

Original Research

Characterization of Size Distribution and Concentration of Atmospheric Particles During Summer in Zhuzhou, China

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Abstract

In order to provide a sound basis for the performance evaluation of a local government's air pollution prevention and abatement measures, the mass concentrations of PM_{2.5} and PM₁₀ and the concentrations of different sized particles were measured in August 2013. Meanwhile, the influencing factors and sources of atmospheric particulate matters were analyzed by combining meteorological factors and gaseous pollutants. The results indicated that PM_{2.5} and PM₁₀ in Zhuzhou were (30.3±12.2) µg·m⁻³ and (56.4±26.6) µg·m⁻³, respectively. The mean values of the particle number concentration in the size range of 0.5~1 µm and 1~2.5 µm on sunny days were (121.2±75.9) cm⁻³ and (6.8±3.7) cm⁻³, respectively, whereas in the rainy days they sharply increased to (868.7±262.1) cm⁻³ and (347.9±238.6) cm⁻³, which accounted for 71.0% and 28.4% of the total number of concentrations, respectively. Backward trajectory analysis demonstrated that the air masses in Zhuzhou city mainly came from three directions: southeast (52.2%), southwest (24.7%), and northeast (23.1%). Although accounting for a minor fraction, the northeast air mass had the largest effect on particle concentrations in Zhuzhou and also carried a lot of gaseous primary pollutants.

Keywords: size distribution, number concentration, mass concentration, hygroscopic growth, backward trajectory, cluster analysis, Zhuzhou City

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Introduction

The mass and number concentration and size distribution of atmospheric particulate matters are an important basis for assessing ambient air quality and provide significant information for understanding the impact of particulate matters on climate, ecology, and human health [1-6]. Based on the differences among the specific concentrations of particulate matter in each city, the effectiveness of the environmental measures being implemented can be evaluated and adjusted scientifically. Particle concentration and size distribution are closely related to its physical and chemical properties, residence time, and transport distance in the atmosphere. The proportion of particles with different sizes was significantly different in the mass concentration and number concentration. At present, it is difficult to provide comprehensive information about particulate matter based on the conventional monitoring of mass concentration that are normally operated by provincial government ambient air quality monitoring stations. Therefore, the study of particle number concentration is of great significance and has received widespread attention [7-9]. Over the past several years, the size distribution and concentration of atmospheric particles have been widely studied in urban or rural areas at home and abroad. Bullard et al. [8] observed the characterization of the particle size distribution and related air quality at a rural location in the continental U.S. (Bondville, Illinois 40.05° N and 88.37° W) for the period July 2013-June 2014. Zhang et al. [10] and Shen et al. [3] studied the particle number size distribution and the new particle formation (NPF) events in Lanzhou and Jiaying, China; Zhang et al. [11] studied the size distribution of particles and their impact on visibility in urban Guangzhou during

spring; Lang et al. [12] and Zhao et al. [13] analyzed the characteristics of the particle size distribution and the influence of meteorological conditions; Li et al. [14] observed the concentration of atmospheric particulate matter in different sizes in Qingdao, China, and discussed the variation characteristics under different weather conditions and air mass sources.

Zhuzhou is a traditional heavy industry city and one of the most important transportation hubs in China. Belonging to the Chang-Zhu-Tan Metropolitan Region (a group of cities of which Changsha, Zhuzhou, and Xiangtan are the cores), Zhuzhou is the focus of inter-regional joint air pollution prevention and control. After implementation of the new national ambient air quality standard (GB3095-2012, China), the main pollutants in Zhuzhou are found to be mainly $PM_{2.5}$ and PM_{10} ; moreover, fine particulate matter has become the main factor affecting air quality. Yang et al. [15] and Zhang et al. [16] studied the particulate matter in autumn and winter in Zhuzhou, but the related research in summer is insufficient, and more importantly the information is rather limited regarding particle size distribution and the underlying variation trends. In addition, most of the current research only focuses on the particle mass concentration and the lack of complementary studies on the number, concentration, and size distribution of particulate matter, especially in central China. Therefore, it is important to measure the concentration and size distribution of particles in summer in Zhuzhou, a typical inland heavy industry city in an established city cluster in central China.

In the present study, based on the data of particle concentration, gaseous pollutant concentrations and local meteorological conditions, a scientific basis is provided for assessing the effectiveness of air pollution prevention

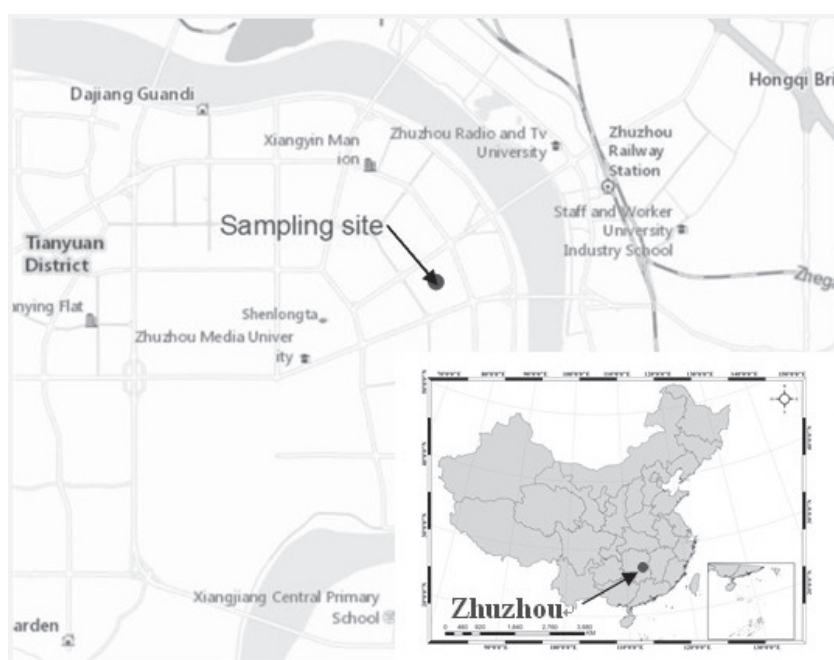


Fig. 1. Location of sampling site in Zhuzhou.

measures for the local government, and supporting data for optimizing inter-regional joint environment policies for air pollution prevention and control and as well for air quality improvement in big city clusters, taking the Chang-Zhu-Tan Metropolitan Area as an example. The objectives of this study were detailed as follows: 1) to investigate atmospheric particle number concentration, mass concentration of PM_{10} and $PM_{2.5}$, and particle size distribution in Zhuzhou in good weather during summer; 2) to assess the influence of different weather conditions on the number concentrations of particulate matter; 3) to compare particle pollution levels of Zhuzhou with other cities in China; and 4) to identify potential pollution sources and transport of atmospheric particulate matter in Zhuzhou using the backward trajectory cluster analysis method.

Material and Methods

Sampling Site and Time

The sampling site in this study was located in Zhuzhou (26°N-28°N, 112.6°E-114°E), the second largest city in Hunan Province, situated in central mainland China with local weather greatly influenced by the prevalence of subtropical humid monsoon. Zhuzhou, belonging to the Chang-Zhu-Tan Metropolitan Region, is a typical heavy industry base and one of the most important transportation hubs in China, and has been undergoing accelerated industrialization and urbanization over the past several decades, which also led to severe air pollution.

The sampling site was on the 6th roof of a building named Tiantai Shanzhuang (113°08-05"E, 27°49-28"N) located in the center of Tianyuan District, Zhuzhou, as illustrated in Fig. 1. There is also one ambient air quality automatic monitoring sub-station on Tiantai Shanzhuang. The instrument (APS3321) was situated approximately 15 m above ground and the air intake was 2 m above the roof. The measurement campaign was conducted during summer in Zhuzhou 8-22 August 2013.

Instruments and Methods

The instrument APS3321 (Aerodynamic Particle Sizer Spectrometer 3321, produced by TSI corporation, U.S.A) was used to continuously observe the particle number concentration and size distribution. The work principle is that particle size information can be obtained by the relationship between the particle aerodynamic size and its velocity after vacuum expansion. The measured particle size ranged from 0.5 to 20.0 μm with a sampling flow rate of 5 $\text{L}\cdot\text{min}^{-1}$ and gas flow rate of 1 $\text{L}\cdot\text{min}^{-1}$ [17-18]. During the experiment, the concentration data of SO_2 , CO , NO_2 , O_3 , PM_{10} , and $PM_{2.5}$ were also collected from the online monitoring instruments at the sampling site. More information associated with the instruments (detection limit, precision, and calibration) and data processing can be found in previous reports [16]. The meteorological

parameters (temperature, relative humidity, wind speed, wind direction, and pressure) were measured hourly by the local meteorological department of Zhuzhou, Hunan Province.

36-h backward trajectories of air masses arriving at the sampling site were calculated using the hybrid single particle lagrangian integrated trajectory (HYSPLIT) model to investigate the influence of different air masses from distant sources on aerosol composition in Zhuzhou [19]. The meteorological data used to run the model are available at global data assimilation system (GDAS). For each sampling day, the model was run with the interval of every hour and the starting height was set as 500 m. The method used in trajectory clustering was based on the GIS-based software TrajStat to determine the main source direction and passage area of air mass [20-21].

Results and Discussion

Concentrations of Particles in Different Sizes

The $PM_{2.5}$ and PM_{10} concentrations in Zhuzhou were $(30.3\pm12.2) \mu\text{g}\cdot\text{m}^{-3}$ and $(56.4\pm26.6) \mu\text{g}\cdot\text{m}^{-3}$, respectively, not exceeding Grade II level of the daily average value recommended by the latest National Ambient Air Quality Standards (GB3095-2012, China – daily average value for PM_{10} Grade I level: $50 \mu\text{g}\cdot\text{m}^{-3}$; Grade II level: $150 \mu\text{g}\cdot\text{m}^{-3}$; daily average value for $PM_{2.5}$ Grade I level: $35 \mu\text{g}\cdot\text{m}^{-3}$; Grade II level: $75 \mu\text{g}\cdot\text{m}^{-3}$). Furthermore, the mean ratios of $PM_{2.5}/PM_{10}$ in our tests were calculated at about 53.7% (Table 1), revealing that the fine particles and coarse particles accounted for similar proportion on mass concentration in Zhuzhou. In addition, the proportion of the particle number concentration of 10~20 μm was very small and therefore could be neglected. In order to conveniently compare the particle number concentration and mass concentration of $PM_{2.5}$ and PM_{10} , the particles were divided into groups of different sizes (0.5~1.0 μm , 1.0~2.5 μm , and 2.5~10 μm , respectively), represented by $PN_{0.5-1}$, $PN_{1-2.5}$, and $PN_{2.5-10}$. During the observation period, the mean value of the particle number concentration of 0.5~10 μm was $(461.9\pm127.8) \text{cm}^{-3}$. $PN_{0.5-1}$, $PN_{1-2.5}$, and $PN_{2.5-10}$ were $(334.3\pm74.2) \text{cm}^{-3}$, $(124.6\pm55.5) \text{cm}^{-3}$, and $(3.0\pm1.8) \text{cm}^{-3}$, accounting for 72.3%, 27.0%, and 0.7% of the total number concentration, respectively (Table 1). The above data demonstrated that the particle number concentration in Zhuzhou was mainly localized in particle size of below 2.5 μm , and the proportion of particles below 1 μm is the largest. In this study, the ratio of fine particle number concentration measured on sunny days was similar to that in other literature [3, 11, 22-24], and the concentration of particles below 1 μm exceeded 90% of the total concentration (Table 2). However, compared with the data of sunny days, the proportion of $PN_{1-2.5}$ in rainy days is significantly increased, which is probably due to the hygroscopic growth of aerosol particles [12, 25].

Table 1. Hourly values of particle number concentration and mass concentration in different sizes.

Particle size range	Mean value	Standard deviation	Maximum value	Minimum value	Ratio
PN _{0.5-1} [*]	334.3	74.2	507.4	244.9	72.3%
PN _{1-2.5} [*]	124.6	55.5	245.5	63.3	27.0%
PN _{2.5-10} [*]	3.0	1.8	7.9	1.0	0.7%
PN _{0.5-10} [*]	461.9	127.8	760.8	309.2	100.0%
PM _{2.5} ^{**}	30.3	12.2	55.5	13.1	53.7%
PM ₁₀ ^{**}	56.4	26.6	121.0	31.3	100.0%

Note: ^{*}Unit (cm⁻³), ^{**} Unit (μg/m³)

Table 2. Hourly values of particle number concentration and mass concentration in different sizes.

Particle size range	Mean value		Ratio	
	Sunny days	Rainy days	Sunny days	Rainy days
PN _{0.5-1} [*]	121.2	868.7	94.20%	71.00%
PN _{1-2.5} [*]	6.8	347.9	5.30%	28.40%
PN _{2.5-10} [*]	0.7	7.8	0.50%	0.60%
PM _{2.5} ^{**}	24.1	48.9	57.10%	48.10%
PM ₁₀ ^{**}	42.2	101.6	100%	100%

Note: ^{*}Unit (cm⁻³), ^{**} Unit (μg/m³)

Compared with data on sunny days, the particle number concentration and mass concentration in different size range obviously varied in rainy days, and the variety of the fine particle number concentration was more significant than that of other sizes (Table 2, Fig. 2). From sunny to rainy days, the number concentration of particles in each size range all showed a tendency to increase.

PN_{0.5-1} increased to (868.7±262.1) cm⁻³ on rainy days, reaching about 7 times (121.2±75.9) cm⁻³ on sunny days, while PN_{1-2.5} increased to (347.9±238.6) cm⁻³ on rainy days, reaching about 51 times (6.8±3.7) cm⁻³ on sunny days. However, the ratio of the particle number concentration in each size range was different. The proportion of PN_{0.5-1} decreased from 94.2% on sunny days to 71.0% on rainy days, while the proportion of PN_{1-2.5} increased from 5.3% to 28.4%. From sunny to rainy days, the concentration of PM₁₀ increased to 101.6 μg/m³, reaching about 2.5 times 42.2 μg/m³ on sunny days, and PM_{2.5} increased to 48.9 μg/m³, reaching about 2 times 24.1 μg/m³ on sunny days. In addition, the PM_{2.5}/PM₁₀ ratio decreased from 57.1% to 48.1%. The significant difference of particle size distribution between the sunny and rainy days was mainly related to the variety of meteorological factors that affected the concentration of atmospheric particulate matter by the process of accumulation, diffusion, and removal of particulate matter, as well as atmospheric chemistry processes [26-28]. The air mass of the south direction 19-21 August caused rainfall in Zhuzhou, resulting in an increase in relative humidity and decrease in temperature. When the relative humidity does not exceed 60-70%, the increase of the particle concentration

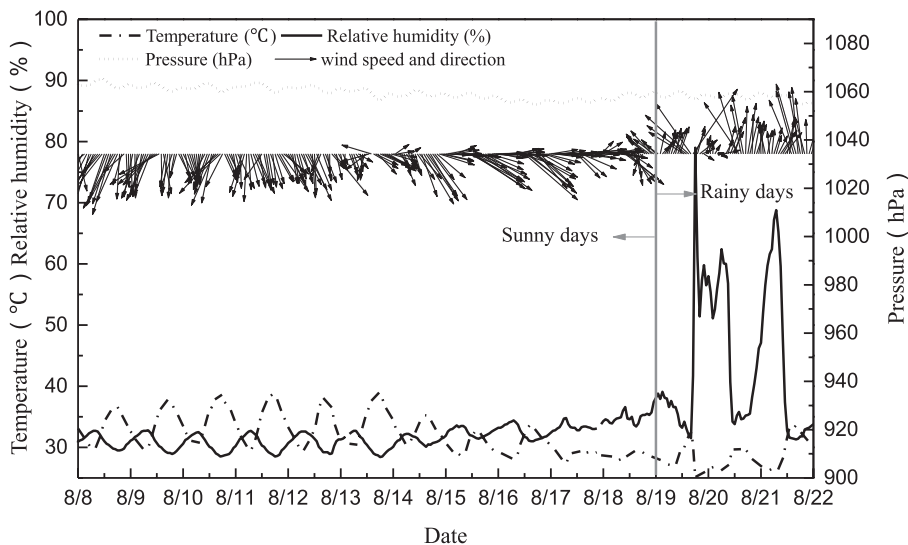


Fig. 2. Variation of meteorological factors during sampling period.

Table 3. Comparison of particle number concentration in different regions.

Region (sampling time) instrument	Particle size range			References
	PN _{0.5-1}	PN _{1-2.5}	PN _{2.5-10}	
Nanjing (July-Dec. 2006) WPS	666.0	7.0	N/A	(Qian et al., 2008) [39]
Guangzhou (March-May 2008) APS	127.7	10.3	1.0	(Zhang et al., 2010) [11]
Shijiazhuang (May 2015) WPS	535.0	19.0	0.7	(Zhuo et al., 2011) [40]
Lanzhou (Aug.-Oct. 2010) APS	98.1	8.5	1.5	(Zhao et al., 2012) [13]
Mount Huang (June-Aug. 2011) APS	19.6	1.0	0.1	(Wang et al., 2014) [24]
Zhuzhou (Aug. 2013) APS	334.3	124.6	3.0	This study

Note: Unit (cm⁻³)

Table 4. Comparison of particle mass concentration in different regions.

Region (Sampling time)	Particle size range			References
	PM _{2.5}	PM ₁₀	PM _{2.5} /PM ₁₀	
Beijing (July 2013)	146.0	214.0	68.2%	(Dao et al., 2015) [32]
Guangdong (Aug. 2012)	38.1	57.8	66.0%	(Yin et al., 2017) [41]
Nanjing (July-Sept. 2014)	34.6	70.6	49.0%	(Chen et al., 2016) [26]
Hefei (Aug. 2014)	70.4	110.8	63.5%	(Chen et al., 2016) [34]
Wuhan (June-Aug. 2013)	52.0	59.0	88.1%	(Zhang et al., 2015) [35]
Changsha (Aug. 2014)	52.5	84.0	62.5%	(Xie et al., 2016) [37]
Xiangtan (Aug. 2014)	49.5	70.0	70.7%	(Xie et al., 2016) [37]
Nanling (June 2013)	5.2	9.6	54.2%	(Zhang et al., 2015) [33]
Haikou (April 2012)	20.7	35.1	59.0%	(Fang et al., 2017) [20]
Zhengzhou (July 2015)	102	164	62.2%	(Wang et al., 2017) [22]
Zhuzhou (Aug. 2013)	30.3	56.4	53.7%	This study

Note: Unit (μg/m³)

is caused by hygroscopic growth and secondary reactions [12, 29-30]. Chemical compositions of particles in different sizes can cause a difference of hygroscopicity, especially strong hygroscopic substance such as water-soluble ions accounting for a larger proportion in fine particles, particularly at the size of 1-2.5 μm, which could explain the sharp increase of particle concentrations on rainy days in Zhuzhou [31].

Comparison with Other Cities

The level of particle concentration in Zhuzhou was compared with megacities in the Beijing-Tianjin-Hebei Metropolitan Region and the Pearl River Delta Metropolitan Region, which are the most developed regions in China, and with background area and other important industrial cities (Table 3, Table 4). The concentration of particulate matter in Beijing-Tianjin-Hebei Region (for example in Beijing) was 4-5 times that in Zhuzhou City, and the proportion of PM_{2.5} (68.2%) was much higher than that in Zhuzhou [32]. The level of

particle concentration in the Pearl River Delta (belonging to Guangdong Province) was not high, and the air quality was relatively better than Zhuzhou. For example, in Guangzhou (capital city of Guangdong Province), PN_{0.5-1} and PN_{1-2.5} were 127.7 cm⁻³ and 10.3 cm⁻³, and the particle mass concentration was similar to that in Zhuzhou [11]. Compared with Mount Huang, Nanling, Haikou, and other background city sites, the particle concentration in Zhuzhou is slightly higher than these cities [20, 24, 33].

Zhuzhou is an industrial city similar to other industrial cities (Nanjing, Wuhan, Hefei and Zhengzhou) in terms of industrial structure, among which particle concentration and particle size distribution of Zhuzhou is most similar to that of Nanjing [22, 26, 34-35]. Within the Chang-Zhu-Tan Metropolitan Region, the concentration of particulate matter in Zhuzhou is the same as that in Changsha and Xiangtan [36-37] (these 3 cities are contiguous and all belong to the same metropolitan region), but the PM_{2.5}/PM₁₀ ratios are significantly different. Because the particle concentration and size distribution changes a lot in different cities within one whole region,

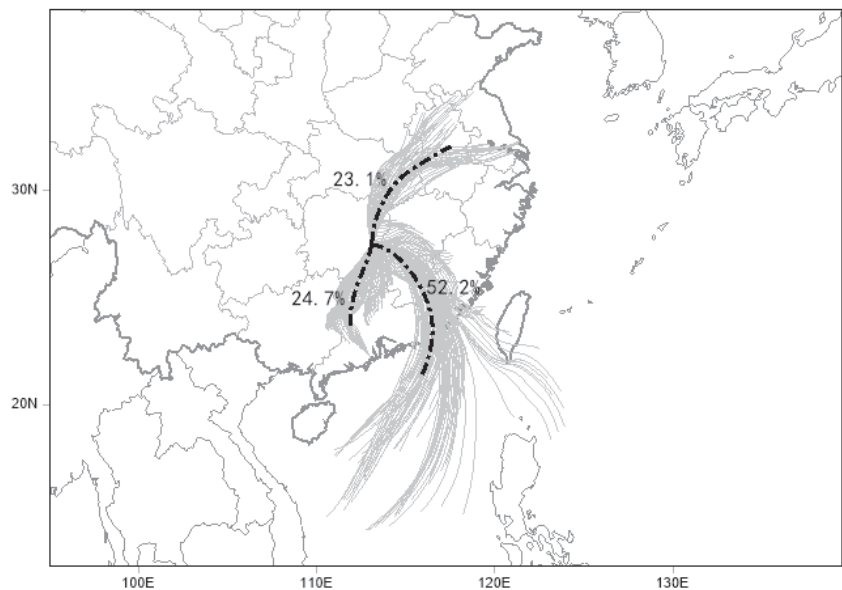


Fig. 3. Backward trajectory cluster analysis result of air mass sources in Zhuzhou.

the inter-regional joint air pollution prevention and control measures should take full account of their different levels of particle concentration in adjacent cities. According to the different sources and proportions of particulate matter in different size ranges, the reasonable formulation and adjustment of environmental measures will play an important role in improving air quality in local city and adjacent regions [38]. The findings presented here can provide an improved scientific basis on particulate matter for the local government of Zhuzhou to develop further environmental regulations for air quality improvement.

Backward Trajectory Clustering Analysis

Backward trajectory analysis showed that the air masses in Zhuzhou mainly came from 3 directions (southeast, southwest, and northeast), going through different areas. The air mass from southeast of Zhuzhou had the highest proportion (52.2%), the second was the air mass from the southwest passing the center of Guangdong Province (24.7%), and the last was the air mass from the northeast (23.1%; Fig. 3). The clustering analysis of the air pollutants (CO, NO₂, SO₂, O₃, PM_{2.5}, and PM₁₀) and the mean values of PN_{0.5-1}, PN_{1-2.5}, and PN_{2.5-10} (Tables 5-6) showed that PN_{0.5-1}, PN_{1-2.5}, and PN_{2.5-10} from the northeast air mass were maximum among 3 cluster directions, namely 868.7 cm³, 347.9 cm³ and 7.8 cm³, respectively; the concentrations of CO, NO₂, SO₂, PM_{2.5}, and PM₁₀ from the northeast air mass also reached the highest, which are 1.0 μg·m⁻³, 36.5 μg·m⁻³, 36.7 μg·m⁻³, 49.0 μg·m⁻³, and 101.6 μg·m⁻³, respectively. This revealed that although accounting for the smallest proportion, the air mass from the northeast had the largest effect on particle concentration and also carried a lot of gaseous pollutants. The air masses from the southwest and southeast show similarities on the concentration of particle and air

pollutants except that the PN_{0.5-1} from the southeast is about twice that from the southwest. In conclusion, compared with the particulate mass concentration and number concentration in Zhuzhou, the air masses from the southwest and southeast were relatively clean, while the “dirty” air mass from the northeast direction seriously affected the particle concentration in Zhuzhou. The air mass from the northeast going through Nanjing, Hefei, and Wuhan in sequence and other important cities eventually arrived in Zhuzhou, and the particulate matter levels of these cities were much higher than

Table 5. Statistics of particle average number concentration in different sizes under different directions.

Direction	PN _{0.5-1}	PN _{1-2.5}	PN _{2.5-10}
	Mean value ± standard deviation	Mean value ± standard deviation	Mean value ± standard deviation
Southwest	75.2±31.2	7.0±2.5	0.7±0.2
Northeast	868.7±264.8	347.9±241.2	7.8±6.7
Southeast	135.4±80.4	6.8±4.1	0.7±0.3

Note: Unit (cm³)

Table 6. Statistics of atmospheric pollutants’ average concentrations under different directions.

Direction	PM _{2.5}	PM ₁₀	CO	NO ₂	SO ₂	O ₃
Southwest	23.6	35.1	0.8*10 ³	22.6	9.3	77.2
Northeast	49.0	101.6	1.0*10 ³	36.5	36.7	76.6
Southeast	24.7	46.9	0.8*10 ³	22.6	14.6	67.4

Note: Unit (μg/m³)

Zhuzhou (Tables 3-4) [26, 34-35]. This was consistent with the results of the backward trajectory clustering in northeastern air mass, which contributed to particle pollution in Zhuzhou. Among these cities, Hefei had the largest contribution to atmospheric particles in the northeast air mass, and $PM_{2.5}$ and PM_{10} are $70.4 \mu\text{g}/\text{m}^3$ and $110.8 \mu\text{g}/\text{m}^3$, respectively [34].

Conclusions

- 1) In Zhuzhou, the concentration of particulate matter smaller than $2.5 \mu\text{m}$ accounted for 99.3% of the total concentration during summer, and the proportion of the fine particle with the size range below $1 \mu\text{m}$ accounted for 72.3%, and the $PM_{2.5}/PM_{10}$ ratio was 53.7%, showing that Zhuzhou was mainly affected by fine particles.
- 2) Both the number concentration and mass concentration of particulate matter were affected significantly by rain. Because of the increase of relative humidity and the difference of hygroscopicity, the particle number concentration varied distinctly and increased more significantly in fine particles, especially at the size of $1\text{-}2.5 \mu\text{m}$.
- 3) Backward trajectory analysis suggests that the air mass with the highest proportion was from the southeast direction of Zhuzhou City (52.2%), followed by the southwest direction (24.7%) passing the center of Guangdong Province and then the northeast direction (23.1%). However, the air mass from the northeast direction made the greatest contribution to the particle concentration and also carried a lot of gaseous pollutants. The particle concentration in Zhuzhou was significantly affected by higher levels of particle concentration in the cities that the northeast air mass passed through; therefore, it is essential to strengthen the inter-regional joint air pollution prevention and control in the Chang-Zhu-Tan Metropolitan Region.

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