



Spatial and Temporal Variability of the PM_{2.5}/PM₁₀ Ratio in Wuhan, Central China

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ABSTRACT

Fine particles (PM_{2.5}) and coarse particles (PM_{2.5–10}) are generally produced by different sources, so the PM_{2.5}/PM₁₀ ratio reveals characteristics of particle pollution. The ratio can be used to characterize the underlying atmospheric processes and evaluate historical PM_{2.5} pollution in absence of direct measurements. However, application of the ratio needs its varying pattern because PM concentrations change significantly at time and space. Hourly PM_{2.5} and PM₁₀ observations at nine monitoring sites in urban area (Urban-sites) and one remote Background-site in Wuhan in 2013–2015 were collected to investigate both long-term, short-term temporal variation and spatial distribution, spatial disparity of the ratio at a city scale. The results show that annual average PM_{2.5}/PM₁₀ ratio is 0.62 at Urban-sites and 0.68 at Background-site with apparent seasonal, monthly and daily variations. The ratio reaches the maximum in winter because of stable atmospheric conditions. There are apparent night-day differences of daily variation of the ratio, which increases at night in all seasons in consequence of temperature inversion and declines in the daytime with a moderate rise in the afternoon. We find obvious spatial gradients of the ratio that gradually increases from urban core to urban fringe and to suburban. This study provides further insights to the spatio-temporal variability of PM_{2.5}/PM₁₀ ratio. The evidence indicates that the variability of PM_{2.5}/PM₁₀ should be noticed in its applications.

Keywords: Air pollution; Particulate matter; Ratio; Spatio-temporal variability; China.

INTRODUCTION

Particulate matter (PM) has been a growing public concern due to its severe health impacts and significant visibility reduction (Dockery and Pope, 1994; Dominici *et al.*, 2014; Zhang *et al.*, 2015; Fang *et al.*, 2016). PM is usually divided and nominated by its aerodynamic diameter, and the most widely monitored particles are PM₁₀ and PM_{2.5}, which refer to particles with aerodynamic diameter less than or equal to 10 or 2.5 micrometers, respectively (Nel, 2005). PM_{2.5} is also called fine particle while particle with a diameter between 2.5 and 10 micrometers is coarse particle (PM_{2.5–10}). Coarse particles (PM_{2.5–10}) are mainly produced from natural processing, such as re-suspension of local soil, dust storm, as well as from anthropogenic sources like road dust and

various industrial processes (Querol *et al.*, 2004). Fine particles (PM_{2.5}) comprise largely primary and secondary anthropogenic combustion products originated from traffic and energy production (Li *et al.*, 2004). Compared with PM₁₀, PM_{2.5} gets more attention due to its smaller size, longer atmospheric lifetime and severer health risks (Dominici *et al.*, 2014).

Since fine and coarse particles come from diverse sources and perform different physic-chemical properties, the PM_{2.5}/PM₁₀ ratio can provide crucial information relating to the particle origin, its formation process and its effects on human health (Speranza *et al.*, 2014; Blanco-Becerra *et al.*, 2015). Higher ratios of PM_{2.5}/PM₁₀ attribute particle pollution to anthropogenic sources and smaller ratios indicate considerable involvement of coarse particles, which might be related to natural sources, e.g. dust storm (Sugimoto *et al.*, 2016). Actually, the PM_{2.5}/PM₁₀ ratio is often used to characterize the underlying atmospheric processes within the local environment (Yu and Wang, 2010; Chu *et al.*, 2015). Sugimoto *et al.* (2016) has estimated the fraction of mineral dust in particulate matter in East Asia using the

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PM_{2.5}/PM₁₀ ratio. PM_{2.5} was not systematically observed and reported in United States until 1998, and it has just been introduced into the air quality monitoring system in 2013 in China (Yu and Wang, 2010). Identifying and quantifying the PM_{2.5}/PM₁₀ ratios can provide useful information for retrospective prediction of PM_{2.5} concentration without direct PM_{2.5} measurements, which has been successfully applied in California and Taipei (Yu and Wang, 2010; Blanchard *et al.*, 2011; Chu *et al.*, 2015). However, all of these applications of the PM_{2.5}/PM₁₀ ratio need evidence of its spatio-temporal variability.

PM concentration is influenced by various factors such as land use, population density, meteorology condition (Xu *et al.*, 2016b), and there is obvious spatio-temporal heterogeneity of PM concentrations at intra-city or regional scales. Therefore, it is evident that the fraction of fine particles (PM_{2.5}) in PM₁₀ also varies at time and space (Parkhurst *et al.*, 1999; Zhou *et al.*, 2016). PM₁₀ can be mainly composed of fine particles (PM_{2.5}), for instance, PM_{2.5} contributes 67% of PM₁₀ in three east-central U.S. states and the average PM_{2.5}/PM₁₀ ratio is 0.60 at 20 European areas (Parkhurst *et al.*, 1999; Eeftens *et al.*, 2012). However, the ratio is relatively lower in desert area because of the significant contribution of coarse particles, for example, the average PM_{2.5}/PM₁₀ ratio is 0.33 in Saudi Arabia (Khodeir *et al.*, 2012). Previous studies also showed that the PM_{2.5}/PM₁₀ ratios were generally higher in the eastern (~0.7) than in the central or western (~0.5) United States (USEPA, 2004). The PM_{2.5}/PM₁₀ ratio shows apparent seasonal and diurnal variability as well. Speranza *et al.* (2016) found that the ratio of PM_{2.5} to PM₁₀, rearranged from published data according to seasons of sampling, was more than 0.6 in colder seasons (autumn-winter) with a lower ratio (< 0.5) in warmer seasons (spring-summer). While in Africa, the PM_{2.5}/PM₁₀ ratio is higher during the wet season than dry season due to dust re-suspension, a major contribution to PM₁₀ (Akinlade *et al.*, 2015). As for daily variation, it was found that the minimum values of PM_{2.5}/PM₁₀ ratio in one day occurred during traffic hours in consequence of re-suspended coarse road dust (Querol *et al.*, 2001; Evagelopoulou *et al.*, 2006).

Compared with numerous studies demonstrating the variability of PM concentrations (Eeftens *et al.*, 2012; Hu *et al.*, 2014; Akinlade *et al.*, 2015; Ghim *et al.*, 2015; Huang *et al.*, 2015; Zhang and Cao, 2015; Zhou *et al.*, 2015; Xu *et al.*, 2016a), few studies have reported on the spatio-temporal variability of the PM_{2.5}/PM₁₀ ratio (Blanco-Becerra *et al.*, 2015; Zhang and Cao, 2015; Munir, 2016; Zhou *et al.*, 2016). The goal of this study is to investigate the spatio-temporal variability of the PM_{2.5}/PM₁₀ ratio at a city scale systematically and comprehensively. Taking Wuhan, central China, as an example, both long-term, short-term temporal variation and spatial distribution, spatial disparity of the PM_{2.5}/PM₁₀ ratio will be analyzed based on hourly measurements of PM_{2.5} and PM₁₀ concentrations at nine monitoring sites in urban area (Urban-sites) and one remote Background-site in 2013–2015. To our knowledge, this is the first study aiming at the spatio-temporal variability of the PM_{2.5}/PM₁₀ ratio specially.

MATERIALS AND METHODS

Study Area

Wuhan, located in central China, is the capital city of Hubei province where the Yangtze River and Han River join together dividing Wuhan into three parts, Wuchang, Hankou, and Hanyang (Fig. 1). Wuhan is undergoing rapid industrialization and urbanization over the past decades (Jiao *et al.*, 2017). In 2014, more than 10 million people lived in this city with a total GDP exceeding one trillion RMB (approximately 154 billion US dollars) (Xu *et al.*, 2016b). With the vast economic development, the air quality in Wuhan has been under high pressure. Like other megacities in China, serious particle pollution, especially PM_{2.5} pollution, was identified in Wuhan in recent years (Guo *et al.*, 2015).

PM Monitoring and Measurements

Currently, there are ten national-controlling ambient air quality monitoring sites in Wuhan city, of which nine sites are located in the urban area (Urban-sites) with one Background-site located about 50 km away from the urban area (Fig. 1). The detailed information on monitoring sites is shown in Table 1. PM_{2.5} has not been monitored routinely until 2013 in Wuhan. The real-time mass concentrations of PM_{2.5} and PM₁₀ are measured using the micro oscillating balance method and/or the β absorption method using commercial instruments (Zhang and Cao, 2015). The measurements are available at the website of Wuhan Environmental Protection Bureau (<http://www.whepb.gov.cn/>). In this study, hourly measurements of PM_{2.5} and PM₁₀ at ten national-controlling monitoring sites from January 18, 2013 to December 31, 2015 were collected.

Data Summarizing and Analysis

Only data available for both PM_{2.5} and PM₁₀ concentrations at the same hour has been included resulting in more than 170,000 records in total. The average ratios of PM_{2.5} to PM₁₀ among diverse temporal scales at each site are summarized step by step (Xu *et al.*, 2016a). At each site, daily averages of the PM_{2.5}/PM₁₀ ratios are summarized from hourly ratios. As a result, daily average ratios are used to calculate monthly averages. These are then used to calculate seasonal and annual average ratios at each site. The season division in this study is as follows: spring (March, April, May), summer (June, July, August), autumn (September, October, November) and winter (December, January, February). In order to compare the variability of PM_{2.5}/PM₁₀ ratios with PM concentrations, the average concentrations of PM_{2.5} and PM₁₀ at diverse temporal scales are summarized as well. The spatio-temporal variability of PM_{2.5}/PM₁₀ ratios in Wuhan is analyzed as the flowchart shown in Fig. 2. Since the pollution level in urban and suburban area is apparently different (Xu *et al.*, 2016a), temporal variability of the PM_{2.5}/PM₁₀ ratio at Urban-sites (nine) and Background-site (one) will be compared. Although temporal and spatial variability of the PM_{2.5}/PM₁₀ ratio will be analyzed separately, spatio-temporal variability affects each other, for example, the distinguishing of Urban-sites and Background-site in temporal variation analysis also reveals the spatial disparity of the ratio.

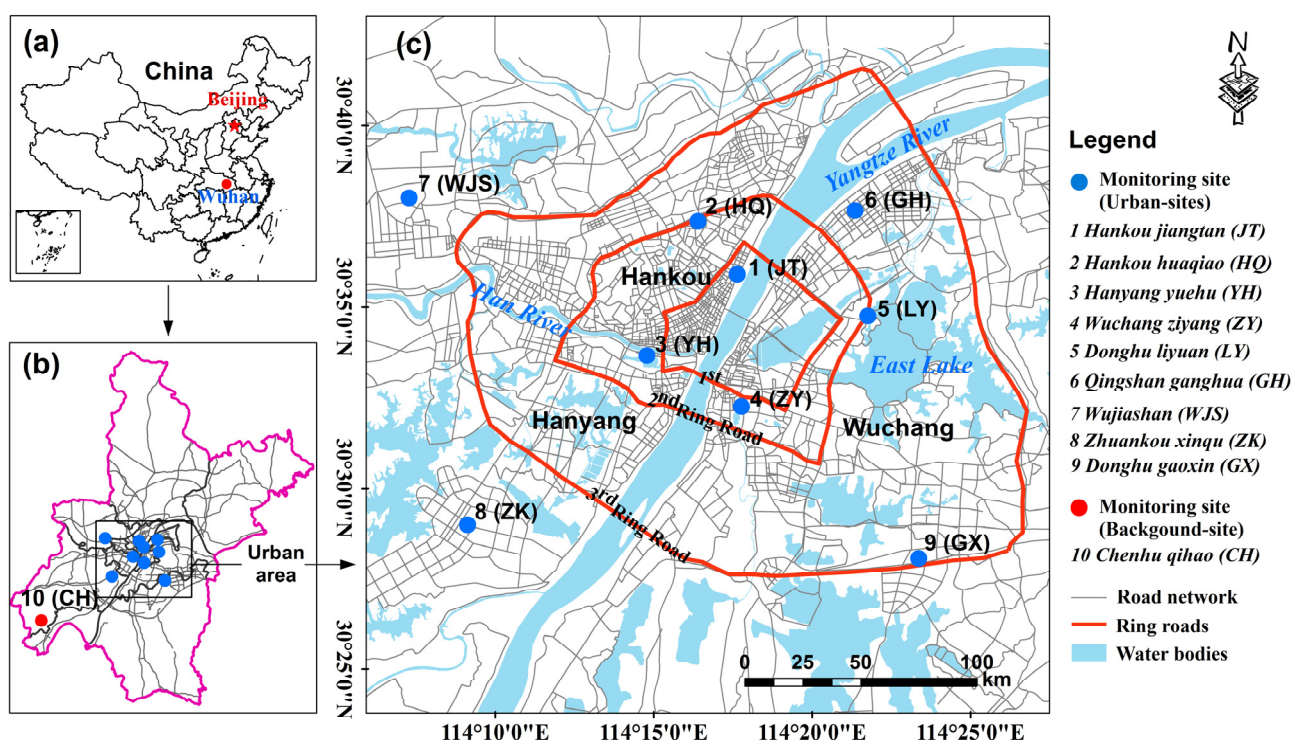


Fig. 1. Study area and the spatial distribution of ten ambient air quality monitoring sites in Wuhan city: (a) location of Wuhan in China; (b) administration boundary of Wuhan city and spatial distribution of ten monitoring sites; (c) detailed distribution of nine monitoring sites in the urban area of Wuhan.

Table 1. Detailed description of ambient air quality monitoring sites in Wuhan.

No.	Site Name	Site Code	Location	Functional Zone	Pop. Density (person km ⁻²)
1	Hankou jiangtan	JT	Inside the 1 st ring road	Scenery, Transportation zone	14,000
2	Hankou huaqiao	HQ	Near the 2 nd ring road	Residential, Transportation zone	14,000
3	Hanyang yuehu	YH	Near the 1 st ring road	Scenery, Transportation zone	5,700
4	Wuchang ziyang	ZY	Near the 1 st ring road	Residential, Transportation zone	14,000
5	Donghu liyuan	LY	Near the 2 nd ring road	Scenery zone	14,000
6	Qingshan ganghua	GH	Outside the 2 nd ring road	Industrial, Transportation zone	7,500
7	Wujiashan	WJS	Outside the 3 rd ring road	Industrial, Residential zone	1,100
8	Zhuankou xinqu	ZK	Outside the 3 rd ring road	Industrial, Residential zone	600
9	Donghu gaixin	GX	Near the 3 rd ring road	Industrial, Transportation zone	400
10	Chenu qihao	CH	About 50 km away from urban area	Natural reserve zone	-

RESULTS

Long-Term Variation of the Ratio

The long-term variation of the $PM_{2.5}/PM_{10}$ ratio will be analyzed from seasonal and monthly perspectives. The daily average $PM_{2.5}/PM_{10}$ ratios and PM concentrations summarized from nine Urban-sites during the whole study period are shown in Fig. 3.

PM concentrations fluctuate prominently day by day, with daily average PM_{10} concentrations varying from 15 to 415 $\mu g m^{-3}$ and $PM_{2.5}$ varying from 7 to 356 $\mu g m^{-3}$. During the whole three years, 40% days exceed the 24-hour Level-II limitation (75 $\mu g m^{-3}$) for $PM_{2.5}$ concentration, and 27% days violate the 24-hour Level-II limitation (150 $\mu g m^{-3}$) for PM_{10} concentration, demonstrating the severity of particle

pollution in Wuhan (China Ambient Air Quality Standards (GB 3095-2012)). The Level-II limitations are applicable to residential areas, commercial-transport-residential mixed areas, cultural areas, industrial areas and rural areas (CAAQS, GB 3095-2012). Due to the noticeable fluctuation of PM concentrations, the daily average $PM_{2.5}/PM_{10}$ ratios vary significantly as well. The daily mean ratios change in 0.2–0.9 with an average ratio of 0.62 for the whole three years in Wuhan urban area, indicating the dominant fraction of fine particles in PM_{10} .

There is a periodical variation for both PM concentrations and $PM_{2.5}/PM_{10}$ ratios in Wuhan urban area. A high level of seasonality can be observed that PM pollution in winter is the most serious and it drops to a comparatively low level in summer. These fluctuations have been broadly

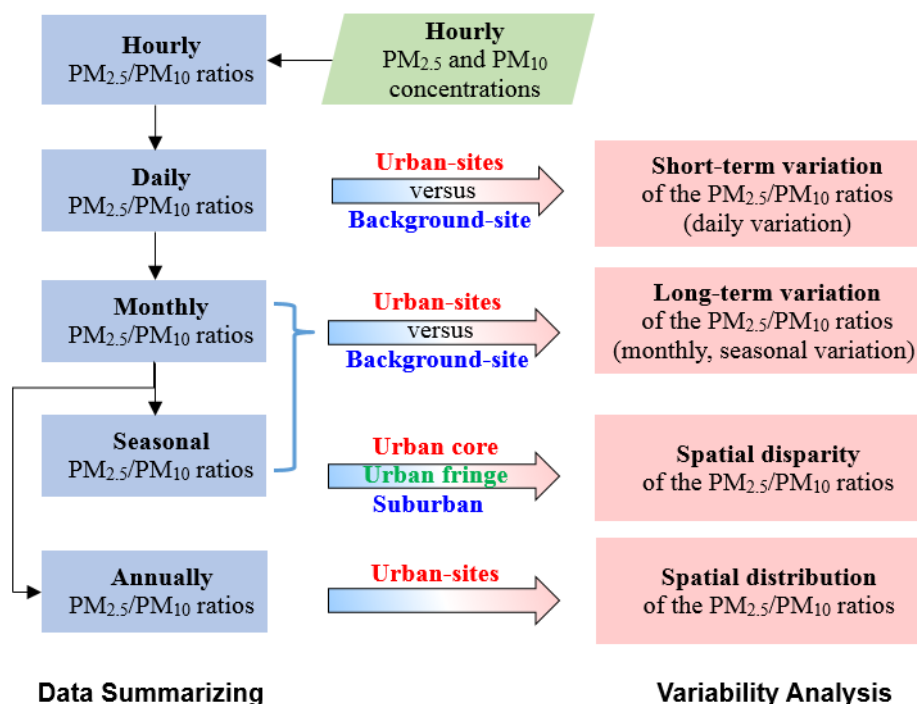


Fig. 2. Workflow of data summarizing and spatio-temporal variability analysis of the $PM_{2.5}/PM_{10}$ ratio.

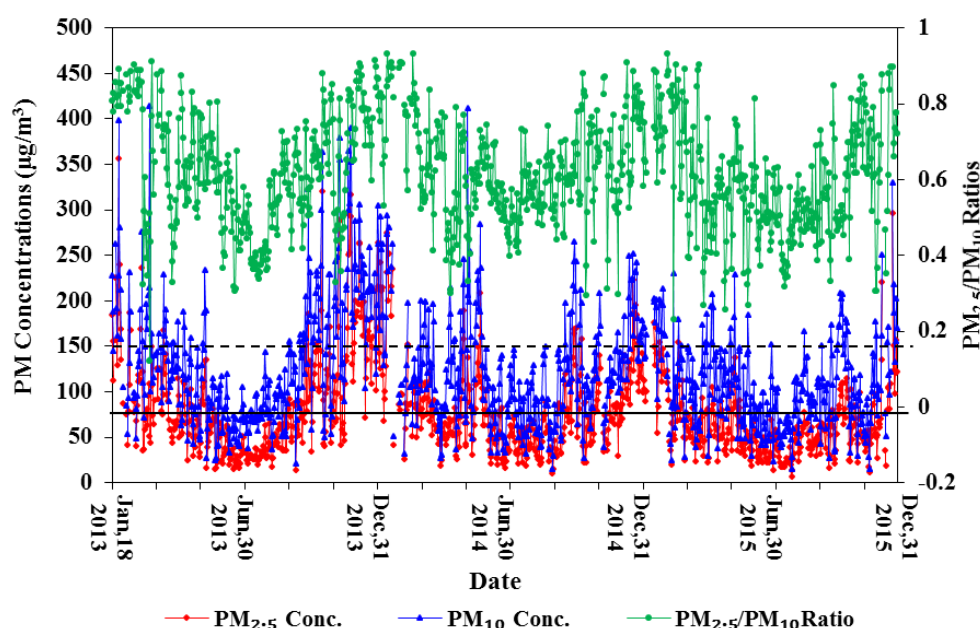


Fig. 3. Daily average $PM_{2.5}/PM_{10}$ ratios and PM concentrations summarized from hourly observations at nine Urban-sites in urban area in 2013–2015 with the solid and dashed horizontal lines representing the 24-hour Level-II limitations for $PM_{2.5}$ ($75 \mu g m^{-3}$) and PM_{10} ($150 \mu g m^{-3}$) concentrations, respectively, according to the China Ambient Air Quality Standards (CAAQS, GB 3095-2012).

reported in China (Zhang and Cao, 2015; Zhou *et al.*, 2015). The daily average $PM_{2.5}/PM_{10}$ ratios display a similar varying pattern to PM concentrations in Wuhan urban areas. Overall, the ratio declines from spring to summer, reaching the lowest, and then it increases to the highest in winter. The seasonal averages of PM concentrations and $PM_{2.5}/PM_{10}$ ratios at nine Urban-sites and one Background-site in

2013–2015 are presented in Table 2.

In terms of seasonality of the $PM_{2.5}/PM_{10}$ ratios, both Urban-sites and Background-site experience the highest ratios in winter, but the ratio drops down to the lowest (0.55) in summer at Urban-sites while Background-site experiences the lowest (0.63) in spring (Table 2). The highest value of $PM_{2.5}/PM_{10}$ ratio in winter was also observed in other

Table 2. Seasonal average PM concentrations and PM_{2.5}/PM₁₀ ratios in Wuhan in 2013–2015.

Periods	Urban-sites (mean ± st. dev, µg m ⁻³)			Background-site (mean ± st. dev, µg m ⁻³)		
	PM _{2.5} Conc.	PM ₁₀ Conc.	PM _{2.5} /PM ₁₀	PM _{2.5} Conc.	PM ₁₀ Conc.	PM _{2.5} /PM ₁₀
Spring	73 ± 37	129 ± 73	0.61 ± 0.20	54 ± 31	93 ± 62	0.63 ± 0.22
Summer	45 ± 30	86 ± 50	0.55 ± 0.19	39 ± 25	60 ± 36	0.66 ± 0.19
Autumn	75 ± 47	127 ± 74	0.62 ± 0.21	65 ± 39	99 ± 57	0.69 ± 0.39
Winter	125 ± 68	170 ± 83	0.75 ± 0.26	98 ± 53	125 ± 57	0.80 ± 0.42
Full year	74 ± 52	122 ± 75	0.62 ± 0.22	59 ± 41	89 ± 57	0.68 ± 0.31

cities in China, such as Beijing, Tianjin, Chengdu (Gu *et al.*, 2010; Huang *et al.*, 2015; Zhou *et al.*, 2015). Higher PM_{2.5}/PM₁₀ ratios relating to colder seasons (autumn and winter) were found as well in a meta-analysis (Speranza *et al.*, 2016). The increasing of fuel consumption for domestic and industrial heating in winter leads to an increase of fine particle emissions (Liu *et al.*, 2016). The secondary aerosol, one of the major source of fine particles, is also accelerated due to the lower mixing height in winter (Huang *et al.*, 2014). On the other hand, the stable atmospheric conditions in winter are favorable for the dry deposition of coarse particles, but also favor the accumulation of fine particles in the air, resulting the domination of fine particles in PM₁₀ in winter (Huang *et al.*, 2015). At nine Urban-sites, PM_{2.5} concentration decreases from the highest to the lowest by 64% but PM₁₀ concentration declines by 49%, indicating a stronger seasonal disparity of PM_{2.5} than PM₁₀.

For more detailed information about the long-term variation, monthly variation of the PM_{2.5}/PM₁₀ ratios and PM concentrations are presented in Fig. 4. The error bars indicate one standard deviation of the ratios at Urban-sites and Background-site. Fig. 4 shows that monthly variation of the PM_{2.5}/PM₁₀ ratios generally share the same variation trend with PM concentrations, especially the ratios at nine Urban-sites. PM_{2.5} and PM₁₀ concentrations decrease from January to July and then increase to December in addition to some local undulations. However, there is obvious difference for the variation of PM_{2.5}/PM₁₀ ratio between Urban-sites and Background-site. The PM_{2.5}/PM₁₀ ratios at

Urban-sites drop to the lowest in July (0.51) while the ratio at Background-site experiences the minimum in April (0.59), although both of them reach to the maximum in February. Another disparity between Urban-sites and Background-site is the variation amplitude of the ratio. The ratio varies more greatly at Urban-sites but there is only imperceptible changes for the ratio at Background-site from March to December, which indicates more complex and changing PM sources in urban area.

Short-Term Variation of the Ratio

Daily variation of the PM_{2.5}/PM₁₀ ratios and PM concentrations at Urban-sites and Background-site in 2013–2015 is presented in Fig. 5. The 19:00 is the turning point of daily variation of the ratios, so the hour begins at 19:00 in Fig. 5. An obvious night-day differences of PM_{2.5}/PM₁₀ ratios can be found in Fig. 5. The PM_{2.5}/PM₁₀ ratios at both Urban-sites and Background-site increase from 19:00 all along to the early morning at 6:00, with the peak value of 0.65 at Urban-sites and 0.72 at Background-site. While in the daytime, the ratios decrease until nightfall (18:00), although there is a modest rise at Urban-sites during post meridiem (12:00–14:00). It is the changes of temperature that should be taken into consideration to understand the evident night-day differences of PM_{2.5}/PM₁₀ ratios. The underground temperature is lower than the air temperature at night, which is called temperature inversion. Temperature inversions lead to stable atmospheric conditions constraining vertical airflow, which is favorable to the dry deposition of

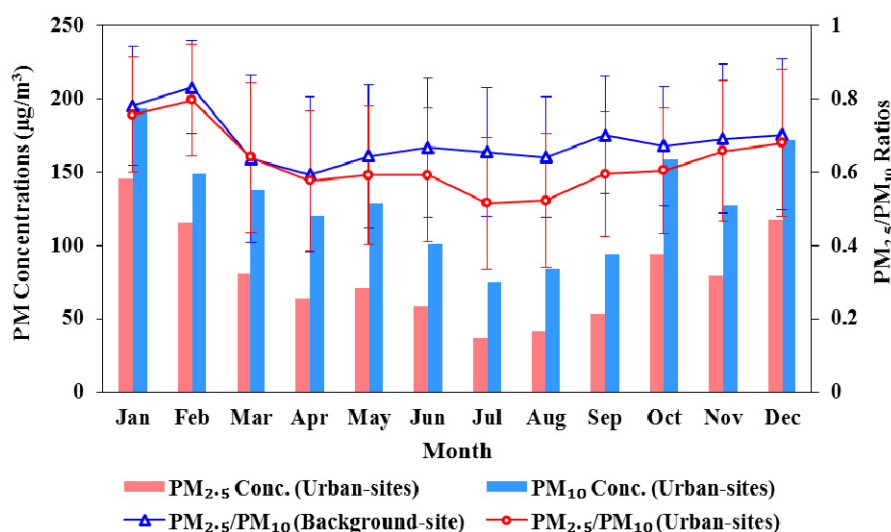


Fig. 4. Monthly variation of the PM_{2.5}/PM₁₀ ratios and PM concentrations at Urban-sites and Background-site.

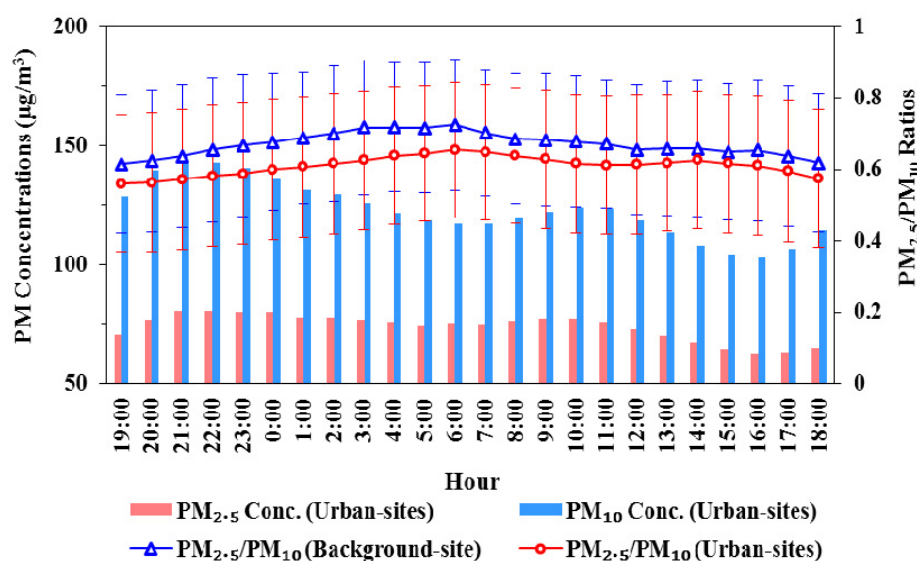


Fig. 5. Daily variation of the $\text{PM}_{2.5}/\text{PM}_{10}$ ratios and PM concentrations at Urban-sites and Background-site in 2013–2015.

coarse particles and the accumulation of fine particles (Wallace and Kanaroglou, 2009). Therefore, the $\text{PM}_{2.5}/\text{PM}_{10}$ ratios increase gradually at night. In the daytime, as a consequence of the re-suspended coarse road dust and dynamic human activities, the $\text{PM}_{2.5}/\text{PM}_{10}$ ratios decline gradually. The daily variation of $\text{PM}_{2.5}/\text{PM}_{10}$ ratios in four seasons are presented in Fig. 6 to identify the seasonality of the daily variation of the ratios.

The turning point of daily variation trend for $\text{PM}_{2.5}/\text{PM}_{10}$ ratios is one or two hours later in summer and autumn, since the temperature inversion occurs later in the two seasons. To keep consistent with Fig. 5, the time in four sub-divisions begins at 19:00 as well. It shows similar daily variation but also disparities of $\text{PM}_{2.5}/\text{PM}_{10}$ ratios in four seasons (Fig. 6). Daily variation of $\text{PM}_{2.5}/\text{PM}_{10}$ ratios at Urban-sites is more stable and smooth. However, there are local fluctuations in daily variation at Background-site, especially during nighttime in winter (Fig. 6(d)), which might be related to the limited number of Background-site (only one) in Wuhan. $\text{PM}_{2.5}/\text{PM}_{10}$ ratios increase dramatically in the nocturnal period except for the ratio at Background-site in winter. The ratios generally share the same decreasing trend in the daytime in four different seasons. The gap of ratios between Urban-sites and Background-site in nighttime is greater than that in the daytime (Figs. 5 and 6). As a consequence of the urban heat islands effect, the ground temperature is higher in urban areas than suburban in nighttime. Therefore, the intensified temperature inversion in suburban areas will lead to more stable atmospheric conditions, which contributes to the rising of $\text{PM}_{2.5}/\text{PM}_{10}$ ratios. Although the ratio varies more like a unimodal pattern in Fig. 5 summarized from all four seasons. The daily variation of ratio in four sub-divisions shows a more obvious bimodal pattern, particularly in summer. Merely, the afternoon peak is not as noticeable as the peak in early morning. Actually, the bimodal pattern of daily variation of $\text{PM}_{2.5}/\text{PM}_{10}$ ratios also have been reported in Catalonia, Spain (Querol *et al.*, 2001) and Kozani, Greece (Evangelopoulos *et al.*, 2006).

Spatial Distribution of the Ratio

The spatial distribution of three-year average $\text{PM}_{2.5}/\text{PM}_{10}$ ratios and PM concentrations in Wuhan urban area are presented in Fig. 7. Apparent spatial disparity of PM pollution level and $\text{PM}_{2.5}/\text{PM}_{10}$ ratios in Wuhan urban area can be recognized in Fig. 7. The three-year average $\text{PM}_{2.5}$ concentrations vary from $68 \mu\text{g m}^{-3}$ at GX to $87 \mu\text{g m}^{-3}$ at GH, and PM_{10} concentrations change in $115\text{--}135 \mu\text{g m}^{-3}$. The most serious $\text{PM}_{2.5}$ and PM_{10} pollutions take place at GH site, where it is a traditional industrial base. The GX site experiences both the lowest $\text{PM}_{2.5}$ and PM_{10} concentrations probably due to lower population density (Table 1). The proportion of fine particles in PM_{10} also shows great variability varying from 58% to 66%. The maximum ratio (0.66) is found at the most polluted site (GH), demonstrating the high contribution of industrial emissions to fine particles, which has also been proved by another industrial site (WJS) with higher $\text{PM}_{2.5}/\text{PM}_{10}$ ratio. ZK and GX sites are located in industrial zones as well, but the ratios at those sites are relatively low. In spite of less population, these two industrial parks are planned for development of high and advanced technologies with lower industrial emission; on the other hand, these two industrial parks are newly built in recent years with much area of bare land resulting in considerable suspension of soil dust (Xu *et al.*, 2016b). As for LY site, located in a scenery zone, PM concentrations and the ratio at this site are still relatively high because it is situated in the downwind of the Qingshan traditional industrial base (GH).

Spatial Disparity of the Ratio

In order to distinguish the $\text{PM}_{2.5}/\text{PM}_{10}$ ratios with different distances to the main urban area, those nine Urban-sites are divided into two parts (urban core and urban fringe) depends on whether located inside or outside the 2nd Ring Road in Wuhan. Obviously, sites 1–5 are located inside the 2nd Ring Road (urban core) while the other four Urban-sites belong to the second part (urban fringe) (Fig. 1), and the

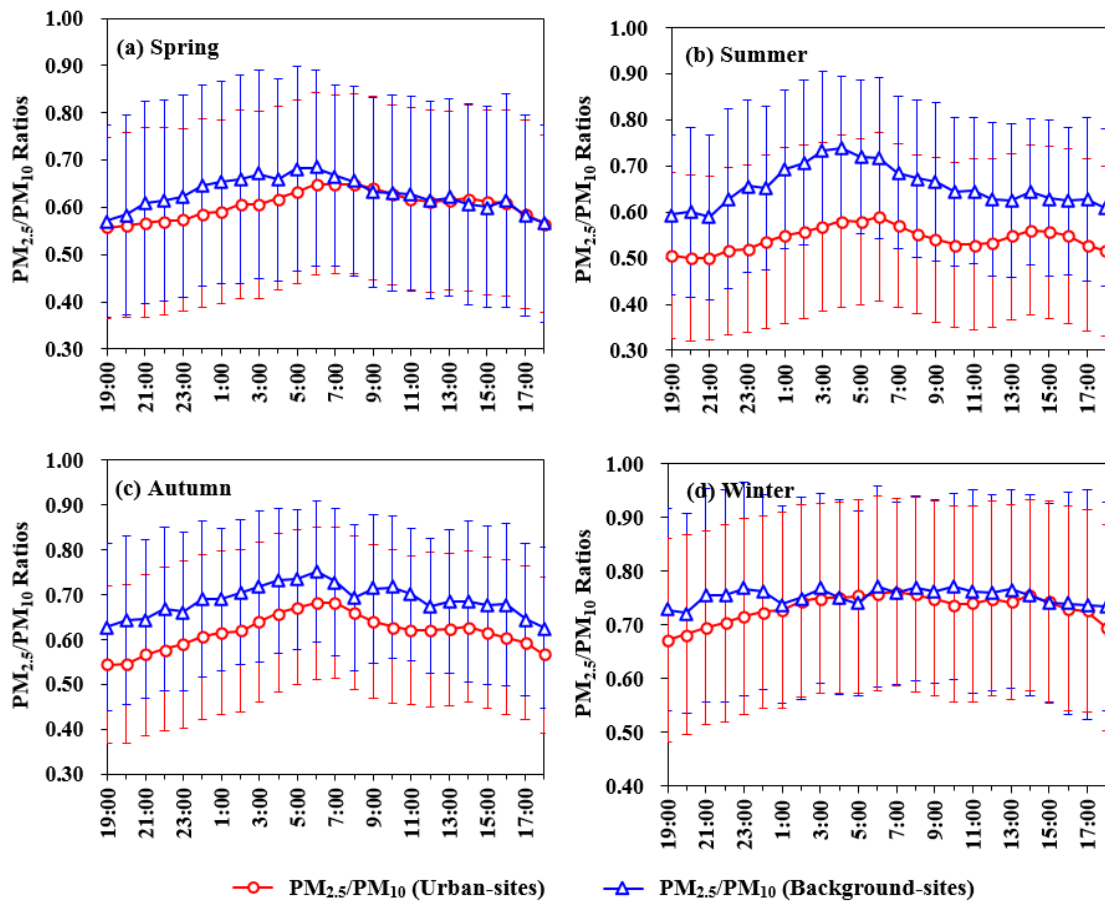


Fig. 6. Daily variation of the $PM_{2.5}/PM_{10}$ ratios at Urban-sites and Background-site in four seasons: (a) spring; (b) summer; (c) autumn; (d) winter.

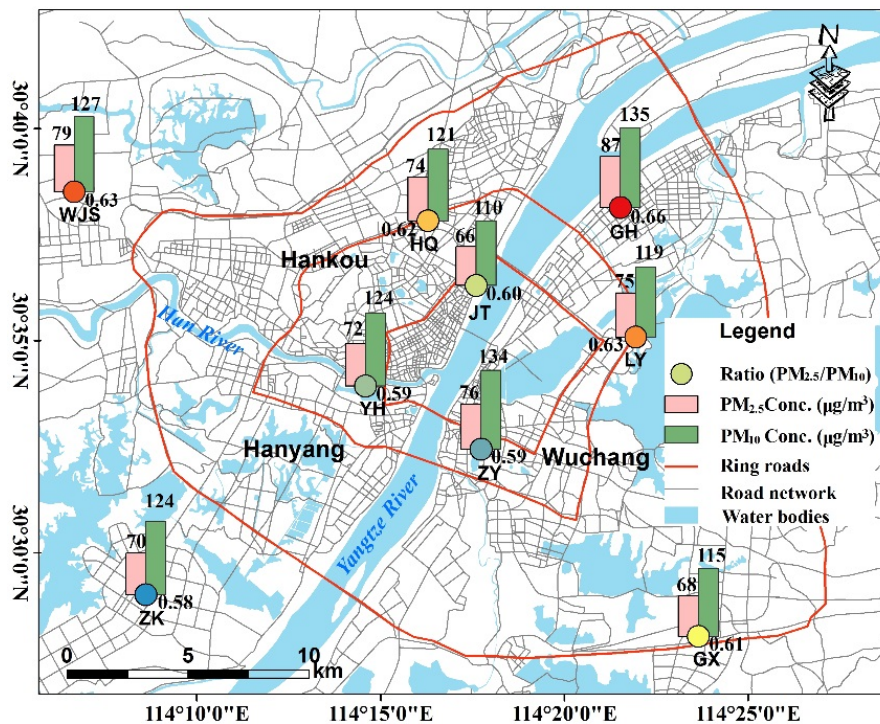


Fig. 7. Spatial distribution of $PM_{2.5}/PM_{10}$ ratios and PM concentrations at nine Urban-sites in Wuhan urban area in 2013–2015.

third part is suburban (Background-site). The ratios at three categories of sites in four seasons are presented in Fig. 8.

An obvious tendency can be found in Fig. 8 that the $PM_{2.5}/PM_{10}$ ratio gradually increase from urban core (0.617, four-season average) to urban fringe (0.630) and further to suburban (0.680). In addition, the magnitude of the spatial disparity varies in different seasons. In winter and spring, the ratios at three categories of sites are similar. The disparity is the most obvious in summer and the ratio at suburban is 16.6%, 23.7% higher than that at urban fringe and urban core, respectively. The ratio at urban fringe is 6.1% higher than that at urban core in summer. But in autumn and winter, the ratio at urban core and urban fringe are almost the same, although the ratio at suburban is significantly higher.

Fine particles as well as coarse particles are emitted and produced due to human activities in urban area, but fine particles can travel further than coarse particles before they are removed from the atmosphere, resulting higher $PM_{2.5}/PM_{10}$ ratios in suburban (Parkhurst *et al.*, 1999; Zhou *et al.*, 2016). In addition, the lower ratios at Urban-sites are partly attributed to the resuspension of construction dust and road dust because of the massive infrastructure construction (e.g., metro and arterial traffic construction) in Wuhan urban area in recent years (Xu *et al.*, 2016b). Parkhurst *et al.* (1999) demonstrated that the fractional contribution of $PM_{2.5}$ to PM_{10} appears greater at remote stations and lower at those stations which are more likely to be influenced by anthropogenic local sources. A previous study in Wuhan also reported higher ratios in suburban site (0.64) than

urban site (0.57) in 1995–1996 (Wei *et al.*, 1999). It was also found the highest $PM_{2.5}/PM_{10}$ ratio at the control site in Ibadan, Nigeria, because of being far from the road and any obvious source of pollutants (Akinlade *et al.*, 2015).

DISCUSSION

This study investigates the spatio-temporal variability of the $PM_{2.5}/PM_{10}$ ratio from both long- and short-term perspectives at a city scale based on hourly PM observations in 2013–2015. The three-year average $PM_{2.5}/PM_{10}$ ratio in Wuhan urban area is 0.62. A wide range of ratios of $PM_{2.5}$ to PM_{10} have been observed across the world. It is reported that 0.6 is regarded as a typical North American $PM_{2.5}/PM_{10}$ ratio (Dockery and Pope, 1994). In Asia, the ratio is generally less than 0.5, indicating higher coarse particle masses; but urban sites in China have mean $PM_{2.5}/PM_{10}$ ratio values greater than 0.5 (Hopke *et al.*, 2008). Studies in China recently reported that the mean ratio of 190 cities is 0.56 in 2014–2015 (Zhang and Cao, 2015). Compared with other megacities or urban agglomeration in China (Table 3), the fraction of fine particles in PM_{10} in Wuhan urban area is relatively high, indicating the severity of fine particle pollution in Wuhan.

This study demonstrates that the $PM_{2.5}/PM_{10}$ ratio varies obviously at time and space. The ratio shares approximate seasonality with PM concentrations at Urban-sites, and the highest ratio observed in winter (0.75) is 20% higher than the minimum ratio (0.55) in summer in Wuhan urban area,

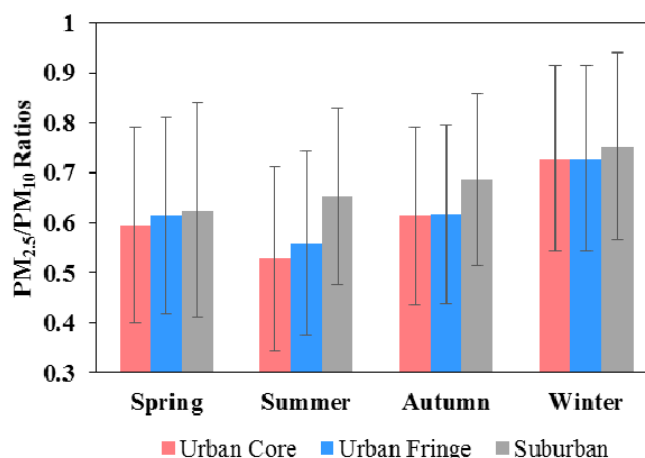


Fig. 8. The average $PM_{2.5}/PM_{10}$ ratios in urban core, urban fringe, and suburban in four seasons from 2013 to 2015.

Table 3. Comparison of the $PM_{2.5}/PM_{10}$ ratio in other cities or regions across China.

Year	City/Region	Location	$PM_{2.5}$ ($\mu g m^{-3}$)	PM_{10} ($\mu g m^{-3}$)	$PM_{2.5}/PM_{10}$ Ratio	References
2013	Beijing	North China	92	119	0.53–0.86	Zhou <i>et al.</i> , 2015
2008	Tianjin	North China	110	196	0.58	Gu <i>et al.</i> , 2010
2013	North China Plain	North China	77	136	0.50–0.64	Hu <i>et al.</i> , 2014
2013	Yangtze River Delta	East China	43	75	0.57–0.64	Hu <i>et al.</i> , 2014
2009–2012	Pearl River Delta	South China	51	76	0.57–0.71	Chen <i>et al.</i> , 2014
2013–2014	Chengdu	Southwest China	100	157	0.64	Huang <i>et al.</i> , 2015
2013–2015	Wuhan	Central China	74	122	0.55–0.75	This study

revealing significant seasonality of particle pollution. Previous study also reported that $PM_{2.5}$ comprised about 80% of PM_{10} during winter months but with 50% in summer months in Birmingham, UK (Harrison *et al.*, 1997). We found apparent night-day difference of the daily variation of $PM_{2.5}/PM_{10}$ ratio. The ratio increases in the nocturnal period all the time due to favorable condition for coarse particle sedimentation caused by inversion temperature, and it declines in the daytime with a moderate rise in the afternoon. In terms of the spatial distribution and disparity, the three year average highest ratio (0.66) in urban area is found at the traditional industrial zone (GH site), indicating the considerable contribution of energy combustion and other industrial process to fine particles. The three-year average $PM_{2.5}/PM_{10}$ ratio at one remote Background-site (0.68) is higher than that at any other Urban-sites (0.58–0.66). We also found an obvious spatial gradient of the ratio, which increases from the urban core to urban fringe, and further to suburban.

The variability of PM concentrations has been reported in many previous studies (Eeftens *et al.*, 2012; Huang *et al.*, 2015; Zhou *et al.*, 2015; Xu *et al.*, 2016a), while this study contributes to the spatio-temporal variability of $PM_{2.5}/PM_{10}$ ratio and its applications. The variation of ratios can reflect the seasonality and type of PM emission sources (Speranza *et al.*, 2016). The considerable variability of the ratio cannot be ignored when we use the ratio in practice. For instance, the seasonal variability of $PM_{2.5}/PM_{10}$ ratio should be addressed in the retrospective prediction of historical $PM_{2.5}$ concentrations without direct measurements. On the other hand, the ratio can be better used if we investigate its variability at first.

There are some limitations in this study as well. At present, only the mass concentrations of PM_{10} and $PM_{2.5}$ are routinely observed at ambient air quality stations in China (Zhang and Cao, 2015). Although PM compositions characterize particle origins and its formation processes, whose significance has been emphasized all the time (Wen *et al.*, 2016), the diverse chemical compositions of fine and coarse particles were not included in this study constrained by data accessibility. In addition, only the arithmetic mean of $PM_{2.5}/PM_{10}$ ratio is used in the data analysis, while other robust statistics, e.g., median, various percentiles, and Theil-Sen trend analysis, which are not sensitive to non-normal distributions and to extreme values in both tails of the distributions, can be applied to better understand the variability of the ratios (Munir, 2016). Finally, the $PM_{2.5}/PM_{10}$ ratio varies at space and time as a consequence of PM fluctuation, but many other factors, such as meteorological condition, affect its value and variability (Blanco-Becerra *et al.*, 2015; Wang *et al.*, 2015). The influencing factors and their relationship with the $PM_{2.5}/PM_{10}$ ratio can be taken into consideration in further work.

CONCLUSIONS

This study contributes to the spatio-temporal variability of $PM_{2.5}/PM_{10}$ ratio. Overall, fine particles make up 62% of PM_{10} in Wuhan urban area in 2013–2015, revealing the

severity of fine particle pollution. The ratio varies obviously at time and space. During one day, the ratio increases in the nocturnal period in all seasons due to favorable condition for coarse particle sedimentation caused by inversion temperature and declines in the daytime with a modest rise in the afternoon. As for the seasonality, the highest ratio is observed in winter (0.75) while it drops to the lowest in summer (0.55) in urban area. There is an apparent spatial gradient of the ratio that gradually increases from urban core (0.617) to urban fringe (0.630) and further to suburban (0.680). The highest ratio is found at industrial zone (GH, 0.66), proving the considerable contribution of energy combustion and other industrial process to fine particles. The implication of this study is that considerable variability of the ratio cannot be ignored when we use the ratio in practice, such as retrospective prediction historical $PM_{2.5}$. In turn, the ratio can be better used to understand atmospheric processes if we investigate its variability at first.

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