Evolution, Transport Characteristics, and Potential Source Regions of PM$_{2.5}$ and O$_3$ Pollution in a Coastal City of China during 2015–2020

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Abstract: The evolution, transport characteristics, and potential source regions of PM$_{2.5}$ and O$_3$ were investigated from 1 January 2015 to 31 December 2020 in the coastal city of Nantong. The annual mean PM$_{2.5}$ concentration declined obviously over the entire study period, and was 34.7 $\mu$g/m$^3$ in 2020. O$_3$ had a relatively smooth decreasing trend, but rebounded greatly during 2017 when the most frequent extreme high-temperature events occurred. Similar trends were observed for PM$_{2.5}$ and O$_3$ polluted hours. No PM$_{2.5}$-O$_3$ complex air pollution happened in 2019 and 2020, likely suggesting the preliminary results from the implementation of emission controls. Notable differences in transport pathways and frequencies were observed from the backward trajectory clusters in four seasons in Nantong. Clusters with the largest percentage of polluted PM$_{2.5}$ and O$_3$ trajectories were transported mostly over short distances rather than long distances. Analysis involving the potential source contribution function (PSCF) and concentration weighted trajectory (CWT) showed that PM$_{2.5}$ polluted sources were from the adjacent western and northwestern provinces, whereas the influence of eastern marine sources was relatively small. O$_3$ had a greatly different spatial distribution of polluted source regions from PM$_{2.5}$, mostly covering the North China Plain, the Bohai Sea, and the Yellow Sea.

Keywords: PM$_{2.5}$; O$_3$; evolution; backward trajectory; transport pathway; potential source region; coastal city

1. Introduction

Fine particulate matter (PM$_{2.5}$) and ozone (O$_3$) are two of the largest contributors to air pollution in the tropospheric atmosphere due to their impact on human health, environmental degradation, vegetation production, and climate change [1–3]. Complex emissions and adverse meteorological conditions normally lead to high PM$_{2.5}$ and O$_3$ concentrations [4–6]. Apart from directly emitted particulate matter, both ground-level PM$_{2.5}$ and O$_3$ are mainly secondary pollutants. Secondary PM$_{2.5}$ and O$_3$ share similar precursors (e.g., nitrogen oxides (NO$_x$) and volatile organic compounds (VOC$_x$)) in photochemical reactions [7,8]. Besides, the secondary PM$_{2.5}$ is also formed by coagulation and nucleation of chemicals from direct emissions. Given the big challenge of controlling both PM$_{2.5}$ and O$_3$ pollution due to their highly nonlinear secondary formation, reducing emissions of NO$_x$ or VOC$_x$ for PM$_{2.5}$ control might lead to unexpected adverse effects on O$_3$ in the photochemical processes [8,9]. In addition, air pollution might worsen due to regional, long-range transport and unfavorable meteorology conditions, even when local emissions are reduced. Thus, both pollutants are of great concern for regional air pollution improvement.

Currently, eastern China is an industrial and urbanized area with the densest population and highest emissions nationwide [10,11]. Due to the complex formation of PM$_{2.5}$ and
O_3 from multiple sources and precursors, integrated tackling of these two pollutants has become one of the largest challenges facing this region. Although PM_{2.5} concentrations have declined in this region compared with previous years after stringent pollution mitigation measures taken since 2013, severe air pollution events still occur under some stagnant weather conditions [12]. In addition, O_3 showed an increasing trend, especially during the summers in the past few years [13,14].

Numerous studies have been conducted to explore the evolution and transport characteristics of PM_{2.5} and O_3, as well as the influence of meteorological conditions in eastern China. However, most of these studies focused on megacities such as Shanghai, Nanjing, and Hangzhou which were severely polluted [15–21]. Few studies of PM_{2.5} and O_3 have been performed in less polluted cities such as Nantong in this region. Nantong is one of many fast-growing coastal cities with a population of 7.72 million in Jiangsu Province, and is adjacent to Shanghai in the south across the Yangtze River. As with other cities in eastern China, Nantong is also suffering problems of PM_{2.5} and O_3 complex air pollution with its rapid growth of industrialization. However, there have only been very limited studies focused on pollutant characteristics and their relationship with meteorological conditions over short periods of one year or less in Nantong [22]. To achieve better synergic control strategies for the pollution of PM_{2.5} and O_3 in Nantong, it is urgent to strengthen the understanding of their long-term pollution characteristics, transport pathways, and potential source regions.

To fill the knowledge gap, in this study an insight into the evolution, transport characteristics, and potential source regions of ground-level PM_{2.5} and O_3 in Nantong during the 2015–2020 period was presented. The evolution of individual air pollutants, as well as nonattainment complex air pollution, were investigated. The transport pathways and potential source regions of PM_{2.5} and O_3 were identified and synthetically analyzed using the backward trajectory cluster, the potential source contribution function (PSCF), and concentration weighted trajectory (CWT). Consequently, these results will provide an important basis for exploring efficient strategies to control both PM_{2.5} and O_3 pollution in Nantong.

2. Data and Methods

2.1. Site Location

Air pollutants of PM_{2.5} and O_3 in the coastal city of Nantong from 2015 to 2020 were investigated here (Figure 1). Located on the north wing of the Yangtze River estuary, and with 206 km of coastline, Nantong (32.01° N, 120.86° E) is one of the vital coastal port cities to foreign investment since the beginning of reform and opening up in Jiangsu Province, located at the Middle-Lower Yangtze Plain. At present, Nantong is one of the many fast-growing and traditionally industrial cities, with its gross domestic product breaking the 1-trillion-yuan threshold in 2020. However, atmospheric environmental problems have brought much attention in the city with the rapid development. Besides, Nantong has a humid subtropical climate, with four distinct seasons influenced by complex climate systems such as seasonal monsoons and changeable weather, which contribute to a substantial influence on air pollutant emissions, formation, and transport pathways.

2.2. Data and Analysis Methods

Real-time, hourly concentrations of air pollutants, including PM_{2.5} and O_3, at seven national air quality monitoring stations in Nantong were published on an online platform by the China National Environmental Monitoring Centre (CNEMC), while historical data are not openly available. Thus, we used historical data of PM_{2.5} and O_3 from 1 January 2015 to 31 December 2020 by one provider (https://quotsoft.net/air/archived, accessed on 10 June 2021). A sanity check was conducted on the hourly data at individual sites to remove problematic data points before calculating average concentrations and parameters. The citywide hourly mean concentrations of PM_{2.5} and O_3 were calculated by averaging hourly data at all sites in the city, which were used in the analysis, as well as daily,
monthly, and annual mean concentrations. The three-hourly meteorological data containing the wind, temperature, and humidity were obtained from the US National Centers for Environment Prediction’s Global Data Assimilation System (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1, accessed on 10 June 2021) with a grid resolution of $1° \times 1°$.

Figure 1. (a) Map showing the locations of Nantong (yellow circle) and (b) the locations of national air quality stations (red circles) in Nantong.

To explore the influence of air mass transport on PM$_{2.5}$ and O$_3$, 72-h air-mass backward trajectories at 500 m arrival height above ground level were calculated by the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) [23]. This height was chosen to represent a well-mixed convective boundary layer for regional investigation [24,25]. The model was set as four times a day at the starting times of 0:00, 6:00, 12:00, and 18:00 local time during the study period. The number of trajectories was 2164 in spring, 2184 in summer, 2160 in autumn and 2144 in winter over the entire study period. The multiple backward trajectories were clustered in four seasons using Euclidean distance in this study. The most representative cluster number was determined as five using the “eye ball” method in the TrajStat software by plotting the percent change in total spatial variance (TSV) against the number of clusters. This graph first presented a monotonic increase, and thereafter a sudden increase. The cluster number before the first sudden increase was chosen for the clustering process [26,27]. PM$_{2.5}$ and O$_3$ concentrations were grouped according to the seasonal trajectory clusters. The hourly PM$_{2.5}$ and O$_3$ concentrations of 75 $\mu$g/m$^3$ and 200 $\mu$g/m$^3$ were defined as the “polluted” thresholds referring to the Ambient Air Quality Standard (GB3095—2012), respectively [28]. PM$_{2.5}$-O$_3$ was defined as the complex air pollution event with mean PM$_{2.5}$ concentration exceeding 75 $\mu$g/m$^3$ and O$_3$ exceeding 200 $\mu$g/m$^3$ simultaneously.

The potential source areas in different seasons were determined using the potential source contribution function (PSCF) and concentration-weighted trajectory (CWT) methods combining with pollutant concentrations of the receptor site [29]. The investigated area was divided into $1° \times 1°$ small grid cells ($i \times j$) with equal size in both methods. The PSCF value of the $ij$th cell was defined as:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}}$$  \hspace{1cm} (1)$$

where $n_{ij}$ represents the total number of trajectory endpoints falling in the $ij$th cell, and $m_{ij}$ is the number of endpoints when the receptor concentrations exceeded the threshold criterion.
set at the mean concentrations of PM$_{2.5}$ and O$_3$ of each season in Nantong (Table 1). The areas with higher PSCF values denoted the greater probability of potential source locations.

**Table 1.** The average concentrations of PM$_{2.5}$ and O$_3$ in each season over the entire study period used as threshold criteria in the PSCF method.

|       | Spring | Summer | Autumn | Winter |
|-------|--------|--------|--------|--------|
| PM$_{2.5}$ (µg/m$^3$) | 43     | 29     | 36     | 62     |
| O$_3$ (µg/m$^3$)       | 62     | 78     | 70     | 52     |

However, the PSCF method failed to distinguish the grid cells with the same $PSCF_{ij}$ when the pollutant concentrations slightly or prominently exceeded the threshold criterion. The CWT method was used to overcome this limitation [30,31]. In the CWT method, a weighted average of pollutant concentration was assigned to each grid cell, as follows:

$$CWT_{ij} = \frac{\sum_{l=1}^{M} C_l \tau_{ijl}}{\sum_{l=1}^{M} \tau_{ijl}}$$

(2)

where $M$ and $l$ represent the total number of trajectories and the index of the trajectory, respectively. $C_l$ represents the observed pollutant concentration with trajectory $l$ arriving in cell $ij$. $\tau_{ijl}$ is the time spent by trajectory $l$ in the $ij$th cell. Additionally, an arbitrary weight function ($W_{ij}$) was applied to minimize the uncertainty of PSCF and CWT values resulting from small $n_{ij}$ values. The $W_{ij}$ was expressed as:

$$W_{ij} = \begin{cases} 
1.00, & n_{ij} > 3n_{ave} \\
0.70, & 2n_{ave} < n_{ij} \leq 3n_{ave} \\
0.42, & n_{ave} < n_{ij} \leq 2n_{ave} \\
0.05, & n_{ij} \leq n_{ave}
\end{cases}$$

(3)

where $n_{ave}$ denotes the average value of the endpoints in each cell. Thus, the weighted PSCF and CWT values were computed as follows:

$$WPSCF_{ij} = PSCF_{ij} \times W_{ij}$$

(4)

$$WCWT_{ij} = CWT_{ij} \times W_{ij}$$

(5)

### 3. Results and Discussion

#### 3.1. Evolution Characteristics of PM$_{2.5}$ and O$_3$

The evolution trends of annual pollutant concentrations in Nantong were investigated first (Figure 2 and Table 2). From 2015 to 2020, PM$_{2.5}$ and O$_3$ presented a net decreasing trend of $-3.7 \ \mu g/m^3$ and $-1.2 \ \mu g/m^3$ per year, respectively. Very different evolution characteristics were observed for PM$_{2.5}$ and O$_3$. PM$_{2.5}$ declined obviously and steadily over the entire period except for a slight rebound in 2018, while O$_3$ in 2017 bounced back to levels higher than those in 2015, which was attributed to the most frequent extreme high-temperature events (14 days above 35 °C) that year. These results are consistent with a previous study [32]. In addition, the O$_3$ trend was relatively smooth over the six years. Although considerable reductions of PM$_{2.5}$ were observed, pollution control measures did little to O$_3$ due to its complicated nonlinear photochemistry formation, which relied on precursor diagnosis and meteorological conditions. Notably, in 2020, the average PM$_{2.5}$ concentration was down to 34.7 $\mu g/m^3$ below the minimum safe level of 35 $\mu g/m^3$ according to ambient air quality standards for residential areas, which was likely due to the drastically reduced emission of primary air pollutants by lockdown measures during the COVID-19 outbreak between January and February 2020 [33].
On each box, the central mark shows the median, and the bottom and top edges of the box mark the 25th and 75th percentiles, respectively. The solid dots represent the annual mean values. Dashed lines show the long-term trends of pollutants.

Table 2. Annual mean concentrations (unit: µg/m$^3$), standard deviations (unit: µg/m$^3$) and corresponding linear trends (lr) of PM$_{2.5}$ and O$_3$ (unit: µg/m$^3$/year) from 2015 to 2020.

|        | 2015    | 2016    | 2017    | 2018    | 2019    | 2020    | lr    |
|--------|---------|---------|---------|---------|---------|---------|-------|
| PM$_{2.5}$ | 56.5 ± 36.1 | 46.1 ± 29.3 | 39.5 ± 23.8 | 41.4 ± 28.8 | 37.2 ± 23.5 | 34.7 ± 24.0 | −3.9   |
| O$_3$   | 72.2 ± 26.7 | 71.2 ± 28.1 | 77.3 ± 28.2 | 69.6 ± 25.2 | 68.0 ± 25.1 | 67.3 ± 22.1 | −1.2   |

The long-term variations of mean PM$_{2.5}$ and O$_3$ concentrations in different seasons were investigated as well (Figure 3). The mean PM$_{2.5}$ concentrations decreased in all seasons over the entire study period except for the rebound in autumn of 2018 related to the unfavorable diffusion conditions of low wind speeds, high relative humidity, and inversion layers. Among the four seasons, the highest concentrations with the most obvious declination of PM$_{2.5}$ was observed in winter. However, the decline of PM$_{2.5}$ slowed down in recent years. In addition, compared with PM$_{2.5}$, the O$_3$ concentrations first increased then decreased in all seasons with peak values in 2017 (spring, summer, winter) or 2018 (autumn) but changed slightly in general. Higher concentrations with larger fluctuations were observed in summer and spring than in autumn and winter. Those results were consistent with the yearly patterns shown in Figure 2.

Figure 4 shows the evolution of polluted hours of PM$_{2.5}$, O$_3$, and PM$_{2.5}$-O$_3$ during different seasons from 2015 to 2020. Generally, hours of PM$_{2.5}$ polluted hours had sharply decreasing trends from 1795 h to 746 h over the entire period, with a seasonal pattern peaking in winter likely resulting from unfavorable meteorological conditions, followed by spring and fall. However, O$_3$ initially increased then decreased, peaking with 200 h in 2017. Unlike PM$_{2.5}$, O$_3$ and PM$_{2.5}$-O$_3$ polluted hours occurred most frequently in summer and none were in winter, which mostly depended on the intensity of solar radiation. PM$_{2.5}$-O$_3$ complex air pollution represented a declining trend with fluctuations, rebounding sometimes such as summer in 2017 and spring in 2018 when the consecutive extreme high-temperature events happened. It is remarkable that no complex polluted hours occurred in 2019 and 2020 all year round, indicating the air pollution controls, as yet, were imperfectly achieved but already having an effect.
3.2. Transport Characteristics

To identify the transport pathways of air masses, back trajectory clustering was utilized. Five major cluster pathways and corresponding statistical results for each season over the entire period were shown in Figure 5 and Table 3. Generally, longer trajectories corresponded to higher velocity of air mass movement. The ratios of clusters during four seasons were relevant to the seasonal monsoons in Nantong, with a prevailing northerly wind in winter, a prevailing southerly wind in summer, and a transition in spring and autumn. In addition, variable weather conditions had a substantial impact as well.
Table 3. Statistical results of the air pollutant concentrations for each cluster in the four seasons of Nantong. The Ratio denotes the percentage of trajectory numbers in all trajectories of each cluster, and P_Ratio is the percentage of polluted trajectory numbers in each cluster.

| Season | Cluster | Ratio (%) | PM$_{2.5}$ Mean ± Std (µg/m$^3$) | P_Ratio (%) | O$_3$ Mean ± Std (µg/m$^3$) | P_Ratio (%) |
|--------|---------|-----------|-----------------------------------|-------------|-------------------------------|-------------|
|        |         |           | Mean ± Std (µg/m$^3$)             |             | Mean ± Std (µg/m$^3$)         |             |
|        |         |           | 20.00 ± 30.50                     | 30.61       | 87.00 ± 45.37                 | 25.53       |
|        |         |           | 30.91 ± 31.22                     | 46.62       | 77.14 ± 55.97                 | 59.57       |
|        |         |           | 29.67 ± 21.53                     | 14.23       | 82.57 ± 36.82                 | 8.51        |
|        |         |           | 9.52 ± 26.87                      | 7.47        | 80.04 ± 38.29                 | 6.38        |
|        |         |           | 7.90 ± 17.71                      | 1.07        | 82.15 ± 28.27                 | 0.00        |
|        |         |           | 11.08 ± 24.43                     | 26.76       | 101.6 ± 54.8                  | 21.25       |
|        |         |           | 31.55 ± 20.02                     | 38.03       | 82.02 ± 64.63                 | 58.75       |
|        |         |           | 16.12 ± 9.10                      | 1.41        | 50.32 ± 37.50                 | 2.50        |
|        |         |           | 32.33 ± 17.77                     | 29.58       | 84.34 ± 38.72                 | 10.00       |
|        |         |           | 8.93 ± 14.78                      | 4.23        | 60.47 ± 51.53                 | 7.50        |
|        |         |           | 41.02 ± 27.08                     | 38.24       | 72.31 ± 40.45                 | 46.67       |
|        |         |           | 24.91 ± 24.54                     | 20.10       | 66.16 ± 34.76                 | 26.67       |
|        |         |           | 14.77 ± 25.93                     | 11.27       | 59.31 ± 30.12                 | 0.00        |
|        |         |           | 11.20 ± 17.94                     | 2.45        | 71.49 ± 31.81                 | 0.00        |
|        |         |           | 8.10 ± 42.20                      | 27.94       | 60.15 ± 52.75                 | 0.00        |
|        |         |           | 13.57 ± 36.69                     | 8.98        | 56.88 ± 25.32                 | 0.00        |
|        |         |           | 35.26 ± 50.35                     | 50.08       | 49.24 ± 30.17                 | 0.00        |
|        |         |           | 25.47 ± 45.72                     | 25.27       | 50.98 ± 28.02                 | 0.00        |
|        |         |           | 19.45 ± 30.21                     | 6.24        | 56.76 ± 23.30                 | 0.00        |
|        |         |           | 6.25 ± 39.00                      | 9.44        | 32.57 ± 27.85                 | 0.00        |

In spring, cluster 2 was the predominant pollution pathway accounting for 46.62% (59.57%) of PM$_{2.5}$ (O$_3$) polluted trajectories, respectively, followed by cluster 3. In addition, the mean PM$_{2.5}$ concentration of cluster 2 was the highest among all clusters at 53.66 µg/m$^3$, while cluster 1 had the maximum mean O$_3$ concentration at 87.00 µg/m$^3$. Cluster 2 air masses were short-range sources moving slowly from the nearby industrial provinces of Zhejiang and Jiangxi to the southwest likely picking up considerable anthropogenic aerosols. Cluster 3 originated from South Korea and then traveled southerly over the Yellow Sea. Clusters 1, 4, and 5 represented long-range transport and fast-moving trajectories from Russia and Inner Mongolia with air masses containing soil and dust.

In summer, clusters 2 and 5 were both from the southwest, but traveled short and long pathways from nearby provinces and the South China Sea, respectively. Air masses in cluster 3 were the cleanest with the lowest PM$_{2.5}$ and O$_3$ loadings (16.77 ± 9.10 µg/m$^3$, 50.32 ± 37.50 µg/m$^3$) originating from the Pacific Ocean directly. In general, southerly clusters 2, 3, and 5 contributed 56.60% of all trajectories, which were consistent with the prevailing southerly monsoon. Cluster 4 was from South Korea and passed over the Yellow Sea. Cluster 1 came from inner Mongolia, passing through multiple provinces before arriving at Nantong. In addition, clusters 1 and 2 contributed the largest percentage (80.00%) of polluted O$_3$ trajectories in summer (Table 3). Air masses of most clusters in summer had relatively lower PM$_{2.5}$ and higher O$_3$ concentrations than those in other seasons.

In autumn, all clusters except cluster 5 with a total ratio of 91.90% gathered trajectories from the north. Among all clusters, cluster 1 had the highest ratio of trajectories (41.02%) and polluted PM$_{2.5}$ and O$_3$ trajectories (38.24% and 46.67%), which originated from the Yellow Sea. Cluster 5 originated from Jiangxi Province, passed through Anhui Province with the lowest ratio of trajectories, however, it had the highest mean PM$_{2.5}$ concentration at 63.83 µg/m$^3$. Cluster 3 and 4 were free of O$_3$ polluted trajectories, with air masses from the Mongolia and Japan Sea, respectively.
Figure 5. Mean backward trajectory clusters in (a) spring, (b) summer, (c) autumn, and (d) winter from 2015 to 2020 in Nantong. The number of each cluster and corresponding ratios are also shown. The black dot denotes the location of Nantong. The line colors are used to clarify transport pathways from different clusters.

In winter, north and northwest clusters prevailed, comprising 93.75%. The cluster 1, 3 and 2 were from similar northwest directions but distinct transport distances. Among these clusters, the 2nd cluster showed the greatest occurrence probability as well as ratios of polluted PM$_{2.5}$ trajectories. Besides, the cluster 2 originated from Shandong Province with shorter trajectories, likely picking up more local and anthropogenic air masses. Notably, although cluster 5 had higher PM$_{2.5}$ concentrations than cluster 2, it had a limited impact on PM$_{2.5}$ concentrations in Nantong due to its least ratio among all clusters. There was no O$_3$ pollution event in winter on account of the unfavorable weather conditions for photochemical reactions.

Given the above, the main factors impacting the PM$_{2.5}$ and O$_3$ polluted trajectories in each season of Nantong were sources from nearby short-distance rather than long-distance. Additionally, as a coastal city, marine air masses played a very important role as well as those from the adjacent provinces.

### 3.3. PSCF and CWT Modeling of Source Regions

Figures 6 and 7 show the PSCF and CWT results for different seasons in Nantong. As an auxiliary, the CWT values can help quantify the relative contribution of pollutants in each grid compensating for the weakness of PSCF. Generally, the greater PSCF and CWT values denoted higher contributions to PM$_{2.5}$ and O$_3$ concentrations.

For PM$_{2.5}$, in all seasons, source regions from the western adjacent provinces were with higher PSCF (>0.6) and CWT (>60 $\mu$g/m$^3$) values, compared to the marine source areas with lower PSCF (<0.3) and CWT (<30 $\mu$g/m$^3$) values. As a result, the main factors impacting the PM$_{2.5}$ pollution in Nantong were sources from inland areas, covering the Anhui, Henan, Hubei, Shanxi, and Shaanxi Province, and as far as inner Mongolia, rather than marine areas. Most of the potential source domains were distributed from southeast to northwest clockwise in all seasons, which were consistent with the prevailing wind direction. The largest domain of potential sources exceeding the mean concentration of
PM$_{2.5}$ occurred in autumn according to the PSCF results, followed by winter, then spring and summer. However, the CWT analysis indicated that the concentrations of potential sources were the greatest exceeding 100 µg/m$^3$ in winter. Therefore, a comprehensive analysis using both the PSCF and CWT values is necessary. Besides, polluted air masses mostly came from the northwesterly clusters contributing 84.32% of all polluted trajectories in winter (Table 3).

Figure 6. The PSCF maps of the potential sources of PM$_{2.5}$ and O$_3$ in spring (a,e), summer (b,f), autumn (c,g), and winter (d,h) for the entire period during 2005–2010 in Nantong. The black dot denotes Nantong.
Figure 7. The CWT maps of the potential sources of PM$_{2.5}$ and O$_3$ in spring (a,e), summer (b,f), autumn (c,g), and winter (d,h) for the entire period during 2005–2010 in Nantong. The black dot denotes Nantong.

The O$_3$ potential source regions had a similar pattern with PM$_{2.5}$ in terms of the whole distribution area. However, the locations of more polluted source regions were much different. In addition to the source regions from the North China Plain, air masses over the Bohai Sea and the Yellow Sea also contributed a great deal to O$_3$ concentrations in Nantong. It was likely due to the transport of O$_3$ and its precursors by the transition between land and sea breeze circulation near the northern industrial coastal cities, which is consistent with the results of previous studies [34–36]. The severely polluted source regions varied seasonally. The polluted trajectories traveled roughly northwest-southeast, in spring, autumn and winter. Unlike these seasons, major severe sources of O$_3$ in summer
came mostly from the southwest to northeast clockwise with the largest polluted area and the greatest values exceeding 100 $\mu$g/m$^3$. Meanwhile, these areas accounted for 97.5% of the polluted trajectories in summer as shown in Table 3.

4. Conclusions

A comprehensive characterization of evolution, transport, and potential source regions of PM$_{2.5}$ and O$_3$ were investigated from 1 January 2015 to 31 December 2020 in Nantong. The annual evolution of PM$_{2.5}$ (O$_3$) concentrations and corresponding trends of pollution hours were presented in detail. The transport pathways and potential source regions of PM$_{2.5}$ and O$_3$ were identified and determined by cluster analysis, PSCF, and CWT methods, respectively. The major conclusions were as follows:

The annual mean PM$_{2.5}$ concentration declined obviously from 56.5 $\mu$g/m$^3$ to 34.7 $\mu$g/m$^3$ over the entire study period. O$_3$ had a relatively smooth decreasing trend, but rebounded greatly during 2017 when the most frequent extreme high-temperature events occurred. Similar trends were observed for PM$_{2.5}$-O$_3$ polluted hours with some fluctuations, with a sharp decrease from 2015 to 2016 and then an increase to the peak values in 2018. No PM$_{2.5}$-O$_3$ complex polluted event happened in 2019 and 2020 indicating the preliminary effect of the implementation of emission controls.

Notable differences in transport pathways and frequencies were observed in four seasons in Nantong. Air masses of most clusters in summer had the lowest (highest) PM$_{2.5}$ (O$_3$) concentrations than those in other seasons. Clusters with the largest percentage of polluted PM$_{2.5}$ and O$_3$ trajectories were from the southwest adjacent provinces in spring and summer, but the northwest adjacent provinces in winter and the northeast ocean near Nantong in autumn, which was mostly short-distance sources rather than long-distance transport sources.

The PSCF method mainly focused on sources identification to calculate and describe possible source locations while the CWT method can distinguish the source strength more easily by assigning the concentrations values at the receptor site. The PSCF and CWT results showed that PM$_{2.5}$ sources in Nantong were from the adjacent western and northwestern provinces with higher PSCF (>0.6) and CWT (>60 $\mu$g/m$^3$) values, and the influence of marine sources was relatively small with lower PSCF (<0.3) and CWT (<30 $\mu$g/m$^3$) values. The O$_3$ potential source regions had a similar distribution pattern but significantly different polluted source regions with PM$_{2.5}$. Apart from the source regions of O$_3$ from the North China Plain, potential sources from the Bohai Sea and the Yellow Sea also contributed a great deal, which is attributed to transport of O$_3$ and its precursors by the transition between land and sea breeze circulation near the northern industrial coastal cities. In addition, the severely polluted source regions of PM$_{2.5}$ and O$_3$ varied seasonally. Polluted air masses of PM$_{2.5}$ mostly came from the northwesterly clusters contributing 84.32% of all polluted trajectories in winter, while major severe sources of O$_3$ from the southwest to northeast clockwise accounted for 97.5% of the polluted trajectories in summer.

The results presented here suggest that, despite the effort made, control of PM$_{2.5}$ and O$_3$ emissions from the adjacent provinces will further play a significant role in achieving compliance with the air quality standard in Nantong. Nonetheless, a detailed further investigation of the impact of meteorological conditions on pollution transport pathways is still needed, which will provide an important scientific basis to explore efficient air pollution reduction strategies.

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