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Retrieving historical ambient $PM_{2.5}$ concentrations using existing visibility measurements in Xi'an, Northwest China

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HIGHLIGHTS

- An exponential regression model was established to retrieve historical $PM_{2.5}$ data over Xi'an, China.
- The trends of the retrieved $PM_{2.5}$ are in agreement with the changes of second industry.
- The regression model was confirmed usefulness in retrieving historical $PM_{2.5}$ data when visibility data was available.

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ABSTRACT

Long term fine particulate matter ($PM_{2.5}$) data are needed to assess air quality and climate issues, but $PM_{2.5}$ data have only been monitored in the recent decade in Chinese cities. Considering strong correlations between $PM_{2.5}$ and visibility, regression models can be useful tools for retrieving historical $PM_{2.5}$ data from available visibility data. In this study, $PM_{2.5}$ and visibility data are both available during 2004–2011 in Xi'an, a megacity in northwest China. Data from 2004 to 2007 were used to develop a regression model and those from 2008 to 2011 were used to evaluate the model. An exponential regression model was then chosen to retrieve the historical $PM_{2.5}$ data from 1979 to 2003, which were then analyzed together with the measured data from 2004 to 2011 for long term trends. Seasonal $PM_{2.5}$ increased from 1979 to 2011 with the fastest increase in winter and the slowest in summer. Annual average $PM_{2.5}$ followed into three distinct periods with a slow decreasing trend from 1979 to 1996, a sharp increasing trend from 1997 to 2006, and a slow decreasing trend from 2007 to 2011. These increasing and decreasing trends are in agreement with the evolution of industrial development in Xi'an.

1. Introduction

Aerosol particles play important roles on air quality in rural and urban areas as well as on regional and global climate (IPCC, 2007). Air pollution has become a serious environmental problem in China along with the rapid economic growth over the past several decades. In particular, severe haze pollution in recent winters has caused worldwide attention. Particulate Matter (PM), especially fine PM ($PM_{2.5}$), is a critical pollutant in most Chinese cities (Tie and cao, 2009; Yang et al., 2011; Cao et al., 2012a). Such acute particulate air pollution has resulted in poor visibility and air quality and a sharp increase in respiratory diseases. To improve air quality in China, the central government released the ‘Atmospheric Pollution Prevention and Control Action Plan’ in 2013 which aimed to reduce the $PM_{2.5}$ concentrations by up to 25% by 2017 relative to 2012 levels. Achievement of this ambitious goal depends on optimum emission control strategies, which have to be developed based on knowledge of the relationship between historical $PM_{2.5}$ concentrations and economic/energy structure in the past decades.

However, the historical $PM_{2.5}$ data are very scarce in China because measurement of this pollutant was not compulsory before

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the release of the first national environmental standard for $PM_{2.5}$ in 2012. The lack of long time PM data limits our ability to estimate the effects of PM on human health and climate change. To overcome this problem, some studies have used ambient visibility data to estimate $PM_{2.5}$ or Total Suspended Particulates (TSP), considering that visibility is largely impacted by PM (Abbey et al., 1995; Trijonis, 1983). $PM_{2.5}$ is the most important portion of PM_{10} or TSP in terms of the ability at scattering visible light (Sisler and Malm, 1994; Pitchford, 1991). More recent studies have also demonstrated the strong relationship between $PM_{2.5}$ and ambient visibility and shown that $PM_{2.5}$ was as a key factor on visibility impairment in urban sites (Watson, 2002; Yuan et al., 2006; Cao et al., 2012b).

Xi'an is the biggest city in Northwest China. TSP and PM_{10} have been monitored in Xi'an since 1980 and 1990, respectively, by the local Environmental Agency for air quality index forecasting. However, $PM_{2.5}$ has only been reported since 2003 for research purposes (Cao et al., 2005a, 2012a; Shen et al., 2009, 2011). Although previous studies have already demonstrated that visibility is a useful surrogate for estimating PM level, their relationship varies with locations. In this study, the relationship between $PM_{2.5}$ and visibility was investigated for the city of Xi'an. A regression model between $PM_{2.5}$ and visibility was then proposed, validated and used to retrieve $PM_{2.5}$ concentrations using visibility data collected during the past half century. Long-term seasonal and annual trends of $PM_{2.5}$ were finally analyzed.

2. Data

Data used in this study include $PM_{2.5}$ and TSP concentrations, visibility, and meteorological data. 24-hour (h) integrated $PM_{2.5}$ samples (09:00 to 09:00 the next day, Beijing time) were continuously measured from Jan 1, 2004 to Dec 31, 2011 at an urban site (34.3°N, 108.93°E) in Xi'an. The $PM_{2.5}$ samples were collected on pre-fired (900 °C, 3 h) 47 mm Whatman QM-A quartz-fiber filters by mini-volume air samplers (Airmetrics, Eugene, OR, USA) at a flow rate of 5 L min^{-1}. The samplers were set up on a rooftop 10 m above the ground inside the Institute of Earth Environment, Chinese Academy of Sciences. $PM_{2.5}$ mass was measured by a Sartorius MC5 electronic microbalance with a ±1 μg sensitivity (Sartorius, Göttingen, Germany). Each filter was weighed at least three times before and after sampling. Before weighting, filters were equilibrated for 24-h at a temperature between 20 °C and 23 °C and relative humidity (RH) between 35% and 45%. The difference among the three repeated weightings was less than 10 μg for a blank filter and less than 20 μg for a sampled filter.

Daily TSP concentrations from 1991 to 2000 were calculated based on the Air Pollution Indices (APIs) for TSP, which were obtained from the Xi'an Environmental Protection Bureau and converted to concentrations using the following equation:

$$C = C_{low} + \left[(I - I_{low}) \middle/ (I_{high} - I_{low})\right] \times (C_{high} - C_{low}) \qquad (1)$$

where C is the concentration (mg m^{-3}) and I is the unitless API value. I_{high} and I_{low} are the two closest values approaching the value I in the Table of API graded limited-values (see in http://www.bjmemc.com.cn/g327/s968/t1264.aspx); C_{high} and C_{low} represent the concentrations corresponding to I_{high} and I_{low}, respectively.

24-h averaged ambient visibility and relative humidity (RH) data from 1 January 1979 to 31 December 2011 were obtained from the meteorological bureau of Shaanxi province. The ambient visibility was measured manually eight times daily at 0:00, 3:00, 6:00, 9:00, 12:00, 15:00, 18:00, and 21:00 by Shaanxi meteorological bureau. Daily average (from 9:00 to next 9:00) visibility data were used in this study.

3. Regression model development and validation

3.1. Development of the regression model between $PM_{2.5}$ and visibility

In this study, $PM_{2.5}$ and ambient visibility data from 1 Jan 2004 to 31 Dec 2007 were used to establish the regression model between the two variables, and measured data from 1 Jan 2008 to 31 Dec 2011 were used to evaluate the model. $PM_{2.5}$ concentrations estimated from the model were compared to the TSP concentrations from 1991 to 2000 for further validation. Given that high moisture can significantly influence visibility, some studies excluded high RH data when developing regression models between $PM_{2.5}$ and visibility. For example, Vajanapoom et al. (2001) developed a regression model to estimate PM concentration in Bangkok using paired visibility/PM_{10} observation dataset under low RH (76.5%). In China, RH in foggy days is normally above 90%, while RH in hazy days is lower than 80%. Therefore, paired visibility/$PM_{2.5}$ data under low RH (<80%) days were also used in this study to develop the regression model.

A total of 1164 pairs of measured $PM_{2.5}$/visibility data satisfied the low RH condition. A linear (Fig. 1a) and an exponential regression (Fig. 1b) were both tested. The significance level (P) for these two regression models were both lower than 0.0001, which indicated that $PM_{2.5}$ and visibility were strongly correlated. The square of correlation coefficient (R^2) was higher from the exponential regression (0.4237) than the linear regression (0.3844), implying a better fit between $PM_{2.5}$ and visibility in this city if using exponential regression. A similar exponential relationship but with slightly lower correlation coefficient was reported for the same city by Cao et al. (2012b), despite using only one year data. Yuan et al. (2006) and Wen and Yeh (2010) also established exponential regression models between ambient visibility and light extinction coefficient in Kaohsiung and Taichung, Taiwan. Vajanapoom et al. (2001) developed a linear regression model between PM_{10} and visibility in Bangkok. The regression model differences among the various studies should be mainly due to different weather conditions and emission sources at different locations.

3.2. Regression model evaluation

To evaluate the reliability of the exponential regression model, visibility data under RH < 80% from 1 Jan, 2008 to 31 Dec 2011 - a total of 1041 days, were used to retrieve $PM_{2.5}$ concentrations. The correlation between measured and retrieved $PM_{2.5}$ concentrations is plotted in Fig. 2. A good correlation ($R^2 = 0.3844$, $P < 0.0001$) between the measured and retrieved values was found, suggesting that visibility can be a good surrogate for $PM_{2.5}$ and the exponential regression model can be used for a first-order estimation of $PM_{2.5}$ in Xi'an when only visibility data is available.

To further demonstrate the reliability of the exponential regression model, retrieved $PM_{2.5}$ concentrations during 1991–2000 were also compared with the measured TSP concentrations. As shown in Fig. 3, the retrieved $PM_{2.5}$ followed a similar variation trend as the measured TSP. This finding further proved that the exponential regression model was appropriate in estimating the $PM_{2.5}$ concentrations using 24-h visibility data in Xi'an.

The annual and seasonal mean concentrations, standard deviation (SD), maximum, and minimum values of the measured and retrieved $PM_{2.5}$ during 2008–2011 are summarized in Table 1. It is evident that the retrieved $PM_{2.5}$ levels were a little higher than the measured ones, indicating that the retrieved $PM_{2.5}$ was overestimated by the regression model. The relative standard deviation (RSD) between the measured and retrieved $PM_{2.5}$ mean concentrations in four seasons were also calculated, which showed that

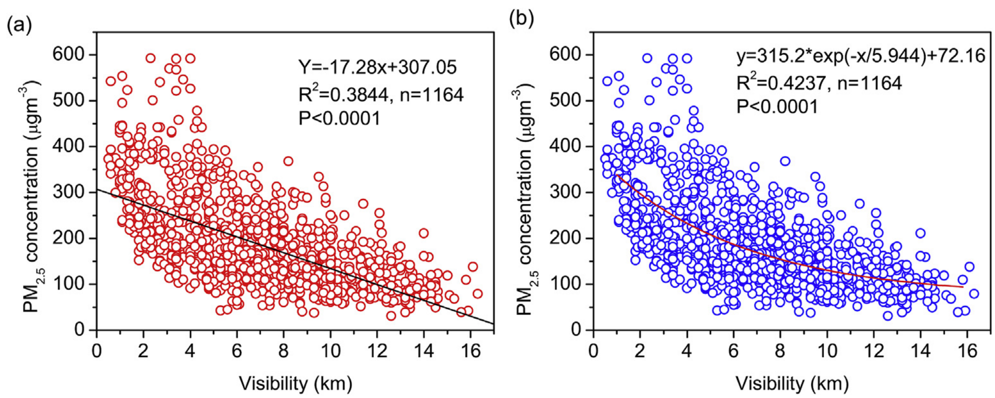

Fig. 1. Regression model of $PM_{2.5}$ and ambient visibility using a linear regression (a) and an exponential regression (b).

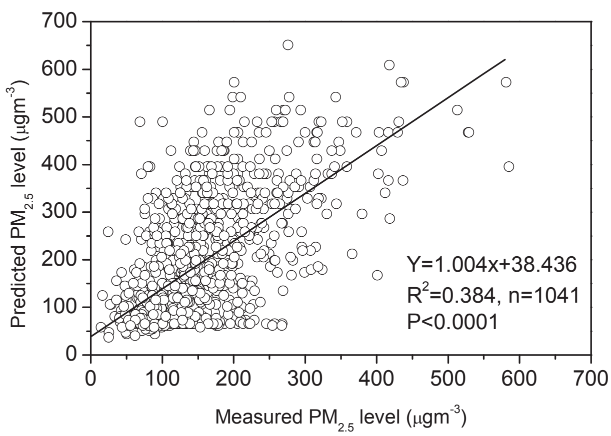

Fig. 2. Correlation between the measured and predicted $PM_{2.5}$ concentrations.

Table 1
Descriptive statistics of the measured and retrieved $PM_{2.5}$ concentration during 2008–2011.

		Average	SD	Max	Min
		μgm^{-3}			
Annual	Measured $PM_{2.5}$	149.2	79.5	585.1	14.0
	Retrieved $PM_{2.5}$	186.3	126.5	651.0	37.2
Spring	Measured $PM_{2.5}$	141.0	14.0	58.1	401.1
	Retrieved $PM_{2.5}$	151.6	44.0	94.4	467.6
Summer	Measured $PM_{2.5}$	111.5	24.4	45.3	294.5
	Retrieved $PM_{2.5}$	154.4	47.4	108.0	489.8
Autumn	Measured $PM_{2.5}$	146.0	25.1	71.1	431.1
	Retrieved $PM_{2.5}$	211.9	37.2	129.0	541.7
Winter	Measured $PM_{2.5}$	200.9	16.4	101.1	585.1
	Retrieved $PM_{2.5}$	237.7	40.6	147.2	651.0

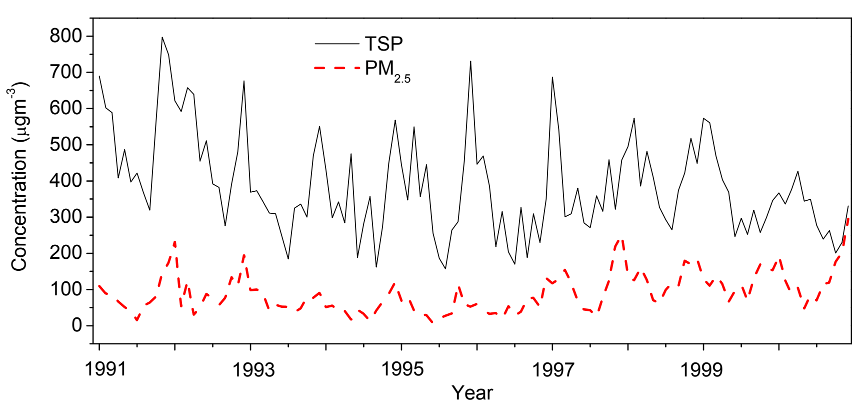

Fig. 3. Predicted $PM_{2.5}$ versus measured TSP during 1991–2000 over Xi'an.

the RSD followed an decreasing order of autumn (36.9%) > summer (32.3%) > winter (16.8%) > spring (7.3%). Such a result indicated that the uncertainties of the autumn and summer retrieved data were higher than those of the spring and winter values. Causes of this phenomenon are discussed below.

Visual range degradation is a complicated issue because it is affected by many factors. Thus, regression models should vary with location since PM chemical composition, sources, and meteorological parameters vary with location. Deng et al. (2011) reported that PM, meteorological variables including RH, wind speed, temperature, and pressure, and synoptic scale processes had obvious impacts on visibility. Here, the correlations between visibility and major meteorological factors were calculated using data from 2004 to 2011. Poor correlations were found between visibility and temperature ($R^2 = 0.0625$), pressure ($R^2 = 0.0625$), or wind speed ($R^2 = 0.0064$). In contrast, visibility had a good negative linear correlation with RH ($R^2 = 0.3025$), indicating RH is another important factor affecting visibility. However, a relatively poor correlation ($R^2 = 0.0484$) was found between RH and visibility when RH was under 50%. Thus, we can conclude that only high RH greatly impairs ambient visibility. This effect was well addressed in this study as the regression analysis excluded visibility data collected during high RH (>80%) days.

Emission sources of PM normally include natural processes and anthropogenic activities. Many studies focused on impacts of anthropogenic produced particles on visibility impairment because anthropogenic activities were the major contributors to $PM_{2.5}$ in urban sites such as in Xi'an (Cao et al., 2007, 2012a; Shen et al., 2009, 2011; Zhu et al., 2012). Previous studies also revealed that $PM_{2.5}$ chemical composition affected heavily the visibility impairment. Ammoniate sulfate was one dominant contributor to visual range impairment, followed by organic carbon, ammoniate nitrate, and elemental carbon (Yuan et al., 2006; Yang et al., 2007; Tao et al., 2009; Cao et al., 2012b). Source contributions to the dry particle light scattering coefficient in Xi'an were also extracted by Cao et al. (2012b), who showed that coal combustion was the dominant factor, accounting for 52% of visibility impairment, followed by the engine exhaust (31%), biomass burning (12%), and fugitive dust (5%). Since China's energy production is dominated by coal burning (~70%), the chemical composition of $PM_{2.5}$ in Chinese cites can be very different from cities in other countries (Cao et al., 2012a). The exponential regression models developed in this study and in another similar study (Deng et al., 2008) represent the characteristics of Chinese cites, which were found to be different when compared to other cities like Bangkok in Thailand (Vajanapoom et al., 2001) where motor vehicle emissions were the main sources of $PM_{2.5}$.

Similar to anthropogenic emissions, aerosol particles from natural sources can also influence heavily visibility degradation. Emissions from large scale events, such as dust storms and wildfires affect the visibility around the globe (Hyslop, 2009). Kim et al. (2001) found that visibility in a large area in Kwangju of Korea was greatly impaired for a few days by a dust storm originated from East Asia, since mineral dust has a relatively high scattering efficiency, although slightly lower than that of ammonium sulfate. The study highlighted that fine dust particles can be transported for a long distance and influenced the air quality and visibility on downwind areas. Since Xi'an is located in semi-arid areas in Northwest China, soil and fugitive dust particles can make up relatively high fractions of $PM_{2.5}$ emission compared to those in coastal cities (Cao et al., 2005b; Shen et al., 2011). Therefore, the exponential regression model developed in this study is representative of the characteristics of cities in semi-arid regions.

Most gaseous pollutants including ozone, sulfur dioxide, and carbon dioxide are invisible to human eyes, but indirectly contribute to visibility degradation by increasing the PM levels through gas–particle reaction processes (Tao et al., 2015; Zhang et al., 2015). Many studies pointed out that sulfate dominated light scattering coefficient, noting that a large portion of $PM_{2.5}$ sulfate was formed through atmospheric heterogeneous reactions. Since coal combustion accounts for nearly 70% of the energy consumption in China, high SO_2 concentrations should be one of the key factors affecting visibility impairment.

4. Retrieved $PM_{2.5}$ concentrations and trends during 1979–2011

Retrieved $PM_{2.5}$ data for the years from 1979 to 2011 and measured data for the years from 2004 to 2011 were analyzed together to investigate $PM_{2.5}$ trends in Xi'an. Seasonal $PM_{2.5}$ and visibility averaged during the 33 years period are summarized in Table 2. It is evident that winter had the highest $PM_{2.5}$ levels and summer had the lowest. The highest winter seasonal value have also been reported in many other cities in North China, which was attributed to the large amount of coal burning emissions for heating purpose and the stable weather conditions (Shen et al., 2008, 2012; Cao et al., 2005a, 2007; 2012a). Better air quality and visibility during summer were probably due to abundance of precipitation and better dispersion conditions (Shen et al., 2010, 2012; Cao et al., 2007, 2012a). Long-term trends of $PM_{2.5}$ and visibility from 1979 to 2011 are investigated by comparing year-to-year seasonal values. $PM_{2.5}$ showed the fastest increasing trend in winter, followed by spring, autumn, and summer. As expected, significant decreasing trends were found for seasonal visibility during these years. However, the seasonal contrasts in the trends of visibility were different from those of $PM_{2.5}$. For example, the fastest decreasing trend occurred in summer for visibility instead of in winter when $PM_{2.5}$ increased the fastest. The differences between seasonal $PM_{2.5}$ and visibility could be caused by a combination of factors, such as changes in PM chemical compositions, $PM_{2.5}$/TSP ratios, and meteorological conditions.

Monthly variations of $PM_{2.5}$ are shown in Fig. 4. In general, lower $PM_{2.5}$ levels were found in June (average of 73.8 μgm^{-3}) and July (average of 80.5 μgm^{-3}). In contrast, air quality in winter months such as December and January was the worst with average $PM_{2.5}$ levels of 186.7 μgm^{-3} and 169.2 μgm^{-3}, respectively. Monthly variations of $PM_{2.5}$ levels were the results of changes in emission sources during summer and winter time and weather conditions mentioned above. As shown in Fig. 4, annual average $PM_{2.5}$ followed into three distinct trends during the period from 1979 to 2011. The first one showed an overall decreasing trend in $PM_{2.5}$ concentration with an average decreasing rate of -4.6 μgm^{-3} per year from 1979 to 1996. The second one exhibited a sharp increasing trend with an average increasing rate of 8.8 μgm^{-3} per year from 1997 to 2006. And the third one showed a decreasing trend with an average decreasing rate of -12.1 μgm^{-3} per year from 2007 to 2011. These increasing and decreasing trends are in agreement with the evolution of industrial development in Xi'an. As shown in Fig. 5, the percentage of second industry, which mainly involves industry, construction, and transportation, decreased from nearly 60% in 1979 to 40% in 1996 (Wang, 2011), which explained the decreasing trend of $PM_{2.5}$ during the same timeframe. Luo et al. (2001) also reported a decreasing trend of aerosol optical depth (AOD) during 1979–1990. In agreement with the increasing trend of $PM_{2.5}$ concentration, the percentage of second industry slowly increased from 1997 to 2006 (Wang, 2011). These results further demonstrate that the regression model developed in this study is a reliable tool to estimate $PM_{2.5}$ concentrations in Xi'an. The retrieved historical $PM_{2.5}$ data can be used to evaluate the effects of aerosol particles on climate change and on human health such as epidemiological study. In addition, $PM_{2.5}$ concentration during 1979 and 2011 averaged as 128.2 μgm^{-3} in Xi'an, which was about three times of the class-two Chinese Air Quality Standard. The relationship between $PM_{2.5}$ concentration and second industry percentage infers that changing the industrial structure should be an effective approach to reduce $PM_{2.5}$ concentration in Xi'an or other Chinese cities with high $PM_{2.5}$ levels.

5. Conclusions

Available $PM_{2.5}$ and visibility data collected in Xian were analyzed in this study to first develop a regression model between $PM_{2.5}$ and visibility, and then retrieve historical $PM_{2.5}$ data from visibility data. Long-term trends of seasonal and annual average $PM_{2.5}$ were then analyzed, which showed a general increase in seasonal $PM_{2.5}$ during all the seasons and a decrease-sharp increase-decrease pattern in annual $PM_{2.5}$ during the period from 1979 to 2011. The trends of the annual average $PM_{2.5}$ during the past 33 years period considered in this study were in agreement with the changes of second industry proportion. Validation and

Table 2
Seasonal variations of visibility and $PM_{2.5}$ averaged during 1979–2011.

Visibility (km)						$PM_{2.5}$ (μgm^{-3})				
	Annual	Spring	Summer	Autumn	Winter	Annual	Spring	Summer	Autumn	Winter
mean	9.4	10.0	11.3	8.4	7.9	128.2	132.1	80.1	132.1	162.7
SD	4.3	2.3	3.4	2.5	2.3	51.8	40.5	33.8	40.5	57.5
Trend	−0.17	−0.12	−0.23	−0.17	−0.15	2.10	2.45	1.66	1.70	3.14
R	−0.64	−0.52	−0.67	−0.67	−0.63	0.52	0.54	0.48	0.42	0.53
P	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.015	<0.01
Maximum	14.4	14.8	17.8	12.6	12.3	195.2	206.7	157.1	206.7	274.6
Minimum	4.7	6.2	4.5	3.9	3.0	59.2	67.4	22.4	67.4	65.5

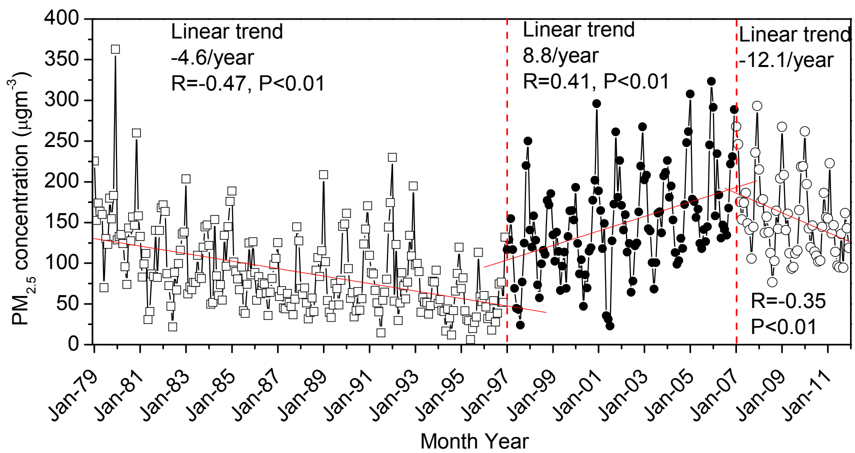

Fig. 4. Predicted monthly $PM_{2.5}$ concentrations for 1979–2011.

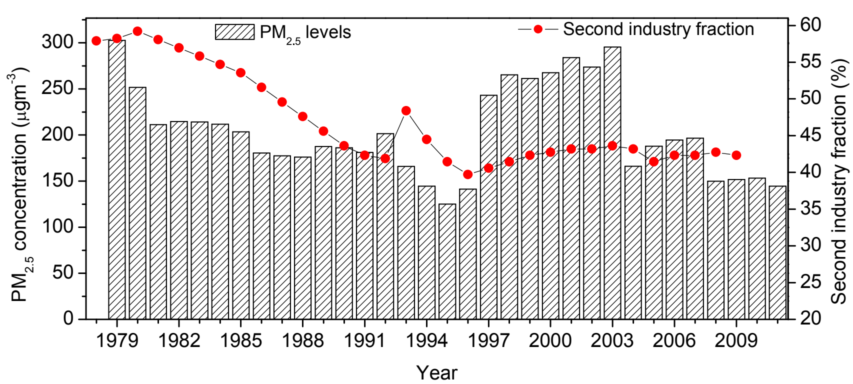

Fig. 5. Predicted annual average $PM_{2.5}$ concentrations and actual percentages of second industry in Xi'an for the past 33 years (1979–2011).

application of the regression model developed in this study confirm its usefulness in retrieving historical $PM_{2.5}$ data when only visibility data was available, which overcomes the difficulties of lacking of long-term $PM_{2.5}$ data that is needed in conducting aerosol effects studies on climate change and human health.

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