

Analysis of regional heavy PM_{2.5} pollution events in Beijing-Tianjin-Hebei and the surrounding area in China during 2015–2018

Zhanshan Wang^{1, +}, Yan Qian^{1, +}, Zhigang Li¹, Kai Wu^{2, 3}, Chen Guo¹, Liang Zhang^{4, +},
Xiaoqian Li^{1, *}, Jian Guo⁵

¹ State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing, China

² Plateau Atmosphere and Environment Key Laboratory of Sichuan Province, School of Atmospheric Sciences, Chengdu University of Information Technology, Chengdu, China

³ Department of Land, Air, and Water Resources, University of California, Davis, CA, USA

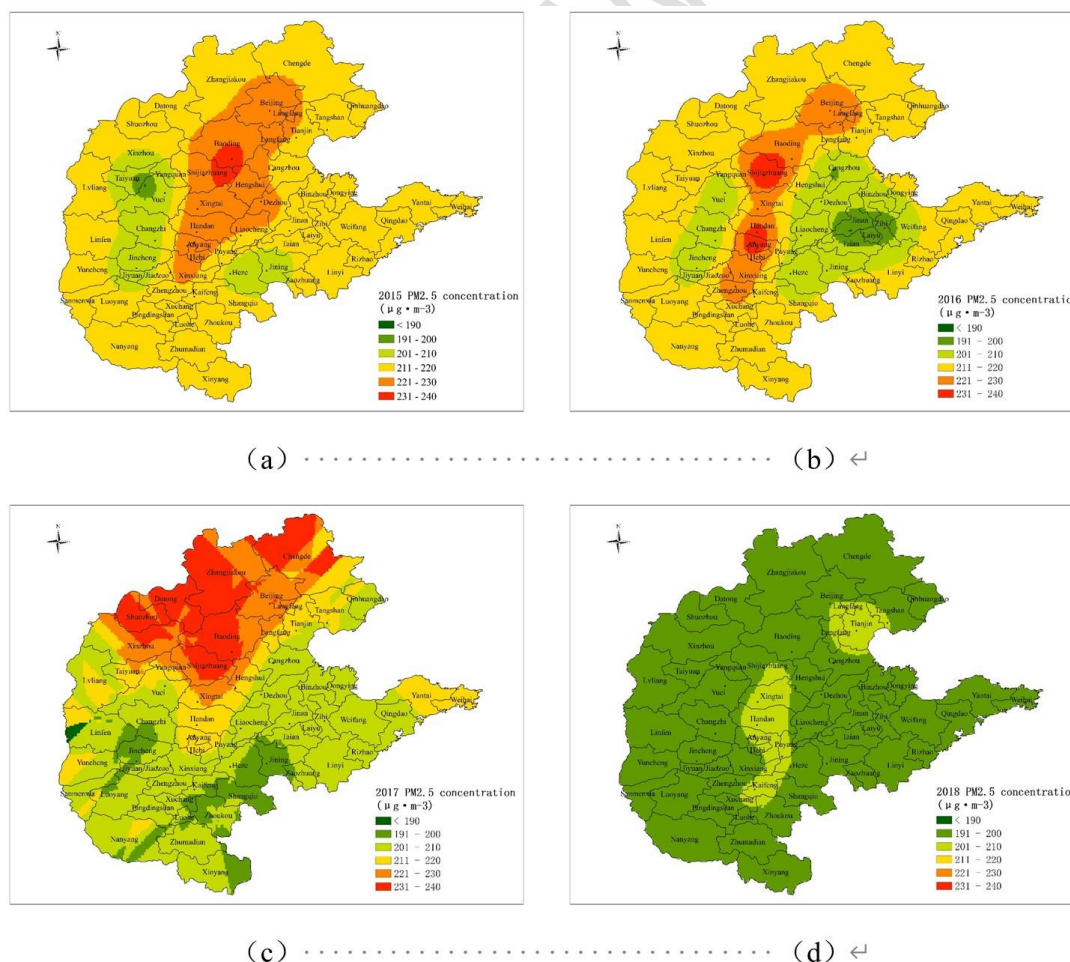
⁴ Center for Environmental Emergency and Heavy Pollution Early Warning, Hebei Environmental Protection Bureau, Shijiazhuang, China

⁵ Jinan Environmental Research Institute, Jinan 250101, China

Correspondence to: Xiaoqian Li (hkyzkjh@sina.com) and Liang Zhang (qingkongweilan@tom.com)

+ These authors contributed equally to this work.

GRAPHICAL ABSTRACT



Abstract.

Beijing-Tianjin-Hebei and the surrounding area (BTHS) experiences the most serious PM_{2.5} pollution in China. To investigate PM_{2.5} pollution characteristics in regional heavy air pollution, forty-three events in BTHS during 2015–2018 were selected in this study. These regional events generally lasted five days and covered nineteen cities, with an average PM_{2.5} concentration of 204 $\mu\text{g}\cdot\text{m}^{-3}$ and a maximum daily average concentration of 358 $\mu\text{g}\cdot\text{m}^{-3}$. The pollution events occurred mainly during October to March of the following year, which is the heating season in this region. The occurrence of pollution events and the PM_{2.5} concentration displayed a decreasing trend during 2015–2018. Of the forty-three events, thirteen started in Baoding city, and the peak PM_{2.5} concentration was recorded in Baoding city eleven times, indicating that it was the most polluted city in BTHS. A comprehensive classification method determined that southwest channel pollution was the main type of pollution event in BTHS. It occurred nine, seven, four, and eight times in 2015–2018, accounting for 56.3%, 53.8% 80.0%, and 88.9% of all pollution events, respectively. A typical regional heavy PM_{2.5} pollution event occurred in March 2016. The analysis revealed that it was initiated by pollutant transport from regions south of BTHS. Local pollutant emissions and regional pollutants accumulated, and secondary pollutant formation occurred in association with weak winds and a high relative humidity, which maintained the heavy pollution levels. Relative humidity had the most significant influence on the two most important stages of a heavy PM_{2.5} pollution event, which were the initial and high concentration stages, respectively.

Keywords : Beijing-Tianjin-Hebei region; PM_{2.5} pollution; spatial-temporal characteristics; classification; relative humidity

1 Introduction

With rapid industrialization and urbanization, and the continuous increase in energy consumption, air pollution has become serious in China during recent years (Hou et al., 2019; Ji et al., 2018; Li et al., 2020; Cheng et al., 2019; Ding et al., 2019). Regional air pollutants, particularly PM_{2.5}, have become the main factor restricting the improvement

of environmental quality in China (Yan et al., 2018; Gong et al., 2019; Wang et al., 2015a; Ji et al., 2019; Wang et al., 2019; Yu et al., 2014).

Beijing-Tian-Hebei and the surrounding area (BTHS) is one of the most PM_{2.5}-polluted areas in China. Geographically, BTHS is located in the north of the North China Plain, with the Yanshan Mountains to the north, the Taihang Mountains to the west, and Bohai Bay to the east. The region consists of semi-enclosed topographic features, with a high elevation in the northwest and a low elevation in the southeast; thus, air pollutants can easily accumulate in this area. In terms of meteorological conditions, BTHS is dry and rainless, leading to weak wet deposition of air pollutants. The mountains in the northwest weaken the pollution-removal effect of the northwest monsoon that prevails in winter, and therefore the diffusion of air pollutants is relatively poor in BTHS. In addition, air pollutant emissions are high due to the heavy industry-based industrial structure, coal-based energy structure, and highway-based traffic structure (Ye et al., 2019; Yi et al., 2018; Wu et al., 2018b).

Studies of PM_{2.5} pollution in BTHS have been conducted during recent years. Some studies have revealed the temporal-spatial characteristics of PM_{2.5} in different years (Lei et al., 2016; Zhao et al., 2017; Hu et al., 2014; He et al., 2020; Xue et al., 2019). Other studies have focused on the source apportionment of air pollutants in this region (Zhang et al., 2017; Wang et al., 2015b; Chen et al., 2017; Zhu et al., 2019). The health effects of PM_{2.5} have also been investigated (Feng, 2017; Song et al., 2011; Yang, 2020; Li et al., 2019; Zheng et al., 2019; Zhao et al., 2019), while some studies have assessed the effects of long-term and temporary measures to reduce air pollutant emissions (Guo et al., 2018; Wang, et al., 2018; Wang et al., 2016).

However, most of those previous studies were limited to few sampling locations or a short sampling period. In this study, PM_{2.5} monitoring data from twenty-seven cities in BTHS during a 4-year period (2015–2018) were analyzed. The main objectives of the study were to (1) define and identify regional heavy PM_{2.5} pollution events in BTHS, (2) characterize the spatial and temporal variations in PM_{2.5} concentrations in regional heavy air pollution, (3) determine a classification system for regional heavy PM_{2.5} pollution events in BTHS, and (4) analyze typical PM_{2.5} pollution events in BTHS.

2 Data collection and analysis method

Twenty-seven cities in BTHS were selected (Figure 1). For each city, PM_{2.5} data were obtained from the China National Environmental Monitoring Center (<http://www.cnemc.cn/>). Quality assurance and quality control of PM_{2.5} data followed the Ambient Air Quality Standards (GB3095-2012) and Technical Regulation on Ambient Air Quality Index (on trial) (HJ 633-2012). Meteorological data and PM_{2.5} chemical composition were obtained for the analysis of typical PM_{2.5} pollution events. Meteorological data from the Guanxiangtai site was obtained from the National Meteorological Information Center (<http://data.cma.cn>).

The PM_{2.5} chemical composition was observed at the Langfang Municipal Ecology and Environment Bureau (LFMEEU), which is located 10 km outside of Beijing (Figure 1). Organic carbon (OC) and elemental carbon were measured using the RT-4 analyzer (Sunset Lab Inc., Portland, OR, USA). Water-soluble cations and anions were measured by ICS-2000 and ICS-3000 ion chromatographic analyzers, respectively (Dionex, Sunnyvale, CA, USA). To ensure the validity and accuracy of the monitoring data, all monitoring instruments were calibrated regularly according to national standards. Volatile organic compounds were removed by inline parallel carbon denuder removing installed on the analyzer. Round 16-mm quartz filters were used for collecting OC and EC samples at a sampling flow rate of 8 L/min. OC and EC samples were collected for 40min and then were analyzed in approximately 15 min. The multiple programmed steps were based on the NIOSH 5040 thermal protocol. A non-dispersive infrared (NDIR) detector was used for quantifying particulate OC and EC concentrations.

The kriging method was selected to obtain the spatial characteristics of PM_{2.5} in BTHS. As one of main contents of geostatistics, kriging interpolation is a method of unbiased optimal estimation for regionalized variables, which is based on variation function theory and structural analysis. Kriging model mainly uses variograms of geographic factors and the structural characteristics of the original data to perform unbiased and linear optimal interpolation estimation of spatial variables. This model can overcome the problem that the interpolation error is difficult to analyze, can

3 Results and discussion

3.1 Definition of regional heavy PM_{2.5} pollution events

According to the definition used by the China National Environmental Monitoring Centre, when the daily average PM_{2.5} concentration in three or more of the twenty-seven cities reached 150 $\mu\text{g}\cdot\text{m}^{-3}$ for three or more days, the pollution episode was classified as a regional heavy PM_{2.5} pollution event. Forty-three such events were detected during 2015–2018, as shown in Table S1 in supplementary material. Start city refers to the city with the highest PM_{2.5} concentration on the day that the regional heavy PM_{2.5} pollution event began. In general, regional heavy PM_{2.5} pollution events, with an average PM_{2.5} concentration of 204 $\mu\text{g}\cdot\text{m}^{-3}$ and maximum daily average PM_{2.5} concentration of 358 $\mu\text{g}\cdot\text{m}^{-3}$, lasted five days. On average, nineteen cities were affected by one regional heavy PM_{2.5} pollution event. Thus, regional pollution events resulted in serious PM_{2.5} pollution conditions.

3.2 Temporal characteristics of regional heavy PM_{2.5} pollution events

Regional heavy PM_{2.5} pollution events occurred sixteen, thirteen, five, and nine times from 2015 to 2018, respectively, indicating a decreasing trend, with a rebound in 2018. These events occurred mainly during October to March of the following year, which is the heating season in this region. The period between April and September was typically a clean period without any regional heavy PM_{2.5} pollution events. During the clean period, air pollutant emission is lower and atmospheric pollution diffusion condition is more favorable than other time period. In general, these events lasted for five, five, eight, and five days during 2015–2018, respectively. In 2017, these events occurred the least frequently but lasted longer than those during other years. The events took two, two, three, and two days to reach the PM_{2.5} concentration peak during 2015–2018, respectively. The average PM_{2.5} concentrations in these events were 211, 196, 215, and 196 $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The maximum PM_{2.5} concentrations in these events were 374, 345, 416, and 308 $\mu\text{g}\cdot\text{m}^{-3}$, respectively, indicating a decreasing trend, with a rebound in

2017. In general, the occurrence and PM_{2.5} concentration displayed a decreasing trend during 2015–2018.

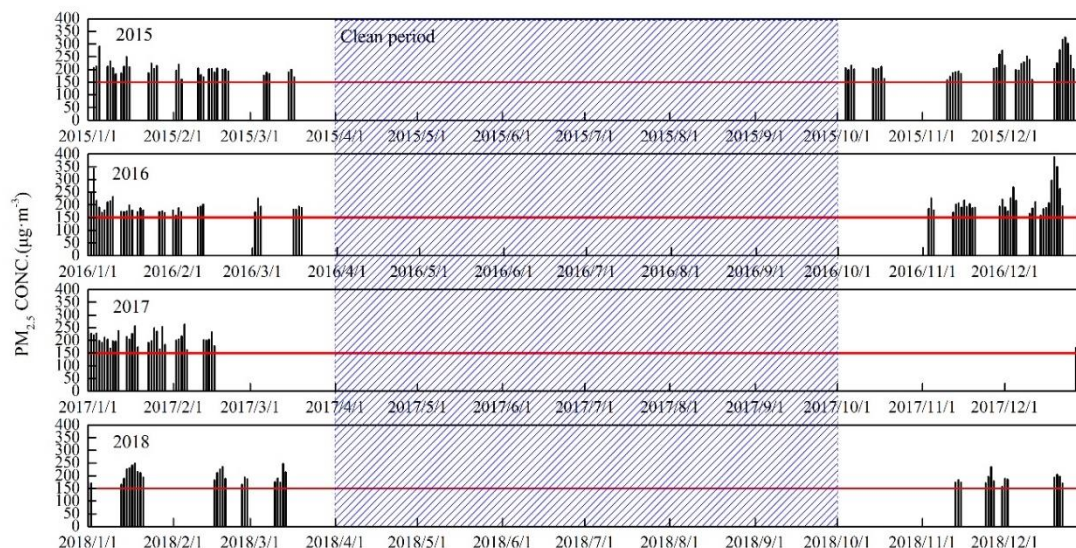


Figure 2. The average daily PM_{2.5} concentrations in the twenty-seven cities during 2015–2018. The time from April to September was defined as a clean period, because no regional heavy PM_{2.5} pollution events occurred.

3.3 Spatial characteristics of regional heavy PM_{2.5} pollution events

Regional heavy PM_{2.5} pollution events affected eighteen, seventeen, twenty-four, and twenty cities from 2015 to 2018, respectively. In 2017, the events occurred the least frequently but had the highest PM_{2.5} concentration, lasted the longest duration, and affected the most cities. Table 1 shows the frequencies at which each city became the start city and had the peak concentration. Of the forty-three events, thirteen started in Baoding city, accounting for 30.2% of the events, followed by Anyang (five events), Shijiazhuang (five events), Handan (five events), and Taiyuan (five events). Of the twenty-seven cities, seventeen were the start city at some time (Figure 3). The peak PM_{2.5} concentration was recorded eleven times in Baoding city, accounting for 25.6% of all peak concentrations, followed by Shijiazhuang (six times) and Beijing (five times). The peak PM_{2.5} concentration was recorded in fifteen of the twenty-seven cities at some point during the study (Figure 3). In general, PM_{2.5} pollution was most serious in

Baoding and Shijiazhuang.

Table 1 Descriptive statistics of regional heavy PM_{2.5} pollution events in individual cities.

| City | Frequency of being the start city | | | | |
|--------------|---|------|------|------|-------|
| | 2015 | 2016 | 2017 | 2018 | Total |
| Baoding | 7 | 3 | 2 | 1 | 13 |
| Zhengzhou | 2 | | | | 2 |
| Beijing | 1 | | | | 1 |
| Beijing | 1 | 1 | | | 2 |
| Liaocheng | 1 | | | | 1 |
| Dezhou | 1 | | | | 1 |
| Jinan | 1 | | | | 1 |
| Shijiazhuang | 1 | 2 | | 1 | 4 |
| Puyang | 1 | | | 1 | 2 |
| Handan | 1 | 1 | 1 | | 3 |
| Anyang | | 2 | 1 | 2 | 5 |
| Xinxiang | | 2 | | | 2 |
| Tangshan | | 1 | | | 1 |
| Taiyuan | | 2 | 1 | | 3 |
| Hengshui | | | | 1 | 1 |
| Kaifeng | | | | 1 | 1 |
| Heze | | | | 1 | 1 |
| City | Frequency of being the city with the peak PM _{2.5} concentration | | | | |
| | 2015 | 2016 | 2017 | 2018 | Total |
| Anyang | 4 | 1 | 1 | 3 | 9 |
| Baoding | 5 | 3 | 2 | 1 | 11 |
| Liaocheng | 1 | | | | 1 |
| Jiaozuo | 1 | 1 | | | 2 |
| Xingtai | 1 | | | 1 | 2 |
| Langfang | 1 | | | | 1 |
| Beijing | 2 | 2 | | 1 | 5 |
| Hengshui | 1 | | | | 1 |
| Xinxiang | 1 | | | | 1 |
| Dezhou | | 1 | | | 1 |
| Changzhi | | 1 | | | 1 |
| Taiyuan | | 2 | | | 2 |
| Shijiazhuang | | 2 | 2 | 2 | 6 |
| Handan | | 1 | | | 1 |
| Kaifeng | | | | 1 | 1 |

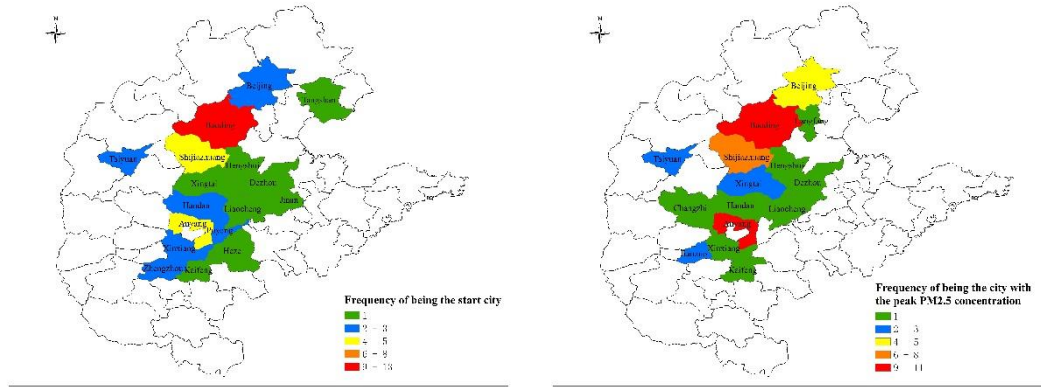


Figure 3. Frequency of being the start city and being the city with the peak PM_{2.5} concentration in the forty-three events evaluated in Beijing-Tianjin-Hebei and the surrounding area (BTHS).

Figure 4 shows the spatial distribution of the PM_{2.5} concentration based on the Kriging method (Young et al., 2016; Wu et al., 2018a; Ma et al., 2019; Jamil and Bellos, 2019) in regional heavy PM_{2.5} pollution events during 2015–2018. In 2015, the region with the highest PM_{2.5} concentration had a zonal distribution alongside the Taihang Mountains, affecting the cities of Beijing, Baoding, Langfang, Shijiazhuang, Hengshui, Xingtai, Handan, and Anyang. The PM_{2.5} concentrations were highest in Baoding and Shijiazhuang. In 2016, the region with the highest PM_{2.5} concentrations also had a zonal distribution, but the extent of the affected region decreased. The PM_{2.5} concentrations were highest in Anyang and Shijiazhuang. In 2017, the spatial distribution changed, and the highest concentrations occurred in the northern area of BTHS, including Baoding, Shijiazhuang, and Beijing. In 2018, PM_{2.5} concentrations were significantly lower than during the first three years. The PM_{2.5} concentrations were higher in Tianjin and the southern region of the Taihang Mountains than elsewhere in the region. The spatial characteristics of PM_{2.5} during the 4-year period also indicated that the air quality was improving.

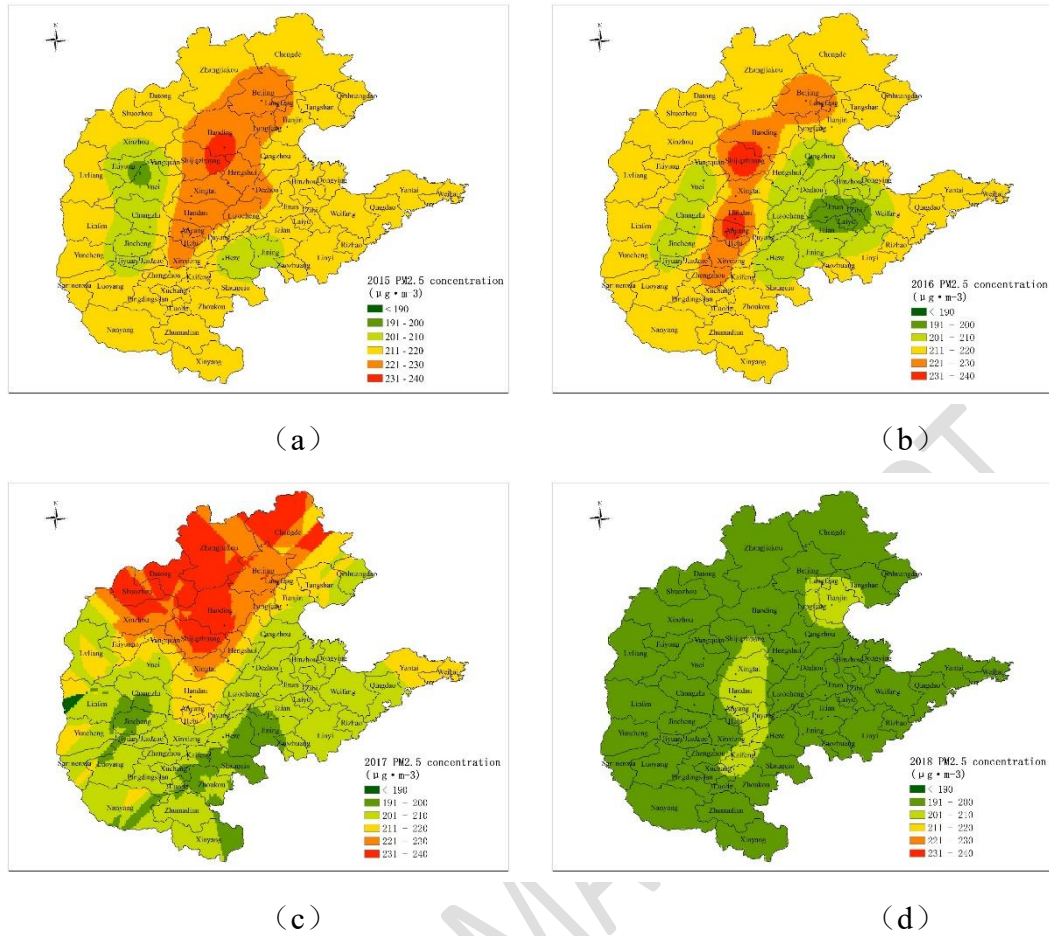


Figure 4. Spatial distribution of the PM_{2.5} concentration during heavy PM_{2.5} pollution events during 2015–2018.

3.4 Classification of regional heavy PM_{2.5} pollution events

According to their geographical orientation and influence on Beijing, the twenty-seven cities were divided into two transmission channels, southwest and southeast channels, as shown in Figure 1.

Three methods were applied to classify the regional heavy PM_{2.5} pollution events, with the preliminary results shown in Table 2. Based on the location of the start city and the city with the peak concentration, it was apparent that most events were of the southwest channel transmission type. According to the number of cities in the different channels, the results were different from those obtained using other methods. More events in 2015 and 2016 were of the southeast channel type, while most events in 2017 and 2018 were of the southwest channel type. A comprehensive classification method

was developed based on this. If more than 80% of the affected cities were in one channel, then the event was defined as the corresponding channel type. Additionally, the classification was conducted according to the location of the start city. The final classification of the forty-three events is shown in Table S1. The southwest channel pollution type was the main type in BTHS; it occurred nine, seven, four, and eight times from 2015 to 2018, respectively, accounting for 56.3%, 53.8%, 80.0%, and 88.9% of all events.

Table 2 Preliminary classification of regional heavy PM_{2.5} pollution events

| Year | Start city | | City with the peak concentration | | Number of affected cities | |
|------|------------|-----------|----------------------------------|-----------|---------------------------|-----------|
| | Southwest | Southeast | Southwest | Southeast | Southwest | Southeast |
| 2015 | 13 | 3 | 13 | 2 | 7 | 10 |
| 2016 | 12 | 1 | 11 | 1 | 5 | 8 |
| 2017 | 5 | 0 | 5 | 0 | 4 | 1 |
| 2018 | 7 | 1 | 8 | 0 | 7 | 1 |

3.5 Analysis of a typical regional heavy PM_{2.5} pollution event

In March 2016, a 4-day regional heavy PM_{2.5} pollution event occurred after the heating season. The event started on 16 March in Beijing and affected nineteen cities. The daily average PM_{2.5} concentration was 186 $\mu\text{g}\cdot\text{m}^{-3}$, and the daily maximum PM_{2.5} concentration was 288 $\mu\text{g}\cdot\text{m}^{-3}$. The peak concentration occurred in Beijing. Figure 5 shows the evolution of the PM_{2.5} concentration in the nineteen cities. The event was divided into three stages: S1 (00:00–21:00 March 16), S2 (22:00 March 16 to 02:00 March 18), and S3 (03:00 March 18 to 23:00 March 19). At the beginning of S1, the PM_{2.5} concentration in Beijing was at an intermediate level among the nineteen cities. On the afternoon of March 16, the PM_{2.5} concentration increased rapidly in Beijing, which became the city with the highest PM_{2.5} concentration in BTHS. The S1 stage involved the accumulation of pollutants and formation of the regional heavy PM_{2.5} pollution event. In S2, the PM_{2.5} concentration fluctuated but was maintained at the highest level in Beijing of all cities in BTHS. At the end of S2, the peak PM_{2.5} concentration (358 $\mu\text{g}\cdot\text{m}^{-3}$) occurred in Beijing. A high PM_{2.5} concentration was maintained throughout S2. During the first half of S3, the PM_{2.5} concentration in Beijing

decreased slowly but increased in other cities. Then, a large mass of cold air reached the area, and the PM_{2.5} concentration in Beijing decreased rapidly. Cities in the northern area of BTHS were affected by the north wind, and the PM_{2.5} concentration decreased to a low level. Cities in the southern part of BTHS were not affected by this cold air, and a high PM_{2.5} concentration was maintained. The regional heavy PM_{2.5} pollution event finally ended at the end of S3, which was considered the clean-up period.

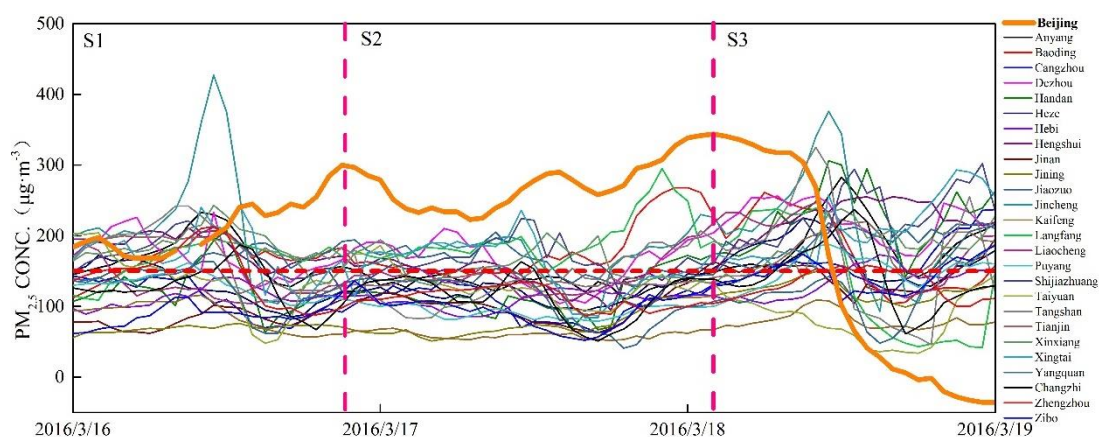


Figure 5. The evolution of the PM_{2.5} concentration in the nineteen cities during this event. The time period S1–S3 refers to the different stages of the event.

Table 3 shows the meteorological data during the different stages. In S1, surface pressure was maintained at a low level, and relative humidity peaked at 57.1%. The wind was weak, with an average speed of 1.7 m/s, and the north wind frequency was only 4.5%. Thus, at the beginning of the event, Beijing was affected by regional transport caused by the persistent southerly wind. In S2, the surface pressure was lower, but the relative humidity was the same, compared with those in S1. Wind speed and visibility kept decreasing, indicating that meteorological conditions were more unfavorable. Although the north wind frequency increased, the wind was too weak to disperse the pollution. In S3, surface pressure, wind speed, and north wind frequency increased, whereas relative humidity decreased to a low level. Due to the arrival of a strong cold air mass, meteorological conditions became favorable, and the event ended.

Table 3 Meteorological data during different stages of the event

| Stage | Surface pressure (hPa) | Temperature (°C) | Relative humidity (%) | Visibility (m) | Wind speed (m/s) | North wind frequency (%) |
|-------|------------------------|------------------|-----------------------|----------------|------------------|--------------------------|
| S1 | 1010.2 | 10.2 | 57.1 | 2840.9 | 1.7 | 4.5 |
| S2 | 1008.1 | 10.9 | 56 | 1517.2 | 1.4 | 37.9 |
| S3 | 1013.6 | 13.1 | 35.1 | 8571.1 | 2.4 | 57.8 |

Figure 6 shows the variation in the chemical composition of PM_{2.5} during this event. The main chemical constituents of PM_{2.5} were SO₄²⁻, NO₃⁻, NH₄⁺, and OC. The location where these samples were collected was 10 km from the border of Beijing and could therefore represent the pollution characteristics of Beijing during a large-scale regional heavy PM_{2.5} pollution event. The SO₄²⁻, NH₄⁺, and OC concentrations were similar. The NO₃⁻ concentration was highest during this event, indicating the contribution of local vehicle emissions. There were several sudden and rapid increases in the NO₃⁻ concentration, indicating that secondary chemical formation had a significant influence on the PM_{2.5} concentrations during this event.

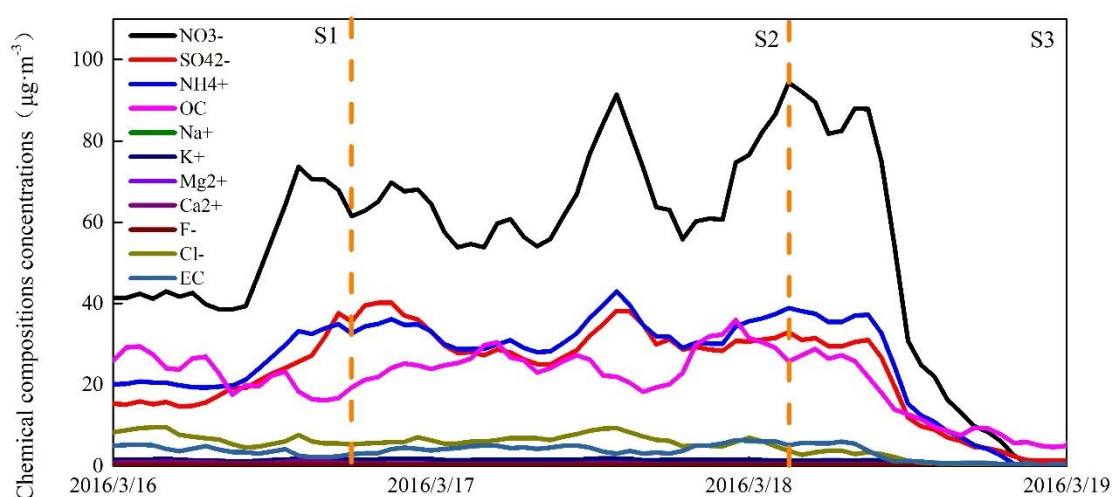


Figure 6. Variations in the chemical composition of PM_{2.5} during this event.

In conclusion, this regional heavy PM_{2.5} pollution event was caused by a combination of local pollutant emissions and regional pollutant transport under unfavorable

meteorological conditions. Pollutant transport from regions south of BTHS initiated the heavy PM_{2.5} pollution in Beijing. Local pollutant emissions and regional pollutants accumulated, and secondary chemical formation occurred, under conditions with weak wind and high relative humidity, which maintained the heavy pollution levels. Finally, the arrival of a strong cold air mass ended this event.

3.6 The relationship between PM_{2.5} and relative humidity

As shown in Table 3, relative humidity and wind speed were the meteorological parameters that had the most significant influence on PM_{2.5}. For the horizontal transport of pollutants, wind from different directions had different effects on PM_{2.5} (Chen et al., 2018; Zhang et al., 2020; Liang et al., 2019; Xie et al., 2019; Liu and Ren, 2019; Chen et al., 2019; Yue et al., 2020). Many studies have confirmed the unfavorable influence of relative humidity on PM_{2.5} (Tai et al., 2010; Fu et al., 2016; Chen et al., 2015; Xie et al., 2020; Sun et al., 2020). The year 2013 was selected for analysis, because the most serious PM_{2.5} pollution occurred during that year. Figure 7 shows the results of a correlation analysis of the relationship between relative humidity and PM_{2.5}. Relative humidity was divided into five ranges to investigate its relationship with the PM_{2.5} concentration. At all ranges, the PM_{2.5} concentration was positively correlated with relative humidity, indicating that an increase in relative humidity aggravated PM_{2.5} pollution. According to the slopes in the five ranges, relative humidity ranges of 80–100% and 20–40% had the most significant influence on the PM_{2.5} concentration. The average PM_{2.5} concentrations within these ranges were 61.6 and 145.6 $\mu\text{g}\cdot\text{m}^{-3}$, respectively. According to the Technical Regulation on Ambient Air Quality Index (on trial), a daily average PM_{2.5} concentration of 75 $\mu\text{g}\cdot\text{m}^{-3}$ is the air quality limit between good and mild pollution. A daily average PM_{2.5} concentration of 150 $\mu\text{g}\cdot\text{m}^{-3}$ is the air quality limit between moderate and serious pollution. Thus, relative humidity had the most significant influence on the two most important stages of a heavy PM_{2.5} pollution event, which were the initial and high concentration stages, respectively. Thus, relative humidity is one of the most important meteorological factors affecting PM_{2.5}

concentrations in Beijing.

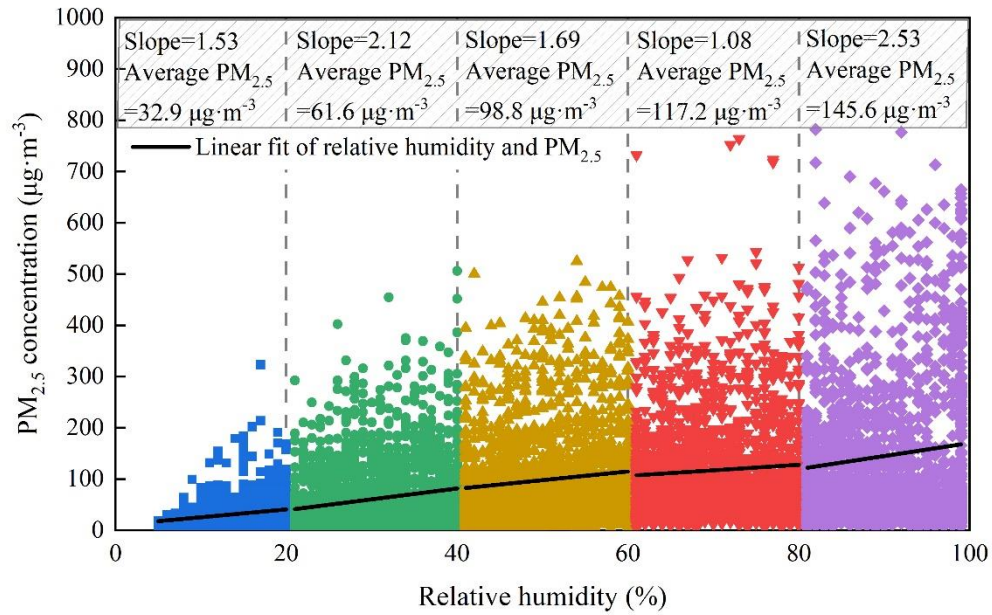


Figure 7. Correlation analysis of relative humidity and PM_{2.5} in Beijing during 2013. The different colors refer to different relative humidity ranges, and the black line refers to the linear fit of relative humidity and PM_{2.5} at different relative humidity ranges.

4 Conclusion and suggestions

- (1) A pollution episode was classified as a regional heavy PM_{2.5} pollution event when daily average PM_{2.5} concentrations in three or more of the twenty-seven cities reached 150 μg·m⁻³ for three or more days. Forty-three events were selected during 2015–2018. In general, regional heavy PM_{2.5} pollution events lasted five days and covered nineteen cities with an average PM_{2.5} concentration of 204 μg·m⁻³ and a maximum daily average PM_{2.5} concentration of 358 μg·m⁻³.
- (2) These events occurred mainly from October to March of the next year, which was the heating season in this region. The occurrence of pollution events and the PM_{2.5} concentration displayed a decreasing trend during 2015–2018. Baoding experienced the beginning of thirteen events and the peak PM_{2.5} concentration eleven times. Shijiazhuang experienced the beginning of five events in Shijiazhuang and the peak PM_{2.5} concentration six times, indicating that Baoding and Shijiazhuang were the

most polluted cities in BTHS.

- (3) Three methods were applied to classify regional heavy PM_{2.5} pollution events. A comprehensive classification method determined that southwest channel pollution was the main type of pollution event in BTHS. Southwest channel pollution occurred nine, seven, four, and eight times in 2015–2018, accounting for 56.3%, 53.8%, 80.0%, and 88.9% of all pollution events, respectively.
- (4) A typical regional heavy PM_{2.5} pollution event, in which Beijing experienced the peak concentration, occurred in March 2016. An analysis of the pollutant characteristics was conducted based on the PM_{2.5} concentration, meteorological data, and chemical composition of PM_{2.5}. The results showed that this event was caused by a combination of local pollutant emissions and regional pollutant transport under unfavorable meteorological conditions. According to a correlation analysis of the relative humidity and PM_{2.5}, relative humidity ranges of 80–100% and 20–40% had the most significant influence on the PM_{2.5} concentration in Beijing.

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