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Analysis of NO_x Pollution Characteristics in the Atmospheric Environment in Changchun City

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Abstract: Nitrogen oxide (NO_x) pollution has become one of the most challenging problems in China in the past 20 years. In this study, on the basis of the Jilin Province Atmospheric Environmental Quality Bulletin and hourly NO_x data from the Atmospheric Environment Automatic Monitoring Station in Changchun, temporal and spatial variations in NO_x concentration in the province and Changchun and their relationships with various pollutants and meteorological factors were analyzed. The results show that Changchun had the highest NO_x concentration of all cities in the province, with a high concentration in the center and a low concentration in the east and west. The areas with high NO_x concentrations in Changchun were mainly distributed in urban centers, and the concentration in the northern part of the city was higher than that in the south. The seasonal variation and average daily variation in NO_x concentration in Changchun had a bimodal distribution, and the NO_x concentration in autumn and winter was higher than that in spring and summer. The maximum monthly average concentrations of NO_x and nitric oxide (NO) were reached in October, and the maximum monthly average concentration of nitrogen dioxide (NO_2) was reached in March. The average daily variation in NO_x concentration first peaked at 07:00–08:00 in the morning, and the second peak occurred between 20:00 and 22:00 at night. The NO_x concentration in Changchun was positively correlated with NO_2, NO, $PM_{2.5}$ (fine particulate matter), PM_{10} (particulate matters), CO (carbon monoxide), and pressure, and it showed a significant negative correlation with O_3, temperature, wind speed, and humidity.

Keywords: nitrogen oxide; spatial–temporal variation; meteorological factors; Changchun

1. Introduction

Among various air pollutants, Nitrogen oxide (NO_x) emissions have the closest relationship with the human use of fossil energy. In the past 20 years, the growth rate of NO_x emissions in China has been the largest compared with other pollutants [1]. With the continuous development of China's economy and the continuous consumption of various mineral resources, the levels of secondary pollutants, such as particulate matter (PM_{10}), ozone (O_3), and nitrogen dioxide (NO_2), have not significantly improved in metropolitan areas [2,3]. Studies have shown that NO_x is a pollutant that has substantially contributed to regional atmospheric pollution and environmental quality, and it has played an important role in the formation of tropospheric O_3, peroxyacetyl nitrate (PAN), and aerosols [4]. Nitrogen oxide emissions and detection concentrations have continually increased every year [5,6]. NO_x includes compounds such as nitrous oxide (N_2O), nitric oxide (NO), NO_2, dinitrogen trioxide (N_2O_3), and dinitrogen tetroxide (N_2O_4). NO and NO_2 are the main atmospheric NO_x compounds that affect human health and the ecological environment, while the nitric acid and nitrate formed by the oxidation of NO_2 (formed by the photo-oxidation of NO and hydrocarbons (HC) and O_3) are the main sources of nitric acid rain [7]. NO can irritate the respiratory system; it can also bind to heme to form

nitrosohemoglobin and is poisonous. NO_2 can severely stimulate the respiratory system and result in heme nitration, and it is more environmentally harmful than NO because it forms acid rain pollution.

In the past, ground-based real-time monitoring [8], aerial survey [9], and satellite remote sensing have been used to study the temporal and spatial variation in NO_x and other atmospheric pollutants on urban and larger spatial scales [10]. Studies have shown that the daily variation in NO_x tends to be significantly bimodal. The NO_x concentration has been found to be higher at night than during the day [11], but the time at which the maximum concentration occurred has differed among studies [11,12]. For example, Robert Cichowicz found that the maximum values appeared at different times between 08:00 and 09:00 in five neighboring provinces in central Poland, Central Europe [13]. Changes in concentrations of atmospheric pollutants such as NO_x during special periods (such as during the Beijing Olympics) or during atmospheric pollution events might vary significantly [14]. Since 2013, there have been several serious air pollution events in Northeast China, and air pollution has been intensifying [15–17]. Thus, research on nitrogen oxides is crucial. The aim of this study was to analyze the characteristics and main causes of NO_x concentration changes in Changchun. The analyzed data were obtained from the Jilin Province Atmospheric Environmental Quality Bulletin and NO_x monitoring data in Changchun in 2018, as well as data on several other pollutants and meteorological factors.

2. Experiments

2.1. Research Area and Experimental Design

Jilin Province is located in the northeastern part of China at a longitude of 122–131° E and latitude of 41–46° N, with an area of 187,400 km^2, accounting for 2% of the total area of the country. The altitude is high in the southeast, with predominantly mountainous hills, and low in the northwest. It is mainly a terraced plain and a central plain in the central and western regions. The cities of Jilin Province are Changchun, Jilin, Siping, Tonghua, Baishan, Liaoyuan, Baicheng, Songyuan, Yanbian Korean Autonomous Prefecture, and Changbai Mountain Administrative Committee. The provincial capital is Changchun, which is located in the hinterland of Songliao Plain in northeastern China. The urban area is 250–350 m above sea level and is located in the continental monsoon climate zone. The most frequent wind direction in summer is southeast. The wind speed in autumn is lower than that in spring. The dominant wind direction is southwest; the wind direction in winter is almost northwest.

The observations used in this study were collected from 10 automatic monitoring stations for the atmospheric environment in Changchun (Figure 1). NO_x was also measured continuously using a NO_x analyzer (Model 42i, Thermo Scientific, Waltham, MA, USA). This instrument analyzes the ambient air using the principle of chemiluminescence at an interval of 1 min. Ozone was also measured using an ultraviolet photometric ozone analyzer (Model 49i ozone analyzer, Thermo Scientific, Waltham, MA, USA), which was regularly calibrated using pure ozone and ozone-free air. Both instruments were regularly calibrated during the study period. In addition, CO was simultaneously measured directly by gas filter correlation analysis technology (Model 48i, Thermo Scientific, Waltham, MA, USA).

Since 2013, the Ministry of Environmental Protection has deployed 1497 state-controlled automatic monitoring stations for atmospheric environmental quality throughout the country, and they are starting to be put into operation. Among the 10 sites in Changchun, 9 are located in the built-up area of Changchun, namely, Daishan Park (DP), High-tech Zone Management Committee (HZMC), Economic Exploitation Zone Sanitation Office (EEZCO), Jingyuetan (JYT), Bus Factory (BF), Labor Park (LP), Food Plant (FP), Institute of Post and Telecommunications (IPT), and Garden Department (GD); a clean control site is located in Shuangyang District of Changchun, namely, Shuaiwanzi (SWZ). JYT and SWZ are first-class ambient air functional zones, while the remaining stations are second-class ambient air functional zones. The instruments and equipment of the automatic monitoring station for atmospheric environmental quality automatically collect samples and analyze and generate data every 15 min. Data

are then automatically uploaded to the national and provincial environmental protection departments. Table S1 shows the details of the sites.

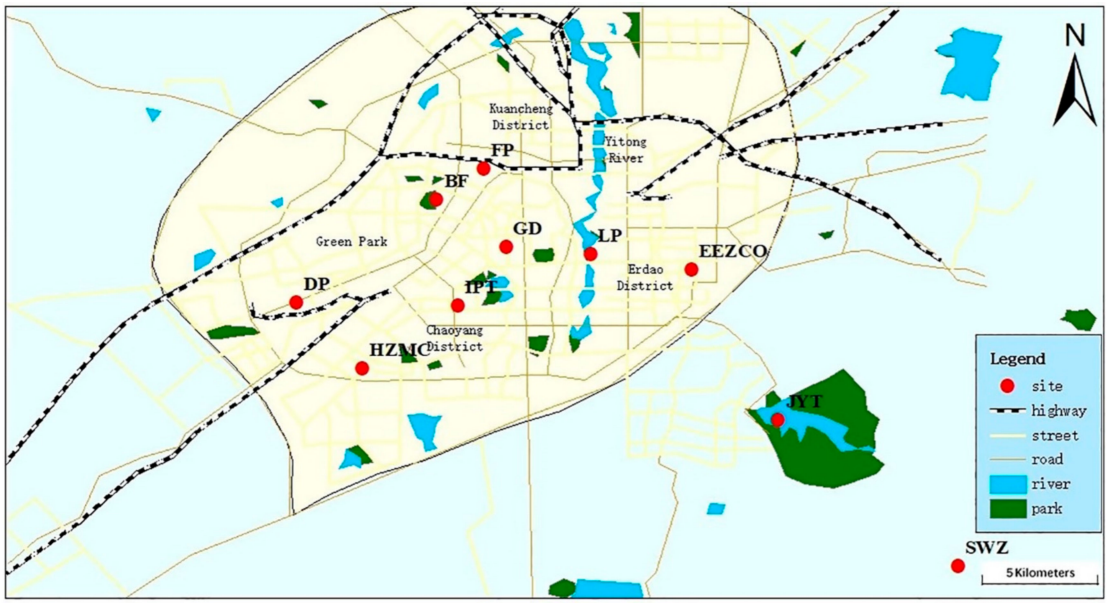

Figure 1. Air monitoring stations of Changchun.

2.2. Meteorological Factors and Pollutant Information

In the continental monsoon climate, the seasonal division of Changchun in 2018 is in accordance with the meteorological industry standard "Climate Season Division" (QX/T152-2012). In Changchun, the annual difference is adjusted from April 17 to June 16 for the spring (61 days), June 17 to August 24 for the summer (69 days), August 25 to October 26 for the autumn (63 days), and October 27 to April 16 for the winter (172 days). Photochemical smog is formed by primary pollutants such as hydrocarbons (HC) and nitrogen oxides (NO_x) discharged into the atmosphere from pollution sources such as automobiles and factories. In winter, because of the frequent occurrence of inversion, the high atmospheric stability causes pollutants to accumulate near the ground, producing a brown haze called photochemical smog [18,19]. In this study, all measured data were selected according to the "Regulations on Ambient Air Quality Monitoring", and unreasonable values were removed to control the data quality. The calculation, statistical analysis, and evaluation of monitoring data of various pollutants were carried out in accordance with the Ambient Air Quality Standard (GB3095-2012) and the Technical Regulation for Ambient Air Quality Assessment (Trial) (HJ663-2013).

3. Results

3.1. Analysis of the NO_x Pollution Degree in the Whole Province

Kriging interpolation is a method based on the theoretical analysis of semi-variograms and the unbiased optimal estimation of variable values in finite regions [20]. Similar studies have applied this method to determine the spatial distribution of O_3 and $PM_{2.5}$ (fine particulate matter) [21,22]. The spatial distribution of NO_x, NO_2, and NO in Jilin Province (Figure 2) was suitable for Kriging interpolation. The high average annual NO_x concentrations were mainly found in densely populated and relatively developed areas, such as Changchun, Tonghua, Liaoyuan, and Siping. In addition, high concentrations of NO_x were found in high-volume areas. For example, a high NO_x concentration was found in Baishan City, which is also rich in mineral resources, accounting for 73% of the minerals found in Jilin Province. In addition, Jiangyuan County and Badaojiang District in this city are among the 60 key coal-producing counties in China. The NO_x concentration in the western and eastern mountainous areas of Jilin Province was low, which may be a result of the high vegetation coverage

and low anthropogenic emissions in these regions. The province's NO_x concentration was high in the middle and low in the east and west.

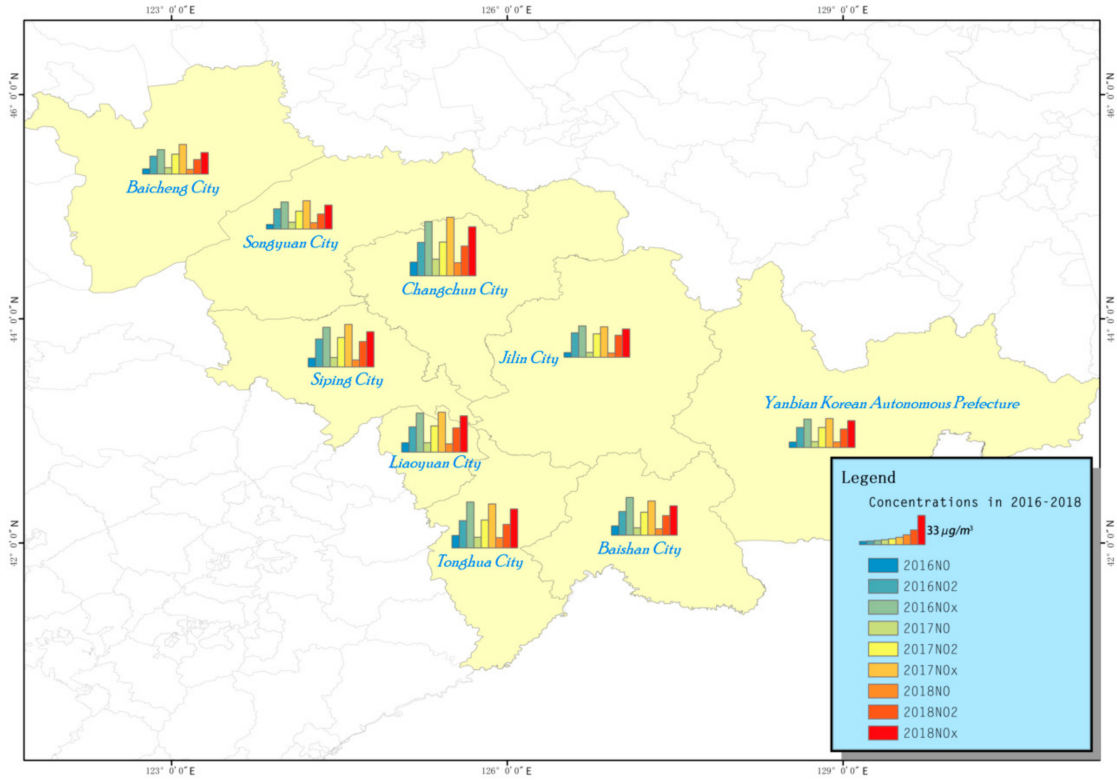

Figure 2. Spatial distribution of NO_x, NO_2, and NO in Jilin Province, 2016–2018.

The trends of NO_x, NO_2, and NO concentrations in the nine cities and states of Jilin Province were similar. The NO concentration fluctuated between 4.5 and 18.7 μg/m^3 in 2016–2018. The NO_2 concentration varied between 16 and 38 μg/m^3 in 2016–2018, which did not exceed the national average secondary air standard (40 μg/m^3). The NO_x concentration varied between 24 and 66 μg/m^3 in 2016–2018, and only Changchun exceeded the national average secondary air quality standard (50 μg/m^3).

3.2. Trend Analysis of NO_x Pollution in Changchun

For the pollutants studied in this work, Changchun had the highest annual average concentration, and it was used as a typical city for the analyses described in the following subsections.

3.2.1. Seasonal Change Analysis

The seasonal variations in NO_x, NO_2, and NO average concentrations are shown in Figure 3. The seasonal patterns of NO_x, NO_2, and NO found in our study are consistent with those reported in other studies [23–25]. The concentrations of NO_x, NO_2, and NO were generally higher in autumn and winter, and the change in the concentration trend was related to meteorological conditions. On the one hand, the low temperature in winter and the long life of NO_2 are not conducive to the conversion of NO_2, so it gradually accumulates. On the other hand, it was also related to winter heating in Changchun. Changchun is a city in Northeast China, and its heating time is earlier and longer from autumn to winter. Heating increases the amount of coal used and increases the concentration of NO_2. For example, the planetary boundary layer (PBL) height over the North China Plain is usually less than 500 m in winter [26], resulting in less efficient vertical transport of particles, as well as their mixing, to higher altitudes. The NO_2 concentrations exhibited a large decrease from winter to summer. During the summer season, the atmospheric temperatures and humidity were relatively high, which are

conditions that accelerate the oxidization of NO_2 in the air [27]. In addition, Changchun features a seasonal monsoon climate with an uneven distribution of annual precipitation, and the frequent rainfall in the summer greatly reduces the concentration of NO_2 in the atmosphere.

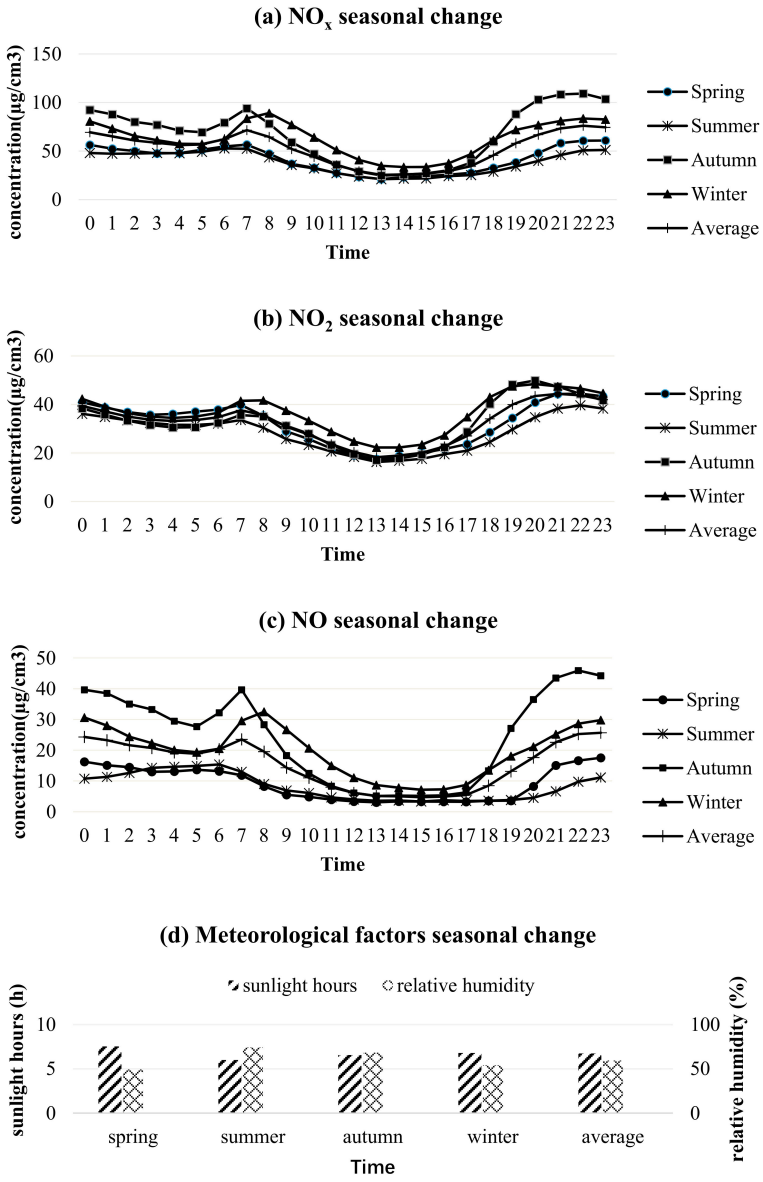

Figure 3. Concentrations of nitrogen compounds and meteorological factors in the four seasons of Changchun in 2018: (**a**) NO_x, (**b**) NO_2, (**c**) NO, (**d**) sunlight hours and humidity.

3.2.2. Daily Average Change Analysis

As shown in Figure 4, the daily average concentration of NO_x, NO_2, and NO in Changchun's various stations had a bimodal distribution: the first peak appeared at 07:00–08:00 in the morning, and the second peak occurred between 20:00 and 22:00 at night. The second peak concentration of NO_x and NO was significantly lower than the first peak; the nighttime peak might be related to higher emissions from diesel vehicles [28]. The lower nighttime peak might be a response to less congested traffic from the traffic rush being spread over 2 h, whereas in the morning, rush hour is shorter but more intense. The second peak concentration of NO_2 was significantly higher than the first peak. The morning peak was consistent with the traffic peak. The peak of NO_2 at night was higher than the peak in the morning; this is a result of a reaction between NO and O_3 to form NO_2 at night when the photolysis reaction of NO_2 is stagnant, resulting in its gradual accumulation.

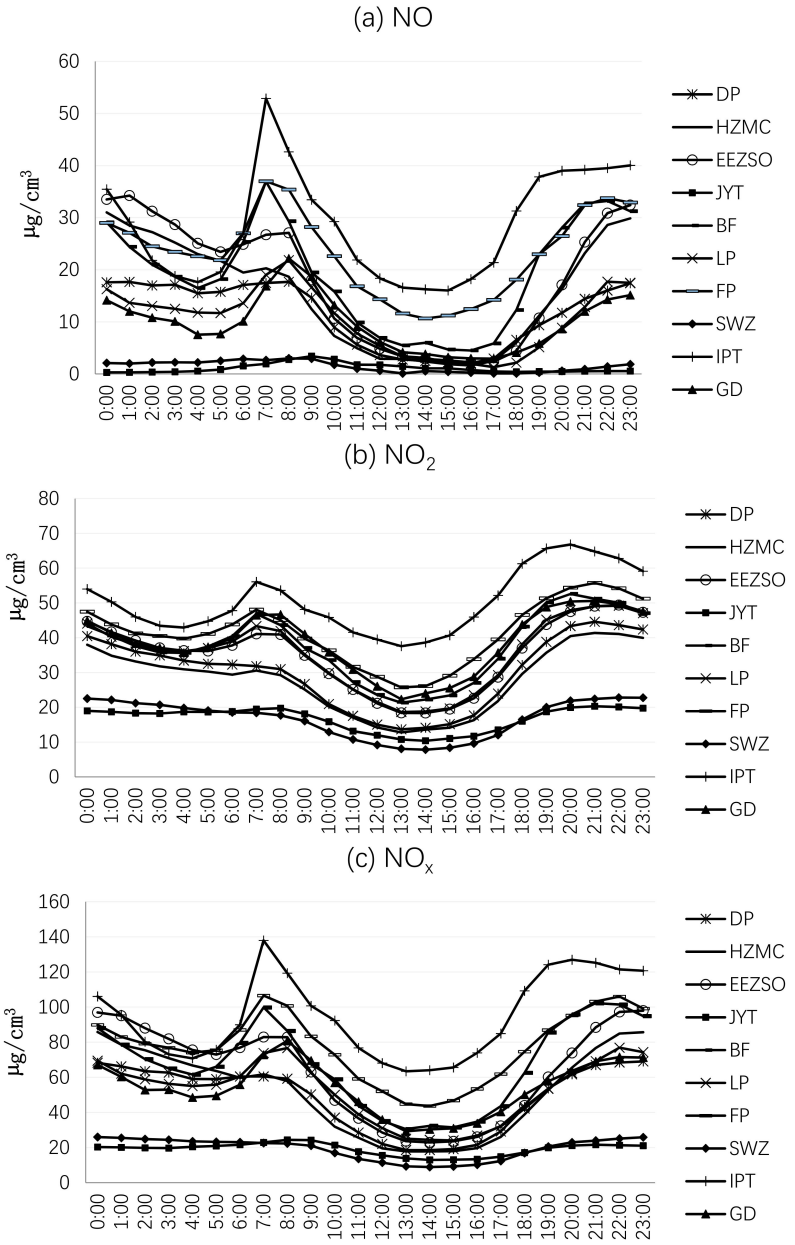

Figure 4. Daily average concentration of nitrogen compounds at each station in Changchun, 2018: (**a**) NO, (**b**) NO_2, (**c**) NO_x.

The current national standard GB 3095-2012 "Environmental Air Standard" does not list NO in the ambient air pollutant project, and therefore, the NO data cannot be compared to this standard. The daily average value of the second-level concentration limit of the NO_2 second-class zone in the ambient air pollutant project was 80 μg/m^3, and the NO_2 concentration did not exceed this limit. The daily average concentration limit of the NO_x secondary zone in the ambient air pollutant project was 100 μg/m^3, and only individual sites (IPT, FP) had some outliers of NO_x concentration that exceeded the limit at 07:00–08:00 in the morning and 18:00 at night. The concentration was consistently higher in the morning and consistent with the morning peak travel. The high concentration at night was dependent on the repeated oxidation of O_3 during the day, indicating that the NO_x emission and distribution phenomena were closely connected to O_3 concentration.

3.2.3. Spatiotemporal Variation

It can be seen from Figure 5 that the NO concentration in October, November, December, and January was higher than that in the other months in Changchun, 2018. The NO concentration decreased in February and continued to decrease in March, April, and May. In June and July, the NO concentration was at its lowest value and remained statistically unchanged. The concentration of NO increased sharply in August, September, and October, and it was the lowest in May, June, and July. This phenomenon is caused by the increase in solar radiation intensity: NO is transformed into NO_2 through a photochemical reaction with atmospheric oxidants. After August, the conversion of NO to NO_2 was weakened with the decrease in solar radiation. The monthly average concentration of NO_2 was higher in October, November, December, January, and March, and it also reached a high concentration in winter in the analysis of seasonal concentration changes. Among the winter months, March had the highest concentration of NO_2, and the main reason is that there were 12 days of mild pollution in March in Changchun, with 1 day of heavy pollution. The concentration of NO_2 decreased in April and May. There was a brief rise in NO_2 concentration in June, followed by a large decrease in July and August, and it rose sharply in September, October, and November. The concentration of NO_x was higher in October, November, December, and January compared with other months, and the spatial distribution and annual average spatial distribution characteristics were the same.

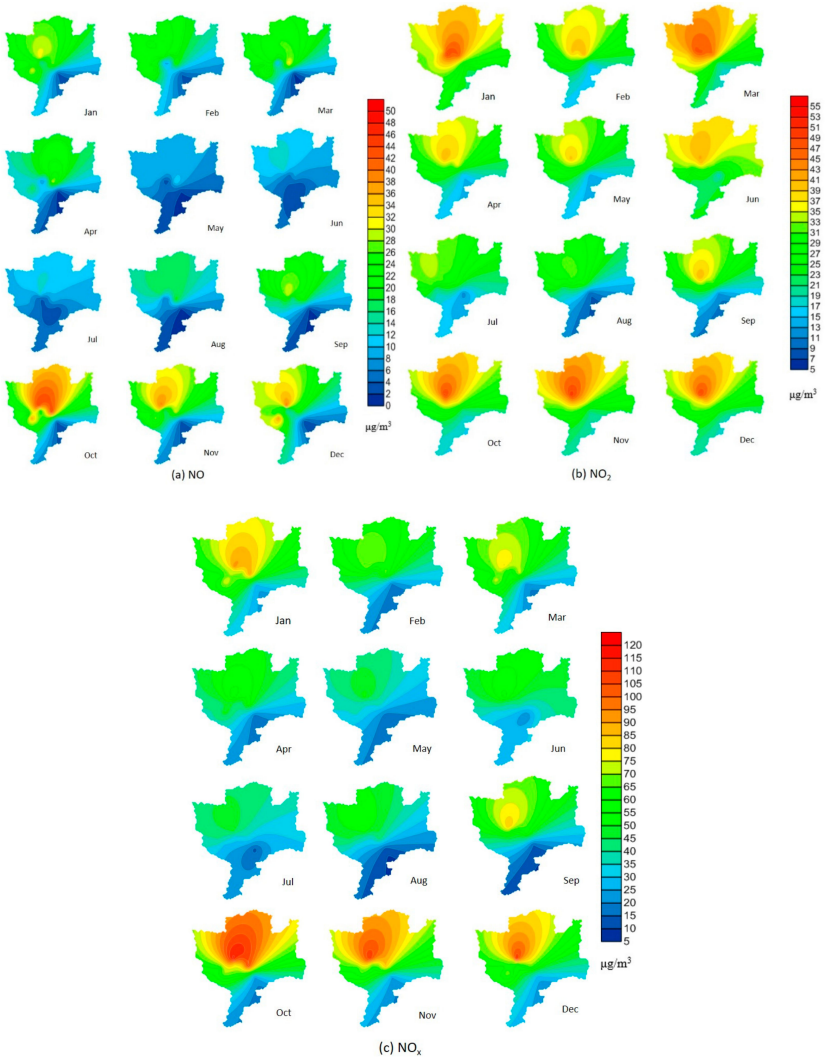

Figure 5. Spatial distribution of the monthly average concentration of nitrogen compounds in Changchun, 2018: (**a**) NO, (**b**) NO_2, (**c**) NO_x.

3.2.4. Spatial Variation Analysis

Figure 6 shows the spatial distribution of NO_x, NO_2, and NO in Changchun, 2018. The overall spatial distribution characteristics of the three pollutants were similar. The concentrations of all the NO_x pollutants were the same in Changchun center and higher than in the periphery, with a clear hierarchical structure. This is most likely related to the relatively extensive development and high population in urban centers. Among the nine stations, IPT had the highest concentration, and JYT had the lowest. The center of the city covered by IPT, FP, and BF was an area of high nitrogen oxide concentration.

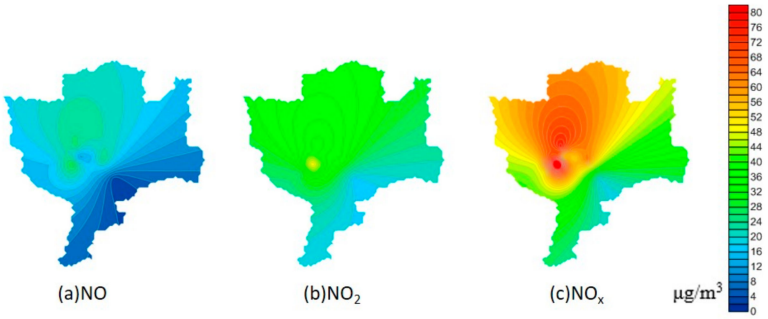

Figure 6. Spatial distribution of the average concentration of nitrogen compounds in Changchun, 2018: (**a**) NO, (**b**) NO_2, (**c**) NO_x.

The coefficient of divergence (COD) method was also used to evaluate the spatial distribution differences in NO_x concentrations at the monitored sites in Changchun. The COD is defined as

$$COD_{fn} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{x_{if} - x_{ih}}{x_{if} + x_{ih}}\right)^2}$$

where x_{if} is the ith concentration measured at the f^{th} site, f and h represent two monitoring sites, and n is the number of observations. The COD is a coefficient used extensively in numerical analyses of ecological data to determine the resemblance either between objects under study or between the variables describing them. For a spatial distribution, the COD approaches zero if the measured values at two monitoring sites are similar. In contrast, if the measured values are very different, then the COD approaches unity. The spatial disparity of NO_x was analyzed from 2016 to 2018 (Table 1). The CODs for NO_x showed an identical trend to that of the GIS analysis above. The COD between IPT, FP, and BF was generally less than 0.2, which means that these sites had the smallest spatial variability. The CODs between JYT and IPT, FP, and BF were generally more than 0.5, while the smallest spatial variability was between JYT and SWZ.

Table 1. Coefficients of divergence for NO_x obtained at the 10 monitoring sites in 2016–2018.

Year	COD										
	Site	**DP**	**HZMC**	**EEZCO**	**JYT**	**BF**	**LP**	**FP**	**SWZ**	**IPT**	**GD**
2016	DP	—	0.11	0.06	0.43	0.13	0.07	0.21	0.42	0.29	0.08
	HZMC			0.11	0.37	0.21	0.10	0.28	0.37	0.35	0.14
	EEZCO				0.44	0.13	0.06	0.20	0.43	0.28	0.08
	JYT					0.51	0.42	0.57	0.11	0.63	0.45
	BF						0.13	0.10	0.50	0.18	0.10
	LP							0.21	0.41	0.29	0.06
	FP								0.56	0.09	0.17
	SWZ									0.62	0.44
	IPT										0.25
	GD										—
2017	DP	—	0.09	0.13	0.40	0.16	0.04	0.22	0.41	0.30	0.05
	HZMC			0.06	0.46	0.11	0.07	0.16	0.45	0.24	0.06
	EEZCO				0.49	0.09	0.10	0.13	0.48	0.21	0.10
	JYT					0.52	0.43	0.56	0.13	0.62	0.43
	BF						0.13	0.09	0.52	0.16	0.13
	LP							0.19	0.43	0.27	0.04
	FP								0.56	0.10	0.18
	SWZ									0.61	0.44
	IPT										0.27
	GD										—
2018	DP	—	0.10	0.15	0.44	0.16	0.08	0.22	0.46	0.32	0.08
	HZMC			0.12	0.45	0.17	0.07	0.22	0.48	0.31	0.08
	EEZCO				0.52	0.08	0.10	0.13	0.54	0.22	0.09
	JYT					0.55	0.46	0.59	0.16	0.66	0.47
	BF						0.13	0.09	0.56	0.17	0.11
	LP							0.19	0.49	0.28	0.02
	FP								0.60	0.11	0.18
	SWZ									0.66	0.49
	IPT										0.27
	GD										—

3.2.5. Relationship Between NO_x and Meteorological Factors

The relationship between NO_x, NO_2, and NO and meteorological factors was slightly different in different regions, but roughly the same conclusions were drawn in terms of temperature (T), wind speed (WS), pressure (P), and humidity (H). The influence of other pollutants on NO_x, NO_2, and NO was analyzed in this study by selecting several typical atmospheric pollutants, such as O_3, $PM_{2.5}$, PM_{10}, and CO (carbon monoxide). On the basis of the selected meteorological factors, 11 factors were analyzed by correlation analysis (Table 2). The total number of data points for the 11 factors was 4015.

Table 2. Correlation of NO_x, NO_2, and NO and other influencing factors.

	NO	**NO_2**	**NO_x**	**O_3**	**$PM_{2.5}$**	**PM_{10}**	**CO**	**T**	**WS**	**P**	**H**
NO	1										
NO_2	0.591	1									
NO_x	0.963	0.779	1								
O_3	−0.43	−0.161	−0.402	1							
$PM_{2.5}$	0.306	0.62	0.427	0.103	1						
PM_{10}	0.217	0.407	0.286	0.353	0.735	1					
CO	0.508	0.725	0.623	−0.085	0.853	0.555	1				
T	−0.274	−0.276	−0.298	0.459	−0.376	−0.096	−0.468	1			
WS	−0.402	−0.336	−0.42	0.528	0.059	0.336	−0.109	0.169	1		
P	0.426	0.341	0.442	−0.499	0.312	0.029	0.433	−0.799	−0.293	1	
H	−0.08	−0.178	−0.11	−0.255	−0.253	−0.442	−0.162	0.387	−0.259	−0.372	1

The correlation coefficient between NO and NO_x was 0.963, and that between NO_2 and NO_x was 0.779. The correlation coefficient between NO and NO_2 was 0.591, which indicates that the correlation between NO, NO_2, and NO_x was very strong. The results also confirm that these pollutants had a common source. The NO_2 concentrations were notably positively correlated with $PM_{2.5}$ concentrations, with a correlation coefficient of above 0.60. It was verified that the secondary conversion of NO_2 had a significant effect on $PM_{2.5}$ concentrations. There was a significant negative correlation between NO_2 concentration and O_3 concentration because the precursors were consumed and produced by photochemical reactions. The correlation between NO_2 concentration and $PM_{2.5}$ concentration was much greater than the correlation with O_3 concentration. The reason may be that Changchun has a lower average annual temperature, higher humidity, less volatile organic compound (VOC) emissions from vegetation, and lower O_3 concentration. The NO_2 concentration was significantly positively correlated with the CO concentration, with a correlation coefficient of above 0.70.

The concentrations of NO_x, NO_2, and NO in Changchun had a strong negative correlation with temperature, wind speed, and humidity, and they had a strong positive correlation with air pressure. The correlation coefficients of NO_x, NO_2, and NO concentrations with T were −0.298, −0.28, and −0.274, respectively. The ground temperature increased, mainly in summer and daytime, and the vertical mixing degree increased, thus minimizing the concentration of nitrogen oxides in the low-altitude atmosphere [29]. The correlation coefficients of NO_x, NO_2, and NO concentrations with WS were −0.42, −0.34, and −0.402, respectively, which could be explained by the fact that higher wind speeds exacerbate the dispersion and mixing of these atmospheric pollutants emitted from local sources, such as automotive engines, thereby minimizing their cumulative concentration in the atmosphere. This result is consistent with the results of several studies in which wind speed showed a negative correlation with NO_x concentration. It has been reported that low wind speeds might increase the impact of local sources of emissions [30,31]. The correlation coefficients of NO_x, NO_2, and NO concentrations with P were 0.442, 0.341, and 0.426, respectively, indicating that high pressure was more likely to cause pollution accumulation during the observation period.

4. Conclusions

On the basis of the Jilin Province Atmospheric Environmental Quality Bulletin and the hourly NO_x (including NO and NO_2) monitoring data of Atmospheric Environment Automatic Monitoring Station in Changchun, the pollution degree of NO_x in the province, temporal and spatial variations of NO_x concentrations in Changchun, and the relationships of NO_x concentration with various pollutants and meteorological factors were analyzed for 2018. The analysis of concentration changes included seasonal changes and daily average changes, and spatial changes included monthly average changes and annual changes. Changchun had the highest NO_x concentration of all cities in the province, with a high concentration in the middle and a low concentration in the east and west. The areas with high NO_x concentrations in Changchun were mainly distributed in urban centers, and the NO_x concentration in the northern part of the city was higher than that in the south.

The seasonal variation and average daily variation in NO_x concentration in Changchun had a bimodal distribution, and the seasonal variation in NO_x concentration was obvious; both analyses showed that the NO_x concentration in autumn and winter was higher than that in spring and summer. The change in the concentration trend was related to meteorological conditions. Both low temperature and heating cause an increase in NO_x concentrations. The low temperatures in winter are not conducive to the conversion of NO_2, resulting in the accumulation of NO_x. Heating increases the amount of coal used and increases the concentration of NO_2. The maximum monthly average concentration of NO_x and NO was reached in October, and the maximum monthly average concentration of NO_2 was reached in March. The spatial distribution characteristics and annual average spatial distribution were the same. The average daily variation in NO_x concentration occurred at 07:00–08:00 in the morning, and the second peak occurred between 20:00 and 22:00 at night. The second peak concentration of

NO_x and NO was significantly lower than the first peak, and the second peak concentration of NO_2 was significantly higher than the first peak.

The NO_x concentration in Changchun was positively correlated with NO_2, NO, $PM_{2.5}$, PM_{10}, and CO, and it had a significant negative correlation with O_3. The NO_x concentration had a significant positive correlation with the $PM_{2.5}$ concentration because the secondary conversion of NO_x had a significant effect on $PM_{2.5}$. There was a significant negative correlation between NO_x and O_3 because the precursors were consumed and produced by photochemical reactions. The NO_x concentration was positively correlated with P and negatively correlated with T, WS, and H. The ground temperature increased, mainly in summer and at daytime, and the vertical mixing degree increased, thus minimizing the concentration of nitrogen oxides in the low-altitude atmosphere. Higher wind speeds exacerbate the dispersion and mixing of these atmospheric pollutants emitted from local sources, such as automotive engines, thereby minimizing their cumulative concentration in the atmosphere. High pressure was more likely to cause pollution accumulation during the observation period.

Supplementary Materials: The following are available online at http://www.mdpi.com/2073-4433/11/1/30/s1, Table S1: Monitoring sites in Changchun.

Author Contributions: All authors have read and agreed to the published version of the manuscript. L.W. worked for the conceptualization, original draft writing, review and editing of the article. X.T. worked for field sampling and filter analysis. J.W. and C.F. worked on the data curation and methodology and also worked as supervisors and directors of this study. All authors have read and agreed to the published version of the manuscript.

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

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