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Original Research

Spatial and Temporal Distribution Characteristics of $PM_{2.5}$ and PM_{10} in the Urban Agglomeration of China's Yangtze River Delta, China

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Abstract

Fine particles ($PM_{2.5}$) and particulate matter (PM_{10}) monitoring data from 2015 to 2016 in 18 major cities in the Yangtze River Delta were analyzed to determine the temporal and spatial distribution characteristics of $PM_{2.5}$ and PM_{10} pollution in the Yangtze River Delta urban agglomeration, China. The results showed that the cities with the most serious $PM_{2.5}$ and PM_{10} pollution were mainly distributed along the Yangtze River in Jiangsu Province, while the lowest concentrations of $PM_{2.5}$ and PM_{10} were measured in Zhoushan of Zhejiang Province. The $PM_{2.5}$ and PM_{10} concentrations in 2016 were lower than those in 2015. In 2015 and 2016, the $PM_{2.5}$ and PM_{10} concentrations were the highest in winter, followed by spring, and the concentrations were the lowest in summer. In 2015 and 2016 the average annual concentrations of $PM_{2.5}$ and PM_{10} were lower than the ambient air quality standard of China (Grade II; $PM_{2.5}$: 35 μg/g and PM_{10}: 70 μg/g) in Zhoushan only, but were higher than the particulate pollutant emission limits of the United States and the European Union. The ratios of $PM_{2.5}/PM_{10}$ in the urban agglomeration were greater than 0.5, indicating that the pollution in the atmospheric particulate matter in the Yangtze River Delta was generally less than 2.5 μm.

Keywords: Yangtze River Delta urban agglomeration, fine particles ($PM_{2.5}$), respirable particulate matter (PM_{10}), pollution level

Introduction

As the effect of atmospheric aerosols on the climate and environment increases, research on atmospheric particulate matter has become an important scientific issue [1, 2]. Fine particulate matter ($PM_{2.5}$) and particulate matter (PM_{10}) have a considerable impact on ecology and human health due to their special physical and chemical properties and are considered the most important air pollutants [3]. The Yangtze River Delta urban agglomeration is an important intersection of China's "Silk Road Economic Belt" and the "21st-Century Maritime Silk Road" (the Belt and Road) strategies, and the Yangtze River economic belt is one of

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China's most economically developed areas [4-5]. With the rapid development of urbanization and the economy, the problem of air pollution in this region is becoming more and more serious [6].

In recent years, research on $PM_{2.5}$ and PM_{10} has been carried out in the Yangtze River Delta region [6-9] Wind direction, wind speed, and air pressure each have a significant effect on the mass concentration of PM_{10} [10]. In addition, regional transmission can affect the concentration of atmospheric particulate matter in Shanghai [10]. Factor analysis and receptor model results show that $PM_{2.5}$ in Nanjing mainly comes from industrial emissions, automobile exhaust, and dust [11]. The spatial distribution and concentration of $PM_{2.5}$ in Hangzhou was correlated with the pattern of urbanization [12]. From a comparison of the $PM_{2.5}$ concentration on weekends and working days, no "weekend effect" was observed in Wuxi [13]. These earlier studies of the Yangtze River Delta region focused on the pollution of atmospheric particulate matter in a single city, while the study of entire urban agglomerations has rarely been reported. In this study, 18 major cities in the Yangtze River Delta urban agglomeration were taken as the research object, and the pollution levels and temporal and spatial patterns of $PM_{2.5}$ and PM_{10} were discussed by using air pollutant monitoring data of 2015 and 2016. The aim of this study was to provide a scientific basis for the monitoring and control of atmospheric particulate pollution in the Yangtze River Delta region.

Materials and Methods

Study Area

The Yangtze River Delta urban agglomeration includes three provinces (Zhejiang, Jiangsu, and Anhui), one municipality (Shanghai; Fig. 1), 26 cities, and 150 million people. This region accounts for approximately 2.2% of the land area of China and is one of the most open, active, and innovative areas in China. In 2016 an Environmental Bulletin issued by the Chinese Ministry of Environmental Protection showed that the percentage of days of poor air quality in the urban agglomerations of the Yangtze River Delta was 23.9%; during these days, the primary pollutant $PM_{2.5}$ accounted for 55.3% of air pollutants. The wind direction in spring and summer is dominated by easterlies, while the autumn and winter are dominated by northerlies.

Data Sources and Processing

The air pollution monitoring data ($PM_{2.5}$, PM_{10}, SO_2, and NO_2) were derived from a monthly average concentration issued by the China Environmental Protection Department (www.mep.gov.cn). In this study, monitoring data from 18 main cities (including Shanghai; Nanjing, Suzhou, Wuxi, Changzhou, Zhenjiang, Yangzhou, Nantong, Taizhou(J), and Yancheng in Jiangsu Province; Hangzhou, Jiaxing, Ningbo, Shaoxing,

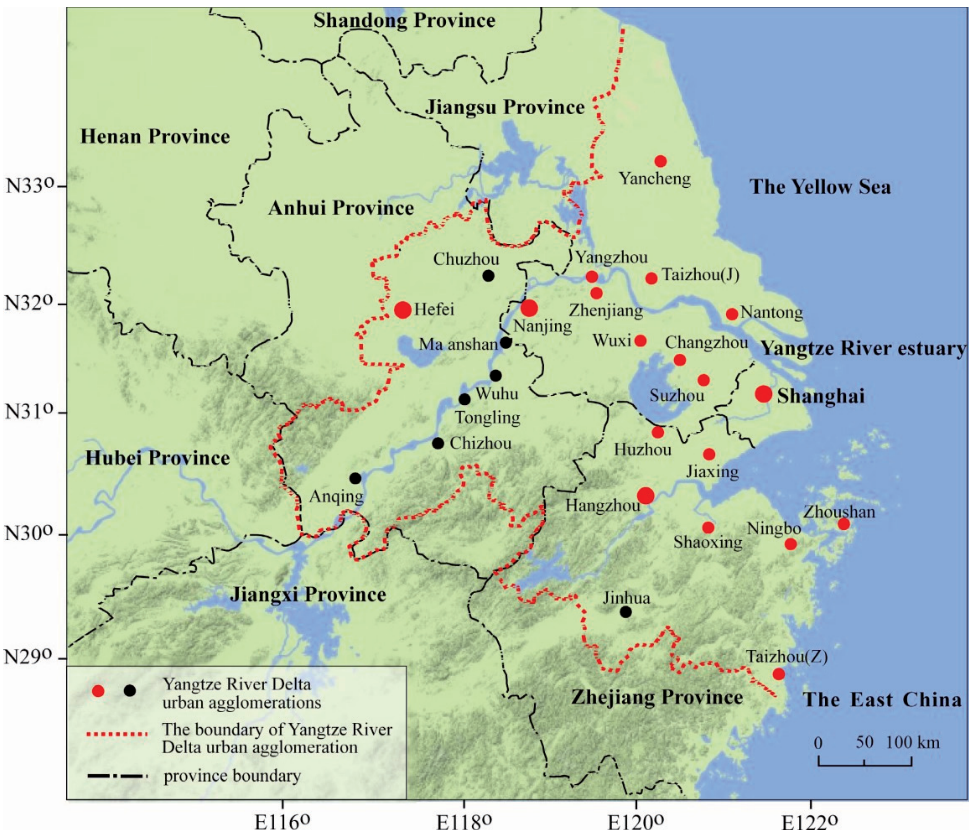

Fig. 1. Distribution of the urban agglomerations in the Yangtze River Delta.

Huzhou, Zhoushan and Taizhou(Z) in Zhejiang Province; and Hefei in Anhui Province (Fig. 1) in the Yangtze River Delta region were analyzed. Freehand and Origin software were used to perform the graphical mapping and analysis of the monitoring data.

Results and Discussion

Interannual Variations

The spatial and temporal distribution characteristics of the concentration of $PM_{2.5}$ and PM_{10} in the Yangtze River Delta urban agglomerations in 2015-2016 are shown in Fig. 2. Generally, the cities with a high concentration of $PM_{2.5}$ and PM_{10} were distributed in Nanjing, Changzhou, Yangzhou, and Taizhou(J) in Jiangsu Province, while the cities with a low concentration were mainly distributed in Zhejiang Province. The concentrations of $PM_{2.5}$ and PM_{10} in 2016 showed a decreasing trend compared with that in 2015, indicating that the atmospheric environment of the Yangtze River Delta region has improved. In 2016 the Yangtze River Delta continued to optimize its industrial structure, deepening energy conservation and emission reduction, which may be the main reason for the reduction of atmospheric pollutants. In 2015, $PM_{2.5}$ and PM_{10} showed high concentrations in the north and low concentrations in the south, with a gradual change in the concentrations between these areas. The highest concentration of $PM_{2.5}$ appeared in Hefei (65.7 μg/m^3), Anhui Province, and the highest concentration of PM_{10} appeared in Taizhou(J) (101.8 μg/m^3), Jiangsu Province. The lowest concentration of $PM_{2.5}$ and PM_{10} appeared in Zhoushan (29.4 μg/m^3 and 45.8 μg/m^3), Zhejiang Province. The high concentrations of $PM_{2.5}$ and PM_{10} are mainly distributed in the cities along the Yangtze River in Jiangsu Province, and the concentrations were 55~61.2 μg/m^3 and 80.2~101.8 μg/m^3, respectively, with averages of 58.6 μg/m^3 and 93 μg/m^3; these averages were higher than those in Shanghai (53 μg/m^3 and 69 μg/m^3) and the 8 cities within Zhejiang Province (47.3 μg/m^3 and 70.9 μg/m^3).

Jiangsu is one of the most developed provinces in China. The primary heavy industrial activities and ports are located on both sides of the Yangtze River. High industrial waste gas emissions are the major contributor to the high concentration of atmospheric particulates [14, 15]. However, the primary heavy industrial activities and ports in Zhejiang Province and Shanghai are located in coastal areas, and sea breeze may dilute the atmospheric

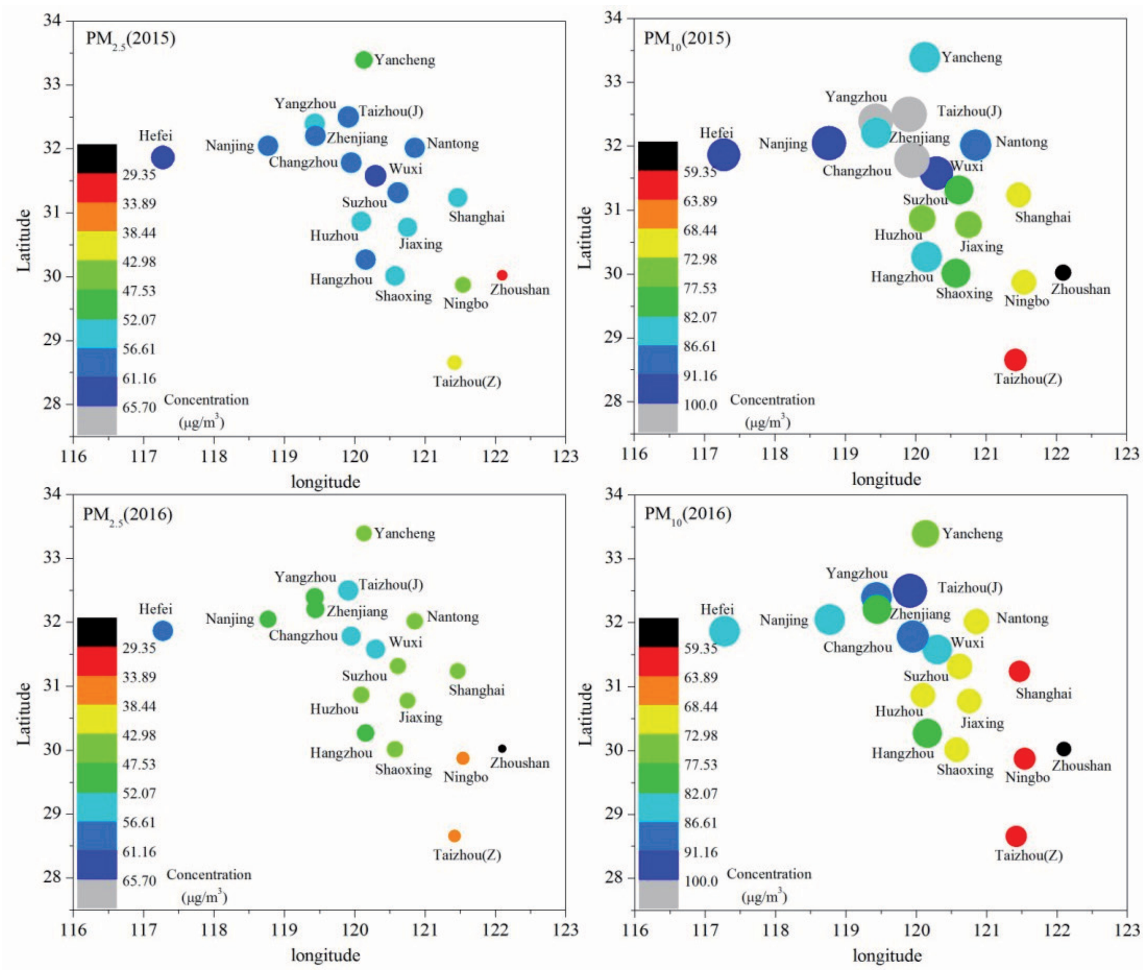

Fig. 2. Interannual variation in $PM_{2.5}$ and PM_{10} in the Yangtze River Delta urban agglomeration.

pollutants. As a result, the concentrations of $PM_{2.5}$ and PM_{10} in Zhejiang Province and Shanghai are lower than those in Jiangsu Province. The spatial distribution characteristics of atmospheric particulate concentrations in 2016 were nearly identical to those in 2015. Similarly, the concentration of $PM_{2.5}$ in Hefei was the highest (57.5 μg/m³); the highest concentration of PM_{10} appeared in Taizhou(J) (94.9 μg/m³), and the lowest concentrations of $PM_{2.5}$ and PM_{10} were found in Zhoushan (25.3 μg/m³ and 42.6 μg/m³). The concentrations of $PM_{2.5}$ and PM_{10} in cities along the Yangtze River in Jiangsu Province were 46.1~54.9 μg/m³ and 70.4~94.9 μg/m³, with an average of 50.3 μg/m³ and 82.8 μg/m³ – clearly lower than those in 2015. In addition, Shanghai (44.7 μg/m³ and 60.1 μg/m³) and the 8 cities in Zhejiang (42.2 μg/m³ and 67.2 μg/m³) were also lower in 2016 than in 2015.

Seasonal Variations

In 2015 $PM_{2.5}$ concentrations in the Yangtze River Delta urban agglomeration were highest in winter (74.6 μg/m³), second highest in spring (57.4 μg/m³), and the lowest in autumn (40.7 μg/m³). The average annual concentration of $PM_{2.5}$ in Zhoushan was lower than the ambient air quality standard of China (Grade II ($PM_{2.5}$: 35 μg/m³), GB3095-2012, hereinafter referred to as China's Grade II standard; Fig. 3), but it was higher than the annual average limit for the air quality standards of the European Union (25 μg/m³) and the United States (15 μg/m³). Other cities were higher than China's Grade II standard as well as the emissions limits of the European Union and the United States. The concentration of PM_{10} was the highest in winter

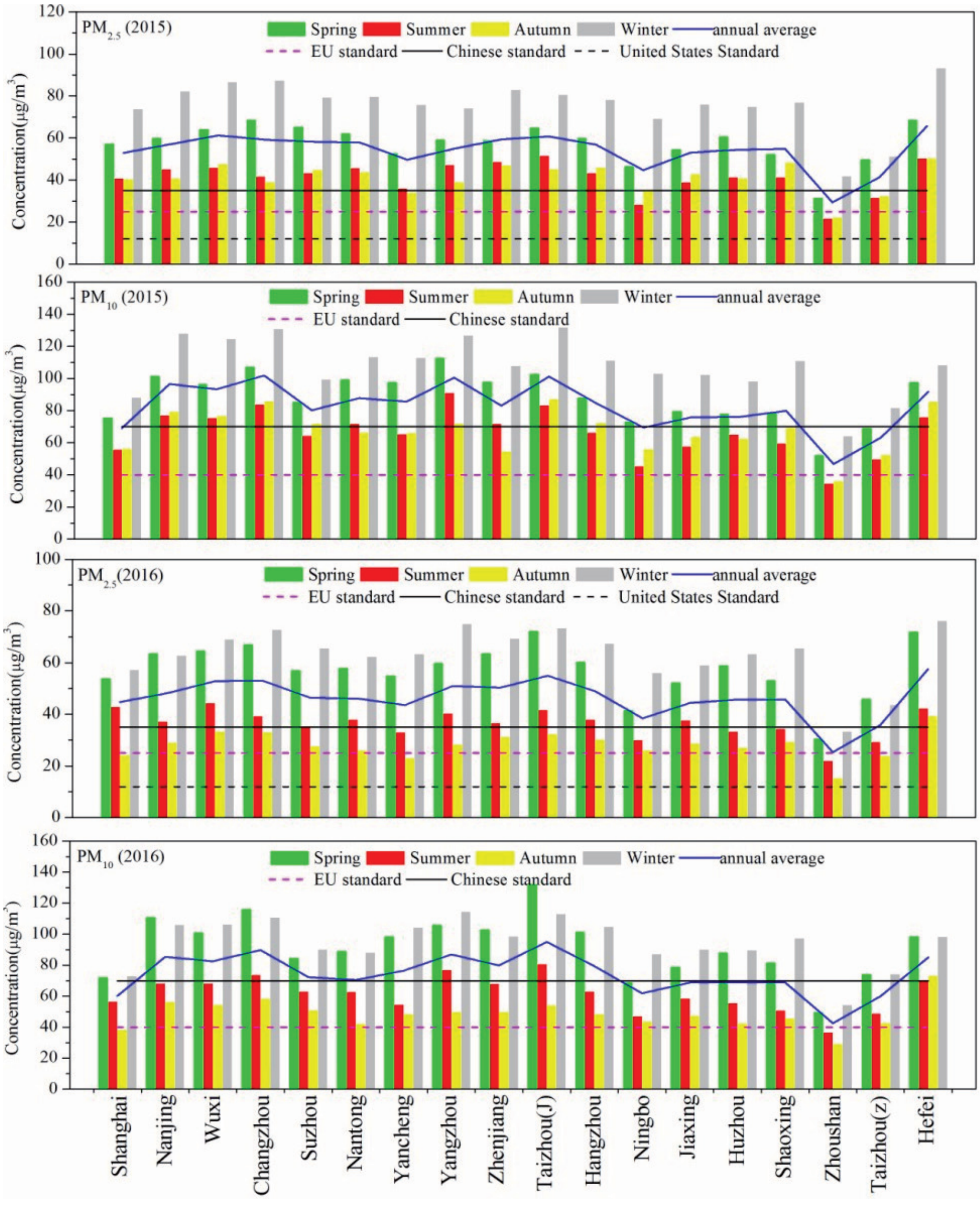

Fig. 3. Seasonal variations in $PM_{2.5}$ and PM_{10} in the Yangtze River Delta urban agglomeration.

(106.7 μg/m³), the second highest in spring (88.2 μg/m³), and lowest in summer (66.2 μg/m³), indicating that the grain size of the atmospheric particles gradually increases from summer to autumn. The average annual concentrations of PM_{10} in Taizhou(Z), Ningbo, Zhoushan, and Shanghai were lower than China's Grade II standard. There was no uniform heating in winter in the Yangtze River Delta area, and the seasonal variation in pollutant emissions was small; the change in meteorological conditions has a considerable influence on the concentration of atmospheric particulates. The atmospheric stability in winter and spring was not conducive to the spread of atmospheric pollutants [16]. In addition, the northwest wind is prevalent in the spring and winter seasons, and numerous particulate pollutants are transported from the upper wind direction to the Yangtze River Delta region, which greatly impacts air quality. Additionally, in summer and autumn the prevailing southeast wind has a significant effect on the dilution of atmospheric pollutants [8].

In 2016 the $PM_{2.5}$ and PM_{10} concentrations were the lowest in autumn (28.1 μg/m³), and the highest concentration of $PM_{2.5}$ occurred in winter (62.2 μg/m³), followed by spring (56.8 μg/m³). The concentration of $PM_{2.5}$ in Zhoushan was lower than China's Grade II standard (35 μg/m³). The highest concentration of PM_{10} was in winter (93.8 μg/m³), the second highest was in spring (91.7 μg/m³), and the lowest was in autumn (48.6 μg/m³). The concentration of PM_{10} in Shanghai, Ningbo, Zhoushan, Wenzhou, Jiaxing, Huzhou, Shaoxing, and Taizhou(Z) were lower than China's Grade II standard.

Specifically, in 2015 the $PM_{2.5}$ concentrations in Ningbo, Zhoushan, and Taizhou(Z) in spring and summer, and those of Yancheng in autumn, were lower than China's Grade II standard. However, PM_{10} concentration of 18 cities in winter were considerably higher than the Grade II standard. In addition to Zhoushan, other cities had spring PM_{10} concentrations that exceeded the Grade II standard; the autumn PM_{10} concentrations in Shanghai, Nanjing, Yancheng, Zhenjiang, Ningbo, Jiaxing, Huzhou, Zhoushan, and Taizhou(Z) were lower than the Grade II standards. Although the summer PM_{10} concentrations were lower than those of the other three seasons, the summer PM_{10} concentrations in Nanjing, Wuxi, Changzhou, Nantong, Yancheng, Zhenjiang, Taizhou(J), and Hefei exceeded the Grade-II standard. In 2016 all cities had autumn $PM_{2.5}$ and PM_{10} concentrations lower than the Grade-II standard. In addition to Changzhou, Yangzhou, and Taizhou(J), other cities had summer PM_{10} concentrations considerably lower than the Grade-II standard. In addition, the summer and autumn PM_{10} concentrations in Zhoushan were lower than the European Union emissions limit (40 μg/m³).

$PM_{2.5}$ and PM_{10} Analysis of Hu-Ning-Hang

Hu-Ning-Hang (where Hu is an abbreviation for Shanghai, Ning is an abbreviation for Nanjing, and Hang is an abbreviation for Hangzhou) are the core cities of the

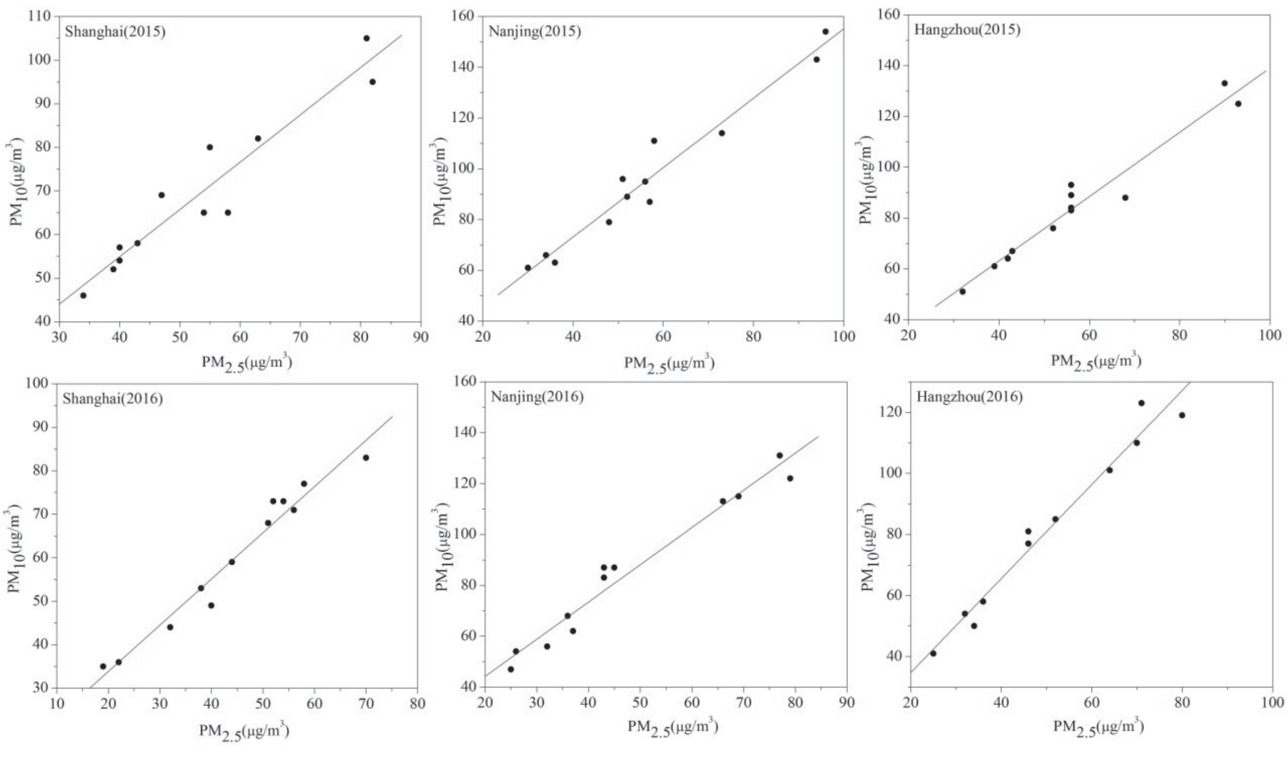

Fig. 4. Correlation analysis between PM2.5 and PM10 concentrations in Shanghai, Nanjing, and Hangzhou.

Table 1. Correlation analysis of atmospheric pollutants in Shanghai, Nanjing, and Hangzhou (correlation coefficient (*r*)).

Time	City		$PM_{2.5}$	PM_{10}	SO_2	NO_2
2015	Shanghai	$PM_{2.5}$		0.955	0.951	0.913
		PM_{10}			0.958	0.875
	Nanjing	$PM_{2.5}$		0.978	0.928	0.804
		PM_{10}			0.97	0.872
	Hangzhou	$PM_{2.5}$		0.98	0.817	0.849
		PM_{10}			0.818	0.884
2016	Shanghai	$PM_{2.5}$		0.981	0.778	0.812
		PM_{10}			0.793	0.814
	Nanjing	$PM_{2.5}$		0.981	0.888	0.851
		PM_{10}			0.84	0.849
	Hangzhou	$PM_{2.5}$		0.99	0.926	0.864
		PM_{10}			0.909	0.879

Yangtze River Delta urban agglomeration. In this study, the $PM_{2.5}$ and PM_{10} concentrations of Hu-Ning-Hang were further studied.

The linear regression analysis results are shown in Fig. 4. In 2015 the regression equations of $PM_{2.5}$ and PM_{10} in Hu-Ning-Hang were y = 1.086x + 11.434, y = 1.367x + 18.460 and y = 1.265x + 12.747, respectively; the correlation coefficients (*r*) are 0.955, 0.978, and 0.975; and the probability values (P) were less than 0.0001. However, in 2016 the regression equations of Hu-Ning-Hang were y = 1.063x + 12.596, y = 1.463x + 14.954, and y = 1.540x + 3.949, respectively; the correlation coefficients (*r)* were 0.981, 0.9881, and 0.985, respectively; the probability values (P) were less than 0.0001. These results show that there were significant linear correlations between $PM_{2.5}$ and PM_{10} concentrations in Hu-Ning-Hang in 2015 and 2016, suggesting that the source of the particulate pollutants was consistent and that the source of the atmospheric particles was relatively stable. However, the results varied between 2015 and 2016, indicating that the pollution of the urban atmospheric particulate matter may be affected by other factors [8].

The correlation of typical air pollutants can be used to determine whether the pollution source was the same. An earlier study showed that SO_2 was mostly derived from coal combustion, which is one of the main sources of $PM_{2.5}$ and PM_{10}, and NO_2 was derived from exhaust emissions from motor vehicles, which contributes greatly to $PM_{2.5}$ concentrations [17-18]. The $PM_{2.5}$ and PM_{10} correlation coefficients with SO_2 between Shanghai and Nanjing in 2015 were larger than those with NO_2, indicating that the atmospheric particulate matter is a substantial pollution source of SO_2, suggesting the the contribution rate of coal combustion was greater than that of automobile exhaust, which is the main source of atmospheric particulate pollutants (Table 1). The $PM_{2.5}$ and PM_{10} correlation coefficients with SO_2 were less than those with NO_2 in Hangzhou, indicating that the vehicle exhaust may be the main source of SO_2. The proportion of heavy industrial activity is smaller in Hangzhou than in either Shanghai or Nanjing. In 2016 the correlation coefficient between the atmospheric particulates and NO_2 in Shanghai was higher than that of SO_2, suggesting that the main pollutant emission sources vary greatly. The change in sources may be due to Shanghai's energy and industry remediation and the increasing industrial emission control efforts. The correlation coefficient between $PM_{2.5}$ and SO_2 in Nanjing was less than that between the $PM_{2.5}$ and NO_2, and the correlation coefficient between PM_{10} and SO_2 was greater than that between PM_{10} and NO_2. Therefore, there were differences in the contributions of the emission sources for different particle sizes. Automobile exhaust may contribute more $PM_{2.5}$, while industrial coal combustion produced more PM_{10}. The Hangzhou atmospheric particulate emissions were mainly controlled by automobile exhaust [17].

$PM_{2.5}/PM_{10}$ Mass Concentration Ratio of Hu-Ning-Hang

Studies have shown that in addition to direct emissions from pollution sources, secondary particles produced by gas-particle conversion and other chemical processes are also one of the main sources of $PM_{2.5}$ [6]. In contrast, the coarse particles (PM_{10}) were derived from industrial emissions and natural processes (dust, construction, etc.). Therefore, the $PM_{2.5}/PM_{10}$ concentration ratio can reflect the type of air pollution and possible sources of pollution [19]. Table 2 shows the Hu-Ning-Hang $PM_{2.5}/PM_{10}$ annual average values in 2015 and 2016. $PM_{2.5}/PM_{10}$ ratio was greater than 0.5, indicating that the air pollution was mainly fine particles. The smaller ratios indicate that the particulate matter is derived from traditional soot, and the larger ratios represent a

Table 2. Interannual variation in the concentrations and ratios of $PM_{2.5}$ and PM_{10} (μg/m^3).

City	2015			2016		
	$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$	$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$
Shanghai	53	69	0.77	45	60	0.77
Nanjing	57	97	0.59	48	85	0.56
Hangzhou	57	85	0.67	49	79	0.62

Table 3. Seasonal variation in concentrations and ratios of $PM_{2.5}$ and PM_{10} (μg/m³).

Time	City	Spring			Summer			Autumn			Winter		
		$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$	$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$	$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$	$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$
2015	Shanghai	50	67	0.75	41	55	0.75	46	60	0.77	75	94	0.80
	Nanjing	53	93	0.50	39	69	0.56	48	86	0.56	88	137	0.64
	Hangzhou	56	87	0.64	38	59	0.64	50	77	0.65	84	115	0.73
2016	Shanghai	53	71	0.75	32	46	0.70	33	46	0.72	61	78	0.78
	Nanjing	55	102	0.54	32	57	0.56	35	66	0.53	71	117	0.61
	Hangzhou	54	94	0.57	31	51	0.61	39	63	0.62	71	110	0.65

compound source. The ratios of $PM_{2.5}/PM_{10}$ in Hu-Ning-Hang in 2015 were 0.77, 0.59, and 0.67, respectively. Shanghai had a higher $PM_{2.5}/PM_{10}$ ratio than those of Nanjing and Hangzhou. By the end of 2015, the number of cars were Shanghai>Hangzhou>Nanjing, indicating that car exhaust emissions were the main source of $PM_{2.5}$ in Shanghai. In addition, meteorological conditions and human activities have a considerable impact on atmospheric particulate matter [6]. In 2016 the ratios of $PM_{2.5}/PM_{10}$ in Hu-Ning-Hang were 0.77, 0.56, and 0.62, respectively. The Shanghai $PM_{2.5}/PM_{10}$ ratio did not change, while those in Nanjing and Hangzhou were slightly lower than their 2015 ratios. Therefore, the source of atmospheric particulate matter in Nanjing and Hangzhou may have changed.

The $PM_{2.5}$ and PM_{10} concentrations in Hu-Ning-Hang were the highest in winter, followed by spring and autumn, and the lowest in summer (Table 3), indicating that the pollution of atmospheric particulate matter in Hu-Ning-Hang clearly has seasonal characteristics. The atmospheric stratification in winter is not conducive to the spread of atmospheric pollutants, while sea breeze and precipitation play a role in scouring dilution of the atmospheric particles in summer [13].

In 2015 the ratio of $PM_{2.5}/PM_{10}$ concentrations in Hu-Ning-Hang is highest in winter and lowest in spring, indicating that the pollutant particles in winter are mainly fine particles. $PM_{2.5}$ easily enters the body, causing a variety of heart and lung disease. In addition, the toxic components in $PM_{2.5}$ will dissolve into blood and have more serious effects on the human body [20]. Monitoring of fine particulate contaminants should be increased in winter. In 2016 the seasonal distribution of $PM_{2.5}$ and PM_{10} concentrations in Hu-Ning-Hang was similar to that in 2015. It should be noted that the 2016 concentration of $PM_{2.5}$ in Hangzhou was slightly lower than in 2015. The $PM_{2.5}$ and PM_{10} concentrations in Shanghai and Nanjing in spring and the PM_{10} concentration in Hangzhou increased from 20015 to 2016 in different ways, unlike the changes in the other three seasons. The ratio of $PM_{2.5}/PM_{10}$ was highest in winter and lowest in spring. In summer, winter, and autumn, the $PM_{2.5}/PM_{10}$ ratios decreased from 2015 to 2016, indicating that the influence of $PM_{2.5}$ was decreasing. The ratios of $PM_{2.5}/PM_{10}$ in each season is Shanghai>Hangzhou>Nanjing, showing that fine particles have the greatest impact on air pollution in Shanghai; this relationship further explains the differences in the main atmospheric pollutants and pollution sources in Hu-Ning-Hang.

Conclusions

1) The concentrations of $PM_{2.5}$ and PM_{10} in the Yangtze River Delta urban agglomeration decreased from 2015 to 2016. The $PM_{2.5}$ and PM_{10} concentrations were higher in Nanjing, Changzhou, Yangzhou, and Taizhou(J) in Jiangsu Province, and lower in the cities mainly in Zhejiang Province. During 2015-2016, the highest concentration of $PM_{2.5}$ was measured in Hefei, Anhui Province. The highest concentration of PM_{10} was measured in Taizhou(J), Jiangsu Province, and the lowest concentrations of $PM_{2.5}$ and PM_{10} were in Zhoushan, Zhejiang Province.
2) The average annual concentrations of $PM_{2.5}$ were highest in winter and lowest in autumn. There was a significant linear correlation between $PM_{2.5}$ and PM_{10} in Hu-Ning-Hang, suggesting that the sources of particulate pollutants were very consistent.
3) During 2015-2016 the ratio of $PM_{2.5}/PM_{10}$ in Hu-Ning-Hang was greater than 0.5, indicating that the main pollutants in the atmosphere were fine particles. The comparison between cities shows that the ratio of $PM_{2.5}/PM_{10}$ in each season follows Shanghai>Hangzhou>Nanjing, which shows that fine particles have the greatest impact on air pollution in Shanghai.

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Conflict of Interest

The authors declare no conflict of interest.

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