

# Distribution Characteristics of Six Criteria Air Pollutants Under Different Air Quality Levels in Cangzhou City, China

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**Abstract:** The problem of urban air pollution has caused widespread concern and solving the problem of air pollution has become a primary research focus. Cangzhou is one of the "2+26" cities in the air pollution transmission channel in the Beijing-Tianjin-Hebei (BTH) region, and its regional advantage is obvious. To study the distribution characteristics of major air pollutants, the air quality index (AQI) and mass concentrations of six criteria air pollutants, including PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub>, CO and O<sub>3</sub>, from 2014 to 2018 were used. Furthermore, by employing the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, the air pollutant concentration level, temporal variations and air mass trajectory characteristics under different air quality levels in Cangzhou city were analysed. The results showed that the mass concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub> and CO and PM<sub>2.5</sub>/PM<sub>10</sub> increased successively with increasing pollution level, while the mass concentration of O<sub>3</sub> was at a level of slight pollution, which first increased and then decreased. In the case of serious pollution, PM<sub>2.5</sub> and PM<sub>10</sub> were 3.3 and 2.4 times the Chinese Ambient Air Quality Standard (CAAQS) Grade II standard, respectively, and PM<sub>2.5</sub>/PM<sub>10</sub> was 0.71 times the standard, indicating that as pollution increased, the air pollution gradually became composed of mainly fine particles. The air quality was dominantly good and light, accounting for 73.4% to 84.7% of the total air quality from 2014 to 2018, respectively. The ambient air quality improved annually; the proportion of excellent and good days increased from 42.9% to 63.8%, and the proportion of severe and serious pollution days decreased from 12.2% to 3.7%. The diurnal variations in air pollutants were different under different air quality levels. The air mass trajectory analysis showed that as the pollution level increased, the proportion of eastern and easterly air masses decreased, and the proportion of western and westerly air masses increased gradually. Compared with the CAAQS Grade II standard, the excessive levels of particulate matter increased, and PM<sub>2.5</sub> was the most serious.

**Keywords:** Air Pollutants, Air Quality, Backward Trajectory, Temporal Variations

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## 1. Introduction

The rapid development of industrialization, acceleration of urbanization and continuous increase in automobile ownership has resulted in the discharge of a large number of pollutants into the atmosphere, causing a continuous air quality deterioration in Chinese cities [1, 2]. The problem of air pollution has shifted from local to regional, and pollutants have gradually shifted from single to complex types, so the air pollution control goal has transformed from single pollutant to overall air quality improvement [3, 4]. The Ministry of Environmental Protection and the General Administration of Quality Supervision, Inspection and Quarantine jointly issued the Ambient Air Quality Standard (AAQS, GB 3095-2012) in

February 2012, which stipulated that the basic ambient air pollutants included PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub>, CO and O<sub>3</sub>. These air pollutants not only cause air quality deterioration but also threaten public health [5, 6]. Particulate matter, especially PM<sub>2.5</sub>, can be deposited in different parts of the respiratory tract, accumulating more harmful components and penetrating into the alveoli of the human body, while SO<sub>2</sub>, NO<sub>2</sub> and O<sub>3</sub> stimulate the respiratory tract and cause inflammation, resulting in respiratory diseases, chronic bronchitis, asthma, pulmonary heart disease and lung cancer [7-9]. The problem of urban air pollution has attracted widespread attention from many different groups and governments at all levels. Solving the problem of air pollution has become a primary research focus.

Many studies have focused mainly on the mass concentration levels, temporal and spatial distribution, regional transport and relationship between atmospheric pollutants and meteorological factors [2, 10, 11]. Studies have shown differences in gaseous pollutant concentrations, atmospheric particulate matter concentrations and chemical compositions under different air quality levels [12-15]. In recent years, air pollution in the Beijing-Tianjin-Hebei (BTH) region has had complex and regional characteristics, and haze pollution events have occurred frequently. Cangzhou is located in southeastern Hebei Province and is an important open coastal city in the Bohai Rim region. It is one of the "2+26" cities in the air pollution transmission channel in the BTH region, and its regional advantage is obvious. Studies have been carried out on the air pollutant changes in Cangzhou city [16, 17]. However, in general, studies on the distribution of various air pollutants in a long time series with different air quality levels as the starting point have rarely been reported. For this reason, based on the monitoring data of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub>, CO, O<sub>3</sub> and the air quality index (AQI) in

Cangzhou from January 2014 to December 2018, this article discusses the distribution of air pollutants and air mass trajectory characteristics under different air quality levels to provide a reference for the prevention and control of urban air pollution.

## 2. Materials and Methods

### 2.1. Data Source and Air Quality Levels

The monitoring data were obtained from the observed urban air quality data released in real time by the China National Environmental Monitoring Center and include the AQI, PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub>, CO and O<sub>3</sub> from January 2014 to December 2018. According to the National Environmental Protection Standard of the Technical Regulations on Ambient Air Quality Index (on trial, HJ 633-2012), the classification of air quality levels and the concentration limits of corresponding air pollutants are shown in Table 1.

**Table 1.** AQI and limits for the mass concentrations of atmospheric pollutants under different air quality levels.

AQI	Levels	PM <sub>2.5</sub> , 24 h (μg/m <sup>3</sup> )	PM <sub>10</sub> , 24 h (μg/m <sup>3</sup> )	SO <sub>2</sub> , 24 h (μg/m <sup>3</sup> )	NO <sub>2</sub> , 24 h (μg/m <sup>3</sup> )	CO, 24 h (mg/m <sup>3</sup> )	O <sub>3</sub> , 1 h (μg/m <sup>3</sup> )
0-50	Excellent	0-35	0-50	0-50	0-40	0-2	0-160
51-100	Good	35-75	50-150	50-150	40-80	2-4	160-200
101-150	Slight	75-115	150-250	150-475	80-180	4-14	200-300
151-200	Moderate	115-150	250-350	475-800	180-280	14-24	300-400
201-300	Severe	150-250	350-420	800-1600	280-565	24-36	400-800
>300	Serious	>250	>420	>1600	>565	>36	>800

### 2.2. Backward Air Mass Trajectory

Backward air mass trajectories that reached the study area were calculated using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model with archived National Centers for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) global reanalysis meteorological data [18, 19]. This mode has a relatively complete process of transport, diffusion and settlement and is an efficient and quick method to analyse the source of pollutants and determine the transmission path. At present, the HYSPLIT trajectory model has been widely used at home and abroad to study the diffusion and transport of pollutants [12, 20, 21]. The author calculated the backward air mass trajectory for 36 h from January 1, 2014, to December 31, 2018, using the HYSPLIT V4.9 model. The backward trajectory data corresponding to different air quality levels were selected from the daily backward trajectory data results, and then the backward trajectory was clustered according to the speed and direction of air trajectory transmission.

## 3. Results and Discussions

### 3.1. Mass Concentration of Air Pollutants Under Different Air Quality Levels

The air quality levels were determined according to the AQI

(Table 1), and mass concentrations of air pollutants under different air quality levels were calculated, as shown in Figure 1. As the air quality level increased, the concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub> and CO increased successively, while the concentration of O<sub>3</sub> increased first and then decreased, and the slight pollution level was the turning point. If the average mass concentration of air pollutants corresponding to the excellent level was used as the reference standard, the amounts of increase in different air pollutants were calculated. From excellent to serious pollution levels, the concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub> and CO increased by 11.0, 8.4, 3.9, 1.9 and 4.5, respectively, and particulate matter displayed the largest increase. Among them, the concentrations of PM<sub>2.5</sub> and PM<sub>10</sub> for severe pollution were 249.0 and 365.3 μg/m<sup>3</sup>, respectively, which were 3.3 and 2.4 times the Chinese Ambient Air Quality Standard (CAAQS) Grade II standard, while the mass concentrations of SO<sub>2</sub>, NO<sub>2</sub> and CO were 82.2, 79.6 μg/m<sup>3</sup> and 3.5 mg/m<sup>3</sup>, respectively, which were lower than the CAAQS Grade II standard, indicating that as the pollution level increased, particulate matter pollution was the most prominent, and the range of increase was the largest.

To further explain the degree of correlation between the air quality level and air pollutants, Table 2 shows the correlation coefficients between the AQI and air pollutant concentrations. As shown in Table 2, the AQI was significantly correlated with particulate matter regardless of air quality level ( $P < 0.01$ ). At the same time, when the air quality level was high (excellent, good and slight pollution), gaseous pollutants were

also strongly correlated with the AQI, but the correlation degree was lower than that of  $PM_{2.5}$  and  $PM_{10}$ , indicating that the AQI was mainly affected by particulate matter.

The correlation between the AQI and different particulate matter also showed some differences. Under excellent and good air quality levels, the correlation coefficient between the AQI and  $PM_{10}$  was higher than that between the AQI and  $PM_{2.5}$ . As the air quality level increased from slight to serious pollution, the correlation between AQI and  $PM_{2.5}$  was higher

than that for  $PM_{10}$ , indicating that as pollution increased, it gradually became dominated by fine particles; in particular, in the case of serious pollution, the correlation coefficient reached 0.84, which indicates the current pollution situation dominated by fine particles in Cangzhou city. Since  $SO_2$  and  $NO_2$  are the main precursors of  $PM_{2.5}$  and can convert into particulate pollutants in the form of sulfate and nitrate through chemical reaction in the atmosphere, it is also necessary to further control the emission sources of  $SO_2$  and  $NO_2$  [22].

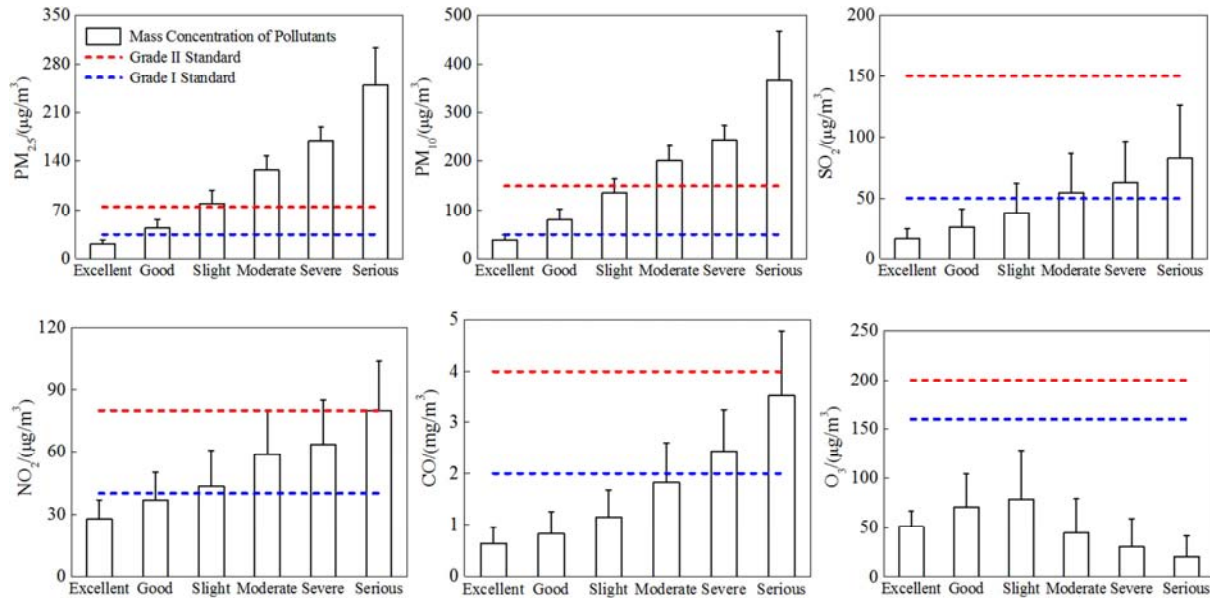


Figure 1. Mass concentrations of air pollutants under different air quality levels.

The mass concentration of particulate matter ratio ( $PM_{2.5}/PM_{10}$ ) can quantitatively represent the pollution degree of a city [23].  $PM_{2.5}/PM_{10}$  varied significantly under different air quality levels, ranging from excellent to severe pollution in the order of 0.55, 0.56, 0.60, 0.65, 0.71 and 0.71. The proportion of fine particles in the particulate matter increased with increasing air pollution levels, which was consistent with the research results [14]. In the cases of severe and serious pollution,  $PM_{2.5}/PM_{10}$  values were higher than 0.70 and were mainly distributed in winter, accounting for 71.2% of the

pollution and indicating that gaseous pollutants such as  $SO_2$ ,  $NO_x$  and  $NH_3$  that are discharged during heating generate secondary aerosols through heterogeneous reactions. At the same time, meteorological conditions were not conducive to the diffusion of air pollutants, resulting in an increase in the mass concentration of  $PM_{2.5}$ , and  $PM_{2.5}/PM_{10}$  was higher than other air quality grades; therefore, attention should be given to fine particle pollution and its harm to human health in the cases of severe and serious pollution [24].

Table 2. Correlation coefficients of the AQI and air pollutants under different air quality levels.

Category	$PM_{2.5}$	$PM_{10}$	$SO_2$	$NO_2$	CO	$O_3$
Excellent	0.721**[a]	0.862**	0.268**	0.268**	0.167	0.128
Good	0.738**	0.770**	0.215**	0.063	0.168**	0.301**
Slight pollution	0.671**	0.579**	0.212**	0.172**	0.326**	-0.159**
Moderate pollution	0.664**	0.454**	0.027	0.292**	0.116	-0.123
Severe pollution	0.684**	0.433**	0.109	0.086	0.162	-0.124
Serious pollution	0.840**	0.555**	0.06	0.267	0.326*[b]	-0.095

[a] \*\* denotes a significant correlation at the 0.01 level (double-tailed). [b] \* denotes a significant correlation at the 0.05 level (double-tailed).

### 3.2. Temporal Variations in Air Pollutants Under Different Air Quality Levels

#### 3.2.1. Interannual and Seasonal Variations

Figure 2 shows the interannual and seasonal changes in the days corresponding to different air quality levels from 2014 to

2018. From 2014 to 2018, the air quality was dominated by good and slight pollution, with a sum of 265 to 309 days, accounting for 73.4% to 84.7% of the whole year. If the levels of excellent and good are used as the air quality standard, the total excellent and good days were 155 and 229 days, respectively, with the proportion increasing from 42.9% in

2014 to 63.6% in 2016, then decreasing to 62.7% in 2017 and increasing to 63.8% in 2018, showing an overall increasing trend. The sum of severe and serious pollution days showed the opposite pattern of change, namely, decreasing from 44 days (2014) to 13 days (2018), and the proportion decreased from 12.2% to 3.7%, which was an overall decreasing trend overall. Overall, from 2014 to 2018, the ambient air quality in Cangzhou improved annually, indicating that the air pollution prevention and control measures implemented by relevant departments improved the air quality in recent years.

The four seasons were defined as follows: spring is from March to May, summer is from June to August, autumn is from September to November, and winter is from December to February of the following year. The air quality in the four seasons was dominated by good and slight pollution levels, accounting for 88.2%, 89.6%, 73.6% and 61.5% in

summer>spring>autumn>winter, respectively, which was similar to the distribution throughout the year. Different seasons had obvious differences in ambient air quality. The sum of excellent and good days was the highest in summer, which was 324 days, followed by autumn and spring, which were 282 days and 247 days, respectively, and the lowest occurred in winter, which was 194 days. The sum of severe and serious pollution days was in the following order: winter (89 days)>autumn (22 days)>spring (13 days)>summer (1 day). The air quality was the best in summer and the worst in winter, which was related to factors such as more precipitation in summer and meteorological conditions conducive to the removal of pollutants, while less precipitation in winter, poor meteorological conditions and increased likelihood of temperature inversion were not conducive to the diffusion of pollutants.

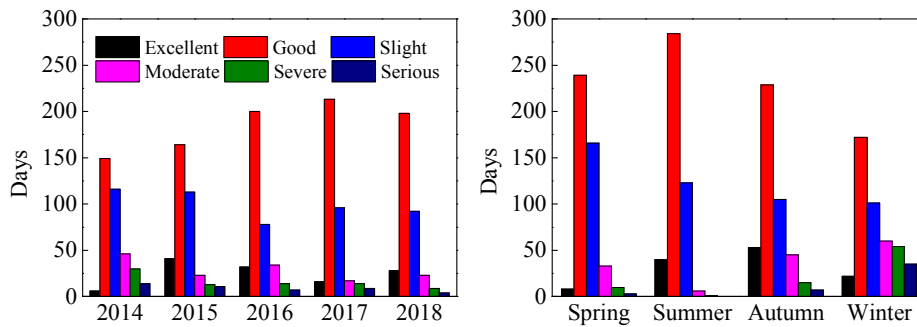
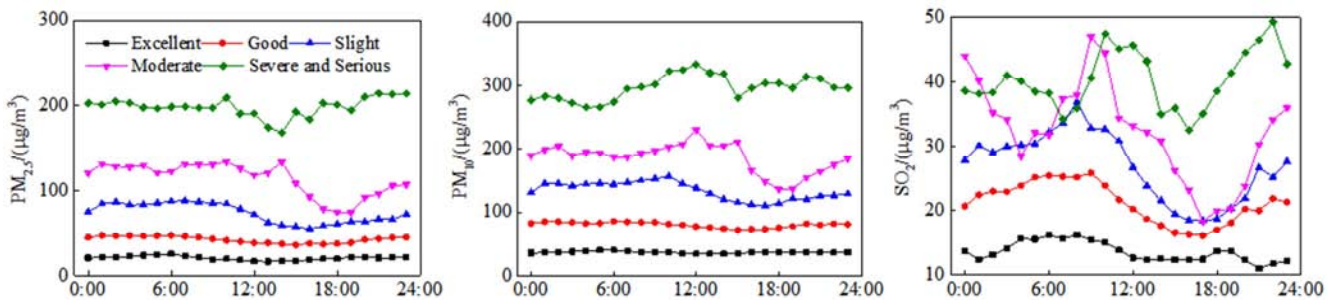


Figure 2. Interannual and seasonal variations in days corresponding to different air quality levels.

### 3.2.2. Diurnal Variations

Since the number of days with severe pollution accounts for a small proportion, the number of days with severe and serious pollution was combined to calculate the daily variations in air pollutant concentrations under different air quality levels, as shown in Figure 3. As the pollution level increased from excellent to moderate pollution, the amplitudes of the diurnal variation in different air pollutants gradually increased. Under different air quality levels, the diurnal variations in  $PM_{2.5}$  and  $PM_{10}$  were similar. When the air quality was excellent and good, the concentrations of  $PM_{2.5}$  and  $PM_{10}$  remained at low levels and changed steadily. With increasing pollution level,

the duration of the low levels decreased gradually. The daily variations in  $SO_2$  concentration showed certain distribution differences. In the cases of moderate, severe and serious pollution, a “double valley” distribution occurred, and the other categories showed a “single valley” distribution. The diurnal variation in  $CO$  concentration formed a single peak. The diurnal variation in  $O_3$  concentration showed a single peak, and as the pollution level increased, the occurrence of the peak was delayed. The diurnal variation in  $NO_2$  concentration showed a “single valley” distribution, and the duration of the trough gradually decreased and showed a good negative correlation with  $O_3$ .





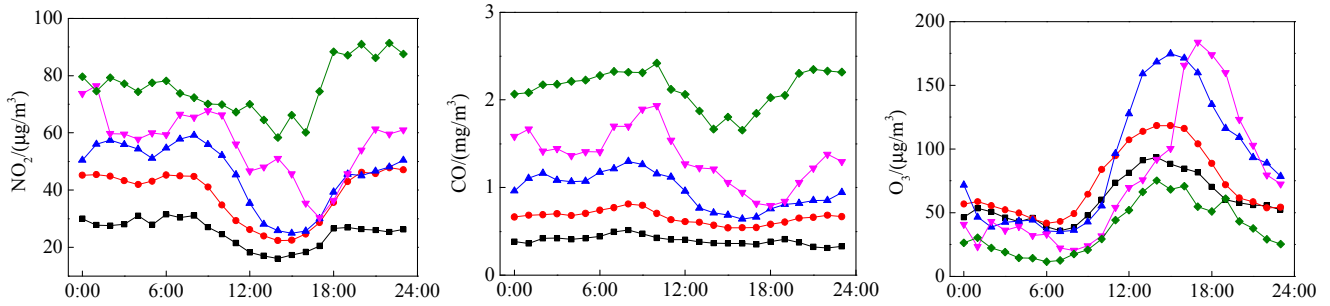


Figure 3. Diurnal variations in air pollutants under different air quality levels.

### 3.3. Backward Air Mass Trajectory Analysis

Figure 4 shows the backward trajectory and proportion of representative air masses under different air quality levels. There were some differences in the dominant air mass types under different air quality levels. When the air quality was excellent, the northwestern and eastern air masses (trajectories 1, 2 and 3) were dominant, accounting for 79%. When the air quality was good, the southern and northwestern air masses (trajectories 1, 2 and 3) were dominant, accounting for 90%. In the case of slight pollution, the southeastern (trajectory 3), southwestern (trajectory 5) and western air masses (trajectory 2) were dominant, accounting for 69%. For moderate pollution, the southern and northwestern air masses (trajectories 1 and 2) were dominant, accounting for 60%. In

the cases of serious and severe pollution, the northwestern and western air masses (trajectories 2 and 1) were dominant, accounting for 44% and 51%, respectively.

In terms of air mass types, when the air quality was excellent, good and slightly polluted, they were all affected by the northeastern air mass (trajectories 4, 4 and 6), accounting for 14%, 10% and 5%, respectively. Moreover, when the air quality was excellent and there was slight pollution, the air masses were also affected by marine air masses (trajectories 5 and 6). In contrast, when the air quality was moderate, severe and serious, the air mass types were mainly northwestern, southwestern and western. In general, as the air quality pollution levels increased, the proportions of eastern and easterly air masses decreased, while those of western and westerly air masses increased gradually.

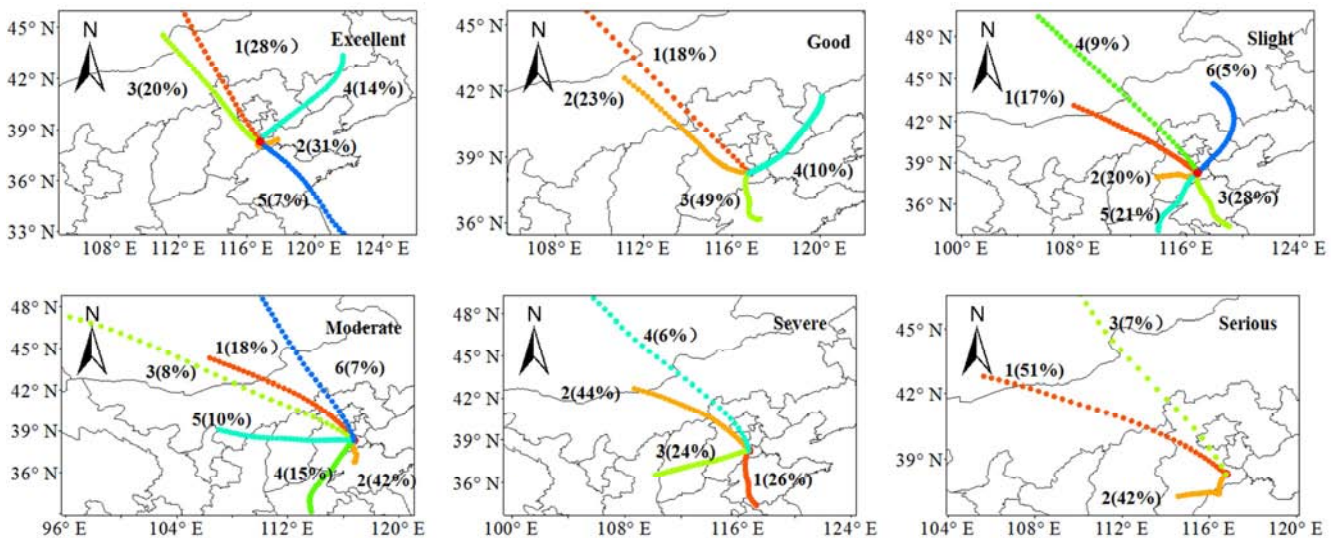


Figure 4. Backward trajectories of air mass under different air quality levels.

Under the excellent and good levels, the concentrations of six air pollutants did not exceed the CAAQS Grade II standard. Therefore, the concentrations of air pollutants corresponding to different types of air masses under slight to severe levels were calculated, as shown in Table 3. Compared with the CAAQS Grade II standard, the particulate matter exceeded the standard to varying degrees. From slight to serious pollution levels, the  $PM_{2.5}$  concentrations that exceeded the standards and corresponded to different air mass trajectories were 0.05-0.16, 0.60-0.76, 1.17-1.33 and 2.10-2.43, respectively.

From moderate to severe pollution levels, the  $PM_{10}$  concentrations that exceeded the standards were 0.29-0.38, 0.58-0.76, and 1.16-1.54, respectively. For the slight pollution level, only the northwestern long-distance air mass corresponding to the  $PM_{10}$  concentration exceeded the CAAQS Grade II standard (0.05 times), but the occurrence frequency was only 9%. As the pollution level increased, the degree to which the CAAQS Grade II standard of particulate matter was exceeded increased, and the amount that the  $PM_{2.5}$  concentration exceeded was the most serious. In the case of

slight pollution, the corresponding PM<sub>2.5</sub> concentration of the northwestern air mass was significantly higher than that of the southwestern and southeastern air masses ( $p < 0.01$ ), and the corresponding PM<sub>2.5</sub>/PM<sub>10</sub> ratio was the highest (0.63), indicating that the air mass was dominated by fine particles. The corresponding PM<sub>10</sub> concentration of the northwestern long-distance air mass was significantly higher than that of other air masses ( $p < 0.01$ ), and its corresponding PM<sub>2.5</sub>/PM<sub>10</sub> was the lowest (0.53), indicating that this air mass was mainly composed of coarse particles. For gaseous pollutants, only in

the case of serious pollution did the corresponding NO<sub>2</sub> concentration of the northwest long-distance air mass exceed the CAAQS Grade II standard, by a multiple of 0.04 and accounting for 51%. In other cases, it did not exceed the CAAQS Grade II standard. This result showed that air pollution was mainly particulate matter pollution under different air quality levels, which was consistent with the result that the particulate matter seriously exceeded the standard.

**Table 3.** Mass concentrations of air pollutants corresponding to the backward trajectories of air masses under different air quality levels.

Types	Levels	1	2	3	4	5	6	Levels	1	2	3	4
PM <sub>2.5</sub> (μg/m <sup>3</sup> )	Slight	86.8	82.1	76	81.5	78.6	79.4	Severe	174.9	167.6	170.1	162.4
PM <sub>10</sub> (μg/m <sup>3</sup> )		141.1	134.6	123.9	157.4	139.4	133.5		237	248.5	238.4	263.6
SO <sub>2</sub> (μg/m <sup>3</sup> )		47.6	37.9	34.2	42.5	35.7	31.7		57.3	59.9	74.9	59
NO <sub>2</sub> (μg/m <sup>3</sup> )		53.2	45.3	37.5	48.1	37.7	46.7		50.5	67.5	67.9	74.4
CO (mg/m <sup>3</sup> )		1.5	1.2	0.9	1.5	1	1.2		2.2	2.6	2.3	2.9
O <sub>3</sub> (μg/m <sup>3</sup> )		42.3	69.8	105.6	39.4	101.9	54.7		40.2	25.5	31.6	20.2
PM <sub>2.5</sub> /PM <sub>10</sub>	Moderate	0.63	0.62	0.62	0.53	0.57	0.6	Serious	0.74	0.68	0.73	0.62
PM <sub>2.5</sub> (μg/m <sup>3</sup> )		128	127.3	131.8	131.9	127	120.1		244.7	256.9	232.3	/
PM <sub>10</sub> (μg/m <sup>3</sup> )		193.6	202.4	206.7	203.6	196.3	206.6		358	380.7	324	/
SO <sub>2</sub> (μg/m <sup>3</sup> )		66.5	53	56.5	52.9	43.8	56.2		75.9	88.5	90.3	/
NO <sub>2</sub> (μg/m <sup>3</sup> )		64.8	57.3	60	55.1	59.9	54.7		83.6	76.6	70.3	/
CO (mg/m <sup>3</sup> )		2.2	1.7	2.3	1.5	1.8	1.9		3.8	3.3	3.1	/
O <sub>3</sub> (μg/m <sup>3</sup> )		28.1	54.8	24.8	59.3	35.9	36.5		15.1	26.9	22.7	/
PM <sub>2.5</sub> /PM <sub>10</sub>		0.74	0.68	0.73	0.62	0.74	0.68		0.7	0.71	0.72	/

## 4. Conclusion

As the air quality level increased, the concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub>, CO and PM<sub>2.5</sub>/PM<sub>10</sub> increased, while the concentration of O<sub>3</sub> first increased and then decreased, and the slight pollution level was the turning point. From excellent to severe pollution, the concentrations of PM<sub>2.5</sub> and PM<sub>10</sub> increased by 11.0 and 8.4 times, respectively, and the particulate matter concentration increased the most. For severe pollution, the concentrations of PM<sub>2.5</sub> and PM<sub>10</sub> were 249.0 and 365.3 μg/m<sup>3</sup>, respectively, which were 3.3 and 2.4 times the CAAQS Grade II standard, and the PM<sub>2.5</sub>/PM<sub>10</sub> ratio was 0.71, with fine particle pollution dominating.

The ambient air quality in Cangzhou city mainly reached good and slight pollution levels, accounting for 73.4% - 84.7%. From 2014 to 2018, the ambient air quality improved annually. Among them, the total excellent and good days increased, and the proportion increased from 42.9% to 63.8%, while the total severe and serious pollution days decreased, and the proportion decreased from 12.2% to 3.7%. The number of excellent and good days was the highest in summer, the air quality was the best, the number of days of severe and serious pollution was the highest in winter, and the air quality was the worst.

As the air quality level increased, the diurnal variations in different air pollutants were different. The diurnal variations in PM<sub>2.5</sub> and PM<sub>10</sub> concentrations were similar, and the duration of the low level decreased gradually as the pollution degree increased. As the pollution degree increased, the

concentration of SO<sub>2</sub> changed from a “single valley” distribution to a “double valley” distribution. The diurnal variation in O<sub>3</sub> concentration shows a single peak, and as the pollution level increased, the occurrence of the peak was delayed. The diurnal variation in NO<sub>2</sub> concentration showed a “single valley” distribution, and the duration of the trough gradually decreased and had a good negative correlation with O<sub>3</sub>.

The backward trajectories of the air masses showed differences in the type and proportion of dominant air masses under different air quality levels. As the pollution degree increased, the proportion of eastern and easterly air masses decreased, and the proportion of western and westerly air masses gradually increased. Compared with the CAAQS Grade II standard, the degree to which the particulate matter exceeded the standard increased, and the PM<sub>2.5</sub> concentration showed the most serious exceedance.

This study mainly focused on the distribution of air pollutants and the characteristics of air mass trajectory under different air quality levels, providing a basis for the study of air pollution characteristics and control measures in Cangzhou area. Future research should take into account the impact of different emission sources on pollutants and the change of chemical composition of particulate matter under different air quality levels.

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