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To cite this article: Y F Ma *et al* 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **351** 012030

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Analysis of the atmospheric pollution transport pathways and sources in Shenyang, based on the HYSPLIT model

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Abstract. The transport pathways and potential sources distribution of air pollutants are statistically analyzed based on the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model and the TrajStat method driven by the meteorological data of the Global Data Assimilation System (GDAS), and the concentration monitoring data of PM₁₀, PM_{2.5}, SO₂ and NO₂ in Shenyang, Liaoning Province in 2017. The trajectory classification method is used to analyze the main transport pathways affecting air masses during the research period. It is considered that the air masses in the northeast and northwest directions are the main reason for the high concentration of pollutants in the study area, while the air masses in the south direction are considered to be clean. Furthermore, the results of PSCF (potential resource contribution function) are consistent with the results of CWT (concentration-weighted trajectory). The potential sources that result in high concentrations of PM₁₀ and PM_{2.5} are mainly located in southern Hebei Province, northern Shandong Province and the central and eastern Inner Mongolia Autonomous Regions. The corresponding potential source area shows a concentration of PM₁₀ exceeding 150 $\mu\text{g}\cdot\text{m}^{-3}$ and PM_{2.5} exceeding 75 $\mu\text{g}\cdot\text{m}^{-3}$, while the source of nitrogen sulfide is mainly influenced by local sources.

1. Introduction

Among the 14 cities in Liaoning Province in 2017, Shenyang has the highest concentration of PM₁₀ [1]. As the 2017 Shenyang Environmental Quality Bulletin showed the from Shenyang Municipal Bureau of Ecology and Environment: the annual average concentration of inhalable particulate matter (PM₁₀), the main ambient pollutant in urban air is 88 $\mu\text{g}\cdot\text{m}^{-3}$, and the average annual concentration of fine particles (PM_{2.5}) is 51 $\mu\text{g}\cdot\text{m}^{-3}$, exceeding the national environmental air quality secondary standard by 0.3 times, and 0.5 times, respectively; the annual average concentration of sulfur dioxide (SO₂) is 37 $\mu\text{g}\cdot\text{m}^{-3}$, and the annual average concentration of nitrogen dioxide (NO₂) is 40 $\mu\text{g}\cdot\text{m}^{-3}$, both of which reach the air quality standard. In addition to being affected by local sources, the degree of air pollution is also affected by regional transmission. Air pollution cross-regional transportation is particularly obvious in urban-intensive areas [2]. Contaminants are transported between cities through airflow movements, resulting in the superposition of pollution above cities and increasing the difficulty of identifying potential source areas [3]. By studying the regional transport of pollutants, this paper analyzes the influence of different regions on the contribution of pollutants in Shenyang.

For the analysis of the sources of Shenyang pollutants, many researchers focus on the use of air quality models and receptor models [4-6], at the same time, the researches on Shenyang air quality mainly focus on the study of the temporal and spatial variation of pollutants and the influencing factors of meteorological conditions [7-9]. Combining the calculation methods of



atmospheric horizontal and vertical motion, the HYSPLIT model is relatively complete in the aspects of pollutant transport [10], diffusion and sedimentation, but is widely used in pollution trajectory simulation, pollution source identification and quantitative estimation of pollutant concentration [11-13]. In the application of HYSPLIT model, Li et al. [14] believe that the main reason for the Beijing haze is the increase of PM_{2.5} concentration caused by regional transportation. Lei et al. [15] analyzed the transport route and its potential sources of PM₁₀ from 2003 to 2009, Wehner et al. [16] analyzed the relationship between atmospheric particle size distribution and regional meteorological transport in Beijing from 2004 to 2006. Yankun [17] believed that atmospheric PM_{2.5} pollution in Shanghai was mainly affected by short-distance migration of airflow. In recent years, the research on inter-regional transport of air masses focuses on the Beijing-Tianjin-Hebei, Pearl River Delta and other places. At the same time, other application fields are included in the analysis of the transport paths of water vapor and sand-dust [18-23]. The present study pays attention on analyzing the main transport channels and potential sources of atmospheric pollutants in Shenyang City.

2. Study area and data sources

2.1. Overview of the study area

As the provincial capital, Shenyang (41°11'51"N~43°2'113"N, 122°25'9"E~123°25'31.18"E) is located in the central part of Liaoning Province, in the south of Northeast China, the south of Liaodong Peninsula, and its northern parts (Changbai Mountain). The landscape is dominated by plains, mountains and hills, while the southeast direction of Shenyang is dominated by hills, with an average elevation between 18 and 270 m (AGL), and it was mainly temperate monsoon continental climate.

2.2. Data sources

The meteorological data required for the model calculation are the Global Data Assimilation System (GDAS) provided online by the National Center for Environmental Prediction (NCEP), recorded once every 6 hours at 00:00, 06:00, 12:00, and 18:00 UTC, respectively. The data processing module of GDAS converts the meteorological field of Sigma coordinate coefficients into pressure coordinate data with 1°×1° resolution. The vertical grid of data is divided into 14 layers, which are near ground level, 1000, 925, 850, 700, 500, 400, 300, 250, 200, 150, 100, 50, 20 hPa, respectively [24]. The concentration PM₁₀, PM_{2.5}, SO₂ and NO₂ in Shenyang City in 2017, were derived from the routine monitoring data of atmospheric pollutants provided by the Shenyang Environmental Monitoring Center.

3. Research methodology

3.1. Backward trajectory calculation and cluster analysis

The HYSPLIT model is a professional model developed by the Australian Meteorological Agency and the National Oceanic and Atmospheric Administration (NOAA) to calculate and analyze the transport and diffusion of atmospheric pollutants. This model is a hybrid diffusion model, where advection and diffusion are calculated by the Lagrangian method, while the Euler method is used for concentration calculations [25,26].

Assuming that the air parcel flutters with the wind, taking the motion of the air parcel in a time step as an example, the final position of the air parcel is calculated from the average speed between its initial position (P) and the first guess position (P'). The first guess position of the air parcel is:

$$P'(t+\Delta t)=P(t)+v(P, t)\Delta t \quad (1)$$

The final position of the air parcel is:

$$P(t+\Delta t)=P(t)+0.5\times [v(P, v)+v(P', t+\Delta t)]\Delta t \quad (2)$$

where, Δt is the time step, and the moving distance of the air parcel within a time step is

required to be no more than 0.75 grid distance, that is to say, $\Delta t < 0.75 u_{\max}$, and u_{\max} is the maximum wind speed. The pattern uses terrain following coordinates; the input meteorological data maintains the original format in the horizontal direction coordinates, and is vertically interpolated to the terrain following coordinate system:

$$\sigma = \frac{(z_{\text{top}} - z_{\text{mst}})}{(z_{\text{top}} - z_{\text{gl}})} \quad (3)$$

where, z_{top} is the top value of the trajectory mode coordinate system, z_{gl} is the terrain height, and z_{mst} is the boundary height under the coordinates.

The trajectory cluster analysis classifies the samples according to the statistic, namely, to classify a large number of trajectories according to the transport speed and direction of the air mass trajectories [27]. The total spatial minimum variance method cluster analysis is used to calculate the Spatial Variance (SV) of each cluster, that is, the sum of the squares of the distances between points in each trajectory in each cluster and the points corresponding to the average trajectory of the cluster. The number of clusters corresponding to the total variance value (TSV) in the minimum increase is the clustering result [28].

$$SV_{i,j} = \sum_k (P_{i,j} - M_{i,j})^2 \quad (4)$$

Equation (4) is summed up according to the number of endpoints of the trajectories in each cluster, P is the vector position of the point (j) on each trajectory, and M is the vector position of the corresponding points on the average trajectory of the cluster i . In equation (5), the Cluster Spatial Variance (CSV) of cluster i is the SV sum of all the trajectories in the cluster:

$$CSV_i = \sum_j SV_{i,j} \quad (5)$$

TSV is the CSV sum of all clusters:

$$TSV = \sum_i CSV_i \quad (6)$$

3.2. Potential source contribution function (PSCF) and concentration-weighted trajectory (CWT) methods

The PSCF algorithm is a method for analyzing the potential source regions based on air mass trajectories, also known as the retention time analysis method, which can be used to initially determine the potential source regions that affect the receptor site [29,30]. The method assumes that the air mass carries pollutants over a region. The value of PSCF on a unit grid is defined as [31]:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \quad (7)$$

where, n_{ij} is the number of trajectory endpoints in the ij^{th} grid, and m_{ij} is the number of endpoints in the ij^{th} grid for all trajectories corresponding to a contaminant concentration above a certain standard limit. Since PSCF is a conditional probability function, when there are fewer trajectory endpoints in a certain grid, its value obtains dramatically greater uncertainty. To reduce the uncertainty, the weighting factor W_{ij} is introduced as [32]:

$$W_{ij} \begin{cases} 1.00 & n_{ij} > 3 \cdot Avg \\ 0.70 & Avg < n_{ij} \leq 3 \cdot Avg \\ 0.42 & 0.5 \cdot Avg < n_{ij} \leq Avg \\ 0.17 & 0 < n_{ij} \leq 0.5 \cdot Avg \end{cases} \quad (8)$$

where, Avg is the average number of trajectory endpoints per grid. The high PSCF value is reduced by the weight function when n_{ij} is less than three times the number of endpoints of the average trajectory in each grid in the study area. The PSCF method, which is a conditional probability function, only represents the proportion of the number of trajectories with pollutant concentration higher than a certain standard limit in a grid to the total number of trajectories. In

order to quantitatively represent the pollutant concentration value in the potential source area, the CWT analysis method [33] was introduced to assign a weight concentration to each grid on average through the pollution concentration carried by the trajectory of a certain grid:

$$C_{ij} = \frac{1}{\sum_{l=1}^M \tau_{ijl}} \sum_{l=1}^M C_l \tau_{ijl} \quad (9)$$

where, C_{ij} is the weight concentration of the ij^{th} grid, M is the total number of trajectories, C_l is the concentration of trajectories l , τ_{ijl} is the retention period of the trajectories l on the grid, and the weight function W_{ij} in the PSCF analysis method is also applicable to the CWT analysis.

4. Results and discussion

4.1. Trajectory calculation and clustering

Advection transport has an important impact on the formation of pollution in certain regions. Affected by the uplift of the atmospheric boundary layer, the retention period of the pollutants becomes longer, and the possibility of spreading to other areas through advection and diffusion also increases, thus forming air quality issues. In the trajectory clustering, the starting point of the trajectories is (41°44'24.00"N, 123°30'0.00"E), starting at 00:00, 06:00, 12:00, 18:00 UTC every day. The push-back time is 48 h. In order to eliminate the influence of disturbances in the near-surface layer, the simulated height is set to 500 m (AGL). The trajectories of the air masses that reached Shenyang in 2017 are shown in Figure 1.

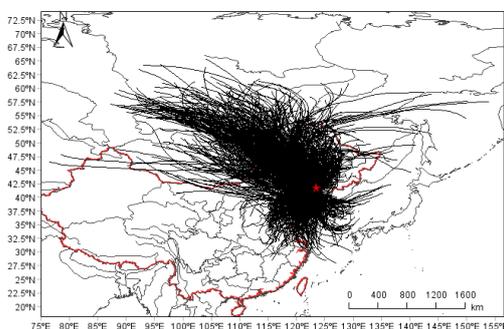


Figure 1. Backward trajectories distribution map of Shenyang in 2017. (The pentagram in the figure represents the starting point or receptor point of the trajectories).

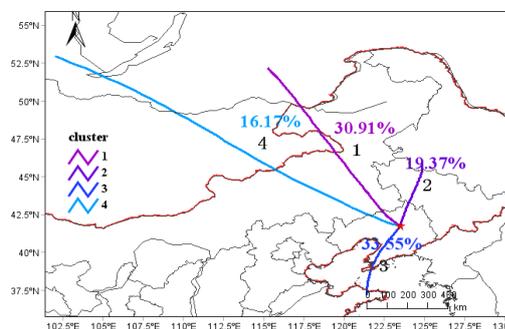


Figure 2. The clustering classification map of Shenyang in 2017.

The average trajectory length of the air masses indicates the moving velocity of the air masses; the longer the average trajectory of the air masses, the faster the transportation moving speed. The clustering results obtained by the clustering algorithm are shown in Figure 2.

Four air mass transport routes in Figure 2 are obtained through the clustering algorithm. In backward trajectory clustering, there are trajectory air masses from the south, northwest and northeast directions. The two trajectories from the northwest (clusters 1 and 4) are longer and move faster than those from the northeast direction (cluster 2) and south direction (cluster 3). The lengths and velocities of clusters 2 and 3 are obviously different. The air mass height changes in each diffusion direction are shown in Figure 3.

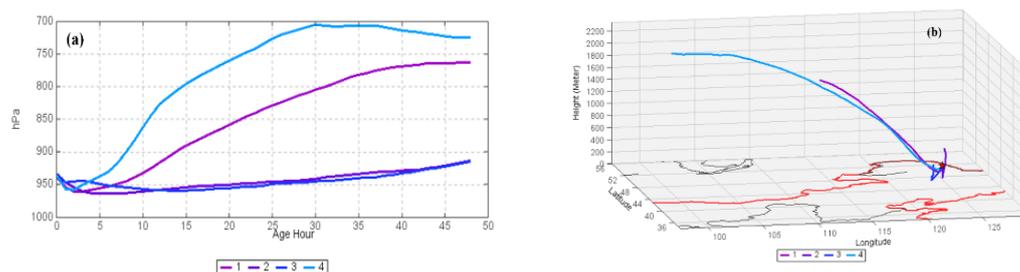


Figure 3. Changes in the pressure (a) and height (b) of the clustering air mass.

The height of clusters 4 and 1 is higher than other tracks (Figure 3a), with the highest pressure reaching 700 hPa, while clusters 2 and 3 have similar height changes, with the highest pressure reaching 920 hPa.

All backward trajectories can be divided into two categories: cleaner trajectories and contaminated trajectories. According to the Ambient Air Quality Standard (GB3095-2012), the basic items of ambient air pollutants are related to the 24-h average concentration limits of PM_{10} , $PM_{2.5}$, SO_2 , NO_2 , i.e. $150 \mu\text{g}\cdot\text{m}^{-3}$, $75 \mu\text{g}\cdot\text{m}^{-3}$, $150 \mu\text{g}\cdot\text{m}^{-3}$, $80 \mu\text{g}\cdot\text{m}^{-3}$, respectively. The trajectories that exceed the concentration limits in all trajectories are selected and defined as the polluted trajectories (Figure 4).

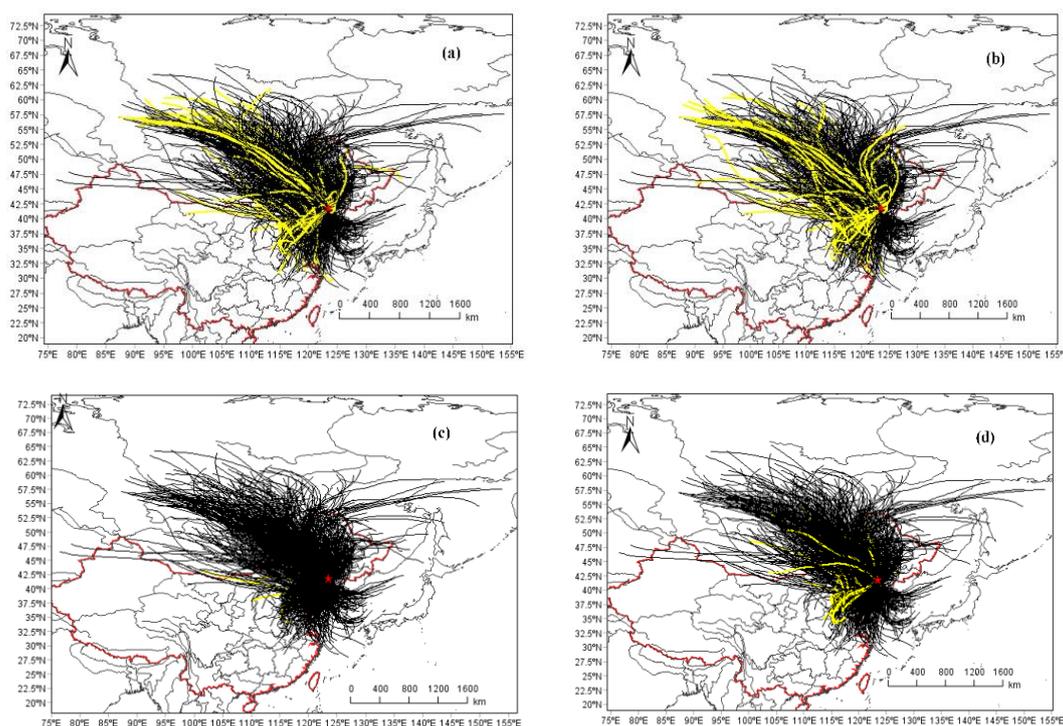


Figure 4. The trajectories of PM_{10} , $PM_{2.5}$, SO_2 and NO_2 concentrations higher than (a) $150 \mu\text{g}\cdot\text{m}^{-3}$, (b) $75 \mu\text{g}\cdot\text{m}^{-3}$, (c) $150 \mu\text{g}\cdot\text{m}^{-3}$ and (d) $80 \mu\text{g}\cdot\text{m}^{-3}$, respectively.

The number of trajectories of pollution in each cluster and the mean concentration for all trajectories and pollution concentrations of each pollutant for polluted trajectories are shown in Table 1. In different clusters, the pollution trajectories' percentages of different pollution items are different. Among all air masses, the trajectories carrying $PM_{2.5}$ are the most polluted, with a total of 248 tracks; the second are the trajectories carrying PM_{10} , with a total of 177 tracks. The trajectories carrying SO_2 are the least polluted, with 21 tracks.

Table 1. The concentration of PM₁₀, PM_{2.5}, SO₂ and NO₂ associated with all trajectories and polluted trajectories in each cluster. (unit : $\mu\text{g}/\text{m}^3$).

Cluster	Mean concentrations for all trajectories					Mean concentrations for polluted trajectories							
	N	PM ₁₀	PM _{2.5}	SO ₂	NO ₂	PM ₁₀		PM _{2.5}		SO ₂		NO ₂	
						P_N	CON	P_N	CON	P_N	CON	P_N	CON
1	39	83.71±	48.15±	38.29±	40.51±	48(12	201.17±	89(22	123.14±	5(1.2	256.00±	12(3	94.42±
	1	54.74	42.21	38.22	20.75	%)	53.75	%)	43.47	%)	36.39	%)	8.75
2	26	90.47±	56.52±	37.19±	43.81±	49(18	196.84±	79(29	130.17±	9(3.3	231.67±	11(4	87.19±
	8	60.19	49.22	48.72	21.62	%)	38.71	%)	41.54	%)	36.13	%)	7.26
3	43	85.09±	50.14±	22.74±	37.13±	47(11	194.60±	100(2	110.28±	4(0.9	201.00±	27(6	91.89±
	8	48.66	33.39	26.54	21.13	%)	39.41	2%)	31.98	%)	19.92	%)	12.88
4	19	99.83±	47.01±	32.58±	37.59±	33(16	241.39±	43(21	106.63±	3(1.5	218.67±	5	90.80±
	9	81.29	35.87	35.10	19.49	%)	106.2	%)	34.65	%)	13.87	(2%	12.13
)							
all	12	89.14±	50.38±	31.93±	39.60±	177(1	205.72±	248(1	118.52±	21(1.	229.76±	55(4	90.95±
	96	59.02	40.25	37.44	21.01	4%)	62.74	9%)	39.33	6%)	46.05	%)	10.96

Note : P_N means polluted number; CON means concentration.

Among the mean concentrations in all clusters, the concentrations of PM₁₀ in cluster 4, PM_{2.5} and NO₂ in cluster 2 and SO₂ in cluster 1 were higher than those in other clusters, and the polluted clusters accounted for 16%, 29%, 4% and 1.2%, respectively. This means that air masses associated with these clusters may reach the receptor point with higher concentrations of contaminants. The average concentration of SO₂ and NO₂ in cluster 3 is lower among the four clusters, and the proportion of the contaminated trajectories is 0.9% and 6%, respectively. Although there are trajectories with the second higher PM_{2.5} concentration, the frequency of the contaminated trajectories in the air masses is lower. Therefore, it is concluded from the above analysis that the pollution level of clusters is lower than that in other clusters, and the direction from Bohai Sea to Shenyang via Dalian is also considered to be relatively clean.

Cluster 2 accounted for 19.37% of all backward trajectories. In addition to the trajectories carrying the highest pollution concentrations of PM_{2.5} and NO₂, there were also the pollution trajectories carrying the second largest value of PM₁₀ and SO₂, with proportions of 3.3%~29%, respectively.

In cluster 2, the source direction of air mass is mainly in the northeast direction, where heavy industrial areas such as Changchun, Harbin produce more particulate matter and nitrogen sulfide. The above analysis suggests that this may be the most important transport path of the four pollutants. In clusters 2 and 3, the trajectories are shorter, the air mass diffusion is slower; therefore, pollutants are easy to accumulate.

The majority of the trajectories of cluster 1 (accounted for 30.91% of the total trajectories) include the second largest number of NO₂ pollution trajectories. It is shown that the northwest direction may be the main transport route for nitrogen oxides. In spite cluster 3 involves a lower NO₂ concentration, 6% of the trajectories are defined as pollution trajectories and the air masses in this cluster have a higher frequency of at 33.55% in the whole year, which may cause slight pollution of NO₂ at the receptor point, it is, therefore, considered that the south-orientation is the potential transport pathway of NO₂.

Cluster 4 accounts for 16.17% of all trajectories. In addition to the highest PM₁₀ concentration, the concentrations of the other three pollutants are relatively low, and the contaminated trajectories account for 1.5%~22%. The trajectory pathways from Mongolia to Inner Mongolia Autonomous Regions carry a large amount of sand and dust, resulting in higher concentration of PM₁₀. So it is considered to be the main source of larger particles.

Generally, cluster 2 is the most important transport pathway leading to an increase of pollutant concentration in Shenyang area; cluster 3 is the cleanest transport path, that is, the concentration of pollutants carried by air masses in this direction is relatively low. The transport paths of clusters 1 and 4 may be the ones that cause the concentration of NO₂ and PM₁₀ to be higher at the receptor points, respectively.

4.2. Results from the PSCF and CWT analyses

The PSCF is a methodology based on the analysis of air flow trajectories to identify the source. The PSCF can be used to preliminarily determine the source area of air pollution qualitatively. According to the area covered by the backward trajectories, the study area is gridded, and the

study area (75 °E~155 °E, 20 °N~68 °N) is divided into 15360 grids with a horizontal resolution of 0.5 °×0.5 °.

Minor changes in contaminants concentration should probably affect the PSCF analysis. Different from the above procedure of dividing the clean and polluted trajectories in Section 4.1, the PSCF analysis was performed based on the 75th percentile of all samples in Shenyang in 2017. The distribution results are shown in Figure 5 where different pollutants are affected differently by the potential source areas. The darker the color in the figure, the higher the PSCF value, that is, the greater the possibility that the grid area to be the pollution source area for Shenyang. The potential pollution sources of PM₁₀ (figure 5a) and PM_{2.5} (figure 5b) in Shenyang are mainly distributed in the south of Hebei Province, the northwest of Shandong Province, the north of Henan Province, and the north of Anhui Province. Affected by southern airflows, the pollutants migrate to Shenyang. The potential source areas of SO₂ (figure 5c) are mainly located in the northeast of Inner Mongolia Autonomous Regions, eastern Mongolia, western Liaoning Province, northern Hebei Province and central Shandong Province, while NO₂ (figure 5d) potential source areas are mainly located in Liaoning Province, Hebei Province, Shandong Province and eastern central Inner Mongolia.

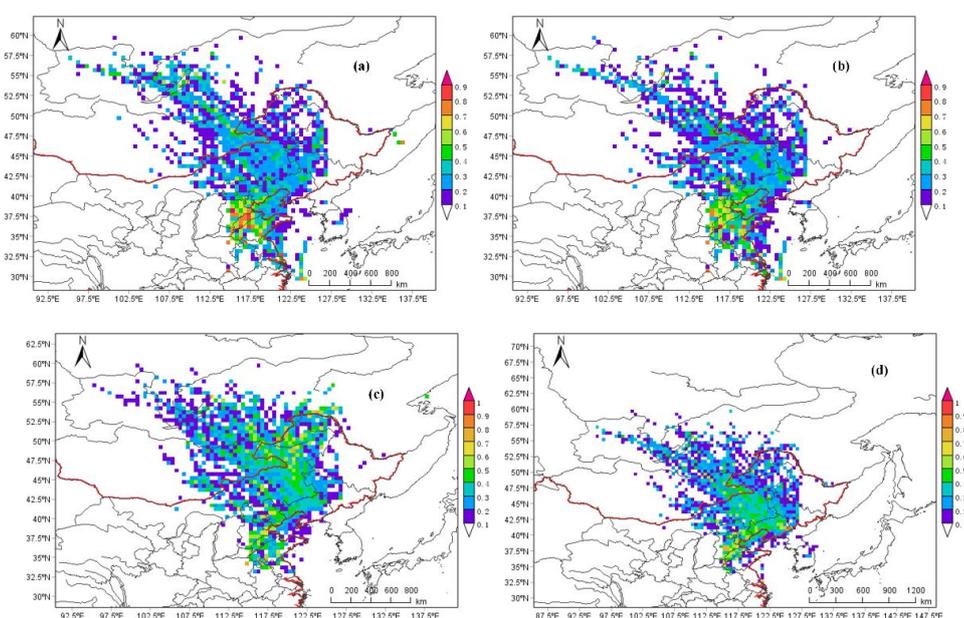


Figure 5. PSCF charts based on the 75th percentile of all samples of (a) PM₁₀, (b) PM_{2.5}, (c) SO₂ and (d) NO₂ in 2017.

The CWT method is a concentration weighting algorithm, which is used to calculate the potential sources concentration based on the airflow trajectories. The results of the CWT are shown in Figure 6, creating a grid with the same resolution as the PSCF analysis. The CWT value of PM₁₀ exceeds 150 $\mu\text{g}\cdot\text{m}^{-3}$ (figure 6a) in southern Hebei Province and northern Shandong Province, and the value of PM_{2.5} exceeds 75 $\mu\text{g}\cdot\text{m}^{-3}$ (figure 6b). The corresponding concentrations of SO₂ and NO₂ are lower than 60 $\mu\text{g}\cdot\text{m}^{-3}$. In the CWT analysis chart of PM₁₀ (figure 6a), most grids with high CWT values are mainly distributed in the central and eastern parts of Inner Mongolia Autonomous Regions, Liaoning Province, southern Hebei Province and western Shandong Province, which results are consistent with the PSCF ones in Figure 4. It can be concluded from the CWT analysis diagrams of SO₂ (figure 6c) and NO₂ (figure 6d) show that most of the grids have lower concentrations, so it is simple to think that local pollution sources in Shenyang may lead to higher density values of the corresponding pollutants. In other words, the nitrogen sulfide carried by external sources does not have a significant impact on the local

nitrogen and sulfur pollution in Shenyang.

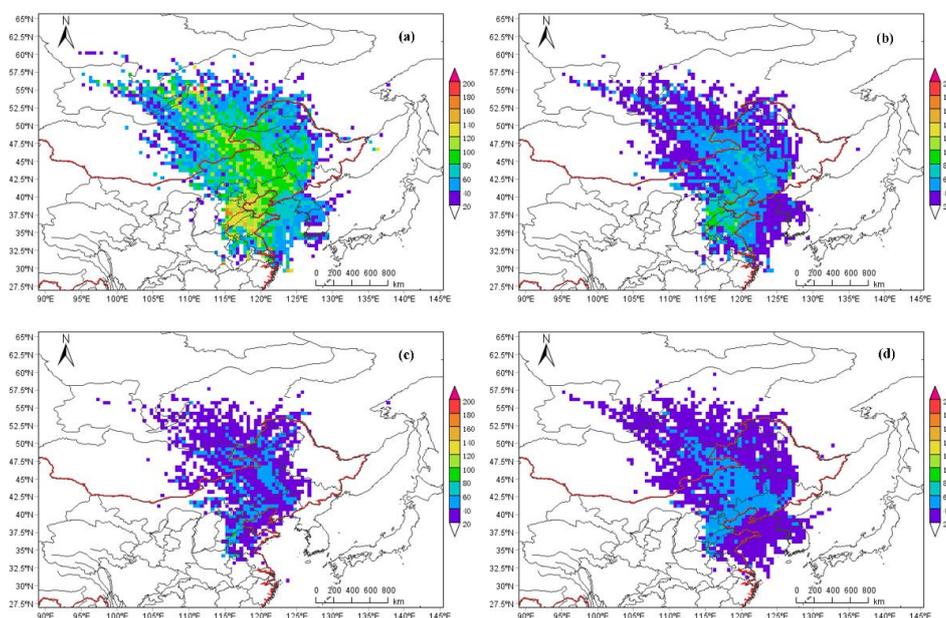


Figure 6. CWT charts of (a) PM_{10} 、 (b) $PM_{2.5}$ 、 (c) SO_2 and (d) NO_2 in 2017.

5. Conclusions

Trajectory clustering, PSCF and CWT analyses were used to study the main paths of air mass transport pathways, the potential pollution source areas and the relative contribution of pollutants to the atmosphere in 2017. The analysis results showed that the trajectory air masses are mainly divided into continental and marine air masses, and contain four different directions of transport. The continental air masses of cluster 2 moving slowly in the direction of Jilin Province and cluster 1 moving quickly in the direction of Inner Mongolia are the main reasons for the increase of pollutants concentration in Shenyang. In comparison, it is likely that the concentrations of pollutants at the receptor points may increase in cluster 4 from Mongolia. The marine air cluster 3 from Dalian and other places through the Bohai Sea to Shenyang is relatively clean and could dilute local pollutants to some extent. From the analysis of PSCF and CWT, it is concluded that Hebei, Shandong and Inner Mongolia Autonomous Regions are the main potential source areas of PM_{10} and $PM_{2.5}$, while the main potential source regions of SO_2 and NO_2 are considered to be from sources local.

Acknowledgments

The research was supported by Aeronautical Science Foundation of China (NO. 2017ZA54001) and Doctor Startup Foundation in the Shenyang Aerospace University (NO.16YB17).

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