

Article

The Impact of Anthropogenic VOC Emissions on Atmospheric Pollution: A Case Study of a Typical Industrialized Area in China

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Abstract: Volatile organic compounds (VOCs) are the main precursors of pollution from ground ozone (O₃) and PM_{2.5}, which cause the deterioration of urban air quality. The emissions of VOCs from industrialized areas are significant and their characteristics are complex, which nowadays contribute significantly to the challenges of investigating the emission inventory. Taking a typical industrialized area in Tianjin as a case study, the anthropogenic VOCs emission inventory for 2020 was established in this study by using the activity data from a large-scale survey and the latest emission factors. The impact of VOCs on the environment was analyzed from the perspective of the combined control of PM_{2.5} and O₃. The results showed that the total emission of VOCs in 2020 was about 1.68 Gg, mainly from industrial processes and mobile sources, which accounted for 38.4% and 36.5% of the total emissions, respectively. The top 10 emitted VOCs were toluene, acetone, ethylbenzene, m/p-xylene, i-pentane, n-hexane, formaldehyde, benzene, ethyl acetate and ethylene. The dominant species of O₃ formation potential (OFP) were almost all aromatic hydrocarbons and alkenes, with m/p-xylene contributing the most to the OFP emissions (8.90%). The top 10 secondary organic aerosols formation potential (SOAP) emission species were aromatic hydrocarbons and long-chain alkanes, and the largest emission came from toluene (39.9%). An analysis of an ADMS diffusion model showed that VOCs emitted from traffic-heavy main roads and industrialized central areas had the greatest impact on the air quality in the surrounding areas. The VOCs concentration was higher in winter due to unfavorable meteorological conditions. Our research updated the VOC inventory of industrialized areas and evaluated VOCs species reactivity and their impact on ambient air quality.

Keywords: VOCs; emission inventory; OFP; SOAP; ADMS



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1. Introduction

In recent years, with the implementation of a series of air pollution control measures, the quality of air in China has significantly improved. Nowadays, PM_{2.5} and ozone (O₃) on the ground have become the main pollutants that restrict the further improvement in air quality and hence have attracted significant attention [1–3]. Studies have shown that the extremely high concentration of PM_{2.5} in China during the high-pollution period was mainly caused by the formation of secondary organic aerosols (SOAs) [4,5]. As is well

known, volatile organic compounds (VOCs) are important precursors for the formation of SOAs and ground O_3 [6,7]. Each species of VOCs reacts at a different rate, through a different reaction mechanism and has a different formation potential, resulting in their varying impacts on the formation of ground O_3 and SOAs in the atmosphere [8,9]. Compared to the relatively low-reactivity VOCs, highly reactive VOCs contribute more to the formation of O_3 and SOAs [10]. VOC emission sources are varied and complex, but in urban areas they mainly comprise anthropogenic emission sources, including industrial production activities, fuel combustion, exhaust emissions from vehicles and solvent utilization [11,12].

Limited by the fact that online monitoring of VOCs has not yet been widely implemented in China, the understanding of VOC emission status has been mainly achieved through establishing emission inventories and fixed-point observations. Establishing an accurate and complete emission inventory cannot only quantify important emission sources, but can also serve as a key input for air quality modeling. Most inventory studies have been conducted at the national, regional and urban levels. For example, Wu et al. [13] established an inventory of anthropogenic VOC emissions in China from 2008 to 2012. Liu et al. [14] and Zheng et al. [15] analyzed the emissions of VOCs from different sources in the Yangtze River Delta and the Pearl River Delta, respectively, and their impacts on air quality. Hua et al. [16] and Zhou et al. [17] established high-resolution inventories of VOC emissions at a city level. However, industrial enterprises in cities are often concentrated in selected districts, and the emission characteristics from these industrialized areas are not only more complex than those from other areas, but also have clear regional differences [18,19]. The existing large-scale emission inventories often cannot meet the requirements of more detailed studies; therefore, in-depth direct studies of industrialized areas are very important for evaluating the regional atmospheric conditions and forecasting and managing local ambient air quality.

Diffusion model simulations are an important method for air pollutant concentration assessment and air quality management, because the concentration of pollutants depends not only on the emissions from different sources but also on diffusion conditions, distance from sources, topography and atmospheric chemistry. Atmospheric pollution diffusion models integrate pollutant emission information and surrounding environment and meteorological data to analyze the transmission and diffusion characteristics of pollutants in the atmosphere. They can be employed to reliably reproduce the transmission and diffusion of pollutants in real urban environments [20]. For instance, Zhang et al. [21] used the WRF model to simulate the NO_x emissions in eastern China during the COVID-19 epidemic, Lonati et al. [22] used the Calpuff model to evaluate the spatial distribution of pollutants, such as NO_x , SO_x and VOCs, generated by activities in a Mediterranean port, while Dèdelé et al. [23] established a seasonal model of PM_{10} concentrations in typical cities in Lithuania based on the ADMS model.

In this study, using the activity-level data obtained from multiple sources, an inventory of anthropogenic VOC emissions in a typical industrialized area in Tianjin (TIA) was established, the key industries and species of VOC emissions were determined, and the potentials of different VOC species for the formation of O_3 and SOAs were evaluated, so as to formulate more effective VOC emission reduction strategies based on species reactivity. This study improved the accuracy of VOC emission estimation and provided a reference example for establishing VOC emission inventories for small-scale, localized areas. The air quality ADMS model was used for a detailed analysis of the impact characteristics of VOC emissions on the surrounding air, which will provide scientific evidence for air pollutant emission reduction measures.

2. Materials and Methods

2.1. Study Area

TIA (Figure S1) is located at the intersection of China's Bohai Economic Belt and Beijing-Tianjin-Hebei (BTH) Metropolitan Area, occupying the most important position on the BTH industrial axis. The TIA is industrialized and large amounts of VOCs are emitted

by major industrial activities, such as power generation, solvent utilization, chemical processing and rubber manufacturing. Furthermore, TIA is connected to the national highway network via more than 10 trunk roads, thus mobile sources have also become one of the main VOC emission concerns in the region [24]. It is therefore urgent to establish a regional emission inventory to understand the emissions from various sources, the emission characteristics of typical industries and spatial distributions of VOC concentrations.

2.2. Building of Emission Inventory

2.2.1. Source Classification

Considering the previous research results and the actual local conditions, this study has developed a three-level source classification (Table S1), which is consistent with the Technical Manual for Compiling Urban Air Pollutant Emission Inventory [25]. There were six classes at the first level, including fossil fuel fixed combustion sources, industrial process sources, solvent utilization, mobile sources, fuel oil storage and transportation sources and waste disposal sources. The first-level sources were divided into 22 sub-sources at the second level according to their characteristics, and then further divided into the third level according to product types, raw materials, fuel types or production technologies. The data of anthropogenic VOC emission activities used in this study came from the environmental statistical databases of the government departments of TIA and field questionnaires.

2.2.2. Emission Estimation

In this study, the emission factor method was used to estimate VOC emissions. The emission factors were selected from the Technical Guide for Compiling Emission Inventory of Road Motor Vehicles [26] and Technical Manual for Compiling Urban Air Pollutant Emission Inventory [25].

The vehicle exhaust emissions were calculated as follows:

$$E_{\text{ex}} = \sum_{i,j} E_{i,j} \times VKT_{i,j} \times VP_{i,j} \quad (1)$$

where $E_{i,j}$, $VKT_{i,j}$ and $VP_{i,j}$ represent the exhaust emission factor, driving mileage and vehicle ownership of the i -th vehicle type (minibuses, buses, light trucks, heavy trucks, and motorcycles) and the j -th emission standard, respectively. This approach also provides a detailed method for the localization of emission factors, considering the influence of geography and meteorology, driving conditions and oil quality on vehicle emissions. The detailed vehicle data used in this study came from [27].

The emissions from sources excluding mobile sources were estimated by multiplying the corresponding activity data with emission factors as follows:

$$E_t = \sum_{x,y,k} A_{x,y,k} \times EF_{x,y,k} \times (1 - \eta_k) \quad (2)$$

where E_t are the emissions from anthropogenic sources except mobile sources, $A_{x,y,k}$ and $EF_{x,y,k}$ represent the activity data and emission factor for the k -th technology source, respectively, and η_k is the removal efficiency while using the k -th technology.

2.3. Calculation of Individual VOC Species Emissions

The emissions of individual VOC species were obtained by multiplying the total VOC emissions by the corresponding species percentage weight as follows:

$$E_p = \sum_q E_q \times f_{p,q} \quad (3)$$

where E_p is the emission of the p -th species, E_q is the emission of the q -th sub-source, and $f_{p,q}$ is the emission percentage weight of the p -th species from the q -th sub-source. The percentage weights of VOCs were taken from [28].

2.4. OFP and SOAP Estimation

The O₃ production potentials were used to quantify the ability of different VOC species to produce O₃. Alkenes, aromatic hydrocarbons, long-chain alkanes and other substances react with hydroxyl radicals quickly, can easily participate in photochemical reactions and promote the generation of O₃. To further characterize the contribution of VOCs to O₃ generation, the most commonly used index to study the OFP of VOCs is the maximum incremental reactivity (MIR) [29,30]. The OFPs were calculated as follows:

$$\text{OFP}_p = \sum_q E_{q,p} \times \text{MIR}_p \quad (4)$$

where OFP_p is the OFP of the p-th species, E_{q,p} is the emissions of the p-th species from the q-th subsector, and MIR_p is the maximum incremental reactivity of the p-th species, which was obtained from [31].

The propensity of individual VOC compounds to form SOAs differs significantly. Previous studies demonstrated that aromatic hydrocarbons and long-chain alkanes are more likely to produce SOAs than other species [32,33]. The SOAs formation potentials (SOAPs) were estimated by using the productivity of different VOC species as follows:

$$\text{SOAP}_p = \sum_q E_{q,p} \times Y_{\text{SOA},p} \quad (5)$$

where SOAP_p is the SOAP of the p-th species, E_{q,p} is the emissions of the p-th species from the q-th sub-sector q, Y_{SOA,p} is the corresponding SOAs yield of the p-th species, provided in [34]. The detailed MIR_p and Y_{SOA,p} values are listed in Tables S2 and S3.

2.5. Data Source and Parameter Setting of ADMS Model

The ADMS model is a Gaussian model which considers the influence of street canyons on diffusion, turbulence and traffic-induced mixing. The model can describe in detail the pollution sources from the street scale to the city scale [35]. The ADMS model has been widely used to simulate the diffusion of VOCs and other air pollutants in airports, ports and cities [36–38]. The simulation area in this study included TIA and its surrounding areas. The simulation period extended from 0:00 on 1 January 2020, to 0:00 on 1 January 2021. Meteorological data came mainly from the observation stations in TIA area, while the remaining necessary data were obtained from the European Meteorological Center (cds.climate.copernicus.eu, accessed on 10 July 2023). The WGS84/UTM region 50N projection coordinate system was used in the simulations and the adopted grid resolution was 100 m × 100 m. The input emission data were obtained from the VOC pollution source emissions of TIA and the grid file of subsequent simulation was further analyzed by ArcGIS.

3. Results and Discussions

3.1. Emission Inventory

3.1.1. Source Contributions

The total anthropogenic VOC emission in TIA in 2020 was 1.68 Gg. As shown in Figure 1, industrial process sources (contributing 38.4%), mobile sources (contributing 36.5%) and solvent use sources (contributing 12.9%) were the top three major emission sources. Among the 22 sub-sources, light-duty gasoline vehicles accounted for the largest proportion of VOC emissions (27.3%), followed by raw chemicals and chemical product manufacturing (16.6%), rubber and plastic production (11.0%), surface coating industry (9.51%) and power generation and supply (6.14%).

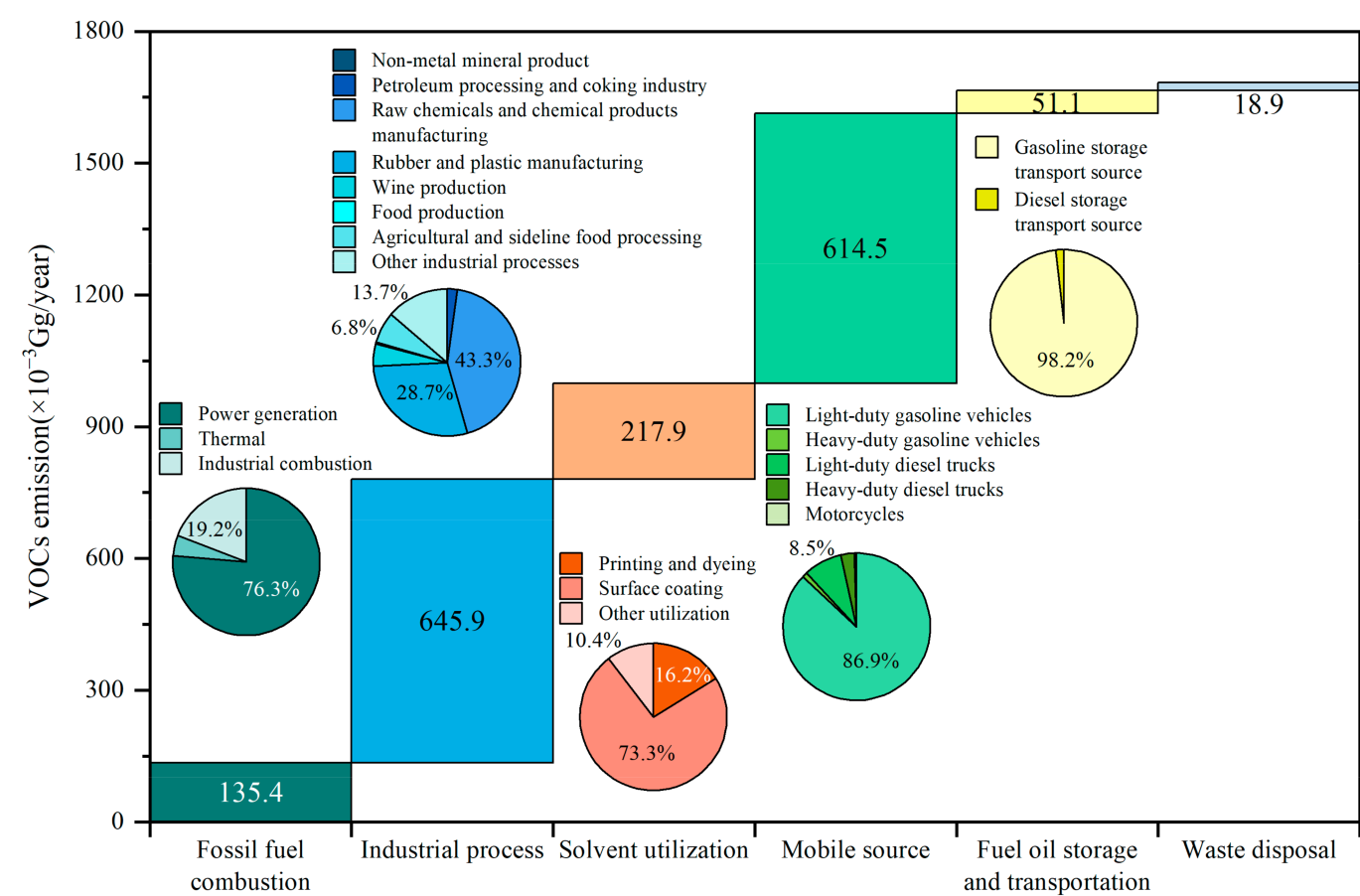


Figure 1. Anthropogenic VOC emissions contributions broken by sub-sources.

A survey of the anthropogenic VOC emission inventories for small-scale regions was conducted. Table 1 shows a comparison of VOC emission intensities and source contributions between TIA and other cities in China. Waste disposal was not included in the comparison.

Table 1. Comparison of VOC emission intensities and source contributions between TIA and other cities in China.

Study Area	Emissions Intensity ($\times 10^{-3}$ Gg/km ²)	Emission Sources Contribution Ratios					Ref.
		Fossil Fuel Combustion	Industrial Process	Solvent Utilization	Mobile Source	Storage and Transportation	
TIA	51.0	8.04%	38.4%	12.9%	36.5%	3.04%	This study
Zhengzhou	12.7	4.30%	24.2%	28.1%	29.7%	7.40%	[39]
Qingdao	13.4	4.10%	49.9%	14.1%	20.1%	/	[17]
Jinan	10.2	7.13%	47.8%	16.8%	17.7%	1.00%	[40]
Chengdu	27.2	1.90%	30.0%	25.9%	36.7%	1.10%	[41]
Huai'an	3.58	6.00%	41.0%	32.0%	13.0%	3.00%	[42]
Yangquan	2.04	31.0%	31.1%	9.50%	23.8%	/	[43].

The contributions of emission sources exhibited obvious regional differences. The research on the inventory of anthropogenic VOC emission in TIA in 2020 showed that the main industries were chemical product manufacturing and rubber and plastic manufacturing. Surface solvent use and industrial solvent use were the dominant industrial activities in Chengdu and Zhengzhou, respectively, with their solvent use sources accounting for a relatively high proportion of emissions, while cities such as Jinan and Qingdao had chemi-

cal product manufacturing as the main emission sources. Only Yangquan, as a coal-based economy, had a high proportion of fossil fuel contribution. Generally, the main sources of VOC emissions in most of the analyzed regions were common and included industrial process sources, solvent utilization and mobile sources as the three major sources, accounting for more than 80% of the total emissions. In terms of VOC emission intensity, in TIA it was 51.0×10^{-3} Gg/km² while in the Zhengzhou high-tech zone it was 46.1×10^{-3} Gg/km², respectively [44]. The emission intensities of these industrialized areas were much higher than those of the cities ($2.04\text{--}27.2 \times 10^{-3}$ Gg/km²). Furthermore, the emission intensities of VOCs in developed cities such as Chengdu and Qingdao were higher than those in ordinary cities such as Yangquan and Huai'an, which confirmed from another angle that the emission problem in industrialized cities and regions was significant.

Among the 430 VOC-emitting enterprises in TIA, the top 10 accounted for 42.5% of the total emissions in the region, while the top 20 enterprises accounted for 57.6%, indicating that a small number of large industrial enterprises contributed a substantial proportion of the VOC emissions. Due to the large number of industrial enterprises in the area and significant electricity consumption, the production and supply of electricity played a leading role in VOC emissions from fossil fuel combustion (76.3%). Among the industrial process sources, raw materials, chemical product manufacturing and rubber and plastic production were the main contributors, accounting for 43.3% and 28.7%, respectively, which was related to the large number of enterprises and high emission factors. The contribution of surface coating industry emission accounted for 73.3% of the total emission from solvent utilization, probably related to the huge number of vehicle manufacturing industries in this area, in which vehicle surface spraying was an important process.

Gas stations form the most important part of the storage and transportation sources in TIA and oil and gas volatilization are important VOC sources. Because of its high volatility, gasoline has become the number one VOC source, accounting for more than 98% of the total emissions. Among the mobile exhaust sources, light-duty gasoline vehicles were the primary VOC source, due to the large ownership and activity levels of these types of vehicles and the volatility of gasoline.

3.1.2. Emission Characteristics of Individual VOCs

This study calculated the emissions of 177 types of VOCs (alkanes, alkenes, alkynes, aromatic hydrocarbons, halogenated hydrocarbons, OVOCs and others) from the anthropogenic VOC sources in TIA. Aromatic hydrocarbons, alkanes and OVOCs were the main contributors to the anthropogenic VOCs, with annual emissions of 516.7, 488.5 and 392.7×10^{-3} Gg, respectively, i.e., accounting for 30.7%, 29.0% and 23.3% of the total VOC emissions, followed by alkenes (7.53%), other VOCs (5.48%) and halogenated hydrocarbons (3.52%), with alkynes accounting for the least mass fraction (0.44%).

Figure S2 shows the chemical proportion of VOCs in emission sources. OVOCs (39.0%), halogenated hydrocarbons (19.8%) and alkanes (16.3%) were the main compounds from fossil fuel combustion sources, while ethyl acetate and acetone in OVOCs were the main types. Aromatic hydrocarbons (32.2%) were the main components emitted from industrial process sources, followed by alkanes (28.2%) and OVOCs (22.7%). Ethylbenzene, n-hexane, styrene, acetone and m-xylene were the largest emission species, among which aromatic hydrocarbons were related to the production of rubber and plastic products and chemical products, whereas n-hexane mainly came from agriculture and sideline food processing. The VOCs produced by solvent sources were mainly aromatic hydrocarbons (51.8%) and OVOCs (30.8%), m-xylene, toluene, ethylbenzene and 1,2,4-trimethylbenzene. Among VOCs emitted from mobile sources, alkanes, aromatic hydrocarbons and OVOCs accounted for 36.9%, 24.7% and 20.2%, respectively, whereas formaldehyde, isopentane, toluene and ethylene were the main emitted species, especially formaldehyde, which accounted for 8.60%. Alkanes (52.6%) and aromatic hydrocarbons (46.2%) accounted for a large proportion in storage and transportation sources, among which isopentane and m-ethyltoluene were the most abundant species. The proportion of alkane emissions from

waste treatment sources reached 72.8%, and the characteristic species were isopentane, n-butane and n-pentane.

Figure 2 lists the top 30 VOC species with the highest emissions, among which the top 10 species were toluene, acetone, ethylbenzene, m/p-xylene, i-pentane, n-hexane, formaldehyde, benzene, ethyl acetate and ethene, with annual emissions of 95.4, 72.4, 67.2, 64.8, 60.2, 59.9, 53.0, 46.9, 44.6 and 40.9×10^{-3} Gg, respectively. Among the 10 species, four belonged to aromatic hydrocarbons, three to OVOCs, two to alkanes and one to alkenes, respectively.

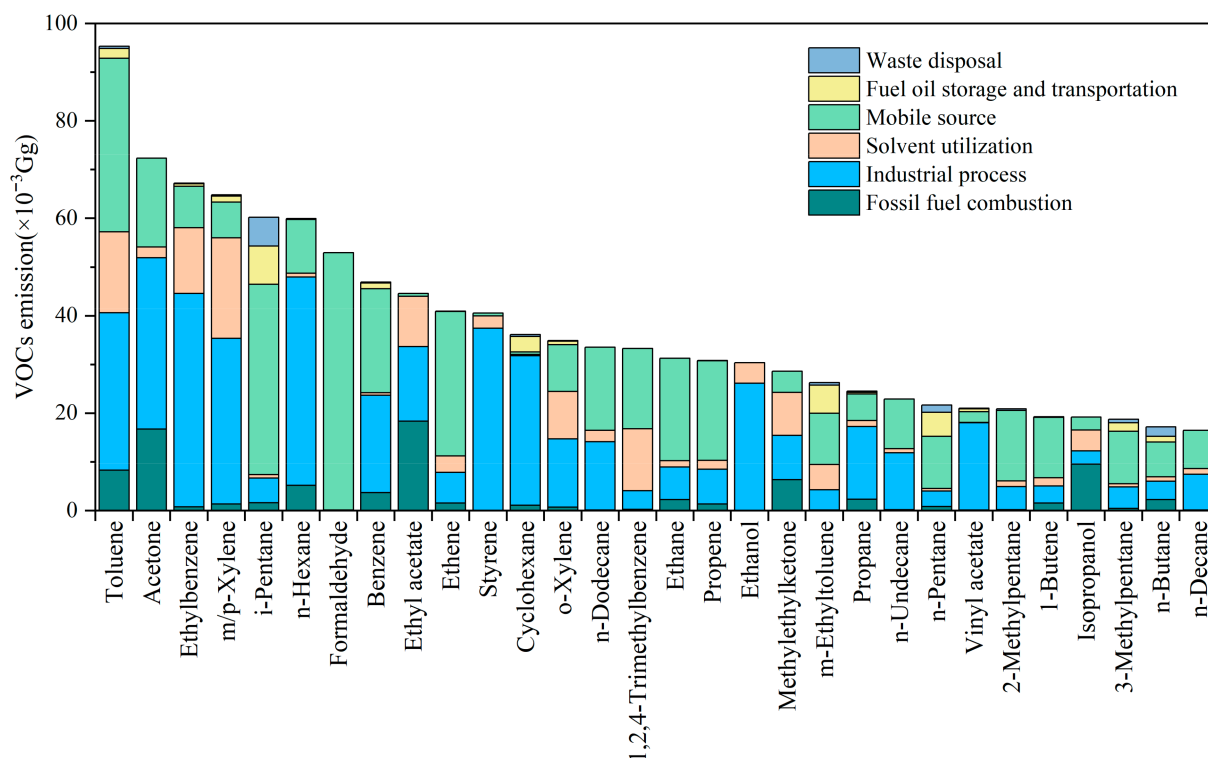


Figure 2. Thirty (30) highest emission VOC species.

Toluene mainly came from mobile sources (37.4%), industrial processes (33.9%) and solvent use (17.4%). The p-ethylbenzene and m-xylene emission from the processes were dominant, accounting for 65.1% and 52.5%, respectively. N-hexane mostly originated from industrial processes, which was mainly related to the high emissions from the agricultural and sideline food processing industries. Acetone emissions mainly come from industrial processes (48.5%) and combustion sources (23.2%). Similarly, the emissions of ethyl acetate were from combustion sources. However, the source distribution of formaldehyde was very different from the other species investigated, as it almost entirely came from mobile sources. This was consistent with previous reports that carbonyl accounted for a large part of VOCs emitted by biomass combustion and vehicles, especially heavy diesel vehicles [45]. Isopentane emissions mainly came from mobile sources (64.9%) and storage and transportation sources (13.0%). In the final analysis, vehicle exhaust and industrial processes were the two sources that contributed the most of the top 10 species, while combustion sources and mobile sources dominated the OVOC emissions.

3.2. Species Activity in VOCs

Based on the VOC species emissions obtained from the composition spectrum of pollution sources combined with the source emission inventory, the OFPs of 140 species and SOAPs of 48 species were quantitatively estimated using their reactive active factors (MIR and Y_{SOA}), the contribution characteristics of each species and pollution sources to OFP and SOAP were analyzed, and the regional O_3 and SOAs pollution potentials were

evaluated. The total OFP of the VOCs emitted by anthropogenic sources in TIA in 2020 was 5.09 Gg. However, using the aerosol generation coefficient method to estimate the SOA generation potential often leads to underestimations [46,47]. Therefore, this study mainly used the relative level of SOA generation potentials to evaluate the impact of various anthropogenic sources on SOAs.

3.2.1. Main Active Species

In terms of individual specie (Figure 3), m/p-xylene had the largest OFP, accounting for 8.90% (453.1×10^{-3} Gg O₃) of the OFP in TIA. Toluene, formaldehyde, ethylene, propylene and 1,2,4-trimethylbenzene were also important species contributing to the formation of O₃, accounting for 7.53%, 7.45%, 6.95%, 6.54% and 5.16% of the total OFP, respectively. In addition, o-xylene, ethylbenzene, 1-butene and m-ethyltoluene also had high levels of potential to form OFP. The OFPs of the 10 major species accounted for 58.6% of total O₃ emissions, while VOC emissions only accounted for about 27.7% of total emissions. All of these compounds except formaldehyde were aromatic hydrocarbons and alkenes, indicating that aromatic hydrocarbons and alkenes were the main O₃ precursors, which was consistent with the composition of OFP of chemical groups. The OFP of aromatic hydrocarbons accounted for 44.9% of the total, while that of alkenes accounted for 22.6%. It should be noted that the significant difference between emissions-based and OFP-based contributions was mainly due to the different tendencies of individual species to form OFP. It can be seen that the importance of species with large emissions to OFP was not necessarily the same. For example, acetone was the second largest single species (accounting for 4.30% of the total VOC emissions), but its contribution to OFP was only 0.49%, ranking 37th among all investigated species. In contrast, propylene accounted for 1.83% of the total VOC emissions, ranking 17th in terms of all emissions, while its potential for O₃ formation ranked fifth, accounting for 6.54% of the total OFP.

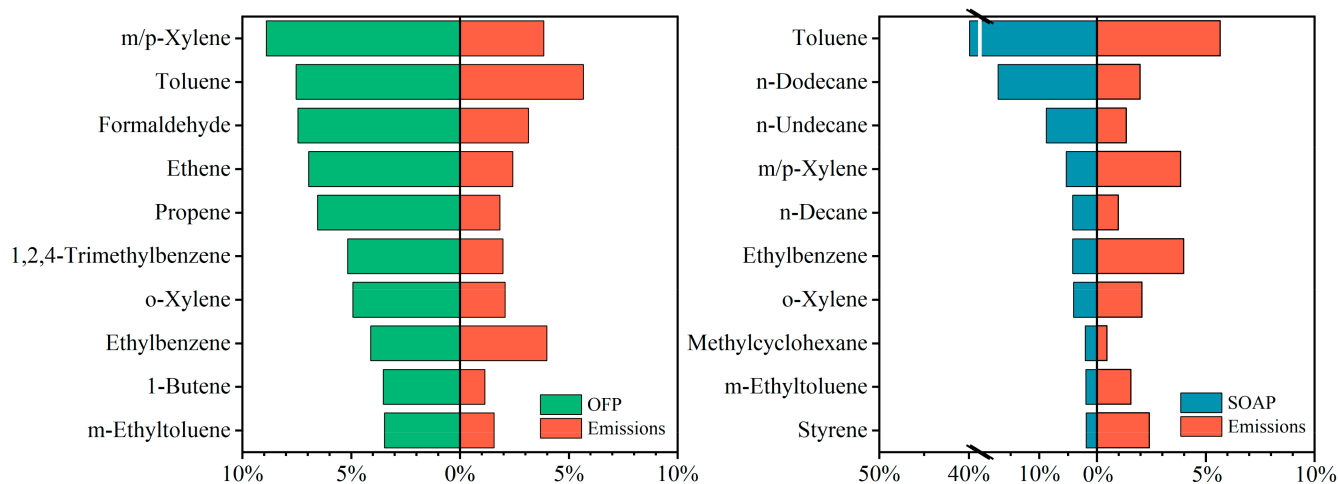


Figure 3. Proportions and emissions of top 10 species contributing to OFP and SOAP.

Aromatic hydrocarbons contributed the largest proportion to SOAP, accounting for 61.0%, followed by alkanes, accounting for 35.7%. Alkenes and OVOCs had relatively low contributions to SOAP, accounting for 1.30% and 0.12%, respectively. The top 10 species in terms of SOAP emissions were all aromatic hydrocarbons and long-chain alkanes, with toluene being the largest contributor (39.9%) due to its high emissions and high potential for SOA production. N-dodecane, n-undecane, m/p-xylene, n-decane, ethylbenzene and o-xylene also contributed significantly to the formation of SOAs, accounting for 17.2%, 8.79%, 5.32%, 4.22%, 4.22% and 4.06% of the total SOAP, respectively. Methylcyclohexane, m-ethyltoluene and styrene also exhibited strong SOA forming tendencies. The total emissions of SOAP from these 10 species accounted for 89.5% of the total emissions. It

should be noted that species with high emissions might not necessarily produce abundant SOAs, which can be attributed to their tendency to form SOAs. For example, the emissions of n-dodecane (1.99%), n-undecane (1.36%) and n-decane (0.98%) were relatively small, but accounted for 17.2%, 8.79% and 5.32% of the total SOAP due to their significantly higher SOAs output (estimated to be 44%, 33% and 22%, respectively). In contrast, acetone, with its second highest emission contribution, provided a lower SOA yield, accounting for only 0.66% of the total SOAP.

3.2.2. Reactive Activity of Emission Source

The top 10 sub-sources with large VOC emissions, and OFP and SOAP productions were selected to analyze the contribution ratios of active components (Figure 4). The source with the largest contribution to the total OFP were light vehicles (31.5%), while rubber and plastic production ranked first among SOAP contribution industries, accounting for 21.5%, whereas its OFP contribution was only 9.60%. Among other major sources with significant contributions to OFPs and SOAPs, the contribution of the surface coating industry to OFP (13.4%) was higher than that to SOAP (9.16%). The contribution of chemical raw material and chemical product manufacturing industry to OFP was also higher than that to SOAP, accounting for 14.7% and 9.32%, respectively. However, light trucks and heavy trucks both showed different trends in comparison. Their contributions to OFP were 5.39% and 3.01%, while those to SOAP were higher, accounting for 8.79% and 8.84%, respectively. The contributions of the VOC emission sources to the O₃ formation were not completely consistent with those to SOAs formation, due to the different composition of VOC active species [48,49].

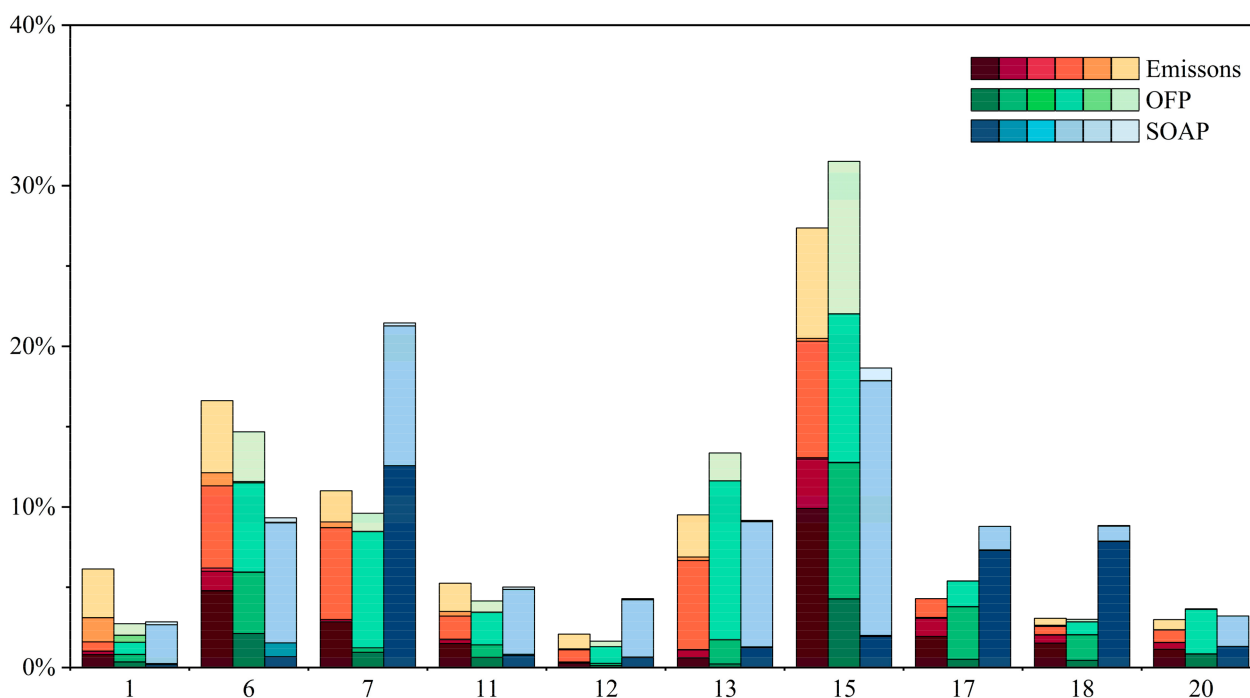


Figure 4. Composition of chemical groups of VOC emissions, OFP and SOAP of 10 key sub-sources. The colors of the bar from dark to light are alkanes, alkenes, alkynes, aromatic hydrocarbons, halogenated hydrocarbons and OVOCs.

The analysis of industrial process sources revealed that the contributions of aromatic hydrocarbons to OFP and SOAP from various industries were dominant. The contribution rates were 37.9–75.4% and 40.6–80.4%, respectively, among which the alkanes from the rubber and plastic manufacturing industry were much higher than those for the other industries in terms of SOAP, because of the high concentration of long-chain alkanes

(n-dodecane, n-undecane and n-decane) in the alkanes discharged and their high SOA-generating capacity. OVOCs played a leading role in OFP from various sub-sources in industrial processes, with the contribution rates of 11.7–21.1%. On the one hand, the high contribution rate of OVOCs to OFP was related to the high reactivity coefficients of OVOCs and the high content of OVOCs in the emissions from industrial process sources. The printing and dyeing industry and the surface coating industry sources, along solvent utilization, had very similar component contributions. Aromatic hydrocarbons were the dominant components in OFP and SOAP, with contribution rates of 63.1–85.0%. The OVOC contributions to OFP from the solvent industry was also noticeable (13.0–20.8%).

The component with a higher contribution to OFP from the mobile sources was alkenes, which was different from all other key industries. In recent years, China has upgraded the quality of its oil products and made some efforts to reduce the alkenes content from below 35.0% at the beginning of the program to below 24.0% at present, but a gap still persists between China and other developed countries. Gasoline and diesel vehicles showed different contribution characteristics to SOAP, because the dominant species of alkanes emitted by diesel vehicles were long-chain alkanes. Aromatic hydrocarbons and OVOCs contributed the most to the OFP of the power generation industry, with contribution rates of 27.6% and 26.2%, respectively, and the contribution rate of aromatic hydrocarbons to SOAP was 84.8%. With the increase in the proportion of electrical automobiles, the contribution of power industry to OFP and SOAP may gradually increase, while vehicle emissions will move in the opposite direction. Therefore, more attention should be paid in future to the emission control of aromatic hydrocarbons and OVOCs from the power generation industry.

3.3. VOCs Impact on Air Quality

3.3.1. Annual Average Concentration Distribution of VOCs

The VOCs diffusion simulation in TIA was conducted using the ADMS model, the VOC emission inventory was used as the input file of the model, and the emissions specific spatial distribution was shown in Figure S3, producing an annual average concentration map of VOCs (Figure 5). The green triangle is the location of the national control station on the Fourth Street, which was input into the ADMS model as a sensitive point.

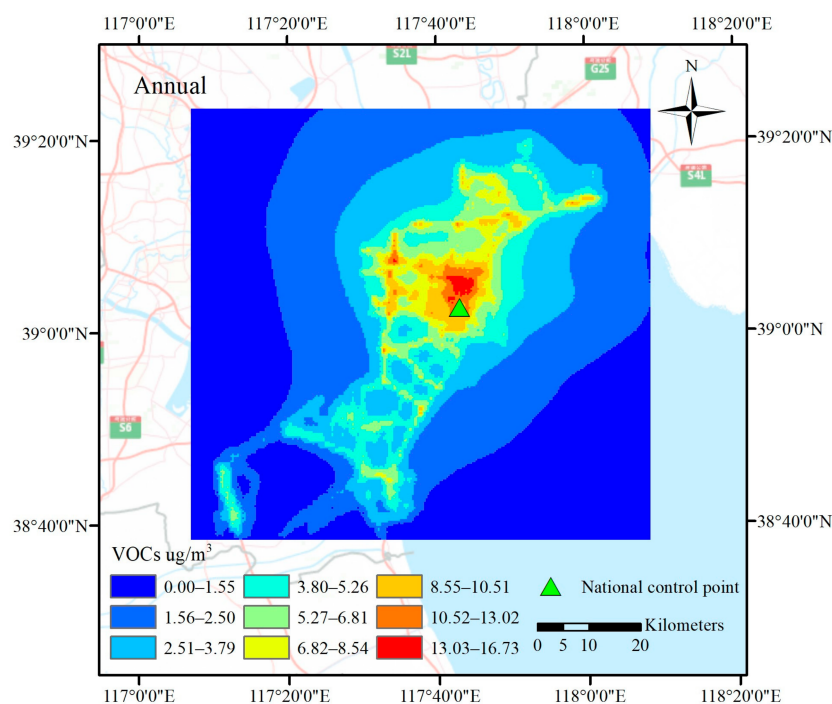


Figure 5. Simulated annual average VOC concentrations (µg/m³) in TIA.

The daily average concentration of VOCs in TIA was $8.04 \mu\text{g}/\text{m}^3$, while that at the national control station in the central area was $10.8 \mu\text{g}/\text{m}^3$, i.e., 34.3% higher than the average concentration. The simulation results showed that the concentration of VOCs was the highest near the main streets with heavy traffic and in core industrial areas. The contribution of VOCs emitted by motor vehicles to the concentration was mainly concentrated within 100 m corridors extending along the roads.

The daily average concentration of VOCs for the Beijing-Tianjin Expressway, Jintang Highway and the area located between them exceeded $8.55 \mu\text{g}/\text{m}^3$ over a large range. The first reason was probably that these roads, as important transport links to the Tianjin Port, served large traffic volumes in the core area of TIA. The second, and more crucial reason, was that most industrial enterprises are located in the central area, causing more VOC emissions. High-density and narrow streets in the central area are surrounded by high-rise buildings, forming a closed air circulation, which reduces the outward diffusion of VOCs and other air pollutants in the area [50,51]. This was consistent with the research results reported in [52] for a typical area in Shenzhen, where the emission concentration of VOCs from mobile sources was $1.04\text{--}4.01 \mu\text{g}/\text{m}^3$, while near the roads with the same grades and traffic volumes, factors such as road width and street canyon height will affect the diffusion of pollutants.

3.3.2. Seasonal Emission Concentration Characteristics

For simulating the seasonal emission characteristics of VOCs, January and July were selected as representative winter and summer months, respectively (Figure 6). The average concentrations of VOCs in winter and summer were $10.9 \mu\text{g}/\text{m}^3$ and $7.58 \mu\text{g}/\text{m}^3$, respectively, i.e., the former was significantly higher, which can be attributed to the effect of meteorological conditions. The ambient temperature in winter suppresses photochemical reactions, while stable meteorological conditions, such as low wind speeds and low boundary layer height, weaken the turbulent exchange and hinder the vertical diffusion of VOCs [53]. This observation was consistent with the environmental measurements from numerous cities, where the concentration of VOCs was even twice as high in winter as in summer [54,55]. However, several points in summer showed high VOC concentrations, which might be related to intense industrial activities.

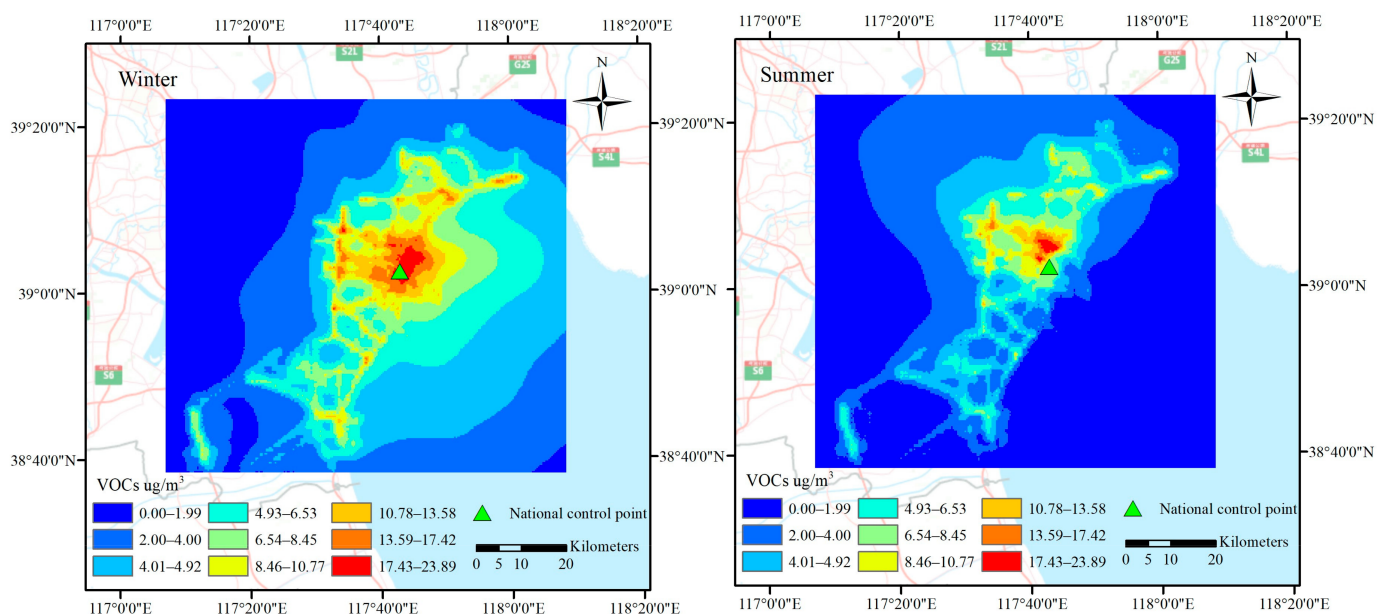


Figure 6. Simulated winter and summer VOC concentrations ($\mu\text{g}/\text{m}^3$).

In addition, we can see that the wind speed and direction had an important influence on the diffusion range of VOCs. In winter, the dominant wind direction was northwest

(Figure S4), while in summer, it was southeast, which led to a reversal of the main areas influenced by VOC diffusion. In winter, the national control point was downwind of the main pollution source, and the concentration of VOCs there reached $19.1 \mu\text{g}/\text{m}^3$.

3.3.3. Diurnal Emission Concentration Characteristics

The period 7:00–19:00 was assumed as the daytime and the rest as the nighttime for this analysis. The VOC emissions during the daytime and nighttime were input into the ADMS model to simulate the daily variations of VOC concentration. The simulation results (Figure 7) showed that the emission concentration of VOCs was $5.20 \mu\text{g}/\text{m}^3$ during the day and $3.57 \mu\text{g}/\text{m}^3$ at night.

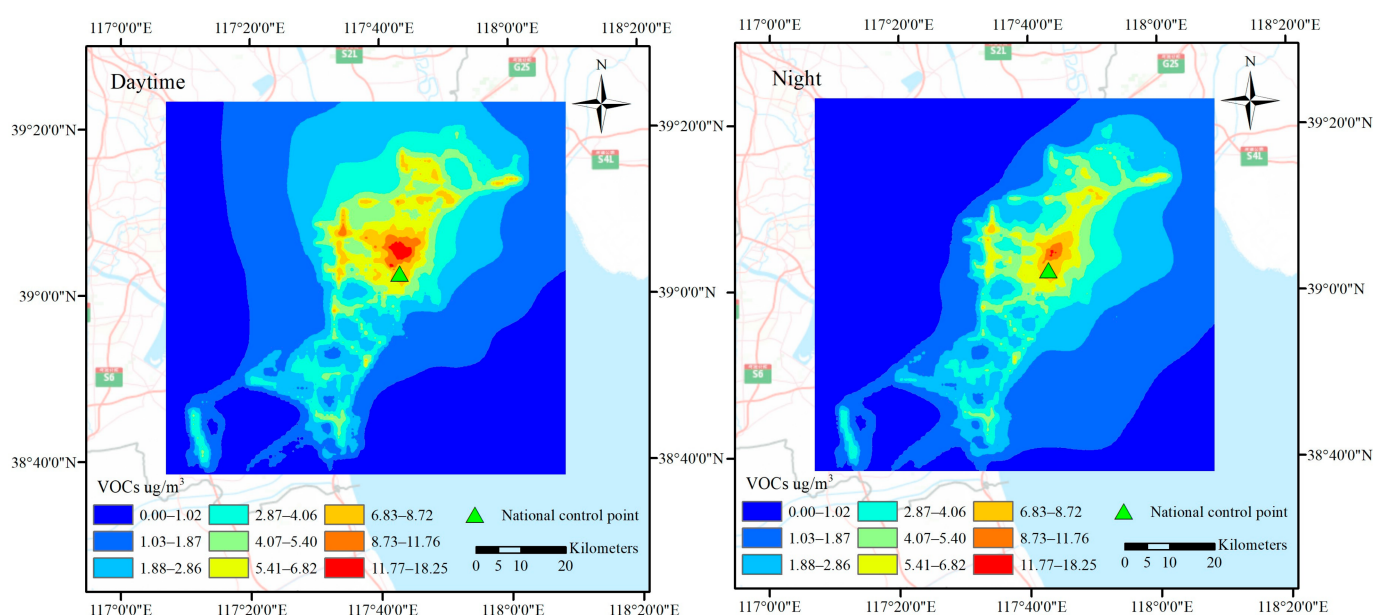


Figure 7. Simulated daytime and night VOC concentrations ($\mu\text{g}/\text{m}^3$).

Mobile source emissions were closely related to human activities, and indeed the emission concentrations from mobile sources at night were significantly reduced. In terms of industrial sources, only some industries in the central area had higher emissions at night. Generally, the overnight emission intensities of industrial sources were much lower than those during daytime, because these industries were inactive at night except combustion sources. In this study, the difference of activity levels between daytime and night was fully considered, and the daytime and night emissions were input into the model separately, which made the characteristics of daytime higher than night more obvious, which showed that the difference of activity levels had a significant impact on the diurnal concentration of VOCs, even exceeding the photochemical reaction. This conclusion has also appeared in previous studies. According to Yeniso-Y-Karakaş's research, the environmental measurement results showed that the concentration of anthropogenic VOCs during the daytime was higher than that at night because of intensive vehicle activities during working hours [56]. Through the long-term observation of urban stations in China from 2014 to 2019, Xia et al. found that human activities related to OVOCs emissions during the daytime were significant and can offset photochemical losses, and the daily peaks of aromatic hydrocarbons appeared at 10:00 and 19:00 during the daytime [57].

In addition, the diurnal distribution of VOC concentrations was strongly affected by the air flow between land and sea [58]. During the day, the hot air over land expands and rises, resulting in a pressure difference between the land surface and the sea surface, thus forming a sea breeze, which blows the emitted VOCs into the land, while at night, land breeze prevails, which moves more VOCs into the sea [59].

3.3.4. Uncertainties and Future Research

In the previous assessments of VOC species, source profiles obtained from foreign source libraries or domestic regional tests were generally adopted because of the limited field measurements available. Although this study adopted the domestic integrated source profiles of VOCs from the latest, more representative review, it still did not reflect the real species emissions, which might negatively influence the result uncertainty. Therefore, local testing of VOC source profiles is needed in future. For VOC concentration simulations, the diurnal emission distribution coefficients were taken from guidebook [25], which could not completely reflect the daily activity levels in TIA. It is also worth pointing out that the ADMS model is usually used to simulate the diffusion over small-scale areas and cannot completely simulate the chemical transformation process of VOCs, so it is necessary to improve the simulation of VOCs secondary pollution in the future.

4. Conclusions

This study considered TIA as a case study to analyze the impact of anthropogenic VOC emissions on the atmospheric pollution in industrialized areas. The established inventory revealed that the emission intensity of VOCs in the industrialized areas was high and the main emission sources were industrial processes (38.4%), mobile sources (36.5%) and solvent utilization (12.9%). The main contributing industries were chemical product manufacturing and rubber and plastic manufacturing. The top 10 VOCs were toluene, acetone, ethylbenzene, m/p-xylene, i-pentane, n-hexane, formaldehyde, benzene, ethyl acetate and ethylene. Mobile sources and industrial processes contributed the most to the top 10 pollutants, while combustion sources and mobile sources dominated the emissions of OVOCs.

Taking OFP and SOAP as evaluation indices of the contributions to VOC emissions by species and industries, we found that m/p-xylene was the largest species in OFP (8.90%) and the dominant emission species were aromatic hydrocarbons and olefins in addition to formaldehyde. The most abundant emission species of SOAP was toluene (39.9%) and the dominant species in SOAP emissions were aromatic hydrocarbons and long-chain alkanes. VOC emission sources had various contributions to the formation of O₃ and SOAs due to the different composition of active components. The contribution rates of the rubber and plastics industry and trucks to SOAP were markedly high because they emitted high concentrations of long-chain alkanes, whereas the mobile sources contributed more alkenes to OFP due to the fuel composition.

In addition, the ADMS atmospheric diffusion model was used to simulate the concentration distribution of VOCs in TIA. The results showed that the daily average concentration of VOCs in TIA was 8.04 µg/m³ and that it was high near the main roads with heavy traffic and the core industrial areas. The emissions from industrial enterprises were a more important reason for the high concentration of VOCs in the region. The stable meteorological conditions in winter led to higher VOCs concentrations than in summer. More VOCs were emitted during the daytime than at night due to the activity levels, but the modeling of photochemical reactions needs to be improved.

The accurate emission characterization of VOCs in TIA will provide a useful reference for further environmental control of VOCs in industrialized areas, which will help the government departments to specify more effective air pollution control measures.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/atmos14101586/s1>, Figure S1. Study area; Figure S2. Composition of VOCs in emission sources; Figure S3. VOC emissions spatial distribution; Figure S4. Windrose; Table S1. Source classification and emission factors [25,26]; Table S2. VOC Species MIR and OFP ($\times 10^{-3}$ Gg); Table S3. VOC Species Y_{SOA} and SOAP (%).

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