Abstract: Fine particulate matter (PM$_{2.5}$) poses a risk to human health. In January 2017, the PM$_{2.5}$ pollution in China was severe, and the average PM$_{2.5}$ concentration had increased by 14.7% compared to that in January 2016. Meteorological conditions greatly influence PM$_{2.5}$ pollution. The relationship between PM$_{2.5}$ and meteorological factors was assessed using monitoring data and the Community Multiscale Air Quality modeling system (CMAQ) was used to quantitatively evaluate the impacts of variations of meteorological conditions on PM$_{2.5}$ pollution. The results indicate that variations of meteorological conditions between January 2017 and January 2016 caused an increase of 13.6% in the national mean concentration of PM$_{2.5}$. Unlike the Yangtze River Delta (YRD), where meteorological conditions were favorable, unfavorable meteorological conditions (such as low wind speed, high humidity, low boundary layer height and low rainfall) contributed to PM$_{2.5}$ concentration worsening by 29.7%, 42.6% and 7.9% in the Beijing-Tianjin-Hebei (JJJ) region, the Pearl River Delta (PRD) region and the Chengdu-Chongqing (CYB) region, respectively. Given the significant influence of local meteorology on PM$_{2.5}$ concentration, more emphasis should be placed on employing meteorological means to improve local air quality.

Keywords: meteorology; CMAQ model; PM$_{2.5}$; impact

1. Introduction

China’s rapid economic development over the past two decades has caused an increase in PM$_{2.5}$ concentrations [1,2]. The frequent occurrence of heavy air pollution in northern cities during the winter heating season [3,4] restricts sustainable economic development and exacerbes human health risks [5–7]. Studies show that PM$_{2.5}$ concentration did not meet the air quality standards set by the EU in the central and southern Poland, the Czech Republic, Slovakia, Bulgaria, Republic of Macedonia, Serbia, Cyprus and Po Valley in Italy. PM$_{2.5}$ concentration was responsible for about 432,000 premature deaths in Europe in 2012 [8]. The major factors affecting ambient PM$_{2.5}$ concentration are emissions of air pollutants and meteorological conditions [9,10]. Large pollutant emissions in the North China Plain are the major reason for high particulate matter concentrations in this region, and unfavorable meteorological conditions can further increase these concentrations [11]. Meteorological conditions
can affect dilution and diffusion, as well as the accumulation and elimination of air pollutants. The air pollution process characterized by a high concentration of PM$_{2.5}$ pollution occurs rapidly under high humidity and stagnant weather conditions. As a result, variations in meteorological conditions impact the spatial and temporal characteristics of air pollution [12]. Many studies have demonstrated the influence of meteorological conditions on PM$_{2.5}$ pollution. With a focus on the heavy PM$_{2.5}$ pollution events during certain periods of time or within certain short periods of time, Chuang et al. (2017) examined the effects of meteorological parameters and general atmospheric circulation on PM$_{2.5}$ in the atmosphere [13]. By analyzing meteorological features, Wang et al. (2014) discovered that the unfavorable meteorological conditions contributed to a prolonged and severe haze episode over central-eastern China in January 2013 [12]. Some researchers have evaluated the variation of PM$_{2.5}$ concentrations and the relationships between PM$_{2.5}$ concentration and meteorological conditions by using statistical methods [14,15]. Chen et al. (2017) examined seasonal variations of PM$_{2.5}$ concentration within the JJJ region and identified meteorological factors were strongly correlated with local PM$_{2.5}$ concentration [16]. Liang et al. (2017) used a linear regression model to establish the relationship between the concentrations of air pollutants and meteorological parameters [15]. Because of the complicated interactions in the atmospheric environment, quantification of the influence of meteorological factors on local PM$_{2.5}$ concentration remains challenging. Studies on the quantitative effects of meteorology on PM$_{2.5}$ concentration are mainly focused on individual cities or specific urban regions [17], but less on the spatial distribution characteristics on national or key areas, especially in the analysis of secondary inorganic aerosols. Quantitative analysis was conducted by various air quality models. Weather Research and Forecasting (WRF) Model and Community Multiscale Air Quality (CMAQ) were widely adopted to simulate variations of PM$_{2.5}$ concentration and regional pollution transport [18]. For the model performance of 74 major cities in Mainland China, the Normalized Mean Bias and the correlation coefficients for winter were 26.1% and 0.6 in Zheng et al. (2015) [19]. Anthropogenic emissions in China derived from the MEIC model (Multi-resolution Emission Inventory of China) and biogenic emissions calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) were of sufficient quality to support regional and global models, which were widely used as important inputs into air quality model [20,21]. The uncertainties (i.e., 95% confidence intervals around the central estimates) of Chinese emissions of SO$_2$, NO$_x$, total PM, PM$_{10}$, PM$_{2.5}$, black carbon (BC), and organic carbon (OC) were estimated to be $-14\%$-$13\%$, $-13\%$-$37\%$, $-11\%$-$38\%$, $-14\%$-$45\%$, $-17\%$-$54\%$, $-25\%$-$136\%$, and $-40\%$-$121\%$, respectively [22]. The average uncertainties of emissions from the JJJ region were estimated through the Monte Carlo method as $-18\%$ to $16\%$ for SO$_2$, $-17\%$ to $15\%$ for NO$_x$, $-24\%$ to $23\%$ for PM$_{2.5}$, $-19\%$ to $18\%$ for PM$_{10}$, $-25\%$ to $23\%$ for CO, $-48\%$ to $44\%$ for NMVOC, $-54\%$ to $48\%$ for NH$_3$, $-54\%$ to $49\%$ for BC, and $-59\%$ to $55\%$ for OC, which was compiled using a bottom-up approach based on detailed data of major air pollution sources [23]. In total, the model predictions of PM$_{2.5}$ generally agree with ambient measured values, but the model performance varies in different regions.

Since the enforcement of the Air Pollution Prevention and Control Action Plan posted by the State Council in 2013, winter PM$_{2.5}$ pollution in China has decreased over the period 2013–2016 in response to reduced coal consumption and pollution prevention efforts. However, in January 2017, the pollution in China was particularly severe. Compared with January 2016, the mean PM$_{2.5}$ increased by 43.8% and 62.5% in the JJJ region and the PRD regions (http://datacenter.mep.gov.cn), respectively. This abnormal phenomenon has not yet been explained and we report here on the influence of meteorology on the PM$_{2.5}$ concentration in these regions.

In the current study, January 2017 was selected as the target investigation period and characteristics of PM$_{2.5}$ pollution and their relationships with meteorological conditions were analyzed in different regions. Based on the CMAQ modeling system, the scenario analysis method was adopted to systematically simulate variations of PM$_{2.5}$ concentration under different meteorological conditions. Finally, the impact of meteorological conditions on PM$_{2.5}$ pollution was quantitatively analyzed both nationwide and in specific regions where high PM$_{2.5}$ concentrations and precursor emissions were
concentrated, such as the JJJ, PRD, YRD, and CYB regions. The results of this study help us to deepen our understanding of the impact of meteorological conditions on PM$_{2.5}$ pollution and provide insights into possible air pollution control strategies.

2. Data and Methods

2.1. Study Area

We carried out impact analysis of meteorological conditions on PM$_{2.5}$ pollution for the entire area of mainland China at three levels: national, urban agglomeration, and individual cities in the heaviest air pollution regions. Figure 1 shows simulation domain and locations of four regions. The analysis can provide an in-depth picture of spatial change on meteorological factors and their influence on air pollution.

The urban agglomerations in this paper focused on four regions, i.e., the JJJ, YRD, PRD, and CYB regions which are the most highly developed areas in China, and probably the major contributors to air pollution. The total urban areas in these regions only accounted for 12% of the total land area in China in 2015, which is much smaller than their gross domestic product (GDP) and population at 32% and 36%, respectively (Table 1). The JJJ region is the most important economic center in northern China. The rate of economic growth in this area exceeds the national average, but air pollutants in this area are the most severe because of the focus on heavy industry.

Table 1. The proportion of several parameters in key regions.

| Regions | Land Area (%) | GDP (%) | Population (%) |
|---------|---------------|---------|----------------|
| JJJ     | 2             | 10      | 8              |
| YRD     | 2             | 6       | 12             |
| PRD     | 2             | 10      | 8              |
| CYB     | 6             | 6       | 8              |
| Total of the four regions | 12 | 32 | 36 |
2.2. Observational Data

2.2.1. Meteorological Data

Daily meteorological data was obtained from the China Meteorological Data Sharing Service System, including major meteorological parameters such as pressure (P), temperature (T), relative humidity (RH), precipitation (PRE), wind direction (WD), and wind speed (WS) in 2423 surface weather stations.

2.2.2. Ambient Air Quality Data

Air quality monitoring data was taken from the data center within the Ministry of Environmental Protection of the People’s Republic of China including six pollutants concentration of air quality monitoring stations in major cities nationwide. The daily average PM$_{2.5}$ concentrations were collected in January from 2014 to 2017.

2.2.3. Sampling Program

To study the characteristics of PM$_{2.5}$ and chemical components, the PM$_{2.5}$ samples were collected during January 2016 and January 2017 in Beijing, Shijiazhuang, and Tangshan, respectively. As shown in Table 2, the three sites represent the urban area condition as location surrounded by mixed commercial, traffic, and residential condition. 24-h (09:00 to 09:00 local time) PM$_{2.5}$ samples were collected by using an atmospheric PM$_{2.5}$ sampling instrument (Wuhan Tianhong Instruments Co., Ltd., Wuhan, China), in which Whatman 41 filters (Whatman Inc., Maidstone, UK) were equipped at a flow rate of 100 L min$^{-1}$. The sample measurements were conducted in a certified lab (Key Laboratory of Beijing on Regional Air Pollution Control, Beijing). All the filters were weighed before and after sample collection in the lab by using a 1 over 10,000 analytical balance. The filters were equilibrated in constant temperature (20$\pm$5°C) and relative humidity (40$\pm$2% RH) chamber for 48 h. Before the filters were placed back into the samplers, all flow meters were calibrated and the samplers needed to pass a tightness test. The inorganic elements were measured by an inductively coupled plasma-mass spectrometry (ICP-MS, 7500a, Agilent). The ions were analyzed by an ion chromatograph (Metrohm 861 Advanced Compact IC). Furthermore, blank filter fiber and standard chemical samples were tested by using ICP-MS and ion chromatograph, so as to ensure data quality. In the end, valid samples of 25 days per month were collected.

| No. | City     | Sampling Site                  | Sampling Height | Surrounding Environment                  |
|-----|----------|--------------------------------|-----------------|------------------------------------------|
| 1   | Beijing  | Beijing Normal University      | 35 m           | Intensive traffic but little industrial activities |
| 2   | Shijiazhuang | Residential building     | 20 m           | Residential buildings and traffic roads  |
| 3   | Tangshan | Environmental monitoring station | 15 m           | Offices and residential buildings        |

2.3. Emission Inventory

The chemicals in the emission inventory needed by the CMAQ model primarily include various pollutants such as SO$_2$, NO$_x$, particulate matter (PM$_{10}$, PM$_{2.5}$, and their components), NH$_3$, and volatile organic carbons (VOCs, which contain various chemical components). The emissions data of SO$_2$, NO$_x$, PM$_{10}$, PM$_{2.5}$, BC, OC, NH$_3$, and VOCs (major components) from different anthropogenic sources were taken from the MEIC emission inventory for 2013 (http://www.meicmodel.org/). The emission inventory of VOCs from biological sources was calculated using the MEGAN natural source emission inventory model. The emission inventory mentioned above has been adopted in related research [24].
2.4. Modeling System

The simulation period was from January 2016 to January 2017, and the time interval of result output was 1 h. The meteorological simulation was conducted by The Weather Research and Forecasting (WRF) Model via the Lambert projected coordinate system, in which the longitude of the central point was 103° E, the latitude of the central point was 37° N, and the two parallel latitudes were 25° N and 40° N, respectively. The horizontal simulation ranges were −3600 km to 3600 km in the X direction and −2520 km to 2520 km in the Y direction from the central point. The grid spacing was 20 km, and the investigation region was divided into a total of 360 × 252 grid cells. A total of 30 pressure layers were configured in the vertical direction, with the distance between adjacent layers increasing gradually from bottom to top. The data for the initial field and the boundary field of the WRF model were the final global analysis data with a resolution of 1° provided by the US National Center for Environmental Prediction (NCEP) every 6 h (http://rda.ucar.edu/datasets/ds083.2/). Furthermore, the initial field was initiated every day, the length of each simulation was 30 h, the time setup of spin-up was 6 h, and NCEP ADP observation data (http://rda.ucar.edu/datasets/ds461.0/) were utilized to perform objective analysis and 4-dimensional data assimilation. The NCEP ADP Global Surface Observational Weather Data were composed of surface weather reports operationally collected by NCEP.

For the air quality simulation, the meteorological field needed by the CMAQ model was provided by the meteorological model (WRF), and the CMAQ model used the same simulation period and the same space projection coordinate system as the WRF model. However, a smaller simulation range was used in the CMAQ model than the simulation range of the WRF model. The CMAQ model applied the Lambert projection coordinate system, the longitude of the central point was 103° E, the latitude of the central point was 37° N, and the two parallel latitudes were 25° N and 40° N, respectively. The horizontal simulation range was −2690 km to 2690 km in the X direction and −2150 km to 2150 km in the Y direction from the central point. The grid spacing was 20 km, which divided the whole country into 270 × 216 grid cells. A total number of 14 pressure layers were configured in the vertical direction, and the distance between adjacent layers increased gradually from bottom to top.

The parameters of the WRF model and CMAQ model are listed in Tables 3 and 4, respectively. The meteorological parameters, such as WS, WD, T, RH, and PRE, simulated by the parameterization scheme of the WRF model have been verified by previous research [19,25].

### Table 3. WRF parameterization scheme.

| Parameterization Scheme                  | Name of the Selected Solution |
|------------------------------------------|-------------------------------|
| Micro-physics scheme                     | WSM6                          |
| Longwave radiation scheme                | New Goddard scheme            |
| Shortwave radiation scheme               | RRTM                          |
| Near-ground scheme                       | Pleim Xiu                     |
| Land surface scheme                      | Pleim Xiu                     |
| Boundary layer scheme                    | ACM2                          |
| Cumulus convection scheme                | Kain–Fritsch                  |

### Table 4. CMAQ model parameterization scheme.

| Model Parameter                        | CMAQ                          |
|----------------------------------------|-------------------------------|
| Model version                          | 5.0.2                         |
| Grid nested mode                       | Single-layer grid             |
| Horizontal resolution                  | 20 km                         |
| Number of vertical layers              | 14                            |
| Gas phase chemical mechanism           | CB05                          |
| Aerosol chemical mechanism             | AERO5                         |
| Photochemical reaction rate            | In-line                       |
| Sand and dust                          | Off                           |
| Boundary condition                     | Default                       |
| Initial condition                      | Restart each day              |
2.5. Model Performance

To evaluate the performance of the WRF-CMAQ modeling system, we compared the simulated PM$_{2.5}$ concentrations with the observation data collected in 338 cities during January 2016 and January 2017. Monitoring data collected in 20 cities in Xinjiang and Tibet were not considered because sandy and dusty weather is relatively frequent in Xinjiang, and the simulation effects of the existing CMAQ model on the sand and dust process are relatively poor. Moreover, the accuracy of the Tibetan pollution source emission inventory is relatively poor, and the value of analysis of the related simulation result is relatively low. Figure 2 presents the correlation analysis results of the average observation data and the average simulation data from CMAQ in the remaining 318 cities. The results indicate that the level of agreement between the modeled and observed data is satisfactory. The statistical values calculated were $n$, correlation coefficient ($R$), normalized mean bias (NMB), and the normalized mean error (NME), as shown in Table 5.

![Figure 2](image-url)

**Table 5.** Performance statistical parameters for PM$_{2.5}$ concentration simulations in China.

| Statistic | January 2016 | January 2017 |
|-----------|--------------|--------------|
| $n$       | 318          | 318          |
| $R$       | 0.76         | 0.75         |
| NMB (%)   | $-11.75$     | $-13.92$     |
| NME (%)   | 31.59        | 30.56        |
2.6. Quantitative Analysis of the Contribution of Meteorology to the PM$_{2.5}$ Concentration

The major factors affecting PM$_{2.5}$ concentration are emissions of air pollutants and meteorological conditions. A comparison between the simulations in both January 2016 and January 2017 with the same emission data was performed, and the difference in the simulated pollutants concentrations can be attributed to the difference impacted by the meteorological conditions. The scenario analysis method was used to quantitatively analyze the degree of impact of the meteorology on the PM$_{2.5}$ concentration. The emission inventory of the pollutants was assumed to be unchanged between January 2016 and January 2017, and two simulation scenarios, S1 and S2, were used, in which S1 adopted the meteorological conditions in January 2016, and S2 adopted the meteorological conditions in January 2017. On the basis of using the WRF meteorological model to simulate the meteorological field in 2016 and 2017, the air quality model was utilized to simulate the PM$_{2.5}$ concentrations in the air under the different scenarios. The results of the two simulations were compared to obtain the quantitative impacts of the variation of meteorological conditions on the air quality in China during January 2017:

$$\eta_m = \frac{C^{S2} - C^{S1}}{C^{S1}}$$

where $\eta_m$ is the degree of the impact of the meteorological conditions on the PM$_{2.5}$ pollution in January 2017 (%), where a positive value indicates that the meteorological conditions become poorer and the negative value indicates that the meteorological conditions improved; $C^{S1}$ is the average simulation concentration under the meteorological conditions in January 2016 (µg m$^{-3}$); and $C^{S2}$ is the average simulation concentration under the meteorological conditions in January 2017 (µg m$^{-3}$).

$$C_m = C^{2016} \times \eta_m$$

where $C_m$ is the variation amount of PM$_{2.5}$ caused by meteorology, and $C^{2016}$ is the observation concentration in January 2016 (µg m$^{-3}$).

3. Results and Discussion

3.1. Distribution Characteristics of the PM$_{2.5}$ Concentration

The PM$_{2.5}$ includes primary particles that are discharged directly and secondary particles indirectly generated by conversion from precursors such as SO$_2$, NO$_x$, VOCs, and NH$_3$ [26]. More attention is now being paid to pollution control in China, and thus the installation rate of the pollution treatment facilities and the removal efficiency of the pollutants has substantially increased. The variations in PM$_{2.5}$ monitoring concentration are shown in Figure 3. As shown in Figure 3, the PM$_{2.5}$ concentration in January 2016 decreased by 15% with respect to that in 2014. However, the PM$_{2.5}$ pollution in January 2017 was severe and the average concentration of PM$_{2.5}$ nationwide was 78 µg m$^{-3}$, which represents an increase of 14.7% over the level in January 2016. In January 2017, the PM$_{2.5}$ pollution displayed substantial spatial variation. The “Hu Line”, which divides China into the east and west parts based on differences in China’s population, geography, climate, and economy, could also be used to describe the geographic division of PM$_{2.5}$ concentrations over China. The regions with heavy PM$_{2.5}$ pollution are primarily located in the east part of the Hu Line, especially in the southeastern areas of Sichuan province where population, industry, and agriculture are concentrated, as well as regions such as Hebei, Henan, Hubei, Hunan, and Shandong. Among the above mentioned four major regions, JJJ suffered from the heaviest PM$_{2.5}$ pollution. The main reason is that January is winter in China, and low temperatures lead to increased domestic heating and related coal consumption, particularly in northern China. As a result, the PM$_{2.5}$ concentration in winter was higher than that in other seasons. In January 2017, the mean PM$_{2.5}$ concentration in the JJJ region was as high as 128 µg m$^{-3}$, which exceeded the national average PM$_{2.5}$ concentration by 64%. Compared with the same period of time in 2016, the PM$_{2.5}$ concentration in the middle to the eastern region of
China in January 2017 increased considerably. Although the mean PM$_{2.5}$ concentration in the YRD region decreased by 16.7%, the other three major regions (JJJ, PRD, and CYB) had increases in mean PM$_{2.5}$ concentration of 43.8%, 62.5%, and 27.5%, respectively (Figure 4).

3.2. The Impact of Meteorology on the PM$_{2.5}$ Pollution

3.2.1. Relationships between Meteorological Parameters and the PM$_{2.5}$ Concentration

The meteorological conditions have various effects on the diffusion and dilution capabilities of particles, the gas-particle conversion process, and secondary formation of particles. The relationships between different meteorological parameters and the PM$_{2.5}$ concentration during January in the four major regions are presented in Figure 5. Precipitation, wind speed, and relative humidity had major influences on the removal or accumulation of air pollutants. Low wind speeds decreased the speed of pollutant diffusion. When the wind speed was less than 2 m s$^{-1}$, the probability of PM$_{2.5}$ pollution...
increased. High humidity and stagnant meteorological conditions accelerated the accumulation process of PM$_{2.5}$ pollution. Differences in environmental factors such as the latitude, topography, land-sea distribution, hydrological characteristics, biological communities, and soil type, influence the response of PM$_{2.5}$ to meteorological parameters in different regions. The JJJ region is located in northern China, where January precipitation is relatively low, wind speed is low (wind speed $<2$ m s$^{-1}$), and humidity is high (the relative humidity $>60$%). These factors have a major influence on the PM$_{2.5}$ concentration. In particular, the growth of the secondary particles was accelerated and the PM$_{2.5}$ concentration was high, with a daily average concentration of PM$_{2.5}$ of $>150$ µg m$^{-3}$. The YRD and CYB regions displayed similar characteristics to the JJJ region in terms of the correlation between meteorological parameters and the PM$_{2.5}$ concentration. The PRD consists of groups of coastal cities, and since the PM$_{2.5}$ is greatly affected by the precipitation and the sea–land breeze, when the daily accumulation precipitation was $>10$ mm, the PM$_{2.5}$ pollution was greatly reduced and the daily mean concentration of PM$_{2.5}$ was $<50$ µg m$^{-3}$. When the wind speed was $>2.5$ m s$^{-1}$, the daily average concentration of PM$_{2.5}$ was usually $<50$ µg m$^{-3}$.

Temperature is an important meteorological factor correlated with air quality. Some studies have shown that temperature was positively correlated with PM$_{2.5}$, but these factors were found to be negatively correlated in other studies [27–29]. The negative relationship between surface-level

![Figure 5. Comparison of daily meteorological parameters and PM$_{2.5}$ concentration in (a) JJJ, (b) YRD, (c) PRD, (d) CYB.](image)
air temperature and PM$_{2.5}$ could be consistent with the importance of inversions. As a result, a complex relationship, further research on the dependencies of mean daily PM$_{2.5}$ concentrations to air temperature would be carried out in future works.

3.2.2. Simulation of the Impact of the Meteorology on the PM$_{2.5}$ Pollution

Nationwide Overall Condition

In January 2017, the general atmospheric circulation in the mid-high latitudes of China was relatively stable and was mainly in a form of zonal circulation. Thus, cold weather in the northern region was inactive. After the cold weather period ended, the temperature increased relatively rapidly to levels that were higher than normal. This progression tends to form unfavorable meteorological conditions such as wide-ranging stagnation, high humidity, and temperature inversion. The weather was relatively mild and stagnant weather occurred in the south-central region of Northern China, the central-western region of Huanghai (Huang and Huai rivers) and the Guanzhong region of Shaanxi province. The simulation results of the air quality model indicated that, compared with January 2016, the variation in the meteorological factors caused the average PM$_{2.5}$ concentration to increase by 9.2 µg m$^{-3}$ in China during January 2017, which was an increase of 13.6%. The unfavorable meteorological conditions counteracted the effects of emission reduction by pollution control, and were the key factors leading to the increase in the mean PM$_{2.5}$ concentration. Except in the YRD region and southern Shandong peninsula, the unfavorable weather conditions decreased the air quality. The mean PM$_{2.5}$ concentration of January 2017 increased by 8 µg m$^{-3}$ or more in most areas east of the Hu line, especially in regions where the pollution was already severe. The JJJ region, the PRD region, the northeastern regions (Liaoning and Jilin province), the plain areas of Hubei and Hunan, Henan, Shanxi, Sichuan, Xinjiang, and Inner Mongolia had worsening meteorological conditions. The impact of the difference in meteorological conditions on the PM$_{2.5}$ pollution between January 2017 and January 2016 is illustrated in Table 6 and the spatial distribution characteristics are shown in Figure 6.

Table 6. Impacts of the meteorological conditions on the PM$_{2.5}$ pollution in each province.

| Province         | Meteorological Impact Degree ($\eta_{\text{mr}}, \%$) | Variation Amount of PM$_{2.5}$ Caused by Meteorology ($C_{\text{mr}}, \mu g m^{-3}$) |
|------------------|-----------------------------------------------|---------------------------------|
| Beijing          | 73.08                                         | 49.70                           |
| Tianjin          | 23.74                                         | 17.50                           |
| Hebei            | 27.46                                         | 25.41                           |
| Shanxi           | 25.58                                         | 21.00                           |
| Inner Mongolia   | 35.78                                         | 14.13                           |
| Liaoning         | 18.63                                         | 11.45                           |
| Jilin            | 6.87                                          | 4.51                            |
| Heilongjiang     | 29.12                                         | 12.91                           |
| Shanghai         | −15.41                                        | −10.78                          |
| Jiangsu          | −10.77                                        | −8.91                           |
| Zhejiang         | −2.29                                         | −1.38                           |
| Anhui            | −0.73                                         | −0.59                           |
| Fujian           | 9.16                                          | 3.01                            |
| Jiangxi          | 22.39                                         | 12.25                           |
| Shandong         | 1.43                                          | 1.60                            |
| Henan            | 17.41                                         | 23.95                           |
| Hubei            | 21.28                                         | 20.72                           |
| Hunan            | 31.34                                         | 22.30                           |
| Guangdong        | 44.29                                         | 14.38                           |
| Guangxi          | 54.11                                         | 21.37                           |
| Hainan           | 20.29                                         | 3.72                            |
| Chongqing        | 11.55                                         | 8.17                            |
| Sichuan          | 7.64                                          | 5.26                            |
The impacts of meteorological conditions on PM$_{2.5}$ pollution in the major regions are presented in Table 7. Comparison of the mean PM$_{2.5}$ concentration in January 2017 with that in January 2016 showed that the PM$_{2.5}$ pollution in the PRD region was slightly reduced, and the mean PM$_{2.5}$ concentration decreased by 16.7% between the two months. However, in the JJJ, PRD, and CYB regions, the PM$_{2.5}$ pollution became more severe, and the mean PM$_{2.5}$ concentration increased by 43.8%, 62.5%, and 27.5%, respectively, compared with January 2016. The simulation results indicate that the meteorological conditions in the JJJ, PRD, and CYB regions all worsened in January 2017 compared with January 2016, inhibiting the diffusion of the PM$_{2.5}$ and processes such as dry and wet deposition. The meteorological conditions were the major cause of the increased PM$_{2.5}$ pollution in these three regions. The frequency of low wind events (wind speed <2 m s$^{-1}$) and the frequency of high humidity events (relative humidity >60%) in the JJJ, PRD, and CYB regions showed an increasing trend and the frequency of large wind events (wind speed >4 m s$^{-1}$) showed a decreasing trend. In particular, the variation in the JJJ region was relatively large. The frequency of small winds and the frequency of high humidity increased by 16.8% and 68.4%, respectively, and the frequency of high wind speeds decreased by 38.9%. The meteorological factors caused the PM$_{2.5}$ concentration in the JJJ, PRD, and CYB regions to increase by 26.4, 13.6, and 5.4 µg m$^{-3}$, respectively, and the corresponding increasing amplitudes were 29.7%, 42.6%, and 7.9%, respectively. On the other hand, it is clear that the meteorological

| Province | Meteorological Impact Degree ($\eta_{\text{m}, \%}$) | Variation Amount of PM$_{2.5}$ Caused by Meteorology ($C_{\text{m}}, \mu g \text{ m}^{-3}$) |
|----------|--------------------------------------------------|--------------------------------------------------|
| Guizhou  | 15.59                                            | 5.76                                             |
| Yunnan   | −0.75                                            | −0.23                                            |
| Tibet    | −17.51                                           | −6.38                                            |
| Shaanxi  | −0.13                                            | −0.13                                            |
| Gansu    | −9.56                                            | −4.78                                            |
| Qinghai  | 2.15                                             | 0.90                                             |
| Ningxia  | −1.54                                            | −0.95                                            |
| Xinjiang | 10.69                                            | 10.66                                            |
| Nationwide | 13.55                                              | 9.17                                             |

Figure 6. Changes in the average PM$_{2.5}$ concentration (January 2017 values–January 2016 values) caused by meteorological factors.
conditions contributed to the improving air quality (reduced PM$_{2.5}$) in the YRD region. As shown in Table 7, the meteorological factors caused the PM$_{2.5}$ concentration to decrease by 6.1 µg m$^{-3}$, which is an improvement of approximately 8.5% from January 2016 to January 2017. In particular, the northern Jiangsu region showed the greatest pollution reduction. 

Table 7. Impacts of the meteorological conditions on the PM$_{2.5}$ pollution in major regions.

| Region | Meteorological Impact Degree ($\eta_m$, %) | Variation Amount of PM$_{2.5}$ Caused by Meteorology ($C_m$, µg m$^{-3}$) |
|--------|----------------------------------------|-------------------------------------------------|
| JJJ    | 29.70                                  | 26.43                                           |
| YRD    | −8.51                                  | −6.13                                           |
| PRD    | 42.60                                  | 13.63                                           |
| CYB    | 7.89                                   | 5.44                                            |

The pattern of development, which has focused on heavy industry, has caused large emissions of air pollutants in the JJJ region and its surrounding areas. The Statistical Yearbook of China, published by the State Statistical Bureau, indicates that the consumption of coal per unit area in this region was 4.8 times the national average. The power generation per unit area, and the production volumes of high energy consumption industrial products including crude steel, concrete, and sheet glass were 4.1, 5.9, 2.7, and 4.9 times the national average, respectively. The JJJ region has the most severe PM$_{2.5}$ pollution in China, and the prevention and control of the PM$_{2.5}$ pollution there is a major concern and focus of research. Among the 13 cities in the JJJ region, the PM$_{2.5}$ pollution in January 2017 showed a worsening trend, and in particular, six cities (Beijing, Shijiazhuang, Tangshan, Qinhuangdao, Handan, Zhangjiakou) all showed an increasing amplitude in the mean PM$_{2.5}$ concentration exceeding 50%, with a worsening of most meteorological conditions (Table 8). During 1–7, 14–18, and 24–28 January 2017, the surface pressure field was mainly controlled by the homogeneous distribution of low pressure in the JJJ region. The ground wind speed was generally slower than 1.5 m s$^{-1}$, and the horizontal diffusion capability of the pollutants was weak. The 500 hPa height-field was controlled by a northwest flow, the cloud cover at high altitudes was relatively sparse, and ground radiation and temperature reduction were high during night. This formed stable inversion stratification near the ground. The decrease of the planetary boundary layer height (PBLH) weakened diffusion capacity and made the air quality deteriorate. Previous research has revealed that the invasion of cold air increases the turbulence flux and the PBLH over the Beijing area [30]. Compared to 2016, the weaker cold air in January 2017 resulted in a decrease of PBLH in the range of 50 to 200 m over the JJJ area (Figure 7), which was one of the main reasons for heavy haze pollution in January 2017. The perpendicular diffusion capability of the air was correspondingly poor. Furthermore, more than 70% of the days had high humidity. Water vapor and pollutants accumulated near the ground and this caused the concentration of secondary inorganic aerosols (SIA; i.e., sulfate, nitrate, and ammonium) increased rapidly. Thus, the daily mean concentration of PM$_{2.5}$ in most cities was higher than 150 µg m$^{-3}$ and the heavy pollution events occurred. Figure 8 shows SIA pollution levels at the Beijing, Tangshan, and Shijiazhuang sampling sites. It is known that SIA makes up about 30–40% of the total PM$_{2.5}$ mass annually [5]. As shown in Figure 8, the total SIA concentrations in the three cities were much higher than those in January 2016, accounting for 53%, 41%, and 54% of PM$_{2.5}$ concentrations at Beijing, Tangshan and Shijiazhuang, respectively. There was an increase in the heavy pollution period compared with the normal levels, accounting for 57%, 44%, and 59% of PM$_{2.5}$ concentrations at Beijing, Tangshan, and Shijiazhuang, respectively. For these reasons, meteorological conditions contributed to the worsening air quality in the 13 cities. The degree of the impact of variations in meteorological conditions on PM$_{2.5}$ pollution ranged between 7.3% and 73.1%, and was relatively large in some central and northern cities, such as Beijing, Qinhuangdao, Zhangjiakou, Chengde and Langfang, where poor conditions increased PM$_{2.5}$ concentrations by >50%.
Table 8. Impacts of meteorological conditions on the PM$_{2.5}$ pollution in 13 cities in JJJ.

| City Name    | Meteorological Impact Degree ($\eta_{m}$, %) | Variation Amount of PM$_{2.5}$ Caused by Meteorology ($C_{m}$, $\mu g m^{-3}$) |
|--------------|---------------------------------------------|-------------------------------------------------------------------------------|
| Beijing      | 73.08                                       | 49.70                                                                         |
| Tianjin      | 23.74                                       | 17.50                                                                         |
| Shijiazhuang | 24.87                                       | 32.75                                                                         |
| Tangshan     | 30.76                                       | 24.16                                                                         |
| Qinhuangdao  | 57.69                                       | 23.33                                                                         |
| Handan       | 15.25                                       | 16.65                                                                         |
| Xingtai      | 19.22                                       | 24.79                                                                         |
| Baoding      | 23.82                                       | 35.59                                                                         |
| Zhangjiakou  | 60.98                                       | 16.60                                                                         |
| Chengde      | 68.93                                       | 31.11                                                                         |
| Cangzhou     | 7.25                                        | 6.17                                                                          |
| Langfang     | 59.94                                       | 52.98                                                                         |
| Hengshui     | 19.62                                       | 26.23                                                                         |

Figure 7. The monthly mean PBLH in JJJ region in (a) January 2016 and (b) January 2017.
Unlike the other three regions, meteorological conditions in the YRD region were favorable. The numerical results showed that meteorological conditions contributed to the improved air quality in most cities in the YRD region. The impact of variations in meteorological conditions on PM$_{2.5}$ pollution was more beneficial in Jiangsu, where good conditions decreased PM$_{2.5}$ concentrations by >10%. The wind rose in January of 2016 and 2017 in major cities (Shanghai, Nanjing, and Hangzhou) is shown in Figure 9. The wind frequency of WNW, NW and NNW reduced by 2–4%, which was more favorable in January of 2017 for the improvements of local air quality than 2016 due to the less transport from upstream areas. Meanwhile, the wind frequency of E and ENE increased by 4–9%, and
more ocean currents had contributed to reduce PM$_{2.5}$ concentration. The wind frequency of NE, ENE and E accounted for over 40% during 15–31 January 2017, which was one of important factors resulting in the lower mean concentration of PM$_{2.5}$.

**Figure 9.** Wind rose in January of 2016 and 2017 in (a) Shanghai, (b) Nanjing, (c) Hangzhou.
4. Conclusions

PM$_{2.5}$ pollution showed an unusually large increase in China during January 2017. Compared with January 2016, the national mean PM$_{2.5}$ concentration increased by 14.7%. Despite the 16.7% decrease in the mean PM$_{2.5}$ concentration in the YRD region during January 2017, the mean PM$_{2.5}$ concentration during January 2017 in the JJJ, CYB, and PRD regions increased by 43.8%, 62.5%, and 27.5%, respectively. In particular, the PM$_{2.5}$ pollution in the JJJ region was the most severe in China, with the mean concentration during January 2017 reaching up to 128 $\mu$g m$^{-3}$, and exceeding the national mean level by 64%. Such severe pollution is a serious threat to human health.

In January 2017, precipitation in the JJJ, YRD, and CYB regions was relatively low, and unfavorable meteorological conditions such as relatively low wind speed and high humidity caused the PM$_{2.5}$ pollution to increase. The variation in the meteorological conditions caused the mean PM$_{2.5}$ concentration in China to increase by 9.2 $\mu$g m$^{-3}$ in January 2017 (relative to January 2016), representing an increase as high as 13.6%. Although the meteorological conditions improved in the YRD region, the meteorological conditions caused the PM$_{2.5}$ concentration in the JJJ, PRD, and CYB regions to increase by 26.4, 13.6, and 5.4 $\mu$g m$^{-3}$, respectively, and the amplitude of this increase was 29.7%, 42.6%, and 7.9%, respectively.

The uncertainty was primarily obtained based on the emission inventory and the air quality model. The MEIC emission inventory was established using the “top to bottom” method and the activity level and the emission factor both showed relatively high uncertainty. The emission of dust is especially difficult to measure directly, and accurate geological information cannot be easily obtained. This leads to a relatively large error in its emission and characteristics of spatial and temporal distribution. The CMAQ model shows a relatively poor effect in simulation of the dust processes and thus a relatively large error exists in the PM$_{2.5}$ concentrations in dusty weather. Finally, while sulfate, nitrate, and ammonium displayed explosive growth during the heavy pollution process, the related chemical reaction mechanism(s) are still under investigation. As a result, the CMAQ simulation results may underestimate the PM$_{2.5}$ concentration during the heavy pollution in January, especially for the sulfate, nitrate, and ammonium in PM$_{2.5}$.

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References

1. Lin, G.; Fu, J.; Jiang, D.; Hu, W.; Dong, D.; Huang, Y.; Zhao, M. Spatio-temporal variation of PM$_{2.5}$ concentrations and their relationship with geographic and socioeconomic factors in China. *Int. J. Environ. Res. Public Health* 2013, 11, 173–186. [CrossRef] [PubMed]

2. Liu, M.; Bi, J.; Ma, Z. Visibility-based PM$_{2.5}$ concentrations in China: 1957–1964 and 1973–2014. *Environ. Sci. Technol.* 2017, 51, 13161–13169. [CrossRef] [PubMed]

3. Zhang, Q.; Quan, J.; Tie, X.; Li, X.; Liu, Q.; Gao, Y.; Zhao, D. Effects of meteorology and secondary particle formation on visibility during heavy haze events in Beijing, China. *Sci. Total Environ.* 2015, 502, 578–584. [CrossRef] [PubMed]

4. Liu, X.; Li, C.; Tu, H.; Wu, Y.; Ying, C.; Huang, Q.; Wu, S.; Xie, Q.; Yuan, Z.; Lu, Y. Analysis of the effect of meteorological factors on PM$_{2.5}$-associated PAHs during autumn-winter in urban Nanchang. *Aerosol Air Qual. Res.* 2016, 16, 3222–3229. [CrossRef]
5. Cao, J.; Xu, H.; Xu, Q.; Chen, B.; Kan, H. Fine particulate matter constituents and cardiopulmonary mortality in a heavily polluted Chinese city. *Environ. Health. Perspect.* 2012, 120, 373–378. [CrossRef] [PubMed]

6. Aishire, J.A.; Clarke, P. Fine particulate matter air pollution and cognitive function among U.S. older adults. *J. Gerontol. B Psychol. Sci. Soc. Sci.* 2014, 70, 322–328. [CrossRef] [PubMed]

7. Liao, Z.H.; Gao, M.; Sun, J.R.; Fan, S.J. The impact of synoptic circulation on air quality and pollution-related human health in the Yangtze River delta region. *Sci. Total Environ.* 2017, 607, 838–846. [CrossRef] [PubMed]

8. Jedruszkiewicz, J.; Czernecki, B.; Marosz, M. The variability of PM$_{10}$ and PM$_{2.5}$ concentrations in selected polish agglomerations: The role of meteorological conditions, 2006–2016. *Int. J. Environ. Health Res.* 2017, 27, 441–462. [CrossRef] [PubMed]

9. Cheng, Y.-H.; Li, Y.-S. Influences of traffic emissions and meteorological conditions on ambient PM$_{10}$ and PM$_{2.5}$ levels at a highway toll station. *Aerosol Air Qual. Res.* 2010, 10, 456–462. [CrossRef]

10. Wang, P.; Cao, J.; Ti, X.; Wang, G.; Li, G.; Hu, T.; Wu, Y.; Xu, Y.; Xu, G.; Zhao, Y.; et al. Impact of meteorological parameters and gaseous pollutants on PM$_{2.5}$ and PM$_{10}$ mass concentrations during 2010 in Xi’an, China. *Aerosol Air Qual. Res.* 2015, 15, 1844–1854. [CrossRef]

11. Zhang, J.P.; Zhu, T.; Zhang, Q.H.; Li, C.C.; Shu, H.L.; Ying, Y.; Dai, Z.P.; Wang, X.; Liu, X.Y.; Liang, A.M.; et al. The impact of circulation patterns on regional transport pathways and air quality over Beijing and its surroundings. *Atmos. Chem. Phys.* 2012, 12, 5031–5053. [CrossRef]

12. Wang, H.; Xu, J.; Zhang, M.; Yang, Y.; Shen, X.; Wang, Y.; Chen, D.; Guo, J. A study of the meteorological causes of a prolonged and severe haze episode in January 2013 over central-eastern China. *Atmos. Environ.* 2014, 98, 146–157. [CrossRef]

13. Chuang, M.-T.; Chou, C.C.K.; Lin, N.-H.; Takami, A.; Hsiao, T.-C.; Lin, T.-H.; Fu, J.S.; Pani, S.K.; Lu, Y.-R.; Yang, T.-Y. A simulation study on PM$_{2.5}$ sources and meteorological characteristics at the northern tip of Taiwan in the early stage of the Asian haze period. *Aerosol Air Qual. Res.* 2017, 17, 3166–3178. [CrossRef]

14. Kozáková, J.; Pokorná, P.; Černíková, A.; Hovorka, J.; Braniš, M.; Moravec, P.; Schwarz, J. The association between intermodal (PM$_{1-2.5}$) and PM$_{1}$, PM$_{2.5}$, coarse fraction and meteorological parameters in various environments in Central Europe. *Aerosol Air Qual. Res.* 2017, 17, 1234–1243. [CrossRef]

15. Liang, P.; Zhu, T.; Fang, Y.; Li, Y.; Han, Y.; Wu, Y.; Hu, M.; Wang, J. The role of meteorological conditions and pollution control strategies in reducing air pollution in Beijing during APEC 2014 and Victory parade 2015. *Atmos. Chem. Phys.* 2017, 17, 13921–13940. [CrossRef] [PubMed]

16. Chen, Z.; Cai, J.; Gao, B.; Xu, B.; Dai, S.; He, B.; Xie, X. Detecting the causality influence of individual meteorological factors on local PM$_{2.5}$ concentration in the Jing-Jin-Ji region. *Sci. Rep.* 2017, 7, 40735. [CrossRef] [PubMed]

17. Liu, T.; Gong, S.; He, J.; Yu, M.; Wang, Q.; Li, H.; Liu, W.; Zhang, J.; Li, L.; Wang, X.; et al. Contributions of meteorological and emission factors to the 2015 winter severe haze pollution episodes in China’s Jing-Jin-Ji area. *Atmos. Chem. Phys.* 2017, 17, 2971–2980. [CrossRef]

18. Hu, J.; Chen, J.; Ying, Q.; Zhang, H. One-year simulation of ozone and particulate matter in China using WRF/CMAQ modeling system. *Atmos. Chem. Phys.* 2016, 16, 10333–10350. [CrossRef]

19. Zheng, B.; Zhang, Q.; Zhang, Y.; He, K.B.; Wang, K.; Zheng, G.J.; Duan, F.K.; Ma, Y.L.; Kimoto, T. Heterogeneous chemistry: A mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China. *Atmos. Chem. Phys.* 2015, 15, 2031–2049. [CrossRef]

20. Zheng, Y.; Xue, T.; Zhang, Q.; Geng, G.; Tong, D.; Li, X.; He, K. Air quality improvements and health benefits from China’s clean air action since 2013. *Environ. Res. Lett.* 2017, 12, 114020. [CrossRef]

21. Geng, G.; Zhang, Q.; Martin, R.V.; Lin, J.; Huo, H.; Zheng, B.; Wang, S.; He, K. Impact of spatial proxies on the representation of bottom-up emission inventories: A satellite-based analysis. *Atmos. Chem. Phys.* 2017, 17, 4131–4145. [CrossRef]

22. Zhao, Y.; Nielsen, C.P.; Lei, Y.; McElroy, M.B.; Hao, J. Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China. *Atmos. Chem. Phys.* 2011, 11, 2295–2308. [CrossRef]

23. Qi, J.; Zheng, B.; Li, M.; Yu, F.; Chen, C.; Liu, F.; Zhou, X.; Yuan, J.; Zhang, Q.; He, K. A high-resolution air pollutants emission inventory in 2013 for the Beijing-Tianjin-Hebei region, China. *Atmos. Environ.* 2017, 170, 156–168. [CrossRef]
24. Hu, J.; Wang, P.; Ying, Q.; Zhang, H.; Chen, J.; Ge, X.; Li, X.; Jiang, J.; Wang, S.; Zhang, J.; et al. Modeling biogenic and anthropogenic secondary organic aerosol in China. Atmos. Chem. Phys. 2017, 17, 77–92. [CrossRef]

25. Xue, W.B.; Wang, J.N.; Niu, H.; Yang, J.T.; Han, B.P.; Lei, Y.; Chen, H.L.; Jiang, C.L. Assessment of air quality improvement effect under the national total emission control program during the twelfth national five-year plan in China. Atmos. Environ. 2013, 68, 74–81. [CrossRef]

26. Dominici, F.; Wang, Y.; Correia, A.W.; Ezzati, M.; Pope, C.A.I.; Dockery, D.W. Chemical composition of fine particulate matter and life expectancy: In 95 US counties between 2002 and 2007. Epidemiology 2015, 26, 556–564. [CrossRef] [PubMed]

27. Zhang, C.; Ni, Z.; Ni, L. Multifractal detrended cross-correlation analysis between PM$_{2.5}$ and meteorological factors. Phys. A Stat. Mech. Appl. 2015, 438, 114–123. [CrossRef]

28. Fu, X.; Wang, S.; Chang, X.; Cai, S.; Xing, J.; Hao, J. Modeling analysis of secondary inorganic aerosols over China: Pollution characteristics, and meteorological and dust impacts. Sci. Rep. 2016, 6, 35992. [CrossRef] [PubMed]

29. He, J.; Gong, S.; Yu, Y.; Yu, L.; Wu, L.; Mao, H.; Song, C.; Zhao, S.; Liu, H.; Li, X.; et al. Air pollution characteristics and their relation to meteorological conditions during 2014–2015 in major Chinese cities. Environ. Pollut. 2017, 223, 484–496. [CrossRef] [PubMed]

30. Liu, X.; Hu, F.; Zou, H.; Cao, X.; Dou, J. Analysis on characteristic of atmospheric boundary layer during a typical heavy fog process in Beijing area. Plateau Meteorol. 2010, 29, 1174–1182.

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