

# Quantitative anatomy of characteristics and influencing factors of PM<sub>2.5</sub> and O<sub>3</sub> in Liaoning province of China\*

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Northeast China is an important region in Asia, bordered by Siberia to the north and Bohai Sea and Yellow Sea to the south. Liaoning province is one of the most important industrial and agricultural bases in China. It is located in the south of Northeast China. Due to the unique characteristics of geography, climate and anthropogenic emissions, it is of great significance to explore the air pollution in Liaoning Province. In this paper, spatial association network feature analysis, spatial interpolation, standard deviation ellipse and exploratory spatial data analysis are used to analyze the temporal and spatial evolution characteristics and influencing factors of PM<sub>2.5</sub> and O<sub>3</sub> in Liaoning Province. The results demonstrate that PM<sub>2.5</sub> concentration has a decreasing trend in all cities, while O<sub>3</sub> concentration has no obvious decreasing trend. The high concentration of PM<sub>2.5</sub> is mainly distributed in central and northern of Liaoning, while high concentration of O<sub>3</sub> is in central-western and coastal cities. PM<sub>2.5</sub> and O<sub>3</sub> show opposite seasonal dynamic characteristic, which is mainly due to their seasonal anthropogenic emissions, meteorological factors conducive to pollutant generation and geographical conditions unfavorable to the diffusion of air pollutants. Moreover, PM<sub>2.5</sub> and O<sub>3</sub> have certain spatial correlation with economic factors, such as agriculture, industry, tertiary industry and population density. The results of this study enable a more comprehensive understanding of the temporal and spatial distribution characteristics and the relative influencing factors of PM<sub>2.5</sub> and O<sub>3</sub> in Liaoning Province. This provides a policy basis for regional joint prevention and control and collaborative air pollution control in Northeast China.

KEYWORDS AND PHRASES: PM<sub>2.5</sub>, O<sub>3</sub>, quantitative anatomy.

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## 1. Introduction

China is the largest developing country in the world and has achieved remarkable economic growth in the past decades. However, this is also accompanied by serious air pollution. In particular, PM<sub>2.5</sub> and O<sub>3</sub> are the two most serious air pollutants [1]. PM<sub>2.5</sub> refers to fine particulate matter with a diameter less than or equal to 2.5  $\mu\text{g}/\text{m}^3$ , which profoundly affects the environment and public health in different ways by hanging in the air for a long time, including human health [2], climate [3], agriculture and other ecosystems [4] and visibility [5]. O<sub>3</sub> is a photochemical pollutant, potentially harmful to human health [2]. In order to cope with the serious air pollution problems, China's State Council has implemented a series of emission control policies to improve air quality in the last decade, including the Action Plan of Air Pollution Prevention and Control (2013–2017) [6], and the Three-Year Action Plan to Win the Battle for a Blue Sky (2018–2020) [7]. Thanks to the measures, the concentration of PM<sub>2.5</sub> in many parts of China reduced greatly during this period [8, 9, 10, 11]. However, it is worth noting that the concentration of O<sub>3</sub> increased significantly in most regions simultaneously [12, 13, 14]. For example, the increase rates of O<sub>3</sub> concentration in the North China Plain, Yangtze River Delta and Pearl River Delta are about 27%, 19% and 8%, respectively [15]. It can be seen from their chemical reaction mechanism that they all share common precursor, i.e., VOCs and NO<sub>x</sub>. And there may be a relationship between their pollution conditions [16]. PM<sub>2.5</sub> and O<sub>3</sub> dominate air quality by interacting with each other through photochemical reactions [17, 18]. PM<sub>2.5</sub> and O<sub>3</sub> are chemically coupled, and this coupling has profound implications for understanding the processes that control their levels [19]. Therefore, PM<sub>2.5</sub> and O<sub>3</sub> are considered to have a significant correlation. They may have collaborative effects and become more complex mixed pollutants, which may bring greater harm to national production and life [20, 21]. Effective reduction of the two pollutants also becomes more complex and difficult. In order to take correct reduction measures and effectively reduce and control these two new complex mixed pollutants, it is very important to deeply understand the spatial-temporal dynamic characteristics, collaborative change rules and possible influencing factors of these two new complex mixed pollutants.

Based on these studies, China is one of the world's worst polluters of PM<sub>2.5</sub> and O<sub>3</sub> [22, 23, 24]. PM<sub>2.5</sub> and O<sub>3</sub> have been studied on their variation, correlation and potential influencing factors at local, regional and national levels, aiming to provide guidance for collaborative control in many aspects [25, 26, 27, 28]. Nevertheless, the collaborative control of PM<sub>2.5</sub> and

O<sub>3</sub> still faces many obstacles, partly because their relationship is complex and difficult to quantify, and the reaction mechanisms have not been fully identified [29, 30]. Many studies concentrated on the characteristics and influence mechanism of PM<sub>2.5</sub> and O<sub>3</sub> in recent years. From the regional perspective, the areas with heavy PM<sub>2.5</sub> pollution in winter were usually the same as those with heavy O<sub>3</sub> pollution in summer. In some areas with obvious PM<sub>2.5</sub> pollution alleviation, the deterioration of O<sub>3</sub> pollution was severe. For instance, the Beijing-Tianjin-Hebei region and its surrounding areas [31], which may be caused by the sensitivity of O<sub>3</sub>-NO<sub>x</sub>-VOCs [32, 33]. From the temporal perspective, it was found that PM<sub>2.5</sub> and O<sub>3</sub> concentrations in many cities were related to seasons. They were positively correlated on the warm season. It was because the higher O<sub>3</sub> concentration on the warm season would significantly promote the generation of secondary aerosols, which was conducive to PM<sub>2.5</sub> generation. In cold season, it was the opposite; they showed as negatively correlated, mainly because atmospheric oxidation was weak, and the high concentration of PM<sub>2.5</sub> in winter weakened solar radiation and inhibited atmospheric photochemical reaction, not conducive to the generation of O<sub>3</sub> [34, 35, 36, 37, 38]. Meteorological factors played a crucial role in the interaction between PM<sub>2.5</sub> and O<sub>3</sub> [39, 40]. Dai et al. [41] found that the co-pollution time of PM<sub>2.5</sub> and O<sub>3</sub> was closely related to relative humidity, surface air temperature and wind speed in the Yangtze River Delta. In addition, numerous experimental studies on PM<sub>2.5</sub> and O<sub>3</sub> have been quantitatively conducted. To study the relationship between air pollution and other factors, including underway measurements, 3D numerical simulation and smog chamber, would require expensive large-scale experiments. Requirements on hardware, operators and time, and insufficient understanding of the complex physical and chemical reactions between PM<sub>2.5</sub> and O<sub>3</sub>, which would inevitably lead to simulation errors [42, 43, 44]. Distributed lag nonlinear model (DLNM) showed good performance when it was used to analyze the effects of various influencing factors on PM<sub>2.5</sub> and O<sub>3</sub> [45, 46]. Collaborative control and effective management of PM<sub>2.5</sub> and O<sub>3</sub> pollution was the top concern of China's 14th Five-Year Plan (2020–2025) [47]. The multi-technology, multi-scale and multi-angle analysis results could provide a basis for the characteristics of air pollution in providing scientific support for the joint prevention and control of PM<sub>2.5</sub> and O<sub>3</sub> in the new stage.

Northeast China is an important region in Asia, bordered by Siberia to the north, Bohai Sea and Yellow Sea to the south. Most of the area is surrounded by three mountains, which are not conducive to the diffusion of air pollutants. Liaoning province lies in the south of northeast China. It is cold

here in winter, and heating is needed for half a year [48, 49]. Heating fuels include coal, biomass, natural gas and burning of agricultural straw, which release more air pollutants than other regions [50, 51, 52, 53]. Liaoning is also the heavy industrial base of China's steel, petroleum, petrochemical, shipbuilding, machine tools and other industries. Most of these heavy industries are traditional industries with high-energy consumption and high-pollution, which is another important cause of air pollution. Although China has implemented a series of air quality control measures, which have greatly improved PM2.5 concentration in northeast, O3 pollution still exists. Based on the unique geographical, climate, anthropogenic characteristics and heavy industrial structure, understanding the distribution characteristics and influencing factors of PM2.5 and O3 in the region have become one of the key problems of air pollution prevention and control in the northeast [54, 55]. Therefore, this paper analyzes the spatial-temporal evolution characteristics and spatial influencing factor of PM2.5 and O3 in Liaoning province, based on spatial association network feature analysis, spatial interpolation, standard deviation ellipse and exploratory spatial data analysis. This is an attempt to provide reference basis for regional joint prevention and control and collaborative air pollution control in Northeast China.

## 2. Materials and methods

### 2.1. Study area

Liaoning province is  $118^{\circ}53' \sim 125^{\circ}46'E$ ,  $38^{\circ}43' \sim 43^{\circ}26'N$ . The terrain of the region is roughly sloped from north to south, from east and west to the middle. The east and the west of the region are mountains and hills, descending into the central plains, taking a horseshoe shape and sloped into Bohai Sea. Liaoning has a temperate monsoonal climate, the winter is cold and long, lasting 4–6 months, the minimum monthly mean temperature is  $-12^{\circ}\text{C}$  to  $-19^{\circ}\text{C}$ , and lots of precipitation in summer. The studies have suggested that heating fuels has become one of the main sources of air pollution in the region, for instance, coal, biomass, natural gas and burning of agricultural straw. Burning of agricultural straw is used for rural heating in late autumn and early spring tillage, leading to a large number of air pollutants being directly released into the atmosphere [50, 51, 52, 53]. Liaoning is the most populous province in northeast China, which is an important base of commodity grain, heavy industry, animal husbandry and forestry in the region. Liaoning Province consists of 14 cities, which are Chaoyang, Fuxin, Shenyang, Tieling, Liaoyang, Anshan, Benxi, Fushun, Huludao, Jinzhou,

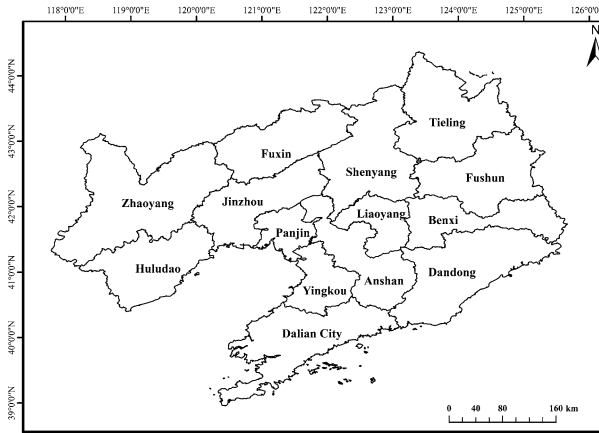


Figure 1: Geographical locations of 14 cities in Liaoning province.

Panjin, Yingkou, Dandong and Dalian. The locations of these 14 cities are showed in Fig. 1 and Table 1.

## 2.2. Data sources

Daily data of PM<sub>2.5</sub> and O<sub>3</sub> in 14 cities from January 1, 2016 to December 31, 2020 were downloaded from the China Air Quality Monitoring Platform. According to the environmental air quality standard (GB 3095-2012) modification list about the requirement in China, Table 2 was the concentration limits of PM<sub>2.5</sub> and O<sub>3</sub> at different levels. PM<sub>2.5</sub> was evaluated by daily average, while O<sub>3</sub> was evaluated by 8h. Economic statistics data in 2019 were from the Statistical Yearbook of Liaoning Province (<http://tjj.ln.gov.cn/>).

## 2.3. Method

Spatial-temporal dynamic analysis is an important method to accurately recognize, evaluate and comprehensively understand the spatial-temporal characteristics and interactions of pollutant distribution [56]. The spatial and temporal distribution characteristics and influencing factors of PM<sub>2.5</sub> and O<sub>3</sub> in Liaoning province were analyzed based on spatial association network feature analysis, spatial interpolation method, standard deviation ellipse method and exploratory spatial data analysis method.

Table 1: 14 cities in Liaoning province

City number	City name	Latitude/degree	Longitude/degree
1	Chaoyang	41.58 N	120.42 E
2	Fuxin	42.00 N	121.65 E
3	Shenyang	41.80 N	123.38 E
4	Huludao	40.71 N	120.84 E
5	Jinzhou	41.13 N	121.15 E
6	Panjing	41.12 N	122.07 E
7	Anshan	41.11 N	122.99 E
8	Liaoyang	41.27 N	123.24 E
9	Benxi	41.3 N	123.73 E
10	Fushun	41.97 N	123.97 E
11	Tieling	42.32 N	123.85 E
12	Dandong	40.13 N	124.37 E
13	Dalian	38.92 N	121.62 E
14	Yingkou	40.65 N	122.18 E

Table 2: The concentration limits at different levels for PM2.5, O3 and AQI. Unit:  $\mu\text{g}/\text{m}^3$  for PM2.5 and O3

AQI levels	AQI value	Air quality level	PM2.5	O3
<i>I</i>	0–50	good	0–35	0–100
<i>II</i>	51–100	moderate	36–75	101–160
<i>III</i>	101–150	lightly polluted	76–115	161–215
<i>IV</i>	151–200	moderately polluted	116–150	216–265
<i>V</i>	201–300	heavily polluted	151–250	266–800
<i>VI</i>	>300	Severely polluted	>250	>800

**2.3.1. The standard deviation ellipse method** The temporal dynamics process of PM2.5 and O3 in Liaoning Province was characterized by standard deviation ellipse method. It was mainly implemented through ArcGIS 10.5. The specific parameters include centrality, distribution, density, orientation and shape in the standard deviation ellipse method [57]. The overall characteristics of pollutants about temporal dynamic process is analyzed by comparing the changes of these parameters over time.

The elliptic spatial distribution range represents the main area of spatial distribution of geographical elements, the center represents the relative position of its distribution, the azimuth Angle reflects the main trend direction of its distribution, and the long axis represents the degree of dispersion in the main trend direction [58]. The comparison of ellipses with different basic parameters such as size and orientation can provide the difference information of different spatial distributions. Calculation of spatial differentiation coefficient can quantitatively describe the degree of spatial differentiation

among different distributions [58]. The specific formula is as follows:

$$\bar{X} = \sqrt{\frac{\sum_{i=1}^n (x_i - \bar{X})^2}{n}}, \bar{Y} = \sqrt{\frac{\sum_{i=1}^n (y_i - \bar{Y})^2}{n}},$$

where,  $x_i$  and  $y_i$  are the spatial coordinate positions of the research elements,  $n$  is the total number of the research elements, and  $(\bar{X}, \bar{Y})$  is the center of gravity of the ellipse. The direction of the ellipse  $\tan \theta$  is based on the  $x$  axis, north (12 points direction) as 0 degrees, clockwise rotation, formula is as follows:

$$\begin{aligned} \tan \theta &= \frac{A + B}{C}, \\ &\sum_{i=1}^n x_i'^2 - \sum_{i=1}^n y_i'^2, \\ B &= \sqrt{\left(\sum_{i=1}^n x_i'^2 - \sum_{i=1}^n y_i'^2\right)^2 + 4\left(\sum_{i=1}^n x_i' y_i'\right)^2}, \\ C &= 2 \sum_{i=1}^n x_i' y_i', \end{aligned}$$

Here,  $x_i'$  and  $y_i'$  are the coordinate deviation between the research elements  $i$  and the mean center. The calculation formula of the axis standard deviation respectively is

$$\begin{aligned} \sigma_x &= \sqrt{\frac{2 \sum_{i=1}^n (\bar{x} \cos \theta - \bar{y} \sin \theta)^2}{n}}, \\ \sigma_y &= \sqrt{\frac{2 \sum_{i=1}^n (\bar{x} \sin \theta + \bar{y} \cos \theta)^2}{n}}. \end{aligned}$$

The greater of the difference between the long axis and the short axis, the more obvious of the directivity of the research elements; otherwise, the directivity is not obvious.

**2.3.2. Spatial association network feature analysis** Spatial association network feature analysis was to explore the spatial action intensity of PM2.5 and O3 between cities, so as to find the region of pollution source similarity and regional association intensity. The points and lines in the spatial association network constitute the association network of pollutants among

cities. The points represent each city, and the lines represent the spatial association degree of pollutants among cities. The association strength in the network reflects the strength of the relationship between cities [59]. The spatial influence degree of each city is obtained by calculating the spatial association degree of each network node.

**2.3.3. Spatial interpolation** The spatial distribution pattern of PM2.5 and O3 could be studied by spatial interpolation method, to obtain the provincial distribution pattern of PM2.5 and O3 in Liaoning province. Spatial interpolation is a kind of unbiased optimal estimation based on semivariance theory. With the help of ArcGIS10.5, ordinary Kriging interpolation method is adopted to interpolate PM2.5 and O3 mass concentration into continuous data surfaces. Based on uneven distribution of spatial data, the attributes of interpolation points can be comprehensively analyzed [60].

**2.3.4. Exploratory spatial data analysis** The spatial agglomeration and correlation characteristics of PM2.5, O3 and influencing factors in Liaoning province were studied by exploratory spatial data analysis method. Since phenomena in space do not exist in isolation, there is often some correlation, exploratory spatial data analysis can describe the potential interdependence among observation data in the same research area, and provide basis for exploring the spatial-temporal aggregation and evolution of each factor. In order to explore the spatial correlation effect of PM2.5, O3 and each impact factor in Liaoning Province, the multi-factor local Moran model was introduced to explore the spatial agglomeration and correlation characteristics of PM2.5, O3 and each impact factor in Liaoning Province. It conducted based on GeoDa1.2.0. [61]. Its calculation formula is as follows:

$$I_i = \frac{n(x_i - \bar{x})}{\sum_{j=1}^n (x_j - \bar{x})^2} \sum_{j=1}^n w_{ij}(x_j - \bar{x}),$$

where  $I_i$  is the local Moran's  $I$  statistic, which represents the spatial correlation degree between the region  $i$  and its neighboring region.  $x_i$  is the observed value of the region  $i$ .  $w_{ij}$  is the spatial weight matrix. The value is 1 if the region  $i$  is adjacent to the region  $j$ , and 0 otherwise. Z test is adopted for the statistical test of local Moran's  $I$ , that is:

$$Z(I_i) = \frac{I_i - E(I_i)}{\sqrt{var(I_i)}},$$



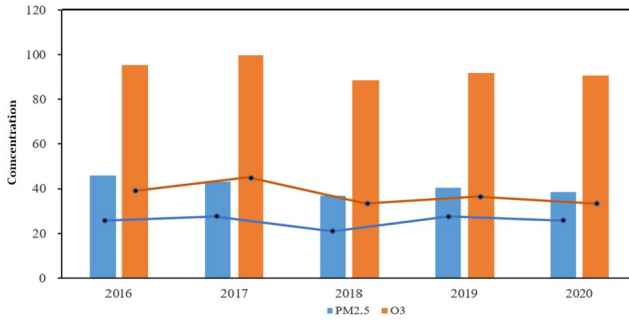


Figure 2: The annual mean values of PM2.5 and O3 in Liaoning province from 2016 to 2020 ( $\mu\text{g}/\text{m}^3$ ).

When given the significance level of the statistic,  $I_i > 0$  and  $Z_i > 0$ , the region is a high-high agglomeration area;  $I_i > 0$  and  $Z_i < 0$ , the region is a low-low agglomeration area;  $I_i < 0$  and  $Z_i > 0$ , the region is a high-low agglomeration area;  $I_i < 0$  and  $Z_i < 0$ , the region is a low-high agglomeration area.

### 3. Results

#### 3.1. Temporal variation characteristics of PM2.5 and O3

The annual mean values of PM2.5 and O3 in 14 cities of Liaoning Province from 2016 to 2020 were calculated (Fig. 2). The annual average concentration of PM2.5 decreased from  $45.9 \mu\text{g}/\text{m}^3$  in 2016 to  $36.7 \mu\text{g}/\text{m}^3$  in 2018, with annual decrease rate of  $4.6 \mu\text{g}/\text{m}^3$ . After fluctuation from 2018 to 2020, it decreased to  $38.6 \mu\text{g}/\text{m}^3$  in 2020. From 2016 to 2018, O3 firstly increased and then decreased. O3 rose again from 2018 to 2020, with large fluctuation range within the year. To sum up, 2018 was an important inflection point for PM2.5 and O3 concentrations in Liaoning Province, reaching the lowest values of  $36.7 \mu\text{g}/\text{m}^3$  and  $88.3 \mu\text{g}/\text{m}^3$ , respectively. The results showed that PM2.5 mass concentration in all cities decreased gradually, and O3 mass concentration in most cities also decreased slightly. With the decrease of PM2.5 mass concentration, the reduction of O3 contribution rate was not obvious, and O3 pollution could not be ignored. According to the Environmental Air Quality Standard (GB3095-2012), PM2.5 and O3 mass concentrations in 14 cities were close to the moderate level.

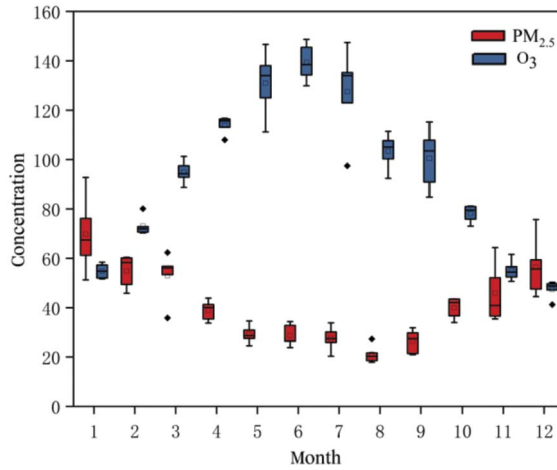


Figure 3: Times series of PM<sub>2.5</sub> and O<sub>3</sub> in Liaoning province from 2016 to 2020 ( $\mu\text{g}/\text{m}^3$ ).

The monthly average concentrations of PM<sub>2.5</sub> and O<sub>3</sub> in Liaoning province from 2016 to 2020 were showed in Fig. 3. PM<sub>2.5</sub> and O<sub>3</sub> concentrations have obvious seasonal patterns, which were winter > spring > autumn > summer, summer > spring > autumn > winter, respectively. The maximum value of PM<sub>2.5</sub> in winter was about 3 times of the minimum value in summer, and the maximum value of O<sub>3</sub> in summer was about 3 times of the minimum value in winter. Monthly variation of PM<sub>2.5</sub> showed an U-shaped trend, while O<sub>3</sub> a significant inverted U-shaped trend (Fig. 3). The monthly O<sub>3</sub> average concentrations presented an increasing trend from January to June and August to September, and a decreasing trend from June to August and from September to December, reaching the maximum value in June eventually. On the contrary, the concentration of PM<sub>2.5</sub> decreased month by month from January to August, rose month by month from August to December, and reaching its maximum value in January. According to the O<sub>3</sub>-8h secondary standard ( $\geq 100 \mu\text{g}/\text{m}^3$ ) and PM<sub>2.5</sub> secondary standard ( $\geq 35 \mu\text{g}/\text{m}^3$ ) in the Environmental Air Quality Standard (GB3095-2012), it could be seen from Fig. 3, the typical period of O<sub>3</sub> pollution was from April to September, and the typical period of PM<sub>2.5</sub> pollution was from January to March and From October to December.

The temporal dynamic process of PM<sub>2.5</sub> and O<sub>3</sub> were quantitatively analyzed by standard deviation ellipse method, through ArcGIS spatial statistics module. The parameter results were showed in Table 3 and Table 4, and the visualization results were showed in Fig. 4.

Table 3: The parameters of standard deviation ellipse with PM 2.5

Item	2016	2017	2018	2019	2020
<i>x</i> coordinate of barycenter (km)	13644971.4	13642724.2	13639876.9	13640191.3	13640482.4
<i>y</i> coordinate of barycenter (km)	5039033.6	5036241.1	5033869.4	5035314.9	5033749.7
Standard deviation along the <i>x</i> -axis (km)	202716.5	208378.7	207785.5	207227.6	206822.7
Standard deviation along the <i>y</i> -axis (km)	159122	157607.9	159777.9	160786.2	160771.9
direction angle (°)	55.8	50.5	49.3	52.2	50.2

Table 4: The parameters of standard deviation ellipse with O3

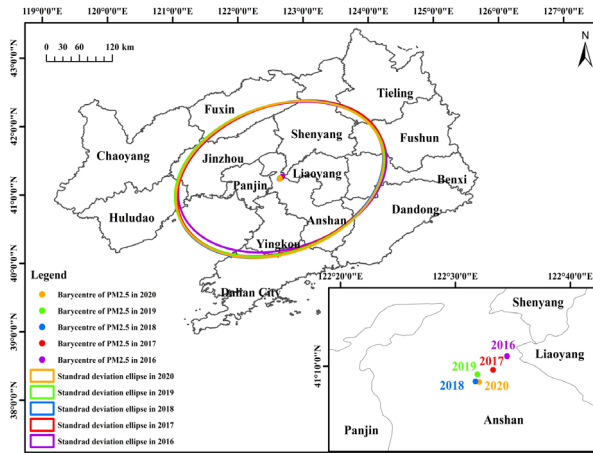
Item	2016	2017	2018	2019	2020
<i>x</i> coordinate of barycenter (km)	13633071.48	13636834.2	13636148.4	13638078.1	13638952.4
<i>y</i> coordinate of barycenter (km)	5037051.2	5035895	5031961	5032725.4	5032935.2
Standard deviation along the <i>x</i> -axis (km)	207267.2	203658.7	203573.7	203957	204839.3
Standard deviation along the <i>y</i> -axis (km)	161767.8	162017.1	163571.6	163297.3	164225.4
direction angle (°)	58.3	61.7	60	59	59.3

The results showed that high concentrations of PM2.5 and O3 were mainly distributed in central and along Bohai Sea of Liaoning province. First of all, there was no significant change with the gravity of PM2.5 and O3 pollutants, which both appeared in northwest Benxi. However, PM2.5 showed a slight trend of southwest shift year by year, while O3 showed a slight trend of southeast shift year by year. The second, the center of standard deviation ellipse of the spatial distribution of PM2.5 and O3 shifted about 5° to the east and 1° to the north, respectively. The overall distribution range of ellipses corresponding to PM2.5 and O3 expanded slightly. The long axis of both of them increased by time. The short axis of PM2.5 increased by time, while the short axis of O3 decreased. The last, the oblateness of the standard deviation ellipse of them gradually increased, which indicated that the direction trend of PM2.5 and O3 was more and more obvious.

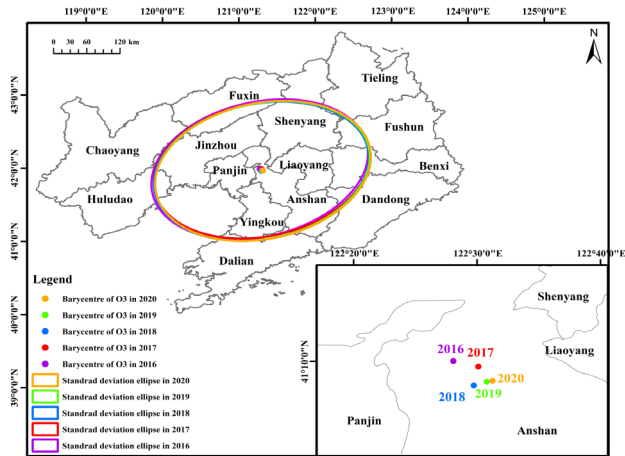
### 3.2. The spatial patterns evolution of PM2.5 and O3

As can be seen from Table 5, areas with high level PM2.5 concentrations were found in the central and southwest of Liaoning Province during 2016–2020, including Jinzhou, Anshan, Shenyang and Huludao. While the areas with high concentrations of O3 were located in coastal cities in the middle and west of Liaoning province, i.e., Panjin, Dalian, Huludao, Jinzhou and Yingkou. The areas with low concentration of O3 were mainly distributed in Benxi, Dandong, Liaoyang and Anshan, which were located in the east and part of the central regions in Liaoning. The areas with low concentration of PM2.5 were found in east and south of Liaoning, such as Dalian and Dandong.

In order to deeply explore the spatial action intensity of PM2.5 and O3 among cities in Liaoning Province from 2016 to 2020, the monthly mean data



(a)



(b)

Figure 4: Spatial distribution pattern and the barycenter of shift path of PM2.5 and O3 in Liaoning province.

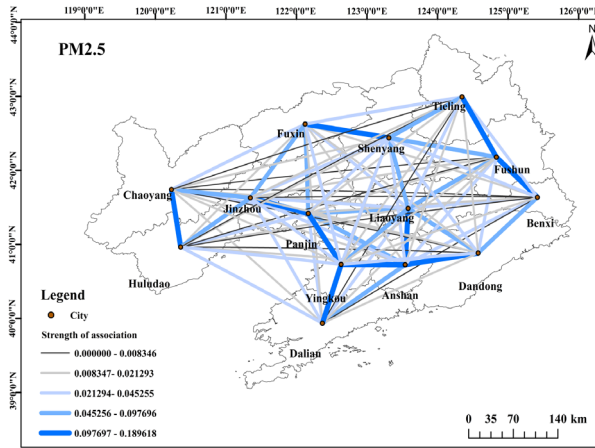
of each city were used for spatial association network analysis, and the similarity of pollution sources and regional association intensity were obtained (Fig. 5). For PM2.5, the four urban agglomerations have strong connections, i.e., Chaoyang-Huludao, Panjin-Yingkou-Anshan-Liaoyang-Dandong-Dalian, Fuxin-Shenyang and Tieling-Fushun-Benxi. For O3, Chaoyang-Huludao-Jinzhou-Panjin-Yingkou-Anshan-Liaoyang-Dandong and Fushun-Benxi had relatively consistent for spatial association. Through the analysis of spa-

Table 5: The annual average concentrations of PM 2.5 and O3 in 14 cities (PM 2.5:  $\mu\text{g}/\text{m}^3$ , O3:  $\mu\text{g}/\text{m}^3$ )

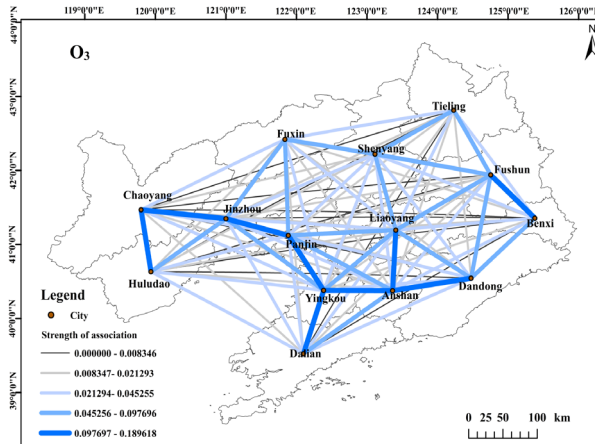
City	PM 2.5					O3				
	2016	2017	2018	2019	2020	2016	2017	2018	2019	2020
Huludao	46.9	46.5	40.5	46.8	42.5	105.9	104.5	92.4	92.4	92.9
Jinzhou	55	47.3	43.8	47	46.8	107.9	103.7	88.3	90.3	89.9
Panjing	39.7	39	35.2	38.9	35.6	113.3	107.3	94.8	95.6	94.3
Yingkou	44.4	42.6	39.5	42.4	40.8	114.2	112.2	103.6	101.4	96.6
Dandong	42.2	35	29	32.5	29	86.2	85.8	76.4	84.7	82.7
Dalian	38.7	33.5	28.8	35.1	30	106.5	105.6	93.6	97.5	95.2
Chaoyang	39.3	40.8	35.2	37	35.6	71.9	100.7	95	96.5	95
Fuxin	44	38.2	32.9	37.3	36.4	83	101.1	92.4	94.5	97.8
Shenyang	53.9	49.3	39.1	42.7	41.9	94.5	99	85	88.8	89.4
Tieling	48.5	48.9	38.4	41.3	39.2	96.7	97.6	81	89.3	90.9
Liaoyang	46.9	46.8	37.7	41.5	40.8	95.1	95.5	84.2	85.2	87.1
Anshan	55.4	47.6	39.6	43.3	43.6	84.4	99.2	85.8	93.3	88.6
Benxi	44.6	40.2	32.8	36.7	34.6	82.8	77.8	75	83.4	80.8
Fushun	43.5	46.5	41.8	43.9	43	96.1	91.3	88.8	93.5	92.2

tial association network, these urban agglomerations have similar pollution sources or have mutual transmission.

Spatial interpolation has been used to analyze the provincial pattern from January to December in Liaoning province (Fig. 6). The monthly variations of PM2.5 and O3 presented clear trends. That was, the highest PM2.5 concentrations were observed in January, February and December. In January, the concentration of PM2.5 was highest in central Liaoning. The PM2.5 concentration in February was lower than that in January. Beginning in March, PM2.5 concentration showed a declining trend, especially from May to September, PM2.5 concentration in the whole province reached the lowest value of the year. In October, PM2.5 concentrations in central and northern Liaoning began to increase. In November, the increase distribution range of PM2.5 concentration was spread across the province. In December, PM2.5 concentration increased significantly, especially in the central and northern regions. Different from the distribution law of PM2.5 concentration, O3 concentration increased significantly in summer, followed by spring and autumn. The O3 concentration reached the lowest value of the year in November, December, January and February. In March, O3 concentration began to rise. O3 concentration increased significantly in most areas in April. O3 concentration reached the highest value from May to September, and the concentration was the highest in the western and Bohai Sea regions. In October, O3 concentration decreased significantly, only in Bohai Sea region was slightly higher than other areas, and the spatial distribution of low concentration increased gradually.



(a)



(b)

Figure 5: Spatial association network feature analysis of PM<sub>2.5</sub> and O<sub>3</sub> in Liaoning province from 2016 to 2020.

### 3.3. The collaborative control of PM<sub>2.5</sub> and O<sub>3</sub>

The ratios of PM<sub>2.5</sub> concentration in 2020 to PM<sub>2.5</sub> concentration in 2016, and O<sub>3</sub> concentration in 2020 to O<sub>3</sub> concentration in 2016 were calculated respectively, as showed in Fig. 7. The concentrations of PM<sub>2.5</sub> and O<sub>3</sub> in Shenyang, Fushun, Benxi, Liaoyang, Tieling, Huludao, Jinzhou, Yingkou, Panjin, Dandong and Dalian showed a downward trend. In Chaoyang, Fuxin

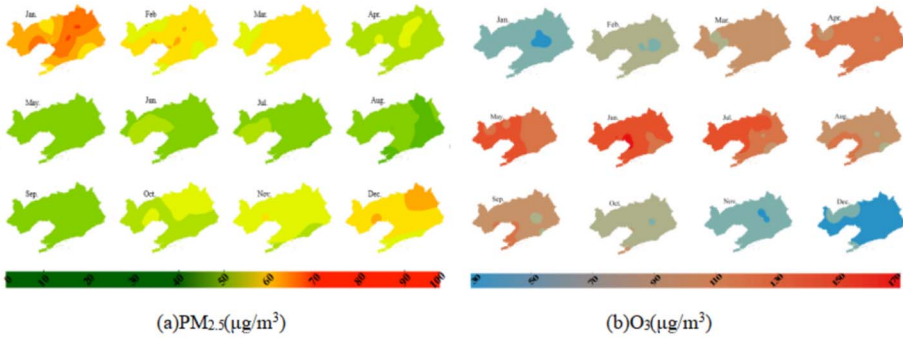


Figure 6: Spatial interpolation diagram of PM2.5 and O3 monthly concentration in Liaoning province.

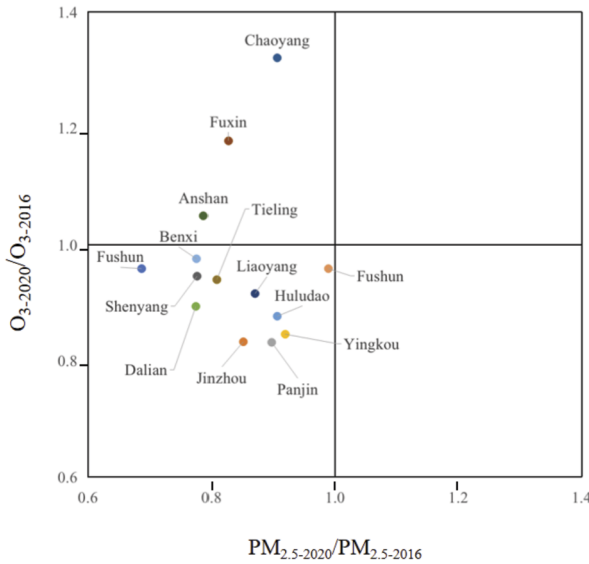


Figure 7: The ratios of PM2.5 and O3 in 14 cities of Liaoning province.

and Anshan, the concentration of PM2.5 presented a downward trend while the concentration of O3 showed an upward trend. It can be seen that from 2016 to 2020, under the influence of a series of policy implementation and human factors, the concentrations of PM2.5 and O3 in all the cities have been improved to some extent. However, it should be pointed out that in Chaoyang, Fuxin and Anshan, it is necessary to strengthen the coordinated control of PM2.5 and O3 to prevent the decline of one pollutant from leading to the rise of another.

Table 6: Moran's analysis of PM2.5 and O3 and economic statistical indicators in Liaoning Province in 2019

Pointer type	Name of economic indicator	Moran's <i>I</i>
Agriculture	Primary industry (hundred million yuan)	-0.7137
Agriculture	Fertilizer usage (ten thousand tons)	0.2108
Industry	Secondary industry (hundred million yuan)	-0.7356
Economics	Gross regional product per capital (yuan/person)	-0.3025
Service Industry	Tertiary industry (hundred million yuan)	-0.9672
Population	Year-end Total (thousands of families)	-0.9771
O3	The annual mean of O3	0.6129
PM2.5	The annual mean of PM2.5	0.1692

### 3.4. The influencing factors of PM2.5 and O3

The Moran's index of PM2.5, O3 and the spatial correlation of each impact factor were calculated to deeply analyze the spatial effects of multiple impact factors of PM2.5 and O3. Because of the availability of data, the main economic statistical indicators, PM2.5 and O3 of 14 cities in Liaoning Province in 2019 were only respectively selected. Related economic indicators were agriculture (primary industry and fertilizer usage), industry (secondary industry), gross regional product per capital, service industry (tertiary industry), and population (year-end total). The spatial analysis of multi-factor Moran model was carried out with PM2.5, O3 and major economic statistical indicators (Table 6), all economic indicators passed the significance test ( $P \leq 0.05$ ), and Z test was conducted. The results showed that there were significant differences in the degree of impact in different cities, which needed to be combined with different regions for specific analysis.

Next, the local Moran indices of the concentrations of PM2.5 and O3 and their influencing factors in 14 cities were calculated (Table 7, Table 8) and visualized (Fig. 8). It could be seen that the factors influencing the concentration of PM2.5 and O3 were gross regional product per capital, agriculture, industry, tertiary industry and population density in a descending order.

The high-high agglomeration area of PM2.5 were mainly located in central and northeast areas, including Huludao, Panjin, Shenyang, Tieling, Jinzhou, Benxi and Dandong. The high-high agglomeration area of O3 was mainly concentrated in central and some areas in Bohai Sea regions. The high-high aggregation area of economic factors was in central and western region. The spatial aggregation of primary industry was relatively scattered.



Table 7: Moran's  $I$  index of PM2.5 and O3 and economic statistical indexes in 14 cities of 2019

Name of economic indicator	Huludao	Chaoyang	Jinzhou	Fuxin	Panjin	Shenyang	Tieling
Primary industry (hundred million yuan)	-0.17	-0.11	-0.04	-0.98	0.28	-4.06	0.00
Fertilizer usage (ten thousand tons)	-0.34	-0.10	1.16	1.95	-0.41	0.62	0.83
Secondary industry (hundred million yuan)	0.53	0.97	-0.14	-0.62	0.00	-5.69	-1.05
Gross regional product per capital (yuan/person)	1.09	1.97	-0.28	0.19	-0.58	-3.95	-1.00
Tertiary industry (hundred million yuan)	0.35	0.60	-0.18	-1.03	0.22	-7.27	-1.09
Year-end Total (thousands of families)	0.01	-0.03	-0.14	-1.81	0.27	-7.38	-0.21
The annual mean of O3	0.31	1.08	-0.69	0.38	0.31	1.09	0.00
The annual mean of PM2.5	1.13	-1.96	1.22	-0.96	-1.04	1.01	0.29

Table 8: Moran's  $I$  index of PM2.5 and O3 and economic statistical indexes in 14 cities of 2019

Name of economic indicator	Fushun	Benxi	Liaoyang	Dandong	Anshan	Yingkou	Dalian
Primary industry (hundred million yuan)	-0.22	0.54	0.26	-0.14	-1.37	-0.88	-3.09
Fertilizer usage (ten thousand tons)	-2.13	1.44	0.39	0.39	0.01	0.13	-0.97
Secondary industry (hundred million yuan)	-0.31	-0.24	-0.37	-1.55	0.26	-0.29	-1.80
Gross regional product per capital (Yuan/person)	-0.03	-0.01	-0.07	-1.56	0.04	0.98	-1.05
Tertiary industry (hundred million yuan)	-0.72	-0.56	-0.77	-0.76	-0.12	-0.51	-1.70
Year-end Total (thousands of families)	-0.76	-1.12	-1.14	-0.44	0.36	-0.31	-0.99
The annual mean of O3	-1.50	5.23	4.64	-0.25	-0.56	0.00	-1.47
The annual mean of PM2.5	-0.12	0.28	-0.38	2.87	-0.50	-0.47	0.98

The high-high agglomeration area of fertilizer usage was in central and eastern region. High-high agglomeration areas of industrial were in the southwest and Bohai Sea regions. The high-high agglomeration areas of tertiary industry were located in southwest region. The areas with high population density were mainly concentrated in Shenyang, Dalian, Anshan, Tieling and Huludao. These high-high agglomerations indicated that these areas were correspondingly high level agglomerations.

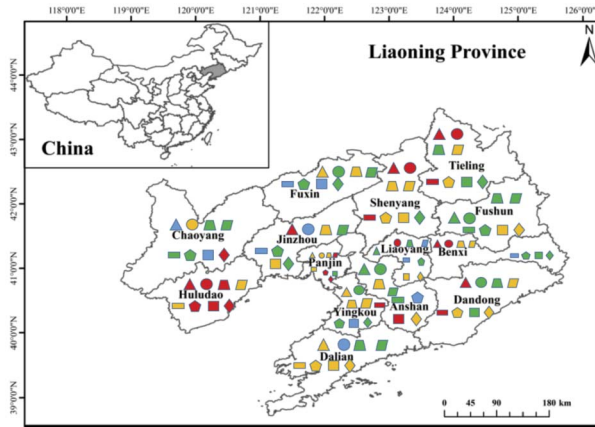


Figure 8: Moran's analysis chart of PM<sub>2.5</sub> and O<sub>3</sub> and various economic statistical indicators in 14 cities of Liaoning Province in 2019, in which the primary industry is represented by parallelogram, fertilizer usage is represented by rectangle, the secondary industry is represented by isosceles trapezoid, per capita GDP is represented by square, the tertiary industry is represented by diamond shape, population is represented by regular pentagon, PM<sub>2.5</sub> is represented by triangle, and O<sub>3</sub> is represented by circle. High-high, low-low, high-low and low-high agglomerations areas are showed in red, blue, yellow and green, respectively.

## 4. Discussions

### 4.1. The influencing factors of the temporal variation characteristics of PM<sub>2.5</sub> and O<sub>3</sub>

From 2013 to 2017, with the great efforts to control air pollution in China, air quality improved significantly, as there was a downward trend in PM<sub>2.5</sub> and O<sub>3</sub> pollutant concentration. After 2018, coordinated emission reduction control measures were taken for PM<sub>2.5</sub> and O<sub>3</sub> in China, thus pollutant concentrations showed a trend of decline after fluctuation. PM<sub>2.5</sub> pollution mostly occurred from October to the following March, and peaked in November and January. This is closely related to the large-scale open-air burning of crop straw and the intensity of coal burning for heating. And during this period, more static and stable weather exacerbated the accumulation of PM<sub>2.5</sub> [22]. The change trend of O<sub>3</sub> concentration was opposite to that of PM<sub>2.5</sub> concentration, with peak values occurring in June and July and

the low values in January and December. Many studies have showed that it was related to the formation mechanism of O<sub>3</sub>, with high temperature, low humidity and strong light in summer, the emission of volatile organic compounds (VOCs) and nitrogen oxides (NO<sub>x</sub>) were high and produce photochemical reactions, producing secondary pollutants while generating O<sub>3</sub> in the atmosphere [12, 56, 62]. Moreover, Liaoning province is located in high latitude area, with short daylight in winter, which leads to short illumination time and weak illumination, which weaken O<sub>3</sub> generation in winter. Therefore, the reverse seasonal distribution characteristics of PM<sub>2.5</sub> and O<sub>3</sub> concentrations should be paid more attention, and the high incidence of PM<sub>2.5</sub> pollution in winter and O<sub>3</sub> pollution in summer should be regarded as the key control period. Specifically, it was necessary to improve the combustion efficiency of coal burning, reduce the emission of pollutants from coal burning. Reduced O<sub>3</sub> generation by decreasing the emission of volatile organic compounds and nitrogen oxides.

#### **4.2. The influencing factors of the spatial patterns evolution of PM<sub>2.5</sub> and O<sub>3</sub>**

The high concentration of PM<sub>2.5</sub> was mainly found in central and southwestern Liaoning, where industrial production was expanding and densely inhabited district [63]. Studies have showed that the spatial distribution of high O<sub>3</sub> concentration was consistent with ground temperature [64]. Moreover, under the influence of high temperature and solar radiation, it was favorable for O<sub>3</sub> generation. The temperature of cities in central and western Bohai Sea in Liaoning was higher than that of other cities, so the O<sub>3</sub> concentration in this region was higher. It could be seen that spatial distribution of high PM<sub>2.5</sub> concentration was different from that of high O<sub>3</sub> concentration in some areas. High concentration of O<sub>3</sub> enhanced atmospheric oxidation, making gaseous pollutants such as sulfur dioxide and nitrogen oxide in the air more easily oxidized into fine particles such as sulfate and nitrate, thus leading to increase in PM<sub>2.5</sub> concentration. High concentration of PM<sub>2.5</sub> would weaken solar radiation and reduced O<sub>3</sub> photochemical reaction, which was not conducive to O<sub>3</sub> generation. There were many factors influencing the correlation between PM<sub>2.5</sub> and O<sub>3</sub>.

#### **4.3. Economic factors affecting PM<sub>2.5</sub> and O<sub>3</sub>**

Studies have shown that PM<sub>2.5</sub> concentration and O<sub>3</sub> concentrations in some areas of China is significantly affected by GDP energy consumption,

per capita GDP and population density [65, 66]. PM2.5 and O3 were significantly positively correlated with economic factors in central region, so effectively solving economic factors in central region and increasing investment in ecological environment would play an important role in alleviating local concentrations of PM2.5 and O3 in this region, while economic factors in western China have no significant effect on reducing PM2.5 concentration. PM2.5 and O3 were positively correlated with primary industry in Shenyang, Huludao, Tieling and Panjin, and significantly positively correlated with fertilizer use in central and eastern regions. This suggested that increased agricultural production and fertilizer use would increase the concentration of pollutants in the area. PM2.5 and O3 were positively correlated with industry in Bohai Sea regions. Therefore, industrial emissions from some cities in Bohai Sea regions needed to be considered to mitigate local air pollutant concentrations. For Shenyang, Huludao, Dalian and Tieling, increasing population density would aggravate local air pollution concentration.

## 5. Conclusions

This study analyzed the spatial and temporal distribution characteristics and influencing factors of PM2.5 and O3 in Liaoning Province, which has important reference for environmental management and air pollution control.

First of all, PM2.5 concentration in all cities showed a downward trend in the past five years, and O3 concentration in most cities also decreased slightly. However, with the decrease of PM2.5 concentration, the decrease of O3 concentration was not obvious. PM2.5 concentration was highest in winter, lowest in summer, followed by spring and autumn. In contrast, O3 concentration was highest in summer, followed by spring and autumn, and lowest in winter. The reasons for the opposite seasonal dynamic characteristics were as follows: PM2.5 concentration varied seasonally, which was related to the seasonal characteristics of emission sources. Mainly coal burning for heating, industrial emissions, straw burning, as well as winter inversion and low atmospheric boundary layer. The emission of volatile organic compounds and nitrogen oxide favorable meteorological conditions such as high temperature and strong solar radiation promote O3 generation in summer.

Secondly, PM2.5 concentration was highest in central and southwestern parts of Liaoning province, especially the industrially developed and densely populated areas of Jinzhou, Anshan, Shenyang and Huludao. The high O3 concentration was mainly distributed in the central-western and some cities in Bohai Sea region, including Panjin, Dalian, Huludao, Jinzhou

and Yingkou. The temperature of these cities was higher than that of other cities in the region, which was more conducive to O<sub>3</sub> generation. The spatial distribution of PM<sub>2.5</sub> and O<sub>3</sub> was strongly correlated with population density, environmental factors and meteorological factors. Chaoyang, Fuxin and Anshan should be pay more attention to the coordinated management of PM<sub>2.5</sub> and O<sub>3</sub>.

Finally, the multivariate Moran model showed that PM<sub>2.5</sub> and O<sub>3</sub> have a certain spatial correlation with economy, agriculture, industry, tertiary industry and population density in a descending order.

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