

Article



# **Estimation and Prediction of Industrial VOC Emissions in Hebei Province**, China

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Abstract: The study of industrial volatile organic compound (VOC) emission inventories is essential for identifying VOC emission levels and distribution. This paper established an industrial VOC emission inventory in 2015 for Hebei Province and completed an emission projection for the period 2020–2030. The results indicated that the total emissions of industrial VOCs in 2015 were 1017.79 kt. The use of VOC products accounted for more than half of the total. In addition, the spatial distribution characteristics of the industrial VOC emissions were determined using a geographic information statistics system (GIS), which showed that the VOCs were mainly distributed the central and southern regions of Hebei. Considering the future economic development trends, population changes, related environmental laws and regulations, and pollution control technology, three scenarios were defined for forecasting the industrial VOC emissions in future years. This demonstrated that industrial VOC emissions in Hebei would amount to 1448.94 kt and 2203.66 kt in 2020 and 2030, with growth rates of 42.36% and 116.51% compared with 2015, respectively. If all industrial enterprises took the control measures, the VOC emissions could be reduced by 69% in 2030. The analysis of the scenarios found that the most effective action plan was to take the best available control technologies and clean production in key industries, including the chemical medicine, coke production, mechanical equipment manufacturing, organic chemical, packaging and printing, wood adhesive, industrial and construction dye, furniture manufacturing, transportation equipment manufacturing, and crude oil processing industries.

Keywords: industrial VOCs; emission inventory; distribution; scenario prediction

## 1. Introduction

Volatile organic compounds (VOCs) are known as one of the main sources of secondary organic aerosols (SOAs) generated through photochemical oxidation and as the key precursors to the formation of ground-level ozone [1]. Additionally, VOCs themselves are harmful to human health. For example, BTEX (i.e., the group of VOCs of benzene, toluene, ethyl-benzene, and xylenes) have inhalation toxicity and cause cancer [2]. In particular, benzene was listed as a first-class carcinogen by the International Institute of cancer (IARC). Therefore, the abatement of VOC emissions has gradually become a significant issue in improving air quality and public health. Before the formulation of effective control strategies to reduce VOC emissions, it is an essential and urgent matter to accurately estimate the present emissions of pollutants and predict the future trend.

The establishment of an emission inventory with a high spatial and temporal resolution could not only help to quantify and characterize pollutant emissions from anthropogenic and natural sources but also be used as input data for air quality models to simulate air quality variation in preparation for further health and cost–benefit analyses that are of

Citation: Guo, X.; Shen, Y.; Liu, W.; Chen, D.; Liu, J. Estimation and Prediction of Industrial VOC Emissions in Hebei Province, China. *Atmosphere* **2021**, *12*, 530. https:// doi.org/10.3390/atmos12050530

Academic Editor: Joel F. de Brito

Received: 24 February 2021 Accepted: 16 April 2021 Published: 21 April 2021

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**Copyright:** © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses /by/4.0/). greater concern. Since the 1990s, Europe and the United States have recognized the harm of VOC pollutants and thus conducted some studies for the establishment of VOC emission inventories and emission factor libraries [3–5]. A global VOC emission inventory of anthropogenic sources was established in 1990, where wood fuel combustion, savannah combustion, gasoline production, waste treatment, organic chemical industry, and other sectors were found to be the main emission sources [6]. Moreover, Rajabi [7] evaluated the overall scale of global VOC emissions from all stages of oil processing. Until now, a number of studies have already established national and regional VOC emission inventories with higher sectoral, temporal, spatial, and chemical species resolutions. For instance, Benjamin [8] conducted a study on the resolved biogenic hydrocarbon emission inventory for the California south coast air basin. Dommen [9] developed a VOC emission inventory for the Lombardy region in Italy with a one hour temporal and 3 km spatial resolution which comprehensively covered emission sources such as biological sources, industrial production, solvent use, waste treatment, transportation, power plants, and oil refineries. Besides this, there are also some studies on VOC emissions focused on other countries and regions, including Spain [10], Greece [11], Denmark [12], India [13], southwest Africa [14], and Korea [15]. The first inventory of anthropogenic sources of VOCs in China from 1990 to 2000 was established referring to the emission factors from Western countries [16]. After that, more studies began to focus on VOC emission inventories from anthropogenic sources in China. For example, Wei [17] and Gong [4] established a VOC emission inventory from 2005 to 2010 in China which was more accurate than its predecessor, owing to its refinement and localization of emission factors. A VOC emissions inventory of anthropogenic sources at the county level in China was compiled in 2000 based on the investigation of activity data for all cities, prefecture-level cities, and counties throughout the country [4,18]. Moreover, a couple of studies focused on VOC emission inventories for some key regions, such as north China, the Yangtze River Delta (YRD), the Pearl River Delta (PRD), and other areas [19–24].

However, there is still lack of VOC emission inventories for some major areas, such as Hebei Province. This region is of particular interest because of its special location and poor air quality. It lies in the key area (Beijing-Tianjin-Hebei region) of pollution control in China. In fact, 6 of the 10 worst air pollution cities of China were found to be located in Hebei Province in 2015 [25]. Therefore, it is of vital importance to establish a VOC emission inventory to further improve air quality and formulate more effective pollution control strategies for Hebei Province.

The VOC emission sources in China generally comprise stationary combustion, road vehicles, solvent utilization, and industrial processes. Some studies have shown that biomass-burning sources cause the largest amount of emissions, but the total amount of industrial VOC emissions is projected to grow substantially in the future [6,26–28]. In addition, the study results of Zhang [29] showed that anthropogenic VOC emissions could dominate on a global level, especially in densely populated urban areas or highly industrialized regions. Moreover, Hebei Province is characterized by its small area, large population density, and developed industry, with a large amount of coal and chemical industries. According to the China Statistical Yearbook [30], the industrial GDP of Hebei Province in 2015 was 144 billion Yuan, accounting for 5.1% of the national industrial GDP, where the output of major industries related to VOC emissions, including chemical medicine, petrochemicals, coke production, machine equipment manufacturing, wood adhesives, organic chemistry, and packaging and printing, accounts for 1%, 2%, 3%, 4%, 5%, 7%, and 8% of the national output, respectively. Hence, it is necessary to fully understand the characteristics of the VOCs emitted from industrial sources in Hebei Province.

In this paper, the emission factor method was used to establish a VOC emission inventory from industrial sources in Hebei Province for 2015. Additionally, the spatial distribution of industrial VOCs was analyzed by applying population density information based on geographic information systems (GIS). Moreover, three emission reduction scenarios were defined based on the presently feasible VOC control strategies in order to predict the changing trend of industrial VOC emissions in Hebei Province for 2020 and 2030. The results could be fundamental and valuable for formulating VOC control policies.

#### 2. Materials and Methods

## 2.1. Study Area

Hebei Province covers an area of 187,700 km<sup>2</sup>, accounting for 1.95% of the total territory, 5.4% of the total population, and 4.3% of the total GDP of China in 2015 [30]. Figure 1 shows the trend of the GDP, urbanization rate, and population of Hebei Province and China over the past 20 years. Although the trends in Figure 1a,b are generally similar, the GDP and urbanization in Hebei Province have grown rapidly and at a faster rate than the nationwide one since 2000. As China's major industrial base, the secondary industry in Hebei Province accounted for 48.3% of the total national value in 2015, forming the industrial structure dominated by seven major industries, including equipment manufacturing, iron and steel, petrochemicals, food, medicine, building materials, and textiles [31]. The pillar industries dominated by steel, coal, and cement have caused Tangshan to rank top in Hebei Province in terms of GDP and GDP per capita for many years. As the second city in Hebei Province in terms of GDP, Shijiazhuang is an important national textile base of China and also has the largest pharmaceutical industry, with its output accounting for about 35% of the total in China. Moreover, the industrial GDP in other cities, such as Cangzhou, Baoding, Handan, also accounted for a relatively large proportion of the Hebei province [32].



Figure 1. The trends of GDP, population, and urbanization rate in Hebei Province (a) and China (b) from 1995 to 2015.

#### 2.2. Methodology

The Technical Guide on Compiling Atmospheric Volatile Organic Compounds Source Inventory (hereafter referred to as the Technical Guide), issued in August 2014 [33], was employed in this study. This methodology includes four steps: the identification of major sources of VOC emissions, activity data collection, emission factor (EF) analysis, and the method of calculation of emissions. The industrial VOC emissions were calculated using the EF approach [11,32], implemented using the following equation:

$$\mathbf{E} = \sum_{i} \sum_{j} A_{i,j} EF_{i,j} (1 - n_{i,j}), \tag{1}$$

where i represents the city, j represents the emission source, E is the annual total emissions of the VOCs,  $\eta$  is the efficiency of the removal of the VOCs by control technologies,  $A_{i,j}$  is the activity level of source j in i city, and  $EF_{i,j}$  is the emission factor of source j in i city.

The classification of emission source is an important basis for the development of emission inventories. In this study, the sources of industrial VOCs emissions were grouped into four types—including the production of VOCs, storage and transport, the

industrial processes using VOCs as raw materials, and the use of VOC products—based on the source tracing method employed in most studies [3,19,34–38].

## 2.3. Data Collection

## 2.3.1. Activity Data

The approach used for the collection of activity data was determined specifically for each industrial emission source. Thus, 74 samples of industrial sources were investigated in 11 cities in Hebei Province. The monthly average information for each city, including fuel consumption and product output, was gathered from the official statistics, such as the Hebei Economic Yearbook [32], the China Statistical Yearbook [30], the China Energy Statistical Yearbook [39], and the Hebei Provincial Bureau of Statistics [40]. For example, the activity data of the storage and transportation of oil were obtained through the Regional Energy Balance Table in the China Statistical Yearbook [30]. Despite this, there were still some activity data that were difficult to acquire due to missing information, including for the textile and dyeing, packaging and printing, wood processing, building decorations, furniture manufacturing, footwear, machinery and equipment manufacturing, and transportation equipment manufacturing industries. In this situation, we collected activity data or product output from national and municipal records to supplement. In addition, some data of industrial sectors were obtained by referring to the literature, websites, and industry reports. The specific types and sources of activity data obtained for Hebei Province are summarized in Table 1.

| Sectors                            | Sectors Sources                              |      |
|------------------------------------|--|------|
|                                    | Crude oil production                         | [32] |
|                                    | Natural gas production                       | [32] |
| Production of VOCa                 | Crude oil processing volume                  | [41] |
| Fibluction of VOCs                 | Methanol production                          | [42] |
|                                    | Benzene production                           | [32] |
|                                    | Output of synthetic ammonia                  | [32] |
| Storage and transport              | Output of petroleum products                 | [39] |
| Storage and transport              | Import and export of oil                     | [39] |
|                                    | Coating production                           | [32] |
|                                    | Ink production                               | [43] |
|                                    | Production of primary form plastic           | [30] |
|                                    | Production of synthetic rubber               | [32] |
|                                    | Output of synthetic fiber                    | [32] |
|                                    | Production of vegetable oil                  | [32] |
|                                    | Yield of finished sugar                      | [32] |
|                                    | Liquor yield                                 | [32] |
| Industrial processes using VOCs as | Beer production                              | [32] |
| raw materials                      | Alcohol production                           | [44] |
|                                    | Production of synthetic detergent            | [32] |
|                                    | Production of chemical medicine raw medicine | [32] |
|                                    | Production of chemical pesticides            | [32] |
|                                    | Tire output                                  | [32] |
|                                    | Cement/lime/gypsum                           | [32] |
|                                    | Flat glass                                   | [32] |
|                                    | Production of sanitary ceramics              | [32] |
|                                    | Steel production                             | [32] |
| The use of VOC products            | Coke production                              | [30] |

Table 1. Source classification and activity level of the major industrial VOC emission inventory.

| Sectors | Sources                                      | Reference |
|---------|--|-----------|
|         | Textile auxiliary consumption                | [30]      |
|         | Dye consumption                              | [30]      |
|         | Consumption of PU slurry                     | [44]      |
|         | Consumption of adhesive                      | [30]      |
|         | Pulp production                              | [44]      |
|         | Paper product output                         | [32]      |
|         | Ink consumption                              | [32,45]   |
|         | Consumption of gasoline detergent            | [32,45]   |
|         | Adhesive consumption                         | [32,45]   |
|         | Adhesive consumption                         | [40]      |
|         | Consumption of wood coatings                 | [40]      |
|         | Paint consumption                            | [32]      |
|         | Assembly adhesive consumption                | [32]      |
|         | Paint consumption                            | [30,46]   |
|         | Adhesive consumption                         | [30,46]   |
|         | Building paint consumption                   | [30,47]   |
|         | Construction adhesive consumption            | [30,46]   |
|         | Landfill amount                              | [48]      |
|         | Amount of waste incineration treatment       | [48]      |
|         | Amount of compost treatment                  | [48]      |
|         | Fossil fuel consumption                      | [32]      |
|         | Heating fuel consumption                     | [39]      |
|         | Industrial and construction fuel consumption | [39]      |
|         | Laundry                                      | [30]      |

# 2.3.2. Emission Factors

As key indicators relating to the quantity (weight) of the pollutants emitted from a unit of activity of the source, emission factors have a direct impact on the estimation of pollutant emissions, as seen in Equation (1). Thus, it is critical to obtain the emission factors with a high accuracy for the preparation of an emission inventory. Considering factors including the representativeness and availability of EFs, localized EFs, and updates of EFs from the latest research, the EFs in this study were mainly obtained from the Technical Guide, the Taiwan Environmental Protection Agency, and other research. Specific details of the emission factors in 2015 for Hebei Province are shown in Table 2.

| Sectors              | Sources                                   | Sources Activity Data                 |        | Unit           | Refer-<br>ence |  |
|----------------------|---|---------------------------------------|--------|----------------|----------------|--|
|                      | Crude oil and natural gas extrac-         | Crude oil exploration                 | 1.5275 | g/kg Crude oil |                |  |
|                      | tion                                      | Natural gas exploration               | 0.5    | g/kg Products  | [33]           |  |
| Droduction of VOCo   | Petroleum refining                        | Crude oil processing volume           | 1.82   | g/kg Products  |                |  |
| Production of VOCs   | Pasia de amigal nove matoriala            | Methanol production                   | 5.55   | g/kg Products  | [40]           |  |
|                      | basic chemical raw materials              | Benzene Production                    | 0.55   | g/kg Products  | [49]           |  |
|                      | manufacturing                             | Synthesis ammonia                     | 4.72   | g/kg Products  | [33]           |  |
|                      | Oil storage                               | Crude oil                             | 0.123  | g/kg Products  |                |  |
| Storage and          | Oli storage                               | Gasoline                              | 0.156  | g/kg Products  | [22]           |  |
| transport            | Oil transportation                        | Crude oil                             | 1.6036 | g/kg Products  | [33]           |  |
|                      | On transportation                         | Gasoline                              | 1.6036 | g/kg Products  |                |  |
|                      | Coating production                        | Paint production                      | 81.4   | g/kg Products  | [22]           |  |
|                      | Ink production                            | Ink production                        | 50     | g/kg Products  | [55]           |  |
|                      |   | Production of primary form plastic    | 5.81   | g/kg Products  | [5]            |  |
|                      |   | Production of synthetic rubber        | 7.17   | g/kg Products  |                |  |
|                      | Production of curthotic materials         | Polyester                             | 0.7    | g/kg Products  |                |  |
|                      | Froduction of synthetic materials         | Nick                                  | 3.3    | g/kg Products  |                |  |
|                      |   | Acrylic                               | 37.1   | g/kg Products  | [33]           |  |
| Industrial processes |   | Other fiber                           | 13.43  | g/kg Products  |                |  |
| using VOCs as raw    |   | Production of vegetable oil           | 2.45   | kg/t           |                |  |
| materials            |   | Yield of finished sugar               | 8      | g/kg sugar     |                |  |
|                      | Food and beverage production              | Liquor yield                          | 16.26  | kg kl-1        | [50]           |  |
|                      |   | Beer production                       | 0.43   | kg kl-1        | [50]           |  |
|                      |   | Alcohol production                    | 32.1   | kg kl-1        | [49]           |  |
|                      | Commodity production                      | Production of synthetic detergent     | 0.025  | kg/t           | [50]           |  |
|                      | Manufacture of chemical drug<br>raw drugs | Production of chemical drug raw drugs | 430    | g/kg Products  | [33]           |  |

# **Table 2.** Emission factors used in this study.

1

|                | Tire manufacturing                | Tire production                               | 0.91  | Kg/a          |      |
|----------------|-----------------------------------|---|-------|---------------|------|
|                | Manufacture of cement, lime and   | Cement/lime/gypsum                            | 0.177 | g/kg Products |      |
|                | Glass and glass product manu-     | Flat glass                                    | 4.4   | g/kg Products |      |
|                | Steelmaking                       | Steel production                              | 0.2   | g/kg Steel    |      |
|                | Cales and dustion                 | Mechanical coking                             | 2.96  | g/kg Coke     | [22] |
|                | Coke production                   | Indigenous coking                             | 5.36  | g/kg Coke     | [33] |
|                | Toutile Drinting and drains       | Textile auxiliary consumption                 | 98    | kg/t          | [40] |
|                | Textue Printing and dyeing        | Dye consumption                               | 81.4  | g/kg dyes     | [49] |
|                | Synthetic Leather Manufacturing   | Consumption of PU slurry                      | 245   | kg/t          | [=0] |
|                | Shoe Making                       | Consumption of adhesive                       | 670   | kg/t          | [50] |
|                | Demographics and manage Droduceto | Pulp production                               | 3.1   | g/kg Pulp     |      |
|                | Papermaking and paper Products    | Paper product output                          | 0.1   | kg/t Products |      |
|                |                                   | Ink consumption(new)                          | 750   | g/kg Ink      | [33] |
|                | Deinting and an density a winting | Ink consumption (traditional)                 | 100   | g/kg Ink      |      |
|                | Printing and packaging printing   | Consumption of gasoline detergent             | 1000  | kg/t          |      |
|                |                                   | Adhesive consumption                          | 1385  | kg/t          | [26] |
|                | Wood processing                   | Adhesive consumption                          | 89    | kg/t          | [50] |
| The use of VOC | Furniture manufacturing           | Consumption of wood Coatings                  | 651   | kg/t          | [51] |
| products       | Mechanical equipment manufactur-  | Paint consumption                             | 0.4   | kg/pieces     | [33] |
|                | ing                               | ing Assembly Adhesive Consumption             |       | kg/t          | [49] |
|                |                                   | Paint consumption (automobile)                | 21.2  | kg/car        |      |
|                |                                   | Paint consumption (motorcycles)               | 1.8   | kg/car        | [22] |
|                | Traffic and transportation equip- | Paint consumption (Sedan)                     | 2.43  | kg/car        | [33] |
|                | ment manufacturing                | Paint consumption (bicycle)                   | 0.3   | kg/car        |      |
|                |                                   | Adhesive consumption                          | 89    | kg/t          | [50] |
|                |                                   | Construction paint Consumption (water-based)  | 120   | g/kg Coating  | [22] |
|                | Architectural decoration          | Construction Paint consumption (solvent type) | 450   | g/kg Coating  | [33] |
|                |                                   | Construction adhesive consumption             | 62    | kg/t          | [50] |
|                |                                   | Landfill Amount                               | 0.23  | g/kg Rubbish  |      |
|                | Waste disposal                    | Amount of waste incineration treatment        | 0.74  | g/kg Rubbish  | [33] |
|                | -                                 | Amount of compost treatment                   | 0.74  | g/kg Rubbish  |      |
|                | Clothes dry cleaning              | Ethylene chloride consumption                 | 1000  | g/kg          |      |

|                              | Fossil fuel (coal)  | 0.15    | g/kg Coal        |      |
|------------------------------|---|---------|------------------|------|
|                              | Thermal power (Fuel oil)                                    | 0.13    | g/kg Fuel oil    |      |
|                              | Thermal power (coal gasification)                           | 0.00044 | g/m³             |      |
|                              | Thermal power (liquefied petroleum gas)                     | 0.034   | g/m³             |      |
|                              | Thermal power (natural gas)                                 | 0.045   | g/m <sup>3</sup> |      |
|                              | Heating fuel (coal)   | 0.18    | g/kg Coal        |      |
|                              | Heating fuel (fuel oil)                                     | 0.2     | g/kg Fuel oil    |      |
| Stationary source combustion | Heating fuel (coal gasification)                            | 0.00044 | g/m³             |      |
|                              | Heating fuel (liquefied petroleum gas)                      | 0.5     | g/m <sup>3</sup> |      |
|                              | Heating fuel (natural gas)                                  | 0.088   | g/m <sup>3</sup> | [33] |
|                              | Industrial and construction fuels (coal)                    | 0.39    | g/kg Coal        |      |
|                              | Industrial and construction fuels (fuel oil)                | 0.35    | g/kg Fuel oil    |      |
|                              | Industrial and construction fuels (coal gasification)       | 0.00044 | g/m³             |      |
|                              | Industrial and construction fuels (liquefied petroleum gas) | 0.48    | g/m <sup>3</sup> |      |
|                              | Industrial and construction fuels (natural gas)             | 0.088   | g/m <sup>3</sup> |      |

## 2.4. Scenario Designed

The future (2020–2030) industrial VOC emissions in Hebei Province were projected in this study, taking 2015 as the reference year. Three scenarios, including a Business As Usual (BAU) scenario, a moderate scenario, and a strict scenario were defined, combining economic development trends, demographic changes, and related control measures and policies in Hebei Province in future years. The trends of GDP, population, and urbanization rate during the period 2015–2030 are shown in Table 3. The population was forecasted by applying the Chinese Population Prediction Software (CPPS), while the urbanization rate was determined based on the growth rate proposed by the 13th Five-Year plan (2016-2020) [52]. Moreover, Chinese policymakers have issued specific objectives and implementation plans for the comprehensive treatment of VOCs emission in recent years, including the Air Pollution Prevention and Control Action Plan (2013–2017) [53], the 13th Five-Year Plan for the Prevention and Control of Volatile Organic Compound Pollution (2017–2020) [54], and the Volatile Organic Compounds Reduction Action Plan for Key Industries (2016-2018) [55]. The specific definitions of the emission scenarios are summarized in Table 4. Some key parameters, such as emission control technologies and removal efficiencies for each industrial sector, are defined in Table 5.

**Table 3.** The predicted GDP, population, and urbanization rate in Hebei Province for the period 2015–2030.

|      | GDP (Billion Yuan) | Population (Million) | Urbanization Rate(%) |
|------|--------------------|----------------------|----------------------|
| 2015 | 2980.6             | 74.24                | 51.33                |
| 2020 | 5280.5             | 76.87                | 54.2                 |
| 2025 | 5863.3             | 80.49                | 57.52                |
| 2030 | 8223.6             | 83.76                | 60.46                |

Table 4. Specific descriptions of the three scenarios designed in this paper.

| Scenarios             | Scenario Description   |
|-----------------------|--|
| <b>DALL</b> according | Based on the 2015 emission level to project future emissions, assuming that the control          |
| DAU Scenario          | technologies maintain unchanged, with no additional measures being implemented                   |
|                       | Key industrial sectors (large-scale enterprises of chemical medicine, coke production,           |
| Moderate<br>scenario  | mechanical equipment manufacturing, organic chemical, packaging and printing, wood               |
|                       | adhesives, industrial and construction dyes, furniture manufacturing, transportation             |
|                       | equipment manufacturing, and crude oil processing) adopt the best available control              |
|                       | technologies and clean production  |
| Strict scenario       | All industrial sectors (100%) adopt the best available control technologies and clean production |

Table 5. Detailed parameters of the control measures of various industrial sources.

| Courses                               |   | <b>Emission Reduction Efficiency</b> |       |  |
|---------------------------------------|---|--------------------------------------|-------|--|
| Sources                               | Control Technologies  | 2020                                 | 2030  |  |
| Chemical medicine                     | Condensation/adsorption/catalytic combustion<br>technology  | 60–70                                | 80–90 |  |
| Coke production                       | Condensation recovery/catalytic combustion/ad-<br>sorption  | 50–65                                | 70-85 |  |
| Mechanical equipment<br>manufacturing | Adsorption and concentration of activated carbon + catalytic combustion/thermal combustion  | 50–65                                | 70–85 |  |
| Organic chemical                      | Condensation/adsorption/catalytic combustion<br>technology/spray absorption + cooling dehumidifi-<br>cation + activated carbon adsorption | 60–70                                | 80–90 |  |

| Packaging and printing                      | Adsorption recovery/catalytic combustion/environ-<br>mentally friendly raw material substitution              | 55–65 | 75–90 |
|---|---|-------|-------|
| Wood adhesives                              | Substitution of environmental protection materi-<br>als/activated carbon adsorption/low temperature<br>plasma | 55    | 75    |
| Industrial and construc-<br>tion dyes       | Adsorption/combustion   | 45-60 | 70–85 |
| Furniture manufactur-<br>ing                | Wheel concentrated combustion/adsorption/envi-<br>ronmentally friendly raw materials                          | 50–65 | 75–85 |
| Transportation equip-<br>ment Manufacturing | Adsorption and concentration of activated carbon + catalytic combustion/thermal combustion                    | 50–65 | 80–90 |
| Crude oil processing                        | Oil and gas recovery system/adsorption concentra-<br>tion + catalytic combustion                              | 55–70 | 75–90 |
| Other sources                               | Adsorption/combustion/biological treatment  | 51–50 | 70–80 |

## 3. Results and Discussion

#### 3.1. Industrial VOC Emission Inventory for 2015

The industrial VOC emissions in Hebei Province in 2015 were estimated by applying the methods described in Section 2, as shown in Table 6. The total industrial VOC emissions were 1017.795 Kt, of which the production of VOCs, the storage and transport, the industrial processes using VOCs as raw materials, and the use of VOC products accounted for 5.04%, 3.58%, 39.26%, and 52.12% of the total, respectively. Referring to the studies of Liang (2017) [56], Ye (2020) [57], and Zhang (2017) [58], the industrial VOC emissions in China were 13,389.4 Kt, 1435.6 Kt, and 10,762.9 Kt, respectively. Compared with these results, it could be concluded that the industrial VOC emissions in Hebei Province accounted for about 7–9% of the total emissions in China.

The industrial VOC emissions of Hebei Province calculated in prior studies from the past few years are summarized and compared in Figure 2. The estimated total industrial VOC emissions in this study were about 22.4–52.1% higher than those of Gong (2013) [4], Yang (2012) [26], and Cheng (2013) [45], and 18.8% lower than the results of Huang (2016) [5]. These differences may have been caused by the following: (1) The selection of activity data: activity data for 81 types of industrial sources were collected by Huang (2016) [5], and this study selected only 74 types, which caused Huang (2016) [5] to show higher emissions. For example, the crude oil processing volume in this study was considered to be activity data of petroleum refining based on technical guidelines, while the tank loss, transport loss, leakage loss, and volatile refining wastewater of Huang (2016) [5] were chosen according to references. (2) The interannual variation in activity levels: The VOC emission inventories of other studies were for periods 3-5 years earlier than that of this study. In fact, VOC emissions have been increasing yearly, owing to the increase in the scale and number of industrial enterprises in Hebei Province. (3) The selection of EFs: For crude oil exploration sources, 1.5275 g/kg crude oil in this study represented the VOC emission factor from crude oil exploration, rather than the 0.6 kg/t found by Yang (2012) [26] and Huang (2016) [5], due to the EF being derived from the latest officially published data, which was highly representative. (4) The determination of key industrial sources: Cheng (2013) [45] estimated the VOC emissions of eight key industries in Hebei Province in 2013, including the chemical medicine, coke production, organic chemical, packaging and printing, wood adhesives, industrial and construction dyes, furniture manufacturing, and crude oil processing industries. The total emissions found by Cheng (2013) [45] were 633.2 kt, while the corresponding emissions of the eight key industries in this study were found to be 677.8 kt. Therefore, the results of this study are reasonable and acceptable.

| Source         | Production<br>of VOCs         | Sto<br>T  | orage and<br>ransport | Industr<br>Using V<br>M | rial 1<br>VOC<br>later | Processes<br>Es as Raw<br>rials | <b>3</b><br>7  | The Use o<br>Produ | f VOC<br>cts       | Total Indus-<br>trial VOCs |
|----------------|-------------------------------|---|-----------------------|-------------------------|------------------------|---------------------------------|----------------|--------------------|--------------------|----------------------------|
| Emissions (kt) | 51.2864                       | ,   | 36.4872               | Э                       | 399.5                  | 582                             |                | 530.43             | 39                 | 1017.795                   |
|                | Industrial VOC emissions (kt) | 1400 -<br>1200 -<br>1000 -<br>800 -<br>600 -<br>400 -<br>200 -<br>0 - | Gong (2013)<br>20     | Yang (201<br>10         | (2)                    | Cheng (201                      | 16) Ht<br>2013 | uang (2016)        | This study<br>2015 |                            |

Table 6. VOC emissions from four types of industrial sources in Hebei Province in 2015.

**Figure 2.** Comparison of industrial VOC emissions in this study with other published results for Hebei province (2010, 2013, and 2015 on the X-axis refer to the years when the VOC emission inventory was established).

#### 3.1.1. Emission Contributions by Source

The contributions of various sources to the total industrial VOC emissions in Hebei Province are illustrated in Figure 3. The use of VOC products was the largest contributor, due to the fact that it involves a wide range of emission sources, including emissions from 14 sectors. The main emission sectors from the use of VOC products were the coke production, mechanical equipment manufacturing coatings, wood adhesives, packing adhesive, and industrial and construction dyes industries, accounting for 38.85%, 17.53%, 8.13%, 7.83%, and 5.52% of the total emissions, respectively. As the second largest VOC contributor, the industrial processes of using VOCs as raw materials has VOC emissions of 399.582 Kt, in which the chemical medicine, coating production, steelmaking, and other industries accounted for 56.09%, 14.65%, 12.64%, and 16.62% of the emissions, respectively. Crude oil and ammonia production accounted for 46.58% and 25.23% of the production of VOCs, respectively. In terms of storage and transport, the lowest emitter sources—other oil, crude oil, and petrol storage and transportation—accounted for 51.88%, 27.98% and 20.09% of all emissions, respectively.



**Figure 3.** Emission contributions of various sources to the total industrial VOC emissions in Hebei Province in 2015 (CNP: crude oil and natural gas extraction; PR: petroleum refining; BCM: basic chemical raw materials manufacturing; CE: chemical medicine; CP: coke production; MEM: mechanical equipment manufacturing; WA: wood adhesives; PA: packaging adhesive; ICD: industrial and construction dyes; WC: wood coatings; TEC: transportation equipment coating).

It could be concluded that there is a large difference in VOC emissions between the contributions from different industrial sectors. Therefore, we summarized the key industrial sources of VOCs emissions in Hebei Province for further analysis, as illustrated in Figure 4. In this study, the phrase "key industrial sources" refers to the top ten sources of industrial VOC emissions in Hebei Province, including the chemical medicine, coke production, mechanical equipment manufacturing, organic chemical, packaging and printing, wood adhesives, industrial and construction dyes, furniture manufacturing, transportation equipment manufacturing, and crude oil processing industries. The emissions from these ten sectors were estimated at 796.8 Kt, accounting for 78% of the total. Thus, it is necessary for national and local government policymakers to focus on strengthening the governance of key industries during the specific formulation of industrial VOC emission reduction policies.

The two largest sources of emissions were the chemical medicine (224.1 kt) and coke production (206.1 kt) industries, accounting for 53% of the total emissions. This may be due to the relatively rapid development of these two sectors in Hebei Province in recent years. The sectors with VOC emissions between 50 and 100 kt included the machinery equipment manufacturing (93.0kt), organic chemicals (67.0kt), and packaging and printing industries (58.2kt), accounting for 9%, 7%, and 6% of the total emissions, respectively.



**Figure 4.** VOC emissions of key industrial sources (CE: chemical medicine; CP: coke production; MEM: mechanical equipment manufacturing; OC: organic chemical; PP: packaging and printing; WA: wood adhesives; ICD: industrial and construction dyes; FM: furniture manufacturing; TEP: transportation equipment manufacturing; COP: crude oil processing).

#### 3.1.2. Spatial Distribution of Industrial VOC Emissions

The spatial distribution of industrial VOC emissions in the Hebei Province for 2015 is shown in Figure 5. Based on the industrial GDP and population density information, the VOC emissions were allocated to each city in Hebei Province using ArcGIS10.2. It can be seen from Figure 5 that the distribution of industrial VOCs in Hebei Province is spatially uneven and generally shows the characteristics of lower emissions in the north and higher emissions in the south. To further examine the characteristics of the spatial distribution of VOC emissions, we calculated the spatial autocorrelation using the global Moran's I index putted forward by Patrick Alfred Pierce Moran [59]. The results showed that Moran's I index is 0.5 > 0 (P < 0.01), which indicates that there is a stronger positive spatial correlation of industry VOC emissions in Hebei Province. That is to say that cities with high (or low) values of VOC emissions often had nearby cities with high (or low) values in Hebei Province, presenting a clear clustering characteristic. The industrial VOC emissions are mainly concentrated on the industrially developed, densely populated cities, such as Handan, Shijiazhuang, and Langfang. In particular, Handan, located in the southern area, was the largest VOC emitter due to its developed coking industry and pharmaceutical enterprises. The distinctive emission characteristics may be attributed to the large differences in geography, industry, economy, and population among different cities.

From the perspective of the spatial distribution of VOC emissions from key industries, the emissions of the chemical medicine sector are concentrated in Shijiazhuang, Baoding, Xingtai, Handan, and Tangshan, owing to the fact that most of the pharmaceutical companies are located in this area. In addition, more than 90% of the coking enterprises in Hebei Province are distributed in Tangshan, Handan, and Xingtai, which has caused the coke production in these cities to emit a large amount of VOCs. Similarly, Machinery and equipment manufacturers are mainly distributed at Cangzhou, Baoding, Tangshan, and Shijiazhuang. Moreover, there are more than 330 organic chemical enterprises in Hebei Province, located in Baoding (26%), Shijiazhuang (18%), Hengshui (14%), and Cangzhou (13%). For packaging and printing enterprises, the distribution is largely concentrated in the Shijiazhuang, Langfang, Baoding, and Tangshan areas. However, there is still a considerable number of small and micro factories that are widely scattered across the province, making it difficult to carry out environmental supervision. Therefore, more attention should be paid to the management of industrial enterprises in central and southern Hebei Province, strengthening technological innovation to increase production capacity, and rectifying small- and medium-sized backward production capacity enterprises.



**Figure 5.** Spatial distribution of VOC emissions from industrial sources and key industries in Hebei Province for 2015.

## 3.2. Scenario Prediction of Industrial VOC Emissions in 2020 and 2030

The trends of industrial VOC emissions and their sectoral distributions in the period 2015–2030 under different scenarios are given in Figure 6. There is an evident difference in emission trends across the three scenarios. Under the BAU scenario, the predicted total VOC emissions in Hebei Province would continue to increase with an average annual growth rate of 10.24%. The predicted total industrial VOC emissions in 2020 and 2030 under the BAU scenario are 1448.95 Kt and 2203.66 Kt, 53.36% and 116.51% higher than those in 2015, respectively. The use of VOC products, one of the most major sources of emissions, contributed greatly to this increasing trend. Moreover, the industrial VOCs emissions gradually increased during the period 2020-2030 under the moderate and strict scenarios, with a relatively lower growth rate than that in the BAU scenario. However, the total VOC emissions could be reduced by as much as 34.15% (494.78 kt) and 47.77% (1052.72 kt) by 2020 and 2030 compared with the BAU scenario, owing to the VOC emission reductions from key industrial sectors. Similarly, for the strict scenario, a greater reduction in total VOCs emissions would be achieved, with a fall of 54.45% (788.99 kt) and 69.89% (1551.29 kt) in 2020 and 2030 relative to the BAU scenario, attributed to the realization of maximum abatement potential from all industrial sectors (100%) adopting the best available control technologies and clean production.

Figure 6 also shows the reduction in VOC emissions from different industrial sectors under the different scenarios. A significant emission abatement would be achieved with the use of VOC products for all scenarios, with the exception of the strict scenario in 2030, where the emissions from industrial processes using VOCs as raw materials were greater. These two large sources collectively accounted for 87.63–89.63% and 89.73–91.97% of the total under the moderate and strict scenarios in 2020 and 2030, respectively.

Figure 7 shows the emission reductions in different cities in Hebei Province under different scenarios. Due to the unbalanced regional economic developments existing in Hebei Province, the VOC emissions projected among the 11 cities showed large differences during the period 2015–2030. These projections revealed that Shijiazhuang and Tangshan were and will continue to be the largest contributors under the BAU scenario. These two cities located in the central and southern areas are developed regions with high population densities and high GDPs. On the contrary, Zhangjiakou, Chengde, and Qinhuangdao, located in the northern area with sparse populations and slow economic growth, have relatively low emissions and growth trends. These results prove that VOC emissions will be out of control if VOC emissions are not governed immediately. A significant decrease occurred in the moderate and strict scenarios. Moreover, relatively high values of reduction were projected in cities that are major contributors to total emissions, and relatively low values were projected in cities with the more minor contributions. Therefore, it can be concluded that the measures defined in both scenarios appear to be effective in reducing industrial VOC emissions.

We tried to compare our results with the actual value in 2020 to ensure the rationality of the prediction results in this study. However, due to the fact that official statistical data are generally published one to two years later in China, we failed to find the actual results for 2020. Nevertheless, we found the latest results of the MEIC (Multi-resolution Emission Inventory of China) [60] developed by Tsinghua University, which is currently recognized as academically credible data in China. The results show that the industrial VOC emissions found for Hebei Province in 2017 in this study (974 Kt) were about 2% lower than those of MEIC (996 Kt). Therefore, it could be concluded that our prediction is reasonable. Moreover, we looked up the latest national policies on VOC control to analyze the rationality of the defined scenario in this study. The Ministry of Ecology and Environment (MEE) issued the Comprehensive Treatment Plan for Volatile Organic Compounds in Key Industries [61] in July 2019, which clearly specified that the petrochemical, chemical, industrial painting, packaging and printing, oil storage, and marketing industries are the key VOC emission sources in China. Additionally, Hebei province is defined one of the key regions of industrial VOC control in China. The present policies demonstrated that the moderate scenario in this study is more realistic.



■ Industrial processes using VOCs as raw materials ■ The use of VOC products



#### Figure 6. Prediction of industrial VOC emissions under the different scenarios in 2020 and 2030.

Figure 7. Spatial distribution of VOC emissions from industrial sources in Hebei Province for 2020 and 2030.

## 3.3. Uncertainty Analysis

Uncertainties are unavoidable during the estimation of pollutant emissions in different scenarios generated from both activity data and emission factors. Moreover, some factors, including the degree of VOC removal by existing pollution control technologies [3], also affected the accuracy of the results in this study. The uncertainties of industrial VOC emissions in Hebei Province in 2015 were quantified by the Monte Carlo simulation method, assuming that both the activity data and emission factors obeyed lognormal distributions [51,62]. In addition, before applying the Monte Carlo simulation, the coefficients of variation (CV, the standard deviation divided by the mean) of the activity data and emission factors were determined according to the different sources of data [63]. With regard to the data from the Statistical Yearbook, the uncertainty was set as  $\pm$  30% [64], which means that these statistical data have relatively less errors than the emission factors and are very reliable for emission estimation at the national and provincial levels [19]. For the data from association statistics obtained by assigning the coefficient, the uncertainty was set as  $\pm 80\%$  or  $\pm 100\%$  [64,65]. Based on the assumption above, the estimated range of uncertainty of VOC emissions for each sector with a 95% confidence interval is shown Table 7. It was found that the biggest uncertainty ranges for the use of VOC products were between -63% and 90%, while those for storage and transport, industrial processes using VOCs as raw materials, and the production of VOCs were -57%–85%, -39%–67%, and - 38%–59%, respectively. Although the predicted results might deviate from the real situation, the values were within a reasonable range.

Table 7. The estimated uncertainty range of industrial VOC emissions for each sector in 2015.

| Sectors                                    | Uncertainty (95% Confidence Interval) |
|--|---------------------------------------|
| Production of VOCs                         | [-38%, +59%]                          |
| Storage and transport                      | [-57%, +85%]                          |
| Industrial processes using VOCs as raw ma- | [_200/+679/]                          |
| terials                                    | [-39 %, +07 %]                        |
| The use of VOC products                    | [-63%, +90%]                          |

#### 4. Conclusions

This study attempted to predict the emissions and mitigation potentials of industrial VOCs under different scenarios based on the industrial VOC emission inventory for Hebei Province established in 2015. We intended to provide optimized strategies for Hebei's VOC emission abatement policy in future years.

The total industrial VOC emissions were estimated to be 1017.795 Kt, with approximately 52.12% and 39.26% originating from the use of VOC products and the industrial processes using VOCs as raw materials, with relatively low proportions of 5.04% and 3.58% caused by the production of VOCs and storage and transport, respectively. The ten largest contributors of VOC emissions in Hebei Province were the chemical medicine, coke production, mechanical equipment manufacturing, organic chemical, packaging and printing, wood adhesives, industrial and construction dyes, furniture manufacturing, transportation equipment manufacturing, and crude oil processing industries, which emitted 796.8 kt VOCs collectively, accounting for 78% of the total. Therefore, these ten sectors should be regarded as the focus for reducing VOC emissions. Moreover, the industrial VOC emissions in Hebei Province were mainly concentrated in the south-central region, with its well-developed industry and large population density, especially Shijiazhuang and Tangshan.

Under the BAU scenario, the total VOC emissions in 2020 and 2030 would increase by 53.36% and 116.51% compared to 2015, respectively. The use of VOC products would continue to dominate the VOC emissions in the future, but the proportion would decrease from 52% in 2015 to 49% in 2030. The share of the second-largest VOC emission source, the industrial processes using VOCs as raw materials, would increase from 39% in 2015 to 52% in 2030.

Compared with the BAU scenario, the total emission reduction from VOCs during the period 2020–2030 was estimated as 494.78–1052.72 kt and 788.99–1551.29 kt under the moderate and strict scenarios, with reduction ratios of 34.15–47.77% and 54.45–69.89%, respectively. Overall, the use of VOC products had the biggest mitigation potential of 78% under the strict scenario in 2030, while other industrial sources would be reduced by 59%–76%. These three scenarios pointed out the reasonable range of future VOC emissions and provided the maximum potential of industrial VOC emissions, which could provide scientific support and significant information to policymakers attempting to establish a complete VOC management system.

According to the above results, it is recommended that measures to control VOC pollution from key industrial sources should be adopted, as they could bring about a considerable reduction. Generally, there are three ways to reduce VOC emissions. The abatement of VOC emissions from industrial sources and processes has always been the most critical aspect of controlling emissions. The strict control of VOC emissions from storage, loading, and unloading processes, including the application of pressure tanks and floating roof tanks instead of fixed roof tanks, is necessary. Implementing exhaust gas collection measures and improving the collection efficiency should be considered to reduce the disorganized and fugitive emissions of exhaust gases. In addition, end-of-pipe governance measures should also be encouraged. It is appropriate for heavy sources of pollution to prioritize the use of condensation and adsorption recovery technologies for recycling. Furthermore, it is necessary to enhance the effectiveness of VOC governance through technological upgrading and innovation. Finally, an integrated VOC management and technology system should be determined specifically according to the emission features of different industrial sectors.

**Author Contributions:** Conceptualization, X.G. and Y.S.; methodology, X.G.; validation, D.C.; formal analysis, Y.S. and W.L.; investigation, W.L.; data curation, W.L. and J.L.; writing—original draft preparation, Y.S.; writing—review and editing, X.G. and D.C. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was funded by the National Key R&D Program of China (No. 2018YFC0213202).

**Acknowledgments:** This paper represents the perspectives of the authors and does not necessarily represent the official views of our sponsors. We would like to thank the anonymous reviewers for their valuable comments and suggestions to improve the manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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