Hindawi
Advances in Meteorology
Volume 2019, Article ID 5295726, 15 pages
https://doi.org/10.1155/2019/5295726

Research Article

$PM_{2.5}/PM_{10}$ Ratios in Eight Economic Regions and Their Relationship with Meteorology in China

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Received 4 October 2018; Revised 4 December 2018; Accepted 16 December 2018; Published 4 February 2019

Academic Editor: Herminia García Mozo

China is suffering severe ambient air pollution in recent decades and particulate matter (PM) has become the major pollutant, especially for $PM_{2.5}$ and PM_{10}, which have highly raised scholars and policy-makers' attention in last few years. The existing research has focused on the characteristics of $PM_{2.5}$ and PM_{10}, respectively, or analyzed the correlation between the two pollutants, while the ratio of $PM_{2.5}$ to PM_{10} has been taken less consideration. In this study, daily mean $PM_{2.5}$ and PM_{10} mass concentrations in 31 provincial capitals from 2014 to 2016 were used to present the temporal variations and spatial distribution of $PM_{2.5}/PM_{10}$ ratios among eight economic regions. And then, statistical method and correlation analysis were adopted to investigate the relationship between the ratios and AQI, the rate of change on the ratios, and the impact of meteorological parameters on the ratios. The results indicated that $PM_{2.5}/PM_{10}$ ratios showed an increasing trend from northwest to southeast due to different economic development and industrial types. The highest values were observed in winter among all regions, and the ratios on weekends were higher than that of on weekdays in most of the regions. Besides, domestic heating in northern China had a significant contribution to the ratios. Moreover, ratios had less changes, and the rate of change was stable in summer. As for air quality, the higher the ratio, the larger the possibility of high AQI so that the air pollution will be more severe. In terms of meteorological factors, the results demonstrated that relative humidity, precipitation, and pressure were the most important factors and had significantly positive impacts, while sunshine duration, temperature, and wind speed had negative effects on the ratios. The findings could identify the pollution sources among PM_{10} and be helpful for making regulation locally to reduce emission which considers anthropogenic sources and meteorological diffusion simultaneously.

1. Introduction

The problem of air pollution has drawn much attention in recent decades with rapid development of economy and urbanization, especially in developing countries [1]. In China, particulate matter (PM) pollution has become the major air pollutant throughout the year [2]. PM originates from natural sources and anthropogenic emissions [3], and the latter is the focus of environmental protection because it can be controlled by emission reduction measures. Among the anthropogenic emissions, they could be divided into stationary source and mobile source; besides, they are emitted directly in the atmosphere (primary particles) or transformed into secondary organic particles by gaseous pollutants such as SO_2 (sulfur dioxide) and NO_x (nitrogen oxides) [4, 5]. Apart from the complexity of PM formation, meteorological conditions play an indispensable role in the diffusion, deposition, and transport of PM [6–8]. Meteorological factors such as temperature, precipitation, wind speed, and wind direction have strong effects on the dilution, nucleation, condensation, and evaporation of particles [9]. It is generally accepted that anthropogenic emissions are source drivers of air pollution, and meteorological variables are diffusion causes [10].

Empirical experience and previous studies have demonstrated that PM has negative impact on public health [11–13], road visibility [14, 15], ambient air quality, and climate [16]. However, different sizes of PM have distinct characteristics and influence due to the varied chemical and physical compositions, emission sources, and location [17]. Among them, the most concerns are $PM_{2.5}$ (particulate matter with diameters less than 2.5 μm) and PM_{10} (particulate matter with diameters less than 10 μm). It is suggested that $PM_{2.5}$ is more harmful to human health than PM_{10} [18]. Moreover, small particle is an essential influential factor to visibility since the mass extinction efficiency of $PM_{2.5}$ is 7 times of larger particles [19, 20]. Furthermore, the deterioration of air quality and haze episodes illustrates that $PM_{2.5}$ has a dominant role in the formation of smog [21, 22]. Therefore, the identification of the fraction of $PM_{2.5}$ in PM_{10} is important owing to its more threats.

Literatures on $PM_{2.5}$ and PM_{10} were often conducted independently, and mainly in health effects, spatial distributions, temporal trends, source apportionment, chemical composition, and influential factors analysis [23–31]. In terms of the relationship between $PM_{2.5}$ and PM_{10}, it is proved that the mass concentration of $PM_{2.5}$ was highly correlated with PM_{10} [32–35]. Nevertheless, the proportion of $PM_{2.5}$ in PM_{10} has been given less attention since just a few studies have mentioned it and only gave the mean ratio during the study period, without any in-depth analysis [36]. Since different sizes of PM originate from different sources, the $PM_{2.5}/PM_{10}$ ratio could be used for identifying their sources [17]. A higher ratio means the overwhelming contribution from $PM_{2.5}$, which is generally ascribed to primary pollution by anthropogenic activities and secondary particulate formation such as NO_3^-, SO_4^{2-}, NH_4^+, and organics, while a lower ratio is mainly contributed to fugitive dust or sand dust from long-distance transport [37]. Varied $PM_{2.5}$ and $PM_{2.5-10}$ (particulate matter with diameters more than 2.5 μm and less than 10 μm, or PM_{coarse}) compose PM_{10} dynamically, and thereby the study of $PM_{2.5}/PM_{10}$ ratio can imply the contribution of $PM_{2.5}$ directly and show the contribution of PM_{coarse} indirectly at the same time. Moreover, the $PM_{2.5}/PM_{10}$ ratio could identify the major pollutants among PM_{10} and then be helpful for making separate regulation locally to reduce emission rather than just controlling $PM_{2.5}$. Munir had done a similar research on the ratio of $PM_{2.5}$ to PM_{10} but in the UK and Saudi Arabia [34, 38]. It is noteworthy that great heterogeneity exists at different regions on the $PM_{2.5}/PM_{10}$ ratio due to the varied ratio from site to site [34].

Thus it is essential and urgent to characterize the $PM_{2.5}/PM_{10}$ ratios in China, to understand how the ratios vary at daily, monthly, seasonal, and annual level in different regions at the national scale. Furthermore, it is crucial to analyze how the ratios are influenced by meteorological factors. Only by combining the uncontrolled factors of meteorology with the controllable factors of anthropogenic emissions can PM pollution be fully understood and effective mitigation measures be developed [39]. Considering the impact of meteorology on PM, statistical approaches such as correlation analysis [40], multiregression [23, 41], neural networks [42, 43], and generalized additive models [44, 45] were widely used. In addition, the study areas of PM pollution in China were several PM concentration monitoring sites [46], or some metropolis [47, 48], may be a well-known region, such as Yangtze River Delta, Pearl River Delta [36, 49]. Recently, the studies at the national scale increased gradually since the monitoring devices were installed all around the country [36, 50]. However, the comparison among different economic regions throughout China has been taken less consideration. Lorenz curve proved that pollutant emissions were sensitive to the economic development and industrial type [51, 52]. Different regions with distinct target of economy and various foundation of industry should make a difference in the process of reducing PM pollution according to the pollution level locally [53].

In this study, the ratios of $PM_{2.5}$ to PM_{10} in eight different economic regions throughout China and the impact of meteorology on the ratios were analyzed. Firstly, temporal trends and spatial heterogeneity of the ratios in various economic regions from 2014 to 2016 were investigated to assess the pollution level as well as the relationship between $PM_{2.5}$ and PM_{10}. Secondly, statistical analysis was conducted between the $PM_{2.5}/PM_{10}$ ratio and air quality index (AQI) to illustrate the influence of the ratio on ambient air quality. Thirdly, the rate of change on $PM_{2.5}/PM_{10}$ was analyzed. Finally, correlation analysis was used to identify the impact of meteorological parameters on the ratios.

2. Materials and Methods

2.1. Study Areas. To evaluate the ratio of $PM_{2.5}$ to PM_{10} in different economic conditions of China, 31 provinces and municipalities (except Hong Kong, Taipei and Macau) were divided into eight economic regions which referred China National Bureau of Statistic, as shown in Table 1 and Figure 1.

2.2. Data Collection. The daily mean $PM_{2.5}$ and PM_{10} mass concentrations from 2014 to 2016 in 31 provincial capital and municipalities were obtained directly from the China National Environmental Monitoring Center (http://datacenter.mep.gov.cn/), and the PM values of the provincial capital were considered as the values of its corresponding province in this study. After that, the $PM_{2.5}/PM_{10}$ ratios of 31 provincial capital and municipalities were calculated separately. In order to ensure accuracy of the study and take full advantage of data, invalid ratios were eliminated, including null values (missing PM values), 0 ($PM_{2.5}$ equal to 0), and ratio greater than or equal to 1 (unreasonable phenomenon that the mass concentration of $PM_{2.5}$ is more than PM_{10}). Furthermore, the regional ratios were calculated by averaging the provincial values according to the partition in Table 1. In addition, meteorological parameters for 31 provinces and municipalities were downloaded from official website of China Meteorological Administration, including daily precipitation amount (PP), pressure (P), relative humidity (RH), sunshine duration (SD), daily mean temperature (T), daily mean wind speed (WS), and daily wind direction of the maximum wind speed (WD). The missing values were deleted and trace amount of precipitation was considered as 0.01 mm. Besides, the categories of WD were the following: N: 348.76°–11.25°; NNE: 11.26°–33.75°; NE: 33.76°–56.25°; ENE: 56.26°–78.75°; E: 78.76°–101.25°; ESE: 101.26°–123.75°; SE: 123.76°–146.25°; SSE: 146.26°–168.75°; S: 168.76°–191.25°; SSW: 191.26°–213.75°; SW:

Table 1: Eight economic regions in China.

Economic regions	Abbreviation	Provinces and municipalities
Northeast Region	NR	Liaoning, Jilin, Heilongjiang
North Coastal Area	NCA	Beijing, Tianjin, Hebei, Shandong
Eastern Coastal Area	ECA	Shanghai, Jiangsu, Zhejiang
South Coastal Area	SCA	Fujian, Guangdong, Hainan,
The Middle Yellow River	MYR	Shanxi, Inner Mongolia, Henan, Shaanxi
Middle reaches of the Yangtze River	MRYR	Anhui, Jiangxi, Hubei, Hunan
Southwest China	SC	Guangxi, Chongqing, Sichuan, Guiyang, Yunan
Big Northwest China	BNC	Tibet, Gansu, Qinghai, Ningxia, Xinjiang

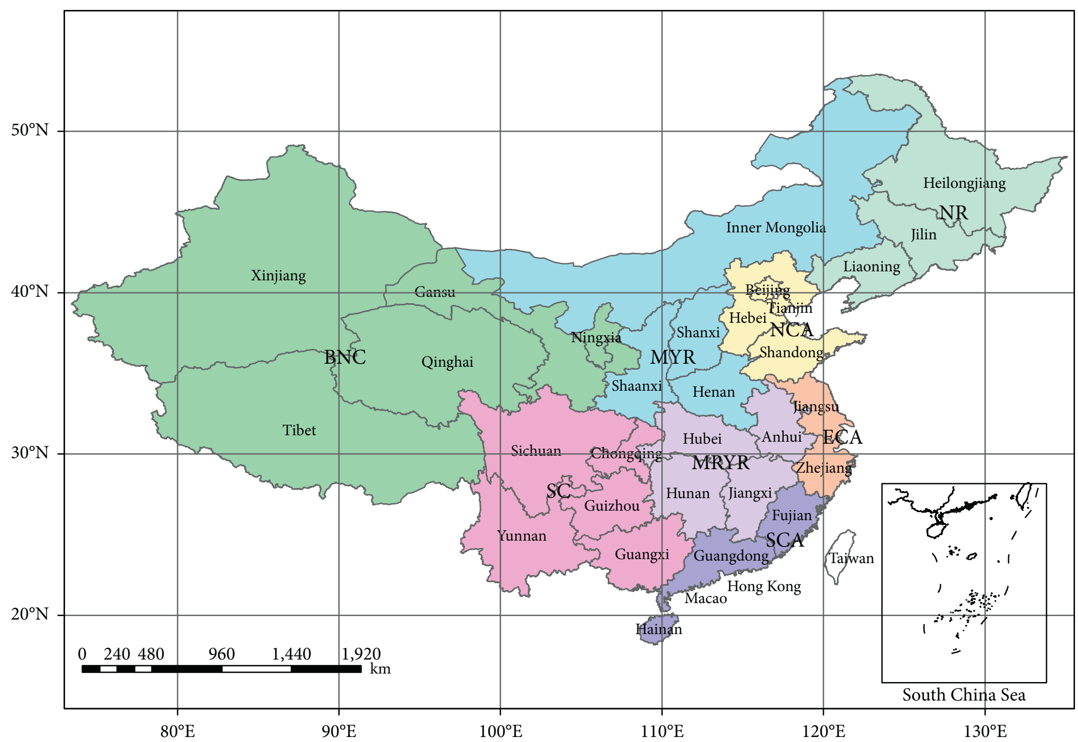


Figure 1: Eight economic regions in China.

213.76°–236.25°; WSW: 236.26°–258.75°; W: 258.76°–281.25°; WNW: 281.26°–303.75°; NW: 303.76°–326.25°; NNW: 326.26°–348.75°, and they were considered as 1 to 16 in turn (N: North; E: East; S: South; W: West). The descriptive statistics of meteorological parameters are given in Table 2.

2.3. Methods. Temporal characteristics of $PM_{2.5}/PM_{10}$ ratios were investigated by annual, interannual, and seasonal variations and the differences between weekday and weekend, domestic heating, and nonheating period. Mean values of the ratios were compared at above time scale to reveal the pollution situation in these years at different parts of China.

Then, a further spatial analysis was conducted. Coefficient of variance (CV) represents the discreteness of data so that the data can be used to reveal internal differences of $PM_{2.5}/PM_{10}$ ratios within one region. It is accumulated as standard deviation divided by mean value [26, 32, 54]. For spatial heterogeneity between two regions, Pearson correlation and coefficient of divergence (COD) were adopted in this study. The former illustrates positive or negative linear correlation statistically, while the latter shows the similarity of ratios between different regions considering spatial geography and some other potential factors [32, 55]. The COD is between 0 and 1, and the larger value is, the greater heterogeneity is. The COD is defined as

$$\mathrm{COD}_{ij} = \sqrt{\frac{1}{n} * \sum_{t=1}^{n} \left(\frac{\left(x_{ti} - x_{tj}\right)}{\left(x_{ti} + x_{tj}\right)} \right)^2}, \quad (1)$$

where COD_{ij} is the coefficient of divergence between region i and region j, x_{ti} and x_{tj} are the $PM_{2.5}/PM_{10}$ ratio during time t in region i and region j, and n is the number of time.

In order to explore the relationship between $PM_{2.5}/PM_{10}$ and AQI, the Spearman correlation coefficient was calculated due to AQI values were nonnormal distribution [34, 56].

Table 2: The mean values (standard deviation) of meteorological parameters.

Regions	PP (mm)	P (hPa)	RH (%)	SD (h)	T (°C)	WS (m/s)	WD
NR	1.58 (5.76)	999.30 (13.36)	62.26 (16.16)	6.67 (4.02)	7.13 (14.21)	2.49 (1.14)	9.50 (4.52)
NCA	1.43 (7.67)	1007.62 (12.47)	55.48 (19.30)	6.09 (4.09)	14.47 (10.71)	2.19 (1.02)	7.92 (4.31)
ECA	4.28 (12.16)	1013.39 (9.33)	73.58 (13.29)	4.52 (3.97)	17.27 (8.56)	2.41 (0.97)	7.02 (4.70)
SCA	5.62 (17.33)	1004.71 (7.02)	78.84 (10.25)	4.70 (3.99)	22.61 (6.29)	2.49 (1.02)	6.91 (4.46)
MYR	1.45 (5.60)	946.48 (44.62)	56.76 (18.47)	6.27 (4.13)	12.62 (11.03)	2.41 (1.17)	8.57 (4.98)
MRYR	3.70 (12.98)	1003.78 (9.99)	77.76 (13.07)	4.36 (4.19)	18.82 (8.39)	2.21 (0.94)	10.95 (5.12)
SC	3.35 (10.76)	924.61 (70.69)	78.37 (11.81)	3.76 (3.91)	17.86 (71.78)	1.81 (8.62)	8.66 (4.81)
BNC	0.98 (3.52)	811.89 (93.49)	51.45 (19.48)	7.62 (3.57)	8.58 (10.54)	1.68 (0.75)	7.64 (4.70)

All of the above present the pollution status and the variation trend of ratios for a long term. However, short-term and instantaneous rate of change, which reflects the stability of the pollutant source, should be also considered. The rate of change (ROC) on the $PM_{2.5}/PM_{10}$ ratio was introduced to reflect the degree of change. Large ROC implies greater changes of emission sources, and it is necessary to be given more concern. The ROC of the ratio is calculated as follows:

$$\mathrm{ROC}_t = \frac{\left[(\mathrm{PM}_{2.5}/\mathrm{PM}_{10})_t - (\mathrm{PM}_{2.5}/\mathrm{PM}_{10})_{t-1} \right]}{(\mathrm{PM}_{2.5}/\mathrm{PM}_{10})_{t-1}}, \tag{2}$$

where ROC_t is the ROC of the $PM_{2.5}/PM_{10}$ ratio at time t and $(PM_{2.5}/PM_{10})_t$ and $(PM_{2.5}/PM_{10})_{t-1}$ are the ratio at time t and $t-1$, respectively.

In terms of meteorological factors, Spearman correlation analysis tested the positive or negative effects of the meteorological variables on $PM_{2.5}/PM_{10}$ ratios. Since it was just from a linear point of view, wind direction cannot be quantified because of category variables, and gray relational method was adopted to identify the dominant factors on $PM_{2.5}/PM_{10}$ ratio. It is an average value of the gray relational coefficients which are calculated as follows [26, 57]:

$$\xi_{a(t)} = \frac{\min_a \min_t \left| X_r(t) - X_a(t) \right| + 0.5 \max_a \max_t \left| X_r(t) - X_a(t) \right|}{\left| X_r(t) - X_a(t) \right| + 0.5 \max_a \max_t \left| X_r(t) - X_a(t) \right|}, \tag{3}$$

where $\xi_{a(t)}$ is the gray relational coefficient between the influencing factor a and the $PM_{2.5}/PM_{10}$ ratio, t is the time, $X_r(t)$ is the sequence of the $PM_{2.5}/PM_{10}$ ratio, and $X_a(t)$ is the sequence of influencing factor a.

The relevant coefficients were calculated by SPSS, and the figures were output by ArcGIS.

3. Results and Discussion

3.1. Temporal Variations of $PM_{2.5}/PM_{10}$ Ratios

3.1.1. Annual Variations. The contribution of $PM_{2.5}$ to PM_{10} varies from time to time, as shown in Table S1. The 3-year mean $PM_{2.5}/PM_{10}$ ratio from 2014 to 2016 in China was 0.562, which ranged from 0.456 (BNC) to 0.633 (ECA), thereby implying the different compositions of particles [58]. An increasing trend from northwest to southeast (regional background color) and the frequency distribution of the ratios (bar chart) could be seen in Figure 2(a), indicated that $PM_{2.5}$ accounted for larger proportion of PM_{10} in southeast districts compared to northwest districts. This phenomenon was consistent with the isohyetal line [59, 60]. The lowest ratios in BNC (0.456) and MYR (0.480) suggested a contribution of more primary PM sources. It was obvious that PM_{coarse} increased due to local dust emission and regional dust transport and also associated with desert climate and less vegetation cover [28, 35, 61]. In contrast, the highest ratios in ECA (0.633) and MRYR (0.625) were related to anthropogenic sources including coal consumption and heavy industries, which were major contributions of $PM_{2.5}$ [62–64]. Therefore, PM emission reduction strategies that consider the reduction of bigger particles at the same time will be more beneficial than just decreasing the high concentrations of $PM_{2.5}$, especially for northwest districts.

3.1.2. Interannual Variations. Interannual variations of $PM_{2.5}/PM_{10}$ ratios from 2014 to 2016 were divided into three categories, as displayed in Figure 2(b) in the regional background color. The first was the ratio dramatically increased from 2014 to 2015, after that decreased slightly, such as NR and NCA. It is referred that these areas have begun to implement measures to reduce $PM_{2.5}$ pollution and have an initial effect. The second was opposite to the first category because the ratio reduced in the previous two years but grew in 2016, and the representative area was MYR, where further effective control for $PM_{2.5}$ was needed. The third category had a continuously decreasing trend in three years for the rest five economic regions, which is contributed to long-term strict emission reduction measures of $PM_{2.5}$ [59].

3.1.3. Seasonal Variations. $PM_{2.5}/PM_{10}$ ratios in spring (March to May), summer (June to August), autumn (September to November), and winter (December to February) were analyzed among eight economic regions, and a remarkable seasonal pattern was observed. The common feature was that the largest ratio emerged in winter among all the regions [61, 65, 66]. That was to say, the severest $PM_{2.5}$ air pollution was in winter throughout the whole country. The large difference on the ratios between winter and other three seasons mainly contributed to heating installations [67], also adverse meteorology with low temperature, less precipitation, and low boundary layer depth, as well as stable atmospheric condition in winter, which were not favorable for $PM_{2.5}$ dispersion [3, 34]. Therefore, improving the performance of the heating installations and choosing clean

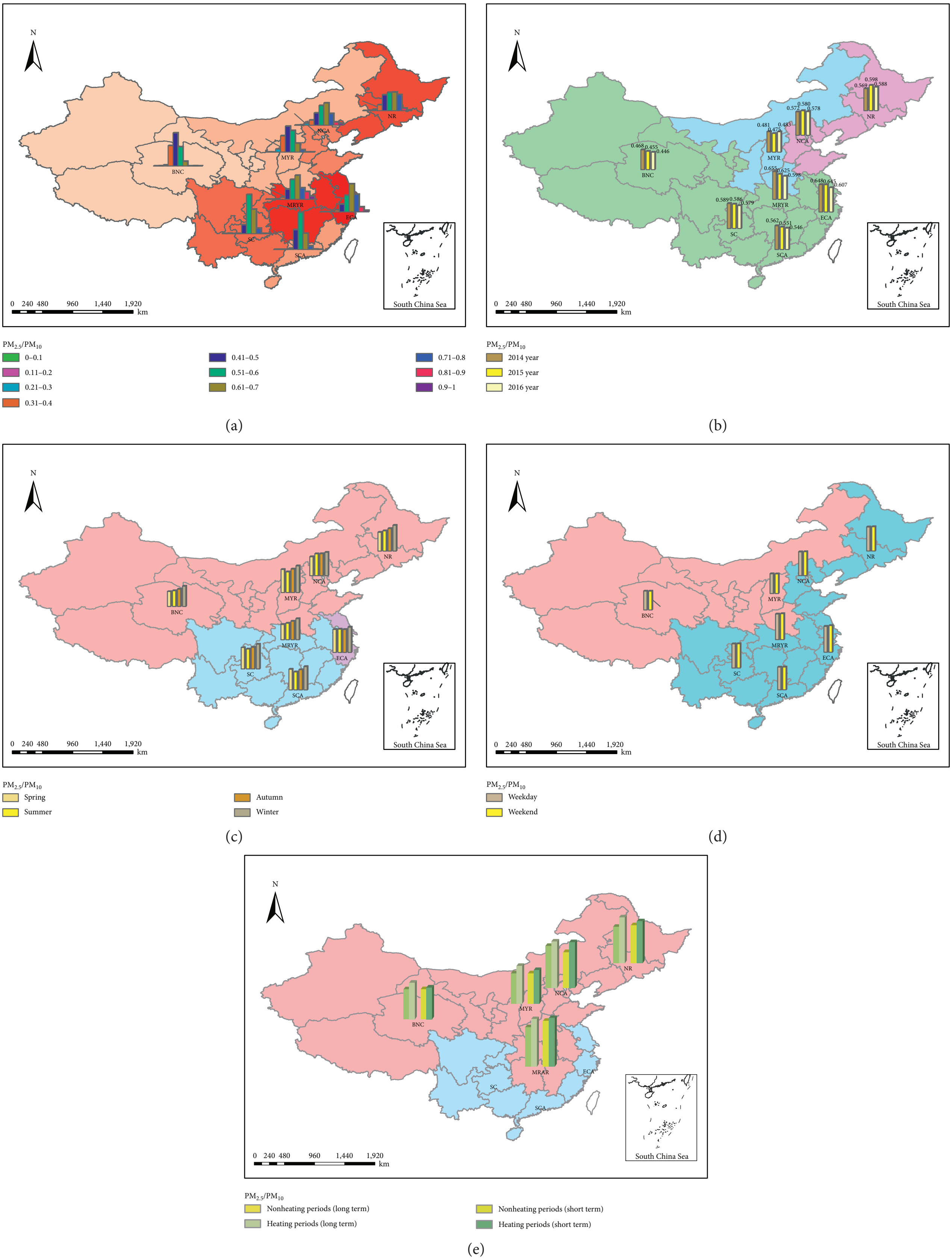


Figure 2: Temporal trends and spatial distribution of $PM_{2.5}/PM_{10}$ ratio. (a) 3-year average ratio with an increasing trend from northwest to southeast (regional background color) and the frequency distribution (bar chart). (b) Interannual variations with three categories (regional background color) and annual mean ratio (bar chart). (c) Seasonal variations with three types (regional background color) and seasonal mean ratio (bar chart). (d) Weekday-weekend variations with two types (regional background color) and corresponding mean ratio (bar chart). (e) Bar chart of mean ratio during heating periods (blue background meant no domestic heating).

energy could be beneficial to reduce small particles in winter. In terms of seasonal variations, they could be split into three types, as shown in background color in Figure 2(c). The ratios in NR, NCA, MYR, and BNC increased gradually from spring to winter, suggesting that PM_{coarse} was the main pollutant of PM_{10} in northern areas during spring because of dust storm, so measures to reduce dust floating such as adding vegetation or using dust remover are necessary [61, 68, 69]. ECA also had the lowest value for $PM_{2.5}/PM_{10}$ in spring, while the ratio in summer which was a little higher than that of in autumn may be contributed to more biomass burning from late May to early June than autumn [21]. Nevertheless, there was a large difference from the above two types, and the third type including SCA, MRYR, and SC, had the least ratio in summer and the second least in autumn, suggesting a higher fraction of coarse particles in summer when may resulted from resuspended and entrainment of dust and sand [32]. On the contrary, the majority of fine particle range was emitted by the biomass burning process in harvest autumn and hence the ratio increased [21, 34]. It can be concluded that the seasonal impact on PM emission and the ratio is mainly related to resuspended dust, biomass burning, and heating devices; therefore, certain emission reduction should be scheduled at different seasons in varied regions.

3.1.4. Weekday-Weekend Variations. There was a clear weekday-weekend pattern in most of the economic regions where weekends had higher values in $PM_{2.5}/PM_{10}$ ratios than weekdays, the same result as other study [38], while only MYR and BNC had the opposite trends, as shown in Figure 2(d). The additional ratios on weekends were connected with additional traffic-related activities, which increased fine particles due to private vehicles and total travel volume [25, 70, 71]. Differently, the ratios on weekends in MYR and BNC were slightly lower than weekdays, which were related to underdeveloped economy and more balanced human activities during a week [72]. It is suggested that traffic behavior has positive impact on small particle emission [73]. Reasonable inducement of traffic travel by improving public transit infrastructure, ticket pricing discount, land use, and layout can reduce vehicle emission by increasing travel efficiency, especially on weekends in south and north of China.

3.1.5. Heating Periods. There is domestic heating phenomenon in northern China during winter, and heating time varies among different cities. Heating periods start normally from October to November, ending in March to April of the next year with a total duration of 4 to 6 months. Where the average temperature is lower, the start of heating period is earlier and the duration lasts longer. Air pollution may result from coal heating so the difference on $PM_{2.5}/PM_{10}$ between heating and nonheating periods is analyzed. ECA, SCA, and SC had no domestic heating and thus were excluded in this section. Moreover, in the rest economic regions, only the cities which had heating phenomenon were analyzed. The heating periods (long term) referred to the date from heating start to end, and the rest of the days were nonheating periods (long term). The comparative results demonstrate in Figure 2(e) that all the heating regions had higher ratio of $PM_{2.5}/PM_{10}$ in heating periods, indicating that domestic heating would indeed increase $PM_{2.5}$ pollution [17, 61]. In order to exclude the seasonal factors and meteorological occasional impact, we further redefined the heating periods (short term) to 15 days after the start of heating period and 15 days before the end of heating; nonheating periods (short term) were 15 days before the start of heating and 15 days after end of heating period. It is also demonstrated that the ratios were higher in heating periods, and $PM_{2.5}$ contributed a lot in PM pollution due to domestic heating.

To sum up, season, traffic, and domestic heating indeed have significant impact on $PM_{2.5}/PM_{10}$ ratios. Winter and heating period need emission control on $PM_{2.5}$ since small particles accounted for a large proportion in PM. Large particles are required to reduce in north of China in spring due to resuspended dust, and other places in summer or autumn due to biomass burning. Furthermore, developed regions with high travel intensity should concentrate on traffic $PM_{2.5}$ emission control, especially on weekends.

3.2. Spatial Variations of $PM_{2.5}/PM_{10}$ Ratios. The method of CV, COD, and correlation analysis reflect spatial heterogeneity from different perspectives. CV represents internal differences of one region while COD and correlation analysis explore the differences between two regions [32, 55]. As shown in the third column of Table 3, the CV of the $PM_{2.5}/PM_{10}$ ratio in NR, NCA, MYR, and MRYR was more than 0.213, suggesting that the ratios changed in a larger range so that there is more heterogeneity in the PM pollution in those regions [35, 74]. Oppositely, CV in SCA and SC were less than 0.150, which meant the ratios in south of China were relatively stable. The value of COD reflects the regional similarity, the larger value means the greater differences of the two regions, and the results were displayed in the upper right of Table 3. The lowest COD value was between SCA and SC (0.069), suggesting that the $PM_{2.5}/PM_{10}$ ratio and variation were similar in those two regions. In contrast, the highest COD value was between ECA and BNC (0.204), resulted from the long distance between the two regions and the large differences in $PM_{2.5}/PM_{10}$ ratios. Correlation analysis reveals the positive or negative correlation of the ratios between the two regions, and Pearson correlation coefficients were presented in lower left part of Table 3 (all coefficients were significant in 99% confidence level). The highest correlation coefficient was between BNC and MYR (0.603), indicating the ratio and its variation were similar. Furthermore, the two regions were adjacent to each other, and a high degree of linear correlation also reflected regional transport of PM to some extent. On the contrary, the lowest coefficient was between NCA and SC (0.069) which meant largest spatial differences.

In summary, the above three methods reached consistent conclusions, SCA and SC had large spatial heterogeneity

Table 3: Spatial heterogeneity of $PM_{2.5}/PM_{10}$ among eight economic regions.

	Mean	CV	NR	NCA	ECA	SCA	MYR	MRYR	SC	BNC
NR	0.586	0.233		0.138	0.151	0.138	0.162	0.151	0.125	0.165
NCA	0.577	0.241	**0.390**		0.145	0.154	0.148	0.158	0.138	0.180
ECA	0.633	0.186	**0.196**	**0.269**		0.136	0.196	0.101	0.113	0.204
SCA	0.553	0.150	**0.154**	**−0.074**	**0.097**		0.150	0.127	0.069	0.137
MYR	0.480	0.233	**0.437**	**0.587**	**0.263**	**0.160**		0.193	0.154	0.100
MRYR	0.625	0.213	**0.294**	**0.171**	**0.488**	**0.330**	**0.401**		0.108	0.199
SC	0.585	0.129	**0.283**	**0.069**	**0.197**	**0.594**	**0.368**	**0.389**		0.153
BNC	0.456	0.190	**0.463**	**0.263**	**0.187**	**0.350**	**0.603**	**0.404**	**0.478**	

Note. All Pearson correlation coefficients were significant at the 0.01 levels (2-tailed). Mean values are in the second column, coefficients of variance (CV) are in the third column, Pearson correlation coefficients are in the lower left (bold), and coefficients of divergence (COD) are in the upper right.

with other regions. The closer the distance between the two regions, the smaller the differences in the $PM_{2.5}/PM_{10}$ ratio distribution and variation due to similar emission sources and diffusion conditions, as well as the distance transport between adjacent regions.

3.3. The Relationship between $PM_{2.5}/PM_{10}$ and AQI. The data of AQI in various regions were of nonnormal distribution, and the skewness is shown in Table 4. Therefore, Spearman correlation analysis was introduced to explore the relationship between the $PM_{2.5}/PM_{10}$ ratio and AQI. The correlation coefficients in Table 4 revealed that the ratio had a significantly positive correlation with the value of AQI. It also can be seen in Figure 3(a) that the higher the degree of AQI, the larger the mean value of the ratio in all economic regions (AQI was divided into six levels: excellent, good, slight pollution, moderate pollution, heavy pollution, and severe pollution) [75]. Higher ratios in severe air pollution may indicate that more small particles or new particles were formatted with the increased emissions [74, 76]. Moreover, it is noteworthy that coefficients in all economic regions did not present a strong linear correlation (coefficients more than 0.5) [77], which meant that for each day, not the higher $PM_{2.5}/PM_{10}$ ratio would inevitably lead to a higher AQI. Nevertheless, in terms of the overall trend, the higher the ratio, the larger the possibility of high AQI and the more serious the air pollution will be. Furthermore, air quality was mainly at the excellent and good levels when the ratio was between 0.3 and 0.6, while AQI was at the moderate and heavy levels when the ratio was more than 0.7, as presented in Figure 3(b).

All the above illustrated that $PM_{2.5}$ contributed more to decline air quality than PM_{10}. It was also consistent with the phenomenon that $PM_{2.5}$ was the major pollutant on severely polluted days like in winter. Therefore, when the air quality become worse, the principal task is to control $PM_{2.5}$ emission sources.

3.4. Rate of Change on $PM_{2.5}/PM_{10}$ Ratio. The spatiotemporal variation of average ROC of every two-day $PM_{2.5}/PM_{10}$ ratios is shown in Figure 4. It is obvious that the ROC of the ratio in the south of China was lower than that in the north but the ratio had the opposite trend. The reason was that $PM_{2.5}$ emission resulted from a few sources in the south of China, was more than PM_{coarse}, so it would not be influenced by other factors easily.

Interannual trends can be divided into two types (background color in Figure 4(a)), one for increasing first and then decreasing with the highest ROC in 2015. Type two was SCA, MRYR, and SC, where the ROC on ratios increased gradually from 2014 to 2016 but the ratios decreased, indicating more sources of PM in recent years but with a decreasing trend of $PM_{2.5}$ emissions compared to large particles.

In terms of seasonal trends, NR, ECA, SC, and BNC had the highest ROC of the $PM_{2.5}/PM_{10}$ ratio in spring, while other regions had the highest rate in winter. Besides, the lowest values were obtained during summer in all the regions except for SC, as displayed in Figure 4(b). It is suggested that $PM_{2.5}/PM_{10}$ ratios had the greater changes, and the proportion of $PM_{2.5}$ was relatively unstable in spring and winter, owing to more diversity and variability of emission sources so may be influenced by various factors. Inversely, the ratios had less changes and were stable in summer because of a few sources with few emissions. Therefore, emission control in spring and winter needs focus on different sources to cope with high ROC and ratios.

The $PM_{2.5}/PM_{10}$ ROC on weekday was dramatically different from weekend. The south of China had the higher ratios and ROC on weekends than those on working days, indicating a variety of emission sources such as additional traffic demand.

ROC on ratios revealed the feature of emission sources to some extent. When compared the variations of the ratios with their ROC, it could be concluded that south of China had more severe small particle pollution and relatively simple source than large particles. On the contrary, PM emission sources were more complex in the north of China due to mixed heavy industries. Moreover, emission sources became more and diffusion conditions became worse in winter due to additional traffic in Spring Festival, bad weather, and domestic heating.

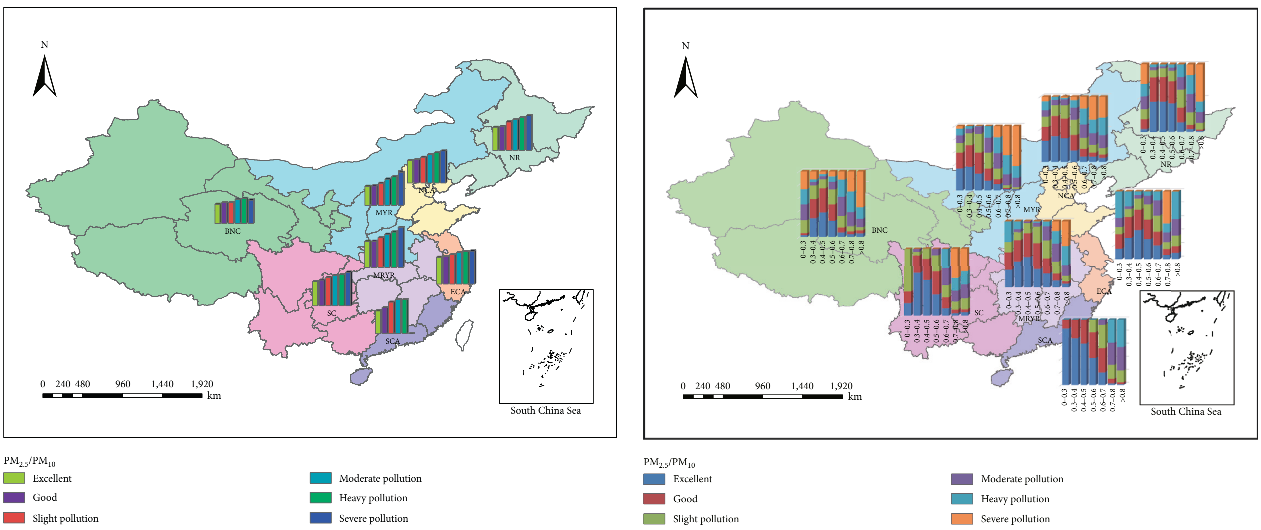
3.5. The Relationship between $PM_{2.5}/PM_{10}$ and Meteorological Factors

3.5.1. Spearman Correlation Analysis. Air pollution results from the final accumulation of particles after time transformation and spatial diffusion [34]. Among them, the

Table 4: The skewness of AQI and Spearman correlation coefficients between the $PM_{2.5}/PM_{10}$ ratio and AQI.

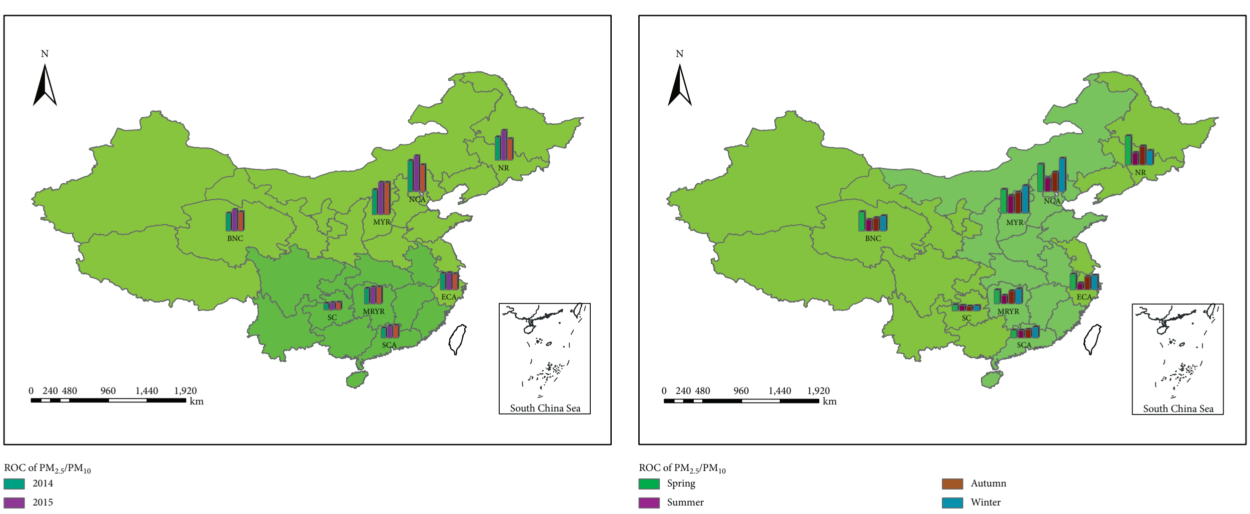
	NR	NCA	ECA	SCA	MYR	MRYR	SC	BNC
AQI skewness	2.142	1.833	1.574	1.549	1.972	1.753	2.245	3.038
Coefficients	0.467	0.388	0.200	0.441	0.376	0.284	0.347	0.068

Note. All Spearman correlation coefficients were significant at the 0.01 level (2-tailed).



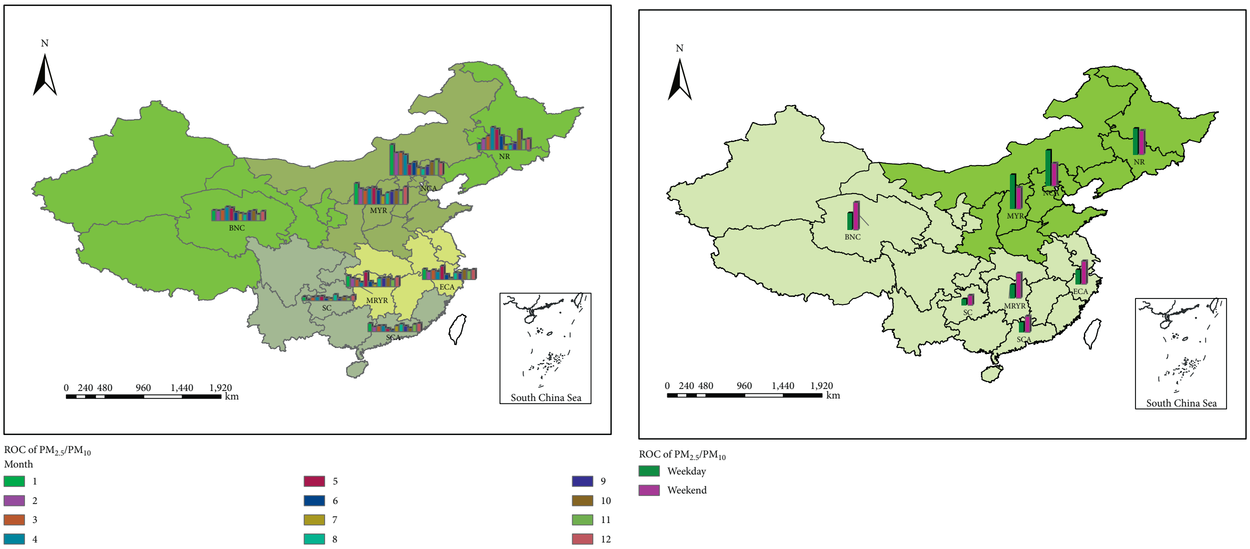
(a) (b)

Figure 3: The relationship between the $PM_{2.5}/PM_{10}$ ratio and AQI. (a) Average $PM_{2.5}/PM_{10}$ ratio in six levels of AQI among eight economic regions; (b) the frequency distribution of AQI levels in different $PM_{2.5}/PM_{10}$ ratios.



(a) (b)

Figure 4: Continued.



(c)

(d)

FIGURE 4: Spatiotemporal variations of average ROC on $PM_{2.5}/PM_{10}$ ratio. (a) Interannual trends with two types. (b) Seasonal variations with two types. (c) Monthly variations with four types. (d) Weekday-weekend pattern with two types.

contribution of meteorological factors cannot be ignored; even with the same source of pollutant, the degree of pollution will be much different due to different meteorological conditions [57, 78]. The correlation analysis was used in this study to explore the impact of meteorological factors on the ratio of $PM_{2.5}/PM_{10}$.

The Spearman correlation analysis was used to test the positive and negative effects of the meteorological variables on the $PM_{2.5}/PM_{10}$ ratio. The correlation coefficients were different for each region, indicating that the meteorological influential factors and the degree of impact varied because of regional heterogeneity, as shown in Table 5. The results demonstrated that PP, P, and RH had a significantly positive impact on ratios in all regions, while SD, T, and WS had a negative effect. Previous studies [9, 74, 79, 80] illustrated that PM concentrations would decrease with the accumulation of precipitation owing to wet scavenging, an essential removal pathway of air pollutants. However, the positive influence of PP on $PM_{2.5}/PM_{10}$ in this study implied that wet scavenging was more effective to coarse particles rather than fine particles. Strong wind was conducive to eliminate air pollutants [78, 81], and the negative correlation with $PM_{2.5}/PM_{10}$ suggested that WS was more beneficial to $PM_{2.5}$ than PM_{10}. Similarly, for sunshine, a signal of great weather with relatively high wind speed or little cloud [82] was favorable for the decrease of the ratio. On the contrary, the decrease in relative humidity was often accompanied by short sunshine time, cloudy, and windless weather [6], which resulted in the increase of pollution. It seemed that RH had a stronger negative influence on PM_{10} than $PM_{2.5}$, so that the positive relationship between $PM_{2.5}/PM_{10}$ and RH was exhibited [34]. Furthermore, high temperature favors the dispersion of secondary pollutants [16], the significant negative effect meant T noticeably affected smaller particles more than larger particles. Pressure was usually inversely proportional to T and positively related to air pollution [14], and the positive correlation implied that P affected $PM_{2.5}$ more strongly than PM_{10}.

In general, the impact of meteorological factors on PM varied with the size of the particle, thus affecting the ratio [44, 83]. Besides, the most important positive factor to the $PM_{2.5}/PM_{10}$ ratio was RH, and the top three negative factors were SD, T, and WS in the majority of economic regions.

3.5.2. Gray Relational Analysis. The correlation coefficients considered the positive/negative impact of meteorological parameters on $PM_{2.5}/PM_{10}$ ratio; however, all coefficients in Table 5 did not exceed 0.5, indicating that meteorology played a complex role, not a simple linear correlation [9]. Moreover, the relationships between various meteorological factors were neglected, and the effect of WD cannot be quantified since WD was a category variable, not continuous variable [80]. Thus, in order to identify the dominant influential factors on $PM_{2.5}/PM_{10}$ ratio, gray relational analysis is further adopted, and the gray relational grades with ranking are shown in Table 6. It is obvious that RH, PP, and P had the most important impact on the $PM_{2.5}/PM_{10}$ ratio throughout the whole country, followed by WD and WS.

It is consistent that RH and P are essential factors to the ratios from both results of Spearman correlation and gray relational analysis. Moreover, wind could have direct effect on the ratios because of the diffusion speed and transformation efficiency of different size of particles.

3.5.3. The Impact of WD on $PM_{2.5}/PM_{10}$ Ratio. There was a weak linear relationship between WD and the $PM_{2.5}/PM_{10}$ ratio since the direction of wind was a categorical variable. Nevertheless, the gray relational analysis and the actual situation showed that WD had a great impact on air

Table 5: Spearman correlation coefficients between $PM_{2.5}/PM_{10}$ and meteorological parameters.

	PP	P	RH	SD	T	WS	WD
NR	0.069**	0.314**	0.392**	−0.450**	−0.475**	−0.169**	−0.059**
NCA	0.196**	0.261**	0.542**	−0.425**	−0.176**	−0.276**	−0.083**
ECA	0.230**	0.098**	0.402**	−0.345**	−0.170**	−0.118**	−0.039*
SCA	0.043*	0.370**	0.125**	−0.308**	−0.464**	−0.157**	−0.183**
MYR	0.173**	0.364**	0.548**	−0.486**	−0.224**	−0.411**	−0.207**
MRYR	0.121**	0.244**	0.191**	−0.312**	−0.429**	−0.071**	0.090**
SC	0.192**	0.289**	0.336**	−0.474**	−0.385**	−0.213**	−0.065**
BNC	0.162**	0.137**	0.566**	−0.382**	−0.457**	−0.409**	−0.107**

Note. * and ** mean correlations were significant at the 0.05 and 0.01 levels (2-tailed).

Table 6: Gray relational grades of meteorological parameters on $PM_{2.5}/PM_{10}$ ratio.

Rank	NR	NCA	ECA	SCA	MYR	MRYR	SC	BNC
1	RH (0.909)	RH (0.949)	RH (0.900)	P (0.903)	RH (0.921)	PP (0.915)	RH (0.917)	RH (0.911)
2	P (0.904)	PP (0.942)	PP (0.898)	PP (0.894)	P (0.908)	P (0.911)	P (0.914)	PP (0.906)
3	PP (0.902)	P (0.935)	P (0.879)	RH (0.886)	PP (0.907)	RH (0.910)	PP (0.912)	P (0.883)
4	WD (0.886)	WD (0.923)	WD (0.873)	WS (0.873)	WD (0.874)	WS (0.906)	WS (0.896)	WD (0.876)
5	WS (0.882)	WS (0.919)	WS (0.871)	WD (0.870)	WS (0.872)	WD (0.903)	WD (0.892)	WS (0.870)
6	SD (0.864)	T (0.916)	T (0.864)	SD (0.861)	T (0.871)	SD (0.883)	SD (0.879)	SD (0.868)
7	T (0.857)	SD (0.909)	SD (0.852)	T (0.856)	SD (0.862)	T (0.879)	T (0.873)	T (0.867)

pollutants. To better understand the effect of WD on the ratio, 16 directions of wind with the ratios were analyzed by wind rose for eight economic regions, as revealed in Figure 5.

The influence of WD on the ratio varied from region to region, mainly determined by topography and distribution of pollution sources [78]. In terms of NR, the smallest ratio occurred in NW, which blew air pollution to the Yellow Sea and Bohai Sea to reduce $PM_{2.5}$, while the largest in SSW which brought $PM_{2.5}$ from the heavily industrial Beijing-Tianjin-Hebei region. Similarly for NCA, the ratios minimized in W to NNW (clockwise) due to the Yellow Sea in the east direction; on the contrary, the ratios maximized in NE and SSW because of the $PM_{2.5}$ sources from Liaoning (iron and steel industry) and Shanxi (coal industry), respectively, as well as Taihang Mountain in the west of the region which prevented the dissipation of pollutants [84]. The East China Sea was adjacent to ECA and SCA so that the wind in ESE to SSE would bring the clean air to the inland [74], while $PM_{2.5}$ was taken by the wind in W to NW from Anhui (coal and steel industry) to ECA and NNW to NE from Jiangxi (metal industry) to SCA. In terms of MYR, the less ratios were in WNW to N which contributed to the east which was relatively flat and open with no high mountains; oppositely, the larger ratios were in ENE to ESE which influenced by NCA and the high Qinling Mountain in the south of the MYR. Besides, there were several mountains in MRYR where it was not favorable for pollutants dispersion, such as Nanling Mountain in the south and Wuyi Mountain in the southeast. Moreover, wind in WNW to NNW from east of SC and south of MYR met the mountains diagonally; hence, the pollution was accumulated, while the wind from SSW to WSW could blow off $PM_{2.5}$ to ECA. It was interesting that there was basin in the north (Sichuan) but plateau with high terrain (Yunan and Guizhou) in the south of the SC. Therefore, SW to W wind from plateau would transfer pollution to MRYR which was a plain in the northeast. In contrast, $PM_{2.5}$ pollution from NW or ENE would accumulate in the basin so that the ratios increased. Furthermore, the slightest pollution observed in BNC was resulted from vast area and less developed industries, and wind in W to NW from higher terrain-brought clean air and diluted $PM_{2.5}$ concentration; on the other hand, wind in ENE would carry $PM_{2.5}$ from MYR, increasing the ratios.

All above phenomena illustrated that $PM_{2.5}$ had longer residence time and larger long-range transport effect than PM_{coarse} [40]. For the stationary sources, the accumulation and dispersion of pollution in the inner region was mainly by the influence of topography as well as the secondary aerosol formation (NO_3^-, SO_4^{2-}, and NH_4^+) by the impact of meteorology [25, 85]. Notably, the effect of $PM_{2.5}$ long-range transport from the neighboring region by wind and its speed would not be neglected, especially for the area where there was great ambient air quality [86].

4. Conclusions

In order to assess the PM pollution with the proportion of $PM_{2.5}$ in PM_{10} throughout China, daily average $PM_{2.5}$ and PM_{10} mass concentrations between 2014 and 2016 were used to analyze the ratios of $PM_{2.5}$ to PM_{10} among eight economic regions. Spatial distribution of $PM_{2.5}/PM_{10}$ ratios showed an increasing trend from northwest to southeast due to economic development and industrial type; adjacent regions had similar characteristics because of the PM spatial transport. Temporally, ratio and its rate of change were high in winter owing to the low temperature and domestic heating, especially for northern China. Besides, the higher ratios were observed on weekends with additional leisure activities and traffic travel in majority of the regions. In terms of ambient air quality, the higher ratio indicated the

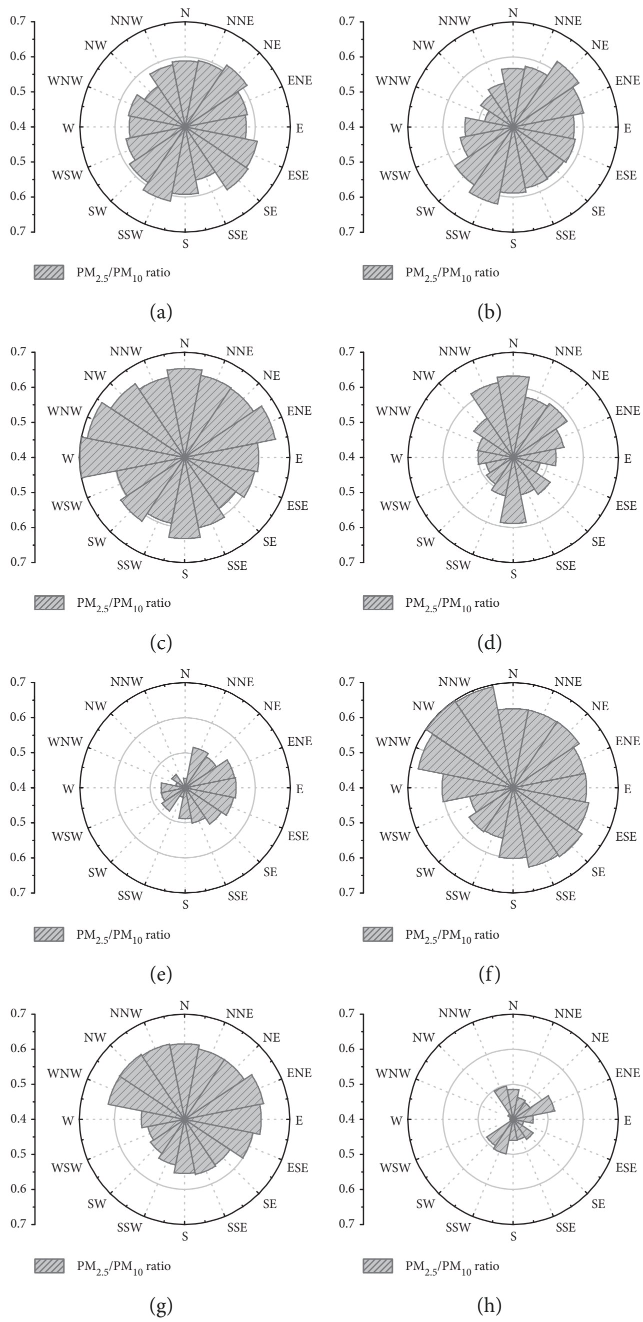


Figure 5: Wind rose of 16 wind directions with the ratios among eight economic regions. (a) NR. (b) NCA. (c) ECA. (d) SCA. (e) MYR. (f) MRYR. (g) SC. (h) BNC.

larger possibility of high AQI, that is, the air pollution will be more severe. Furthermore, meteorological parameters had different impact on the ratios owing to different size of particles, and the phenomena of RH and *P* were the important factors, and both were positively influenced the ratios and illustrated that relative humidity, and precipitation had a stronger effect on PM_{10} than on $PM_{2.5}$, while other parameters had opposite effects.

As small particles are more harmful than large particles, the higher $PM_{2.5}/PM_{10}$ ratios may result in serious air pollution. It is essential to reduce the proportion of $PM_{2.5}$ in PM_{10} rather than decreasing $PM_{2.5}$ simply. Areas with high ratios should concentrate on the reduction by industrial and traffic emissions, which mainly resulted in $PM_{2.5}$ (south of China). In contrast, areas with low ratios could focus on the resuspended dust and sand, mainly lead to PM_{10}, to improve ambient air quality (north of China). Various measures should be conducted simultaneously to cope with multi-sources of the PM emission in winter by considering traffic, domestic heating, meteorology, and spatial location. This study provides a reference for the environmental department and government policy-makers to formulate emission reduction measures by considering the $PM_{2.5}$/PM_{10} ratio and controlling the ratio at a relative low level from the perspective of both anthropogenic sources and meteorological diffusion. The database was from China; however, the method to investigate and evaluate the characteristics of $PM_{2.5}$/PM_{10} ratio is suitable to other places, which have the similar goal to mitigate air pollution and improve ambient air quality. The diurnal variations of $PM_{2.5}$/PM_{10} ratios should be further analyzed to better understand the relationship between air pollution and human daily activities.

Data Availability

The data (daily mean $PM_{2.5}$ and PM_{10} mass concentrations) used to support the findings of this study were obtained from the China National Environmental Monitoring Center (http://datacenter.mep.gov.cn/).

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this article.

Acknowledgments

This study was supported jointly by the Technology Project of Shaanxi Transportation Department (grant number 15-39R) and Special Fund for Basic Scientific Research of Central Colleges of Chang'an University (grant number 300102218409).

Supplementary Materials

Table S1 shows the mean values of $PM_{2.5}$/PM_{10} at different periods of time. (*Supplementary Materials*)

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