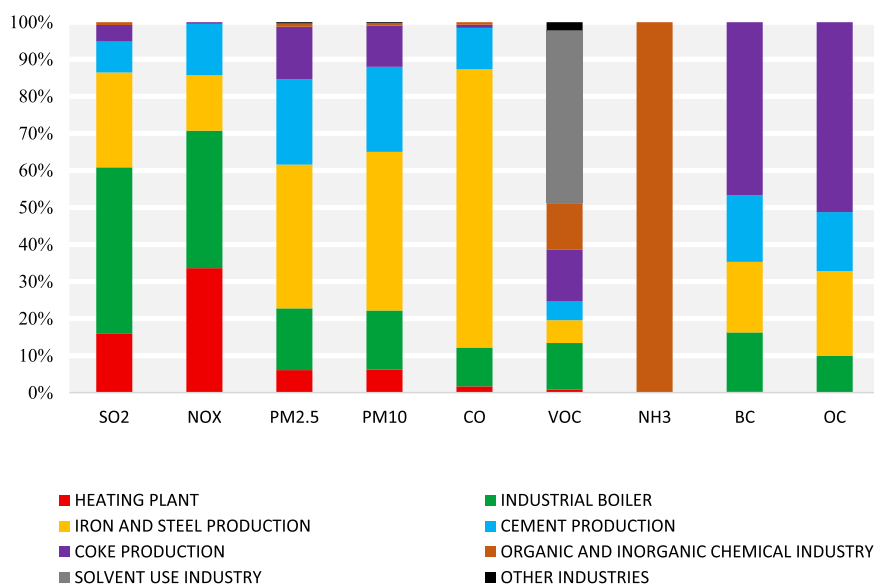


Fig. 3. Emission contributions of major sources in the industry sector in the BTH region for the year 2013.

cities have relatively higher urbanization levels and less farmland area. There are differences in the rank of cities in terms of their emissions contributions for different pollutants, which is owing to the differences in emission sources and industry structure. For example, Beijing ranks high in NMVOC emissions, which is related to solvents used in architecture coating and transport emissions, whereas the SO₂ emissions from Beijing are average because the city does not have many large industrial coal fired units. Another example is Tangshan, whose developed iron production yields the highest emissions of SO₂, NO_x and PM_{2.5}. However, NMVOC emission from Tangshan is less than that of Tianjin and Beijing. Fig. 6 shows the SO₂, NO_x, PM_{2.5} and NMVOC emissions from individual cities by sector.

Fig. 7 illustrates the relations among emissions intensity, emissions per capita and emissions per GDP for SO₂, NO_x, PM_{2.5} and NMVOC at the city level. The average emissions intensity in the BTH

region for SO₂, NO_x, PM_{2.5}, PM₁₀, CO, NMVOC, NH₃, BC and OC were 10.4, 12.1, 4.9, 6.7, 92.8, 10.0, 2.81, 0.7 and 1.1 t/km², respectively. The range of the emission intensities for different cities in the BTH region were 2.3–30.7 t/km² for SO₂, 1.1–36.1 t/km² for NO_x, 1.2–17.5 t/km² for PM_{2.5}, 1.7–26.2 t/km² for PM₁₀, 21.6–387.3 t/km² for CO, 1.5–32.2 t/km² for NMVOC, 0.8–5.5 t/km² for NH₃, 0.1–2.0 t/km² for BC and 0.2–3.0 t/km² for OC. These large variation ranges originate from differences in the level of development, industrial structure, and population. The cities with developed industries, such as Tianjin and Tangshan, have the highest emission intensities for SO₂, NO_x and PM_{2.5}. By contrast, such cities as Chengde and Zhangjiakou have low emissions intensities.

Moreover, the emissions per capita in Tangshan is significantly higher than that of any other city in the BTH region. Beijing has the lowest emissions per capita for SO₂, NO_x, and PM_{2.5}, whereas both the emission intensity and emission per capita of Beijing are high

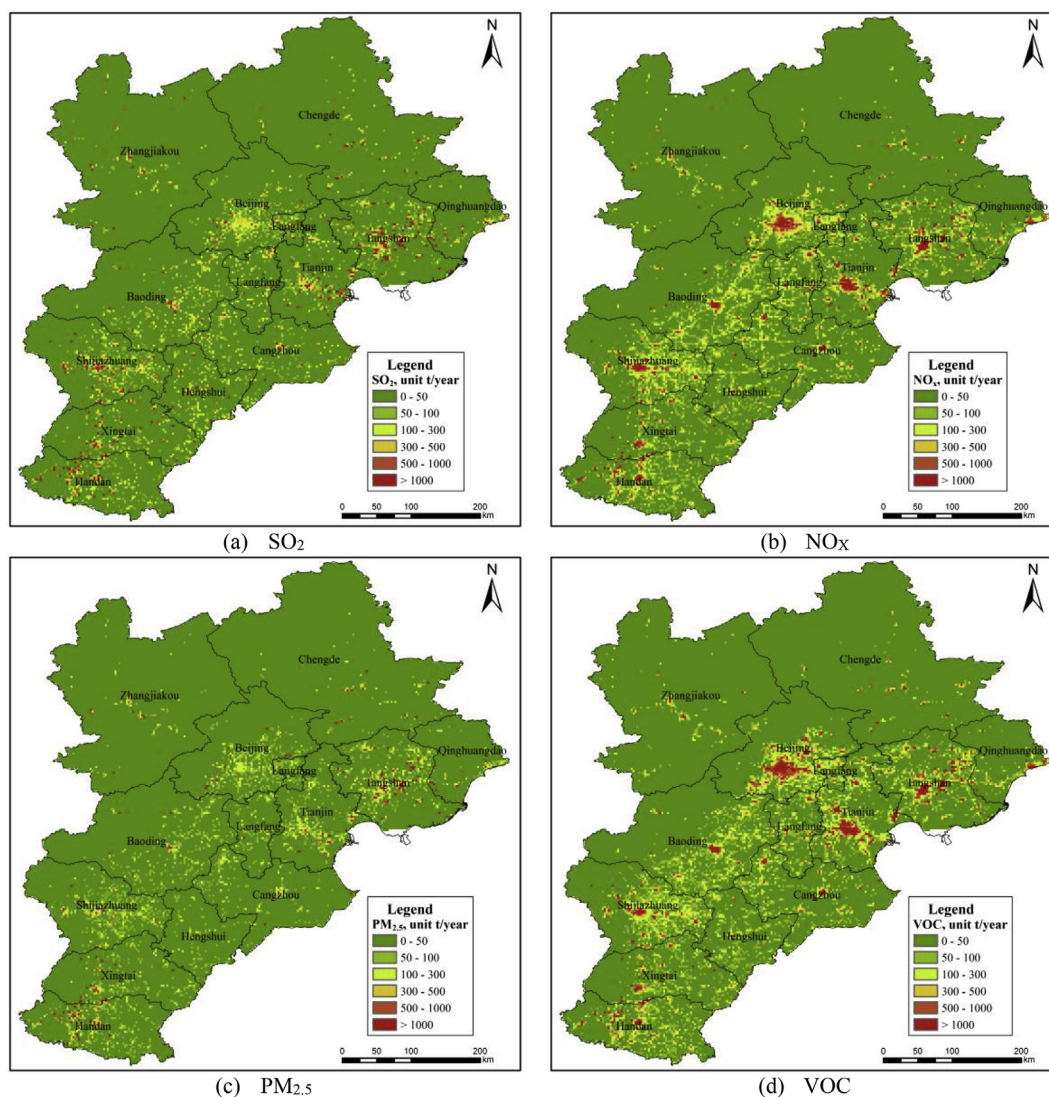


Fig. 4. Spatial distribution of air pollutant in the BTH region for the year 2013.

for NMVOC emissions, which is reasonable because solvent use in building and transport emissions are the major source of NMVOC. In addition, emissions per GDP in Beijing and Tianjin are significantly lower than that for cities in Hebei Province.

3.3. Comparison with other inventories

Some national or regional emission inventories that included BTH region are available for the years between 2003 and 2010. Since our inventory is the first comprehensive emission inventory for the year of 2013, the comparisons with other inventories derived from previous literatures mainly reflect the inter-annual variation of air pollutant emissions in the BTH region. We mainly compared the emission estimates in the BTH region emission inventory with those from the 2006 INTEX-B inventory (Zhang et al., 2009), and the Huabei regional emission inventory (2003 Huabei EI) for the year of 2003 (Zhao et al., 2012b). For SO_2 , NO_x and NH_3 , the research of Su et al. (2011b), Zhao et al. (2013) and Zhou et al. (2015) were also compared with our data respectively. Emission estimates from different studies are presented in Table S1 of the Supplement.

Our estimated SO_2 emission (2 305 Gg) in BTH region for the

year 2013 is between the results from the 2006 INTEX-B (2 865 Gg) and the 2003 Huabei EI (2068 Gg). The estimation of SO_2 in the BTH region for the year 2007 from Su et al. (2011b) is 2 187 Gg, which is lower than the result in this study. The fluctuation in annual SO_2 emission is consistent with the official data. According to the Ministry of Environmental Protection, because of the installation of flue-gas desulfurization devices, SO_2 emission in China decrease for the first time in 2007 after the rapid increase during 2002–2006. While our calculated NO_x emission (2 686 Gg) in BTH is higher than that in the 2006 INTEX-B (2000 Gg) and 2003 Huabei EI (1 578 Gg). The estimates in this study is also higher than that of Zhao et al. (2013) who estimated NO_x emissions in the BTH region for the year 2010 to be 2 510 Gg. The results reflect the continuous increase in NO_x emission in the BTH region during 2003–2013. One reason for the increase is that the activity data (e.g., vehicle population and fuel consumption) in the BTH region increase significantly. For example, the coal consumption and diesel consumption in the BTH region have increased 87.9% and 182% during the same period, respectively. Meanwhile, NO_x control technologies like SCR/SNCR were not widely installed in the power plant and cement industry until 2014, when China upgraded the emission standard of air pollutants for NO_x . The NO_x removal efficiency of power plant and

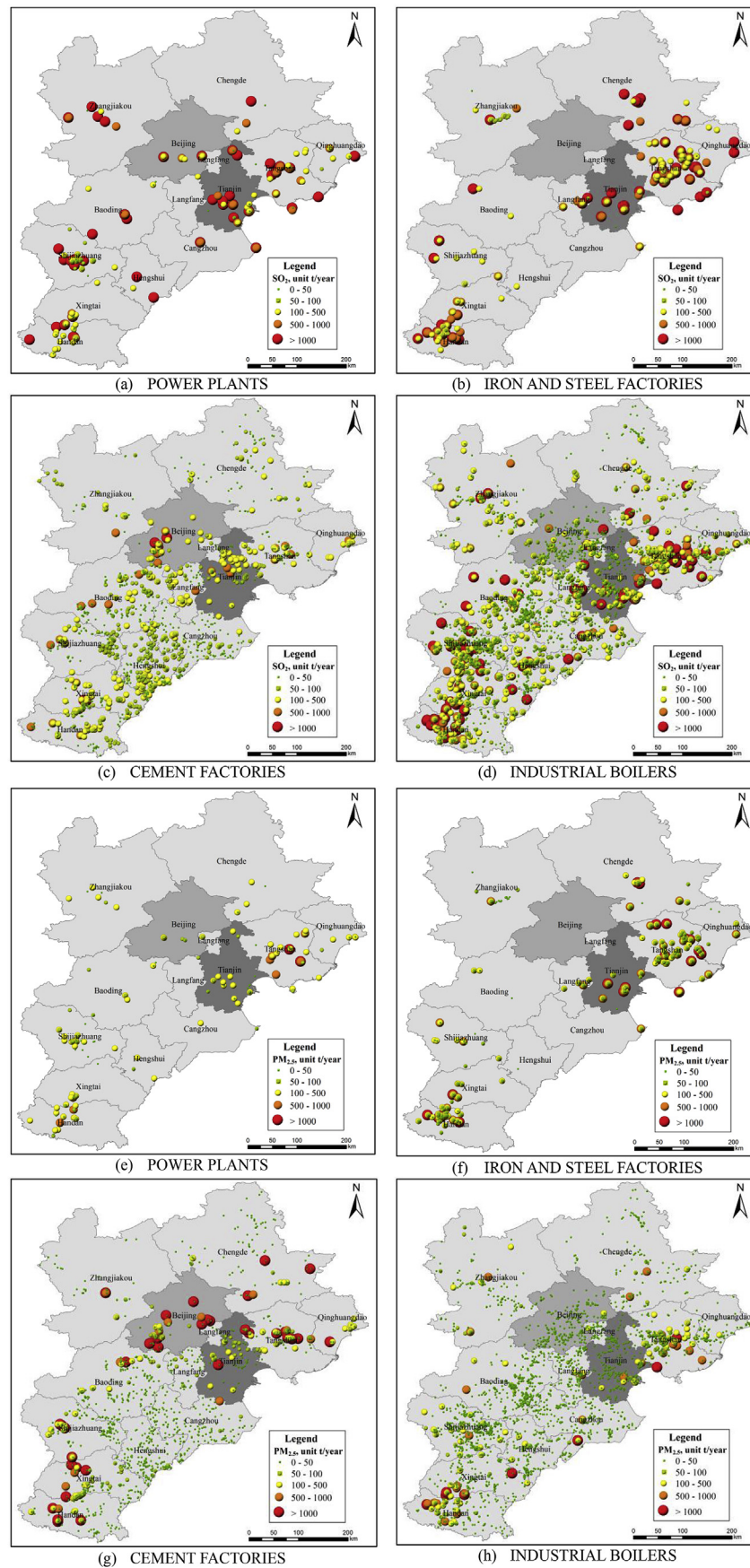
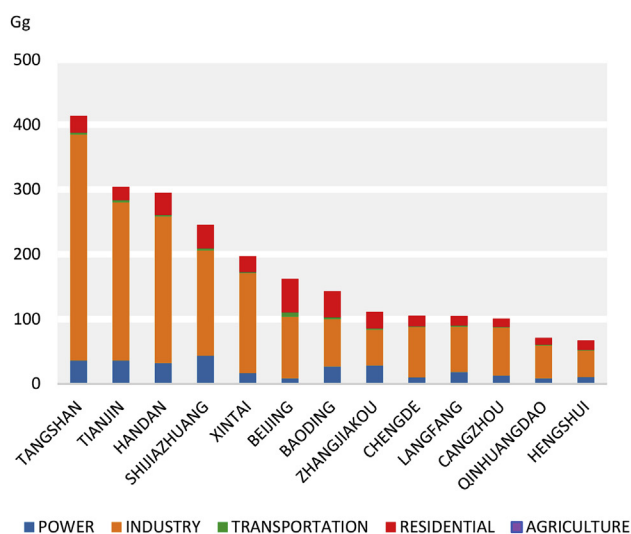


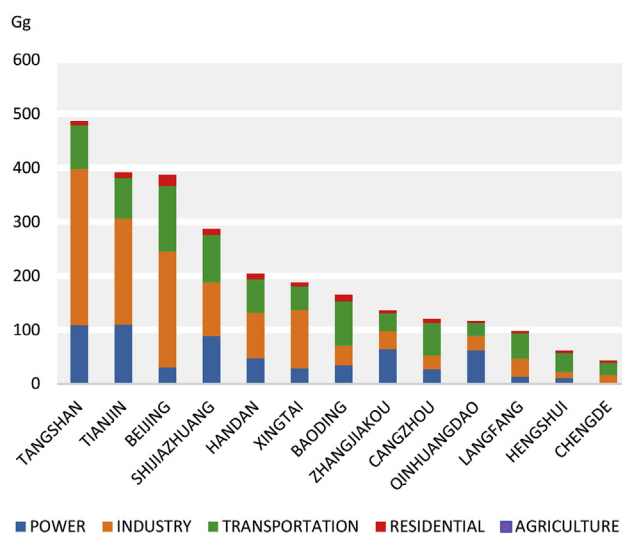
Fig. 5. Major point sources of SO_2 and $\text{PM}_{2.5}$ in the BTH region for the year 2013.

Table 3
Emissions by city in the BTH region for the year 2013.

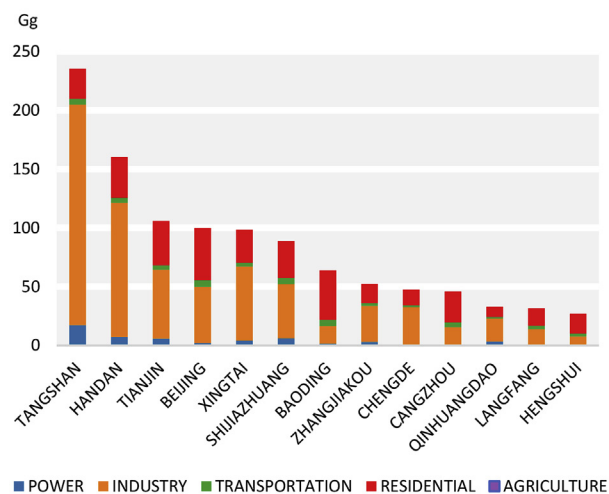
Province	City	Annual air pollutant emissions/Gg								
		SO ₂	NO _x	PM _{2.5}	PM ₁₀	CO	VOC	NH ₃	BC	OC
Beijing		160.3	387.2	99.8	134.1	1592.4	336.5	36.5	17.3	25.3
Tianjin		304.1	391.8	105.8	143.7	2264.1	385.2	41.5	12.5	25.5
Hebei	Shijiazhuang	243.8	287.0	88.9	122.7	1646.3	203.0	68.5	13.2	20.9
	Tangshan	413.6	486.8	235.3	352.3	5218.4	310.3	59.8	24.2	32.2
	Qinhuangdao	87.9	116.7	32.9	47.7	694.1	48.1	24.3	3.3	5.7
	Handan	295.4	203.9	160.2	220.1	2678.8	198.4	65.9	24.3	36.0
	Xingtai	195.7	188.1	98.4	128.5	1283.0	143.5	46.4	14.5	22.3
	Baoding	149.2	165.2	63.8	78.8	1181.8	147.0	68.2	13.8	25.4
	Zhangjiakou	120.3	135.9	52.2	69.1	797.1	70.1	46.2	8.7	13.4
	Chengde	91.4	43.2	47.5	67.4	1084.5	60.4	32.3	5.1	9.1
	Cangzhou	89.2	120.9	45.8	55.8	892.5	164.2	54.7	10.5	17.5
	Langfang	89.6	97.9	31.7	41.1	711.1	81.2	36.0	5.8	9.6
	Hengshui	64.6	61.5	27.1	32.4	523.5	59.5	42.4	6.8	11.3



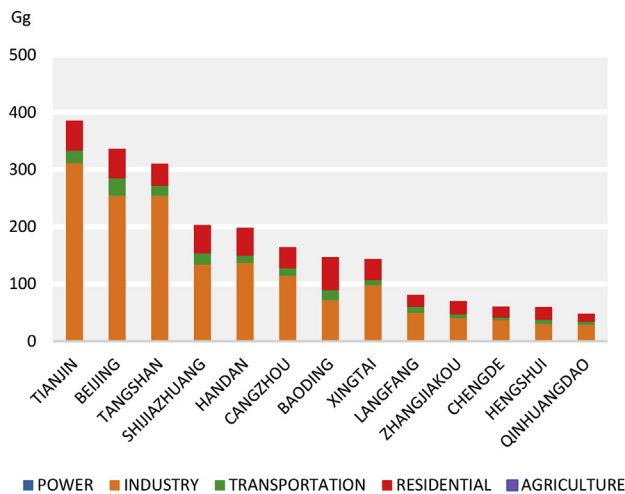
(a) SO₂



(b) NO_x



(c) PM_{2.5}



(d) VOC

Fig. 6. City-level emissions by sector in the BTH region for the year 2013.

cement industry in the BTH region was 22% and 16% in 2013, respectively. Besides, the use of different estimation methods and different data sources is also an explanation to the difference.

For NH₃ emission, our estimates (623 Gg) was lower compared to the results of 2003 Huabei EI (1 192 Gg) and Zhou et al. (2015) who calculated the NH₃ emission in 2010 to be 1 574 Gg in the

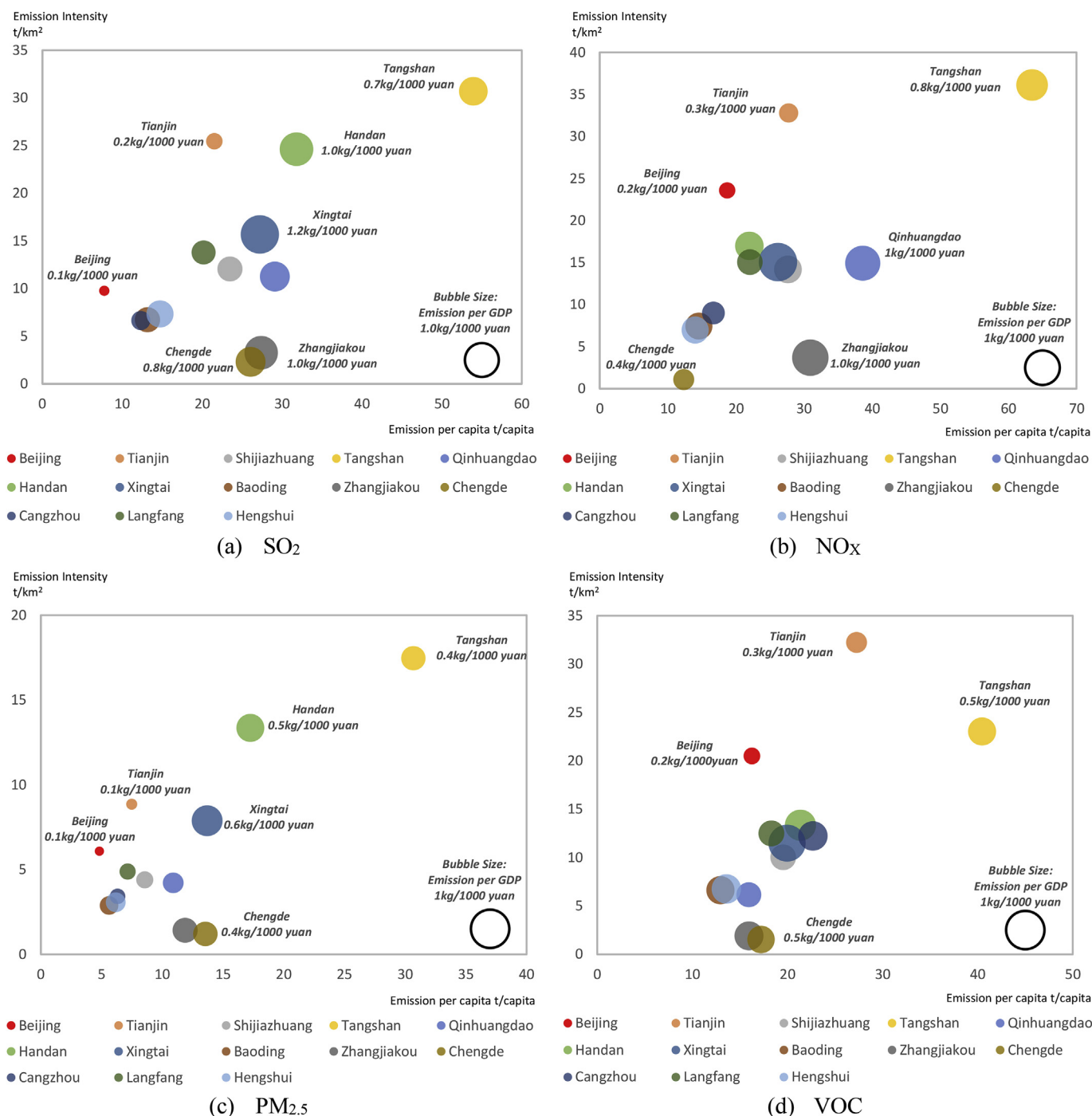


Fig. 7. Emissions intensity (t/km^2), emissions per capita (t/capita) and emissions per GDP ($\text{kg}/1000 \text{ yuan}$) of cities in the BTH region for the year 2013. Emissions intensity and Emissions per capita are marked by Y-axis and X-axis respectively. The size of bubble represents the emissions per GDP.

BTH region. Different non-industrial sources considered is the main reason for the difference in NH_3 emissions. In this study, we mainly estimates the NH_3 emission from livestock and fertilizer application that are the largest emission sources for NH_3 . Other NH_3 emission sources, including the ammonia related-chemical industries, on-road mobile, waste treatment, and the fuel combustion were considered. However, biomass open burning is not included in this study. $\text{PM}_{2.5}$, PM_{10} and NMVOC estimates from the BTH inventory in this study and the 2006 INTEX-B are very comparable, however,

significant different compared to the results in 2003 Huabei EI. The disparity was caused mainly by the differences in the sources considered and emission factors selection which related to the technology improvements. The Multi-resolution Emission Inventory for China (MEIC) was also compared to this BTH regional emission inventory. MEIC is an emission model framework developed by Tsinghua University (<http://www.meicmodel.org>). We chose the emission estimates for the year of 2013 from MEIC. The estimates of SO_2 , NO_x , $\text{PM}_{2.5}$ and PM_{10} from the two emission

inventories are in good agreement with the difference less than 10%.

3.4. NMVOC speciation

We estimated the total NMVOC emissions of Beijing, Tianjin and Hebei Province to be 336, 385 and 1486 Gg, respectively, using the bottom-up approach described in Section 2.1. Owing to the high-energy consumption and a large population, Hebei contributes to 67.3% of NMVOC emissions in the BTH region. Emissions from Beijing and Tianjin are comparable, contributing 15.2% and 17.5%, respectively.

The NMVOC emissions by sector for cities in the BTH region are presented in Fig. 6, demonstrating the differences between Hebei province and the two other cities. All three sub-regions are dominated by industrial sources, contributing more than 70% of the total NMVOC emissions. With a greater population living in rural areas, thus corresponding to greater energy consumption, the contribution from residential sources is considerably larger in Hebei Province (24.8%) than in Beijing (15.5%) and Tianjin (13.6%).

Different individual chemical species have different impacts on the formation of ozone and secondary organic aerosols, which should be characterized by emissions of individual NMVOC species. We speciated the total NMVOC into more than 700 individual chemical species following the profile-assignment approach presented in the methodology section. As presented in Fig. 8, we ranked the species according to the emissions in the BTH region and depicted the emissions contributions by the power, industry, residential, transportation and agriculture sectors for the top 30 species. Dominated by Hebei Province for total NMVOC emissions, the emission characteristics of individual species for the entire region are identical to those of Hebei. The major NMVOC species were ethylene, acetylene, ethane and toluene. Ethylene is the largest contributor in Tianjin (615 Mmole) and Hebei (3 049 Mmole) and the second largest contributor in Beijing (536 Mmole). For Beijing, toluene contributes 616 Mmole, ranking first among all species. All emissions are dominated by industry sources, except for OVOC species, such as methyl alcohol and glyoxal. The differences in significant emission patterns of individual species among the

three sub-regions can be attributed to the differences in industrial structures.

3.5. Uncertainty analysis

The BTH regional emission inventory was compiled using a bottom-up approach based on detailed data of major air pollution sources in the BTH region. Considering the uncertainty information of each parameter, the average uncertainties of emissions from the BTH region are estimated through the Monte Carlo method as –18 to 16% for SO₂, –17 to 15% for NO_x, –24 to 23% for PM_{2.5}, –19 to 18% for PM₁₀, –25 to 23% for CO, –48 to 44% for NMVOC, –54 to 48% for NH₃, –54 to 49% for BC, and –59 to 55% for OC. The uncertainty of our emission inventory is slightly reduced compared to the previous inventories that relied on provincial statistics data (Lu et al., 2011a; Zhao et al., 2011, 2012a). The ranges of uncertainties for varied pollutants and cities are presented in Table S2 of the Supplement.

For SO₂, NO_x, PM_{2.5} and PM₁₀, over 60% of the total emission were originated from point sources according to the results of this study. Point sources including power and key industries sources are relatively reliable because the majority of the activity data, including fuel consumption, sulfur content, ash content, exhaust control efficiency and geographic location, of the sources were obtained from the detailed facility-level census data. Therefore, the uncertainties in SO₂ and NO_x emissions are relatively low, because point sources are dominated contributors. Uncertainties for SO₂ and NO_x emissions are mainly caused by residential sources and mobile sources, respectively. CO, PM_{2.5} and PM₁₀ have relative higher uncertainties than SO₂ and NO_x. Transportation sources, residential sources, and industrial processes are major contributors to the uncertainties in CO, PM_{2.5} and PM₁₀ because we are less confident of the emission estimates of these sources owing to the considerable uncertainty in activity levels and emission factors. Compared to the pollutants above, VOC and NH₃ have higher uncertainties. The majority of VOC emissions come from chemical industry processes and solvent use. These two major emission contributors have high uncertainties because the calculation method simplified the complexity of the actual emission process

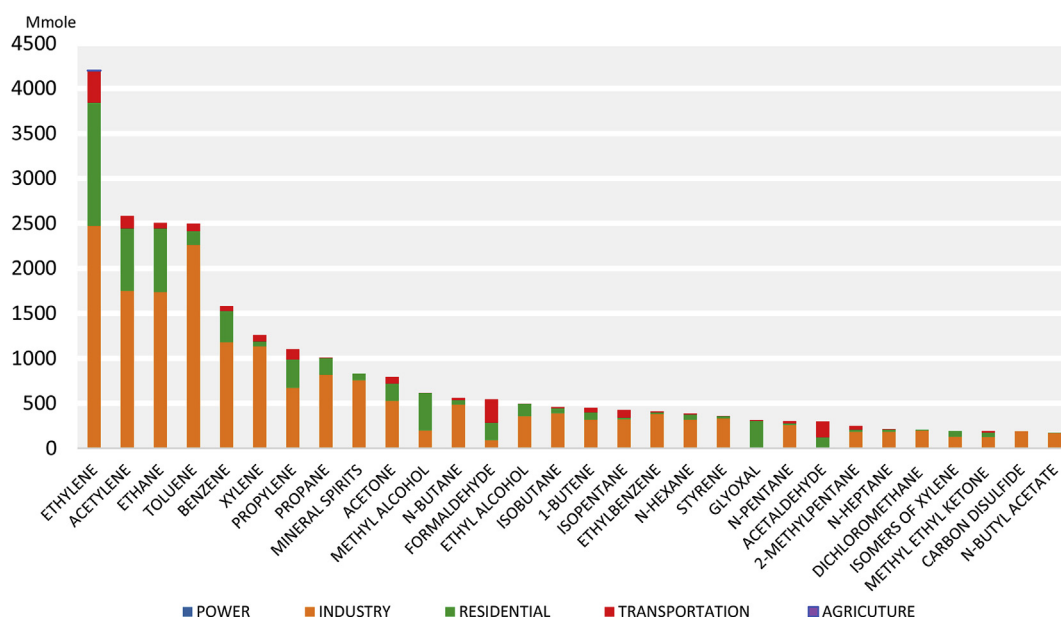


Fig. 8. Top 30 NMVOC species emissions by sector in the BTH region for the year 2013.

and the emission factors are mainly taken from previous research findings, which could not reflect the actual situation in the current BTH region. As the largest NH_3 sources, livestock and fertilizer application have many factors that influence its uncertainty, such as variations in fertilizer, fertilizing time, and local meteorological conditions. In addition, NH_3 escape from SCR/SNCR is also a source of NH_3 emission. Our study did not include this source in the BTH region, and this omission will cause a slight underestimate of the NH_3 emission. Carbonaceous aerosols (i.e., BC and OC) have the highest uncertainties among the air pollutants, because the largest emissions source of BC and OC is the residential sector, especially rural coal use and biofuel use, which are subject to large uncertainties due to lack of sufficient information on the activity level, combustion technologies, and the emissions rates.

4. Implications and discussion

In recent years, the Chinese government has made considerable efforts to solve air quality issues. However, a comprehensive air pollutants emission inventory based on facility-level data is difficult to establish owing to data applicability and availability. Detailed data is the foundation for high-resolution air pollutants emission inventory. Just because of detailed data of key industries were collected from several restricted databases, we can develop the method that estimating emissions on unit level in this study. The use of facility-level data can lower the uncertainty of inventories and improve the accuracy of air quality modeling. Developed countries have already established industrial emission inventories containing all point sources based on facility-level data. By contrast, the existing relative emission databases in China are not designed for developing an emission inventory. Moreover, detailed activity data at the facility level are typically not publicly available in China. In 2017, China will launch the Second-Time China Pollution Source Census. This effort is an opportunity for China to build a detailed database that can be used to develop an emission inventory. Therefore, the designer of the census should consider the data structure and statistical information to meet the needs associated with developing an emission inventory.

The Action Plan for Air Pollution Prevention and Control set several specific targets for the BTH region. It is important to assess all possible paths and select an effective strategy to achieve these targets. Based on the 2013 emission inventory in the BTH region, the industry sector is the primary air pollutant source for all three sub-areas. According to the data used in the paper, the key industries in the BTH region have more than 6000 industrial boilers to provide power and heat for industrial production. Because of the lack of regulations, few of these industrial boilers have emission reduction facilities in 2013. Thus, policy makers should consider enhancing the standards on industrial boiler emissions and require upgrades to the emission control technologies of existing industrial boilers. In addition to the industrial boilers, all fossil fuel fired units in key industries, including iron and steel production, cement production, and coke production, should have desulfurization, denitrification and dust transformation technologies installed in the facilities to reduce the SO_2 , NO_x , $\text{PM}_{2.5}$ and PM_{10} emissions. All dry cement kilns should also be required to implement low- NO_x combustion denitration. Furthermore, VOC-related emissions should have stricter standards, and VOC treatments should be implemented in such industries as coking, chemicals and printing.

In addition to industrial upgrading, it is equally important to focus efforts on emission control from the transportation sector, particularly for such cities as Beijing, in which vehicle ownership is over 5.5 million. Currently, the BTH region has been limiting the vehicle population growth by such measures as car plate lotteries and electric vehicle popularization. Supporting measures, including

the development of public transportation and charging pile construction, should be employed. Policy makers should consider universally upgrading new vehicle emission and fuel standards every few years. In addition, accelerating the phasing out of old vehicles and strengthening vehicle inspection and maintenance would mitigate air pollution.

The emissions per GDP of cities in Hebei Province is significantly higher than that of Beijing and Tianjin. Therefore, limiting high-emissions-intensive but low-GDP industries is an urgent issue in Hebei Province. Currently, Hebei Province is dominated by high-energy and emission-intensive industries with outdated production facilities. Hebei Province should consider reconstructing its industrial structure by reducing its iron-production capacity, accelerating the denitrification of key industries, cutting back on coal use and phasing out small boilers. Moreover, Beijing should be more active in helping cities in Hebei Province upgrade their industrial structure by transferring Beijing's low-emission industries, such as information technology, commercial logistics, and financial services, to cities in Hebei. In turn, the population pressure in Beijing could also be mitigated.

In this study, we developed a data assimilation method combining different statistics to obtain parameters for emission estimating, based on that produced a unit-based emission inventory. We combined city-level vehicle activity, city-level emission factors corrected by ambient factors, and provincial technology turnover model to map vehicle emissions at high resolutions. Compared with the previous studies, industrial and mobile emissions sources considered in this paper were relatively more comprehensive and some emission factors were updated. Therefore, we believe the BTH regional emission developed in this study can provide better support to emission characteristics analysis and fine scale atmospheric modeling than other existing works. There are aspects that can be improved in the development of a future BTH regional inventory. For the industry sector, a comprehensive investigation of solvent type and activity data collection for VOC product-related sources in the BTH region could greatly improve the accuracy of VOC emission estimates. Furthermore, the use of real-time traffic flow data that can be obtained from digital map companies could improve the emission estimates from mobile sources, particularly for NO_x and VOC emissions in the BTH region. Local emission factor development for residential and agriculture sectors could be another crucial path to improve the BTH inventory. Other important sources including road dusts, biomass open burning and NH_3 escape from SCR/SNCR not considered in this study should further be examined.

5. Conclusion

This study sought to develop a high-resolution emission inventory for major air pollutants in the BTH region for the year 2013. A "bottom-up" methodology was adopted to compile the inventory based on the fusion of updated facility-level data from restricted databases. The inventory was prepared for the development of future BTH regional air pollution prevention policy and air quality models. The estimates from the BTH 2013 inventory show that the total emissions of SO_2 , NO_x , $\text{PM}_{2.5}$, PM_{10} , CO, NMVOC, NH_3 , BC, and OC were 2,305, 2,686, 1,090, 1,494, 20,567, 2,207, 623, 160, and 254 Gg, respectively.

The industry sector is the greatest contributor of SO_2 , NO_x , $\text{PM}_{2.5}$, PM_{10} , CO, and NMVOC emissions in the BTH region, contributing 72.6%, 43.7%, 59.6%, 64.7%, 60.3%, and 70.4% of the total emissions, respectively. Power plants contributed 11.8% and 23.3% of the total SO_2 and NO_x emissions, respectively. The transportation sector contributed 28.9% of the total NO_x emissions. The residential sector is a major source of SO_2 , $\text{PM}_{2.5}$, PM_{10} , CO, NMVOC,

BC and OC emissions in the BTH region, contributing 14.4%, 31.3%, 26.5%, 29.2%, 21.5%, 46.6% and 71.7%, respectively, of the total emissions. More than 90% of the total NH_3 emissions originate from the agriculture sector, with 44.2% from fertilizer use and 47.7% from livestock. Beijing, Tianjin, Shijiazhuang, Tangshan, and Handan are the major contributors, accounting for 61.6%, 65.4%, 63.3%, 65.1%, 65.2%, 64.9%, 43.7%, 57.2% and 55.0% of SO_2 , NO_x , $\text{PM}_{2.5}$, PM_{10} , CO, NMVOC, NH_3 , BC and OC emissions in the BTH region, respectively. The major NMVOC species were ethylene, acetylene, ethane and toluene. Ethylene is the biggest contributor in Tianjin and Hebei. The largest contributor in Beijing is toluene.

In general, we have greater confidence in the emission estimates from point sources, including power and key industries from the industry sector, because facility-level data ensure low uncertainty; the uncertainties are mainly from estimates of non-point and mobile sources. Therefore, the uncertainties in SO_2 and NO_x emissions are relatively low, because point sources are dominated contributors. Transportation sources, residential sources, and industrial processes are major contributors to the uncertainties in CO, $\text{PM}_{2.5}$ and PM_{10} . In addition, VOC and NH_3 emissions have high uncertainties because of the lack of key representative emission factors and large uncertainties in activity data.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.atmosenv.2017.09.039>.

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