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Effects of Meteorological Factors and Anthropogenic Precursors on $PM_{2.5}$ Concentrations in Cities in China

Ziyun Jing [1,2], Pengfei Liu [1,2,*,†], Tuanhui Wang [2,3], Hongquan Song [2,3,4,*], Jay Lee [5,6], Tao Xu[7,8] and Yu Xing [9]

1 Key Research Institute of Yellow River Civilization and Sustainable Development & Collaborative Innovation Center on Yellow River Civilization of Henan Province, Henan University, Kaifeng, Henan 475004, China; jzy@henu.edu.cn
2 Institute of Urban Big Data, College of Environment and Planning, Henan University, Kaifeng, Henan 475004, China; thwang@henu.edu.cn
3 Laboratory of Geospatial Technology for the Middle and Lower Yellow River Regions, Ministry of Education, Henan University, Kaifeng, Henan 475004, China
4 Henan Key Laboratory of Integrated Air Pollution Control and Ecological Security, Henan University, Kaifeng, Henan 475004, China
5 College of Environment and Planning, Henan University, Kaifeng, Henan 475004, China
6 Department of Geography, Kent State University, Kent, OH 44242, USA; jlee@kent.edu
7 School of Computer and Information Engineering, Henan University, Kaifeng, Henan 475004, China; txu@henu.edu.cn
8 Henan Key Laboratory of Big Data Analysis and Processing, Henan University, Kaifeng, Henan 475004, China
9 Henan Ecological and Environmental Monitoring Center, Zhengzhou, Henan 450000, China; nikkoyu@126.com
* Correspondence: lpf@henu.edu.cn (P.L.); hqsong@henu.edu.cn (H.S.)
† These authors contributed equally to this work and should be considered co-first authors.

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Abstract: Fine particulate matter smaller than 2.5 μm ($PM_{2.5}$) in size can significantly affect human health, atmospheric visibility, climate, and ecosystems. $PM_{2.5}$ has become the major air pollutant in most cities of China. However, influencing factors and their interactive effects on $PM_{2.5}$ concentrations remain unclear. This study used a geographic detector method to quantify the effects of anthropogenic precursors (AP) and meteorological factors on $PM_{2.5}$ concentrations in cities of China. Results showed that impacts of meteorological conditions and AP on $PM_{2.5}$ have significant spatio-temporal disparities. Temperature was the main influencing factor throughout the whole year, which can explain 27% of $PM_{2.5}$ concentrations. Precipitation and temperature were primary impacting factors in southern and northern China, respectively, at the annual time scale. In winter, AP had stronger impacts on $PM_{2.5}$ in northern China than in other seasons. Ammonia had stronger impacts on $PM_{2.5}$ than other anthropogenic precursors in winter. The interaction between all factors enhanced the formation of $PM_{2.5}$ concentrations. The interaction between ammonia and temperature had strongest impacts at the national scale, explaining 46% (q = 0.46) of $PM_{2.5}$ concentrations. The findings comprehensively elucidated the relative importance of driving factors in $PM_{2.5}$ formation, which can provide basic foundations for understanding the meteorological and anthropogenic influences on the concentration patterns of $PM_{2.5}$.

Keywords: $PM_{2.5}$; particulate matter; interactions; GeoDetector; air quality

1. Introduction

With rapid urbanization and economic development, China has experienced increased emissions of greenhouse gases and atmospheric pollutants [1]. Particulate matter (PM), especially fine PM with a diameter less than 2.5 μm ($PM_{2.5}$), has significant impacts on human health [2–6], atmospheric visibility [7,8], climate [9,10], and ecosystems [11,12]. In 190 Chinese cities, the annual mean $PM_{2.5}$ concentration (57 ± 18 μg m^{-3}) severely exceeds the limited value (35 μg m^{-3}) recommended by the Chinese ambient air quality standards (CAAQS) (GB3095-2012) [13], which is at least 1.6 times the limited value. $PM_{2.5}$ pollution is a severe threat to the economic and social sustainability of China.

$PM_{2.5}$ concentrations could be affected by numerous factors such as meteorological conditions and anthropogenic precursors (AP) [14–17]. Meteorological conditions play important roles in the process of air pollutant emission, transport, dilution, chemical conversion, and deposition [18–20]. AP directly or indirectly affect $PM_{2.5}$ concentrations in the atmosphere through a series of photochemical reactions under specific meteorological conditions [21–23]. Previous studies have explored the effects of AP and meteorological conditions (MCs) on the formation of $PM_{2.5}$ concentrations. They found that temperature affects $PM_{2.5}$ concentrations through atmospheric disturbances and photochemical reactions [24]. For example, as temperature and sulfur dioxide levels (SO_2) rise, this leads to increased sulfate concentrations [25,26]. In addition, temperature has indirect effects on secondary organic aerosols. Under high-temperature conditions, higher biomass volatile organic compound (VOC) emissions can increase secondary organic aerosol concentrations [27]. Wind speed can change the diffusion and transport of $PM_{2.5}$ in the horizontal direction [28,29]. Wet deposition caused by precipitation has scavenging effects on $PM_{2.5}$ in most areas [25,29]. Absolute humidity also affects $PM_{2.5}$ concentrations, and the increase in humidity is conducive to the conversion of semi-volatile substances into an aerosol phase, thereby increasing $PM_{2.5}$ concentrations [30,31]. Moreover, the height of the mixing layer also plays an important role in the dilution of primary and secondary aerosols [32,33].

Due to the limitation of data accessibility before 2013 in China, research on $PM_{2.5}$ was concentrated in Beijing-Tianjin-Hebei [17,34,35], the Yangtze River Delta [36–38], and the Pearl River Delta [39–42]. With the release of national air quality monitoring data, it is possible to study the temporal and spatial variations [43–45] and driving mechanisms of $PM_{2.5}$ concentrations [46–48] throughout China. In recent years, some studies have been conducted to identify spatial distributions and temporal trends in $PM_{2.5}$ concentrations at the national level, and to explore their relationships with meteorological conditions and precursor emissions. A series of models have been used to identify driving mechanisms influencing factors in $PM_{2.5}$ formation, such as the classical ordinary least square model, geographically weighted regression model [49], spatial regression model [50], and land-use regression and other models [51]. The mechanisms of $PM_{2.5}$ formation are very complicated, and there are interactions between influencing factors. The complex coupling of factors remains unclear. Under different driving conditions, $PM_{2.5}$ concentrations have greater heterogeneity at different spatial and temporal scales. Moreover, traditional methods can lead to biased results because the impacting factors can interact closely with each other. Therefore, advanced methods should be adopted to explore the relative importance of the effects of each influencing factor on $PM_{2.5}$.

To solve this issue, we applied a geographical detector q statistical method (GeoDetector) to assess the influence of AP and MCs on the concentrations of $PM_{2.5}$ in major cities of China. This study can provide a better understanding of the contribution of a single impacting factor on $PM_{2.5}$ concentration. The findings can also provide a foundation for the improvement of $PM_{2.5}$ prediction models' performance.

2. Materials and Methods

2.1. Study Area

Given differences in economic development, topography, and climate patterns, mainland China can be typically divided into 10 regions (Supplementary materials, Figure S1). These 10 regions are NE (northeastern region), NC (northern coastal region), EC (eastern coastal region), SC (southeastern coastal region), MUYR (Yangtze River's middle and upper reaches), MUPR (Pearl River's middle and upper reaches), UYR (Yellow River's upper reaches), MYR (Yellow River's middle reaches), XJ (Xinjiang region), and QTP (Qinghai Tibetan Plateau region) [29]. Among them, MUPR, SC, EC, and MUYR are considered to be southern China, while NC, NE, MYR, and UYR are considered to belong to northern China.

2.2. Datasets

According to CAAQS, annual average $PM_{2.5}$ concentrations are limited to 15 μg m^{-3} (Grade I) and 35 μg m^{-3} (Grade II). The daily average concentrations are 35 μg m^{-3} (Grade I) and 75 μg m^{-3} (Grade II) [39]. Grade I refers to the concentration limit required for scenic spots, nature reserves, and other areas requiring special conservation in China. Grade II refers to the concentration limits required for rural areas, residential areas, industrial areas, cultural areas, and mixed-use residential areas. The daily $PM_{2.5}$ concentrations of 366 cities in China were obtained from the China Environmental Monitoring Center. Due to the availability of data, data for Hong Kong, Macau, and Taiwan were not included. Figures S3 and S4 show maps of $PM_{2.5}$ concentrations [29].

We acquired meteorological data (839 sites) from the China Meteorological Data Network throughout the whole year of 2016. The daily meteorological data included surface air pressure (PS, hPa), air temperature (TE, ℃), relative humidity (RH, %), wind velocity (WI, m s^{-1}), sunshine duration (SS, h), and accumulated precipitation (PE, mm) (Figures S5–S11) [29]. The monthly anthropogenic emissions of VOCs ammonia (NH_3), sulfur dioxide (SO_2), and nitrogen oxide (NOx) were monitored in 2016. They were usually considered as AP of $PM_{2.5}$ and were collected from MEIC (multi-resolution emission inventory for China, http://www.meicmodel.org/). The MEIC includes emission data of the four sub-sectors of transportation, power, industry, and residential [52], and has been widely used in the research of air pollution [53–56]. Figure S12 indicates that the highest anthropogenic precursor emissions are mainly distributed in EC, MYR, SC, NC, and MUYR.

2.3. GeoDetector

In this study, the q statistics of GeoDetector were used to quantitatively analyze the impacts of AP and MCs on $PM_{2.5}$ in China. GeoDetector supports a series of statistical methods that can explore spatial difference and identify the driving factors. The main idea is based on the assumption that if an independent variable (X) causes a dependent variable (Y), then the spatial distribution of the independent variable and the dependent variable should be consistent [57–60]. GeoDetector can detect both qualitative data and numerical data. Compared with traditional linear statistical methods, this is a major advantage of GeoDetector. Another unique advantage of GeoDetector is the ability to detect the interaction between two factors acting on the dependent variable. The GeoDetector includes four detectors, which are factor detection, risk area detection, ecological detection, and interaction detection. In this study, factor detection and interaction detection were used.

Factor detector uses q statistic to detect the influence of X (e.g., MCs and AP) on Y (e.g., the $PM_{2.5}$ concentrations). The expression is:

$$q = 1 - \frac{\sum_{h=1}^{L} N_h {\sigma_h}^2}{N\sigma^2} = 1 - \frac{SSW}{SST},$$

$$SSW = \sum_{h=1}^{L} N_h {\sigma_h}^2,\ SST = \mathrm{N}\sigma^2$$

In the formula, $h = 1,\ldots,$ L, which classifies X or Y; N and N_h are the numbers of the whole region and categories in classification h; σ^2 and σ_h^2 are the Y value of the whole region and the variance of strata h, respectively. SST and SSW are the total variance of the whole region and the sum of variance within the strata, respectively. Greater values of q (0–1) indicate more spatial variation in Y. If the

classification is based on X, a higher q value explains the influence of X on Y (i.e., explaining power: $100 \times q\%$).

Interaction detection can identify the impact of the interaction between potential driving factors. Based on that, we can assess whether the interaction between X_1 and X_2 will strengthen or weaken the explaining power of Y. Additionally, the influences of these factors on the dependent variable Y would be independent of each other. There are five types of interactions; please refer to [59] for more information. In addition, in order to identify the positive or negative correlations between $PM_{2.5}$ concentrations and influencing factors, this study calculated their Pearson correlation coefficients at different temporal and spatial scales.

3. Results

3.1. Effects on $PM_{2.5}$ Concentrations at the National Scale

The influence of each driving factor on $PM_{2.5}$ concentrations was acquired by calculating the corresponding q value (the power of determinant, Figure 1a), which indicated the contribution of each impacting factor on $PM_{2.5}$ concentrations. Figure 1 shows that there were obvious seasonal and annual difference in factors' impacts on $PM_{2.5}$. Meteorological conditions were dominant impacting factors in $PM_{2.5}$ formation at the annual time scale. TE (q = 0.27) was the primary impacting factor, followed by PE (q = 0.22) and PS (q = 0.17).

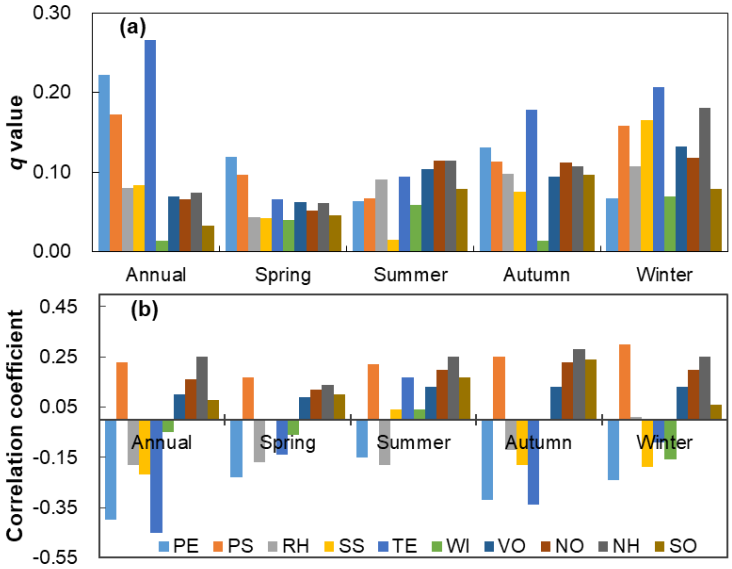

Figure 1. The annual and seasonal q values (**a**) and correlation coefficient (**b**) between $PM_{2.5}$ concentrations and influencing factors at the national scale. Note: VO denotes volatile organic compounds (VOCs); NO denotes NOx; NH denotes NH_3; SO denotes SO_2.

Meteorological factors were dominant driving forces in spring, such as PE (q = 0.12) and PS (q = 0.10). In summer, AP showed stronger impacts on $PM_{2.5}$ concentrations than meteorological conditions, and the NOx, NH_3, and VOCs were the three dominant factors (q > 0.10). In autumn, the meteorological factors and AP showed comparative influence on $PM_{2.5}$. The dominant impacting factor in autumn was TE (q = 0.18), followed by PE (q = 0.13), NOx (q = 0.11), and NH_3 (q = 0.10). Similar to the autumn, meteorological factors and AP had comparative impacts on $PM_{2.5}$ concentrations in winter. TE (q = 0.25) was the dominant factor, followed by NH_3 (q = 0.18), SSD (q = 0.17), PS (q = 0.16), and VOCs (q = 0.13). This indicated that AP were the dominant impacting factor in winter.

3.2. Effects on $PM_{2.5}$ Concentrations at the Regional Scale

Figure 2 shows the effects of AP and MCs on $PM_{2.5}$ significantly varied at regional and seasonal scales in China. In general, meteorological factors were the major driving forces in China. PE and TE were primary driving forces in southern and northern China, respectively.

Figure 2 shows that meteorological factors were primary drivers of $PM_{2.5}$ formation in spring, which is similar to most regions at the annual time scale. The dominant meteorological factor was PE and SS in southern and regions of MYR and NC, respectively. Meteorological factors and AP showed comparative impacts on $PM_{2.5}$ concentrations in summer except for in XJ and QTP. TE was the dominant driving factor on $PM_{2.5}$ concentrations in autumn in UYR, NE, and MYR, but PE and WI played the dominant role in regions of MUYR and SC, and of NC and EC, respectively. WI was the primary driving factor in NC, NE, EC, and UYR in winter, but PS was the major driving factor in MUYR and MUPR. However, in QTP, PS was the major impacting factor on $PM_{2.5}$ concentrations throughout the whole year.

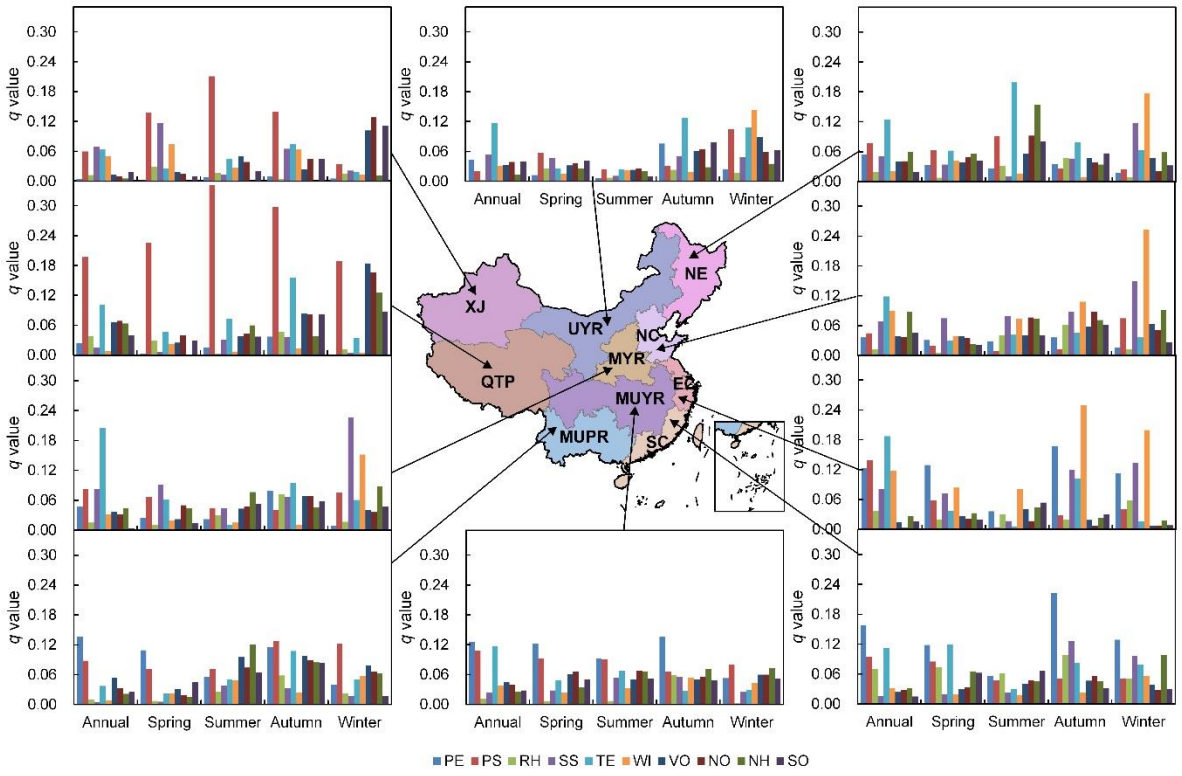

Figure 2. The q values of impacting factors at the annual and seasonal time scales in 10 regions of China. Note: VO denotes VOCs; NO denotes NOx; NH denotes NH_3; SO denotes SO_2.

3.3. Interactive Effects on $PM_{2.5}$

This study explored interactive effects on $PM_{2.5}$ by using the interaction detector with a total of 45 pairs of interactions. The interaction of any two factors was analyzed by comparing their combined contribution with their individual contributions to $PM_{2.5}$ concentrations. Figure 3 shows the q values of each pair of impact factors and their interaction through the whole year at the national scale. Interactions of PE ∩ TE, PS ∩ TE, PS ∩ VO, PS ∩ NO, VO ∩ NO, VO ∩ NH, and NO ∩ NH belong to bivariate enhancements and other interactions belong to nonlinear enhancements (Figure 3). Generally, the interaction between NH and TE (q value = 0.46) was the strongest interaction among all impacting factors. Figure S14 indicates that there were obvious seasonal disparities in the interactive influence. In spring, fall, and winter (but not summer), the interactions between meteorological factors played major roles in $PM_{2.5}$ concentrations. The interaction WI ∩ RH (q value = 0.38) had the strongest effect on $PM_{2.5}$ in spring. In autumn and winter, the interaction between SS

and TE (autumn: q value = 0.54; winter: q value = 0.42) played the strongest role in $PM_{2.5}$ concentrations. However, NH ∩ RH (q value = 0.35) was the highest in summer.

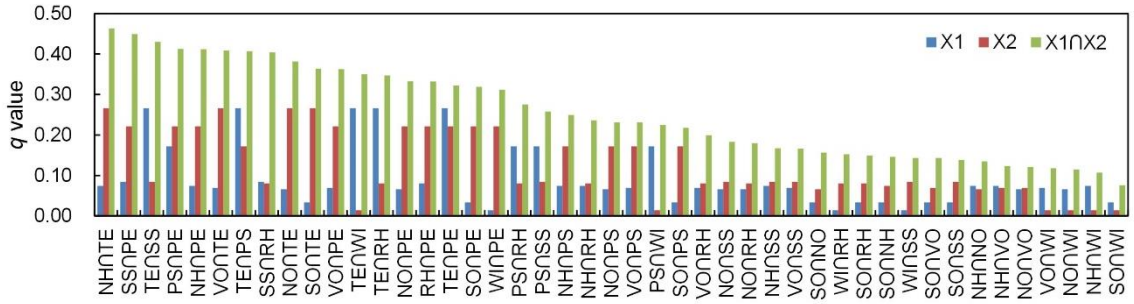

Figure 3. The annual interactive and individual q value of impacting factors in China. Note: VO denotes VOCs; NO denotes NOx; NH denotes NH_3; SO denotes SO_2.

There were obvious regional disparities in all the interactions in China (Figure 4). The interactions between meteorological factors were strongest in all regions. In the interactions between AP and meteorological factors, the interaction of AP∩TE played a primary role in EC, MUYR, and northern China, and the interaction of AP∩PE played a dominant role in MUYR, MUPR, and SC. Figures S15–S18 show the seasonal interactive effects of each pair of driving factors at the regional scale in China, indicating that there were also obvious regional and seasonal differences in all the interactions.

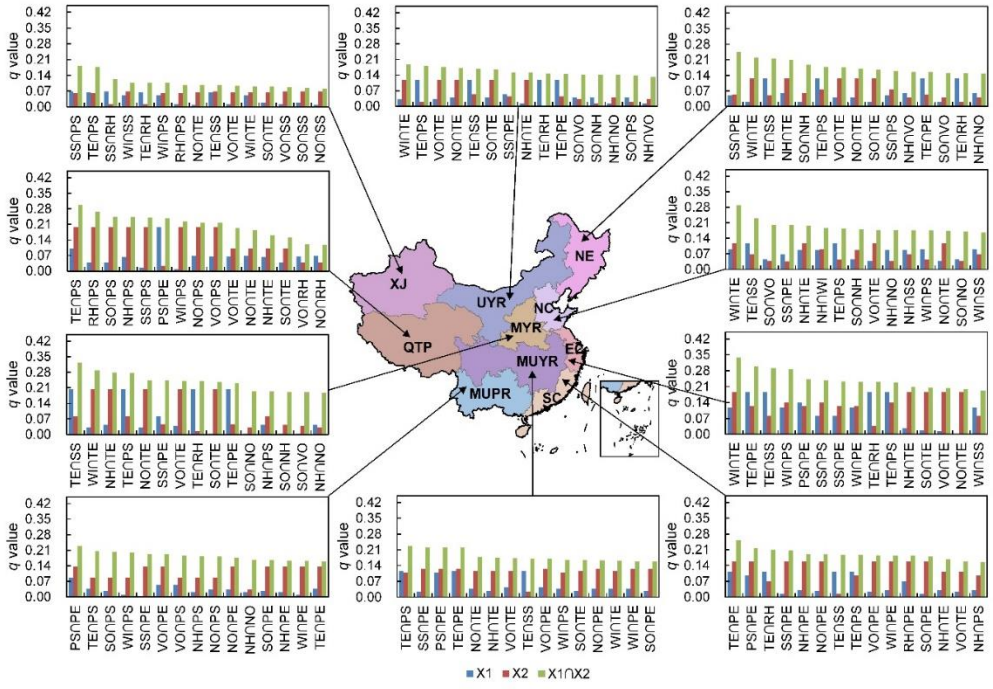

Figure 4. Annual interactions between driving factors at the regional scale (we listed the top 15 interactions). Note: VO denotes VOCs; NO denotes NOx; NH denotes NH_3; SO denotes SO_2.

4. Discussion

$PM_{2.5}$ concentrations were found to be driven by natural conditions and human activities. Previous studies had reported that $PM_{2.5}$ concentrations correlate with both MCs and AP emissions [16,61–65]. We analyzed influences of AP and meteorological factors and their interactions on $PM_{2.5}$ in Chinese cities. Results showed that effects of AP and MCs and their interactions on $PM_{2.5}$ had obvious seasonal and regional variations across China.

In this study, we found that meteorological factors were the leading driving factors at the annual time scale and the national scale, indicating that the meteorological conditions had dominant influences on $PM_{2.5}$ concentrations. TE and PE were the two leading factors affecting $PM_{2.5}$ concentrations. TE was closely related to $PM_{2.5}$ concentrations by affecting atmospheric perturbation and chemical reactions [24]. PE could scavenge $PM_{2.5}$ from the air and had a moisture removal effect [11,66,67]. However, the major influencing factors on $PM_{2.5}$ concentration had significantly seasonal variations. The influences of meteorological conditions on $PM_{2.5}$ concentrations were the strongest in winter, and TE was the leading factor affecting $PM_{2.5}$ concentrations. This is because temperature inversion can weaken the scattering and dispersion of $PM_{2.5}$, resulting in higher local $PM_{2.5}$ pollution [68,69]. In summer, influences of meteorological conditions on $PM_{2.5}$ concentrations were weakest. RH was the leading factor affecting $PM_{2.5}$ concentrations in summer. This was because RH was higher in summer than in the other three seasons, which had a suppression effect on $PM_{2.5}$ under moist air conditions [29].

At the annual time scale, TE was the dominant factor in northern and southern China (except MUPR), which is consistent with previous studies [70,71]. PE was the primary factor affecting $PM_{2.5}$ concentrations in southern China in most seasons throughout the whole year. This was because PE in southern China was higher than in other regions [29]. The increasing PE had scavenging effects on $PM_{2.5}$ by wet deposition, and could lower the $PM_{2.5}$ concentration [66,67]. WI was the main influencing factor in EC and northern China in winter, which is similar to some reports that WI was the most important and negative impacting factor on PM concentrations [72,73]. This was because weaker East Asia winter monsoons could slow wind speeds and increased the frequency of static wind, which had made it more difficult for PM to disperse [74].

Previous studies have indicated that AP were also crucial driving factors on $PM_{2.5}$ [48,63,75]. We found that the influence of AP on $PM_{2.5}$ in winter were higher than in other seasons in XJ and northern China (MYR, UYR, NC). This might be due to the high anthropogenic emissions from winter carbon-fired heating and less surface vegetation cover in winter in northern China, which significantly increased pollutant emissions in the atmosphere [76,77]. In addition, NH_3 had important impacts on $PM_{2.5}$ among AP factors in winter at the national scale and in some individual regions (NC, SC, MUYR, and MYR). Ammonia is an important precursor, and emissions of ammonia had stronger associations with $PM_{2.5}$ concentrations than other anthropogenic precursors; this is similar to results of previous studies [78]. Ammonia participates in photochemical reactions as an atmospheric alkaline gas, which is important in the SIA (secondary inorganic aerosol) formation of compounds such as ammonium salts, sulfate, and nitrate [79–83]. In China, SIA was an important driving factor of $PM_{2.5}$ pollution, especially during severe smog events [80,84]. SIA accounted for 32% of $PM_{2.5}$ mass concentration in China (e.g., Beijing, Guangzhou, Shanghai, and Xi'an) during the 2013 haze pollution events [84]. There was a 5.7% reduction in the annual concentration of $PM_{2.5}$ concentrations when ammonia emissions were cut by 47% in the Beijing-Tianjin-Hebei region of China [79].

$PM_{2.5}$ concentrations are affected by complex interactions between AP and meteorological conditions. This study found that interactions between any driving factors at all time and space scales had significant enhancement effects on $PM_{2.5}$ concentrations. The leading interactive effect between AP and MCs was between AP with RH and PE at the national scale in summer. This might be due to the fact that PE and RH have scavenging and suppression effects on $PM_{2.5}$ by wet deposition in summertime. However, the primary interactive effect between AP and MCs was AP ∩ TE at the national scale in winter. This was due to increased temperature inversion under the lower winter temperature, which weakened the diffusion and dispersion of pollution. The interaction between AP had the dominant effect on $PM_{2.5}$ concentrations over most regions in summer, which indicated that accelerated photochemical reactions between AP occurred under high-temperature conditions. Interactions between AP and factors of PE and PS were important at annual and seasonal scales in southern China. This might be due to the high precipitation and surface pressure in southern China [29].

5. Conclusions

The effects of AP and MCs on $PM_{2.5}$ in Chinese cities were systematically analyzed in this study. The findings revealed significant seasonal and regional disparities in the impacts of examined factors and how they interacted on $PM_{2.5}$. The results can help us to better understanding the relative importance of the driving factors in the formation of $PM_{2.5}$. The study indicated that local AP and meteorological factors had important impacts on $PM_{2.5}$ in China, and had obvious regional and seasonal variations. Meteorological conditions played a leading role in determining $PM_{2.5}$ concentrations at the regional and national scales throughout the whole year. At the seasonal time scale, WI was the primary factor on $PM_{2.5}$ concentrations in winter in northern China and XJ, but PE was the major driver on $PM_{2.5}$ concentrations during most seasons in southern China. However, AP had stronger impacts on $PM_{2.5}$ during winter than in other seasons in XJ and most regions of northern China. NH_3 had a stronger effect on $PM_{2.5}$ concentrations during winter than other anthropogenic precursors. Interactions between all influencing factors have enhanced effects on $PM_{2.5}$ concentrations. In addition, the interaction between MCs and AP played a leading role at the national scale throughout the whole year and in summer and winter. The results could provide a basis for the government to develop more precise air pollution control strategies.

Some limitations to the study should be clarified to assist future studies. First, land use and land cover, socioeconomic conditions, elevation, and topography were not considered to assess their influence on $PM_{2.5}$ concentrations. Second, the uncertainty of MEIC emission inventory may lead to some uncertainties in the research results. When processing emissions inventory data, because the inventory only has monthly emissions data, so as to match the daily $PM_{2.5}$ concentration and daily weather monitoring data, we took the arithmetic average of the monthly inventory emissions data, which also increased the uncertainty of the analysis results. Third, the data used in this study is limited to 2016, and does not include data analysis of other years. This is because the emission inventory data that we could obtain were for 2008, 2010, 2012, 2014, and 2016, but the $PM_{2.5}$ data in 366 cities were available only from 2015 to 2017. In order to maintain consistency between the data, we selected 2016 as the research period in this study. There was no comparative analysis of inter-annual variability. Therefore, we should comprehensively consider other factors on $PM_{2.5}$ concentrations including socioeconomic, land use, terrain, and elevation in the future. In addition, inter-annual change analysis based on multi-year data needs to be added.

Supplementary Materials: The following are available online at www.mdpi.com/2071-1050/12/9/3550/s1, Figure S1: Ten regions in China, including NE, NC, EC, SC, MYR, MUYR, MUPR, UYR, XJ, and QTP. Figure S2: Map of air quality monitoring sites (a) and meteorological sites (b). Figure S3: Annual mean $PM_{2.5}$ concentrations in Chinese cities (2015-2017). Figure S4: Seasonal mean $PM_{2.5}$ concentrations in spring (a), summer (b), autumn (c), and winter (d) in Chinese cities (2015–2017). Figure S5: Spatial distributions of annual mean precipitation (a), surface pressure (b), relative humidity (c), sunshine duration (d), air temperature (e), and wind speed (f) in China during 2015-2017. Figure S6: Spatial distributions of seasonal precipitations in spring (a), summer (b), autumn (c), and winter (d) in China during 2015-2017. Figure S7: Spatial distributions of seasonal surface pressure in spring (a), summer (b), autumn (c), and winter (d) in China during 2015-2017. Figure S8: Spatial distributions of seasonal relative humidity in spring (a), summer (b), autumn (c), and winter (d) in China during 2015-2017. Figure S9: Spatial distributions of seasonal sunshine duration in spring (a), summer (b), autumn (c), and winter (d) in China during 2015-2017. Figure S10: Spatial distributions of seasonal air temperature in spring (a), summer (b), autumn (c), and winter (d) in China during 2015-2017. Figure S11: Spatial distributions of seasonal wind speed in spring (a), summer (b), autumn (c), and winter (d) in China during 2015-2017. Figure S12: (a) NH_3 emissions, (b) NOx emissions, (c) SO_2 emissions, and (d) VOCs in China in 2016 with resolution of 0.25° × 0.25°. Figure S13: Correlations between $PM_{2.5}$ concentrations and impacting factors in 10 regions (a.EC, b. MUPR, c. MUYR, d. MYR, e. NC, f. NE, g. QTP, h. SC, i. UYR, and j. XJ) of China. Figure S14: The seasonal interactive q values and the original q value of each pair of factors. Figure S15: The interactions between impacting factors in spring at the regional scale in China. Figure S16: Interactions between impacting factors in summer at the regional scale in China. Figure S17: Interactions between impacting factors in autumn at the regional scale in China. Figure S18: Interactions between impacting factors in winter at the regional scale in China. Table S1: Effect of various factors on $PM_{2.5}$ in China in 2016. Table S2: Effect of various factors on $PM_{2.5}$ throughout the whole year at the regional scale. Table S3: Effect of various factors on $PM_{2.5}$ in spring at the regional scale. Table S4: Effect of various factors on $PM_{2.5}$ in summer at the regional scale. Table S5: Effect of various factors on $PM_{2.5}$ in autumn at the regional scale. Table S6: Effect of various factors on $PM_{2.5}$ in winter at the regional scale.

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