Aerosol and Air Quality Research, x: 1–13, xxxx

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2011.11.0220

Characterization of Regression Relationship between Recent Air Quality and Visibility Changes in Megacities at Four Haze Regions of China

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ABSTRACT

The Chinese government has put forward a series of aggressive control measures to tackle environmental problems such as visibility degradation since the first year of the 11th five-year plan (2006–2010). Recently recorded visibility, air quality and meteorological data in four major megacities (Beijing, Shanghai, Guangzhou and Chengdu) in different haze regions (and climatic zones) of China were analyzed with the aim of evaluating the extent to which the control measures have affected air quality and atmospheric visibility. The ambient concentrations of three major air pollutants (SO_2, NO_2 and PM_{10}) in these cities all decreased in years 2005–2009. However, improved visibility was observed only in Beijing and Guangzhou; it remained steady in Shanghai, and showed a decreasing trend in Chengdu. The results emphasized that the relationship between air quality and visibility is complex. Optimal empirical regression models were developed, based on measured air quality and meteorological parameter data, to better isolate possible causal relationships between visibility and air quality as well as meteorological conditions. Our results showed that the improvements of visibility in both Beijing and Guangzhou were due mainly to reduced PM_{10} concentration. In Guangzhou, improved atmospheric visibility was also helped by a reduction of SO_2 concentration in winter. In contrast, lower wind speed together with possible changes in fine particle concentration and composition could explain why an increasing visibility trend was not found in Shanghai or Chengdu.

Keywords: Atmospheric visibility; Air quality; Megacity; China.

INTRODUCTION

Atmospheric visibility degradation is a key issue in climatology and air pollution studies. It exerts adverse effects on humans' lives, such as in highway crowding and restricted aircraft movements. Recently, visibility in clear sky was found to be decreasing over land globally from 1973 to 2007 (Wang *et al*., 2009). In urban areas, visibility is a highly relevant visual indicator of air pollution level. Visibility reduction, other than on foggy days, is always associated with poor air quality caused by intensive emissions of air pollutants, which generate adverse health effects in humans (Watson, 2002).

Dramatic economic and industrial developments as well as vigorous urbanization in China have led to increased emission of various pollutants from urban areas, making visibility degradation one of the most severe environmental problems in such a rapidly changing country. Twenty-five years' visibility data (1981–2005) recorded by well-trained observers at 615 meteorological stations across China revealed that visibility in the country was degrading rapidly due to increased fossil fuel usage (Che *et al*., 2007, 2009).

Visibility impairment is generally accompanied by airborne particulate matter (PM), especially fine particles with aerodynamic diameters of 2.5 µm and less ($PM_{2.5}$), due to their light-scattering and absorption capabilities. Numerous studies around the world have focused on the relationship between visibility and aerosol composition (e.g. Dzubay *et al*., 1982; Sisler and Malm, 1994; Singh *et al*., 2008). In China, this relationship has also been widely investigated for individual cities. For examples, visibility degradation in Hong Kong has been found to be mostly due to fine sulfate particles (Lee and Sequeira, 2002; Cheung *et al*., 2005). Chemical speciation of aerosols in Guangzhou has indicated that fine particles, especially elemental carbon (EC), play a dominant role in visibility reduction (Deng *et al*., 2008).

However, most if not all of these studies were focused only on specific visibility degradation cases or seasonal variation. Although visibility is routinely monitored by the

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China Meteorological Administration (CMA), inter-annual variation is rarely examined due to the absence of long-term PM composition data. Routine observations of air quality and meteorological data are vital for interpreting the inter-annual changes of visibility and understanding its underlying causes. For instance, Tsai (2005) investigated the long-term trend of visibility in Taiwan (1961–2003) by analyzing the relationship between meteorological parameters and air quality data, and found that the parameter of ln(PM_{10}) (Nature logarithmic concentration of PM_{10}) had the most significant impact on visibility. Zhang *et al.* (2010) found that the decreasing trend in visibility in Beijing from 1999 to 2007 was attributed to an increase in relative humidity.

Since the beginning of the 11th five-year plan (2006–2010), aggressive steps taken by the Chinese government to tackle air pollution problems are expected to have a strong impact on regional air quality and atmospheric visibility. For example, the government encouraged the use of "cleaner" fuels such as liquefied petroleum gas (LPG) and low sulfur gasoline in industrial, transportation and domestic sectors The Flue-Gas Desulfurization (FGD) technique was widely adopted in power plants. These measures resulted in significant changes in atmospheric composition (e.g. Tang *et al*., 2008, Lu *et al*., 2010, Wan *et al*., 2011). Nevertheless, although decreasing trends of PM_{10} concentration have been recorded by ground-based measurements over the five years, satellite observations continue to show an upward trend in aerosol optical depth (Lin *et al*., 2010). Such discrepancies highlighted the complexity of the air pollution problem in China. A crucial question is whether the control measures really do improve air quality and visibility.

Beijing, Shanghai, Guangzhou and Chengdu are four major megacities located in four economically and industrially developed regions of China with different climates (Fig. 1). Beijing, as the capital of China, is the largest megacity, located at the center of the Beijing–Tianjing–Tangshan city cluster (BTT) in northern China. Shanghai and Guangzhou are the centers serving the Yangtze River Delta (YRD) in the east and the Pearl River Delta (PRD) in the south, respectively. The deltas are the most developed regions in eastern and southern China. Chengdu, located in the Sichuan Basin, is the most important megacity in southwestern China. Each of the four cities is also situated in a different haze region (Chang *et al*., 2009).

The present study presents newly recorded data in these four locations over a recent five-year period (2005–2009) to reveal the changes in visibility and air quality. An empirical regression model was developed to evaluate the effects of air quality and meteorological conditions on visibility, and to predict daily visibility based on air quality data and meteorological parameters.

DATA SOURCES AND METHOD

Descriptions of Measurement Sites and Data

Urban meteorology stations provided data for Beijing (39°48'N, 116°28'E), Shanghai (31°24'N, 121°27'E) and Guangzhou (23°10'N, 113°20'E) were used for analysis. The station in Chengdu (30°42'N, 103°50'E) was situated in a suburban area. Its data was used because no continuous observation was carried out at an urban station during the period. Considering the well-mixed surface layer with a capping inversion in urban mountain basins (e.g. Pataki *et al*., 2005), we suggest that this suburban data adequately

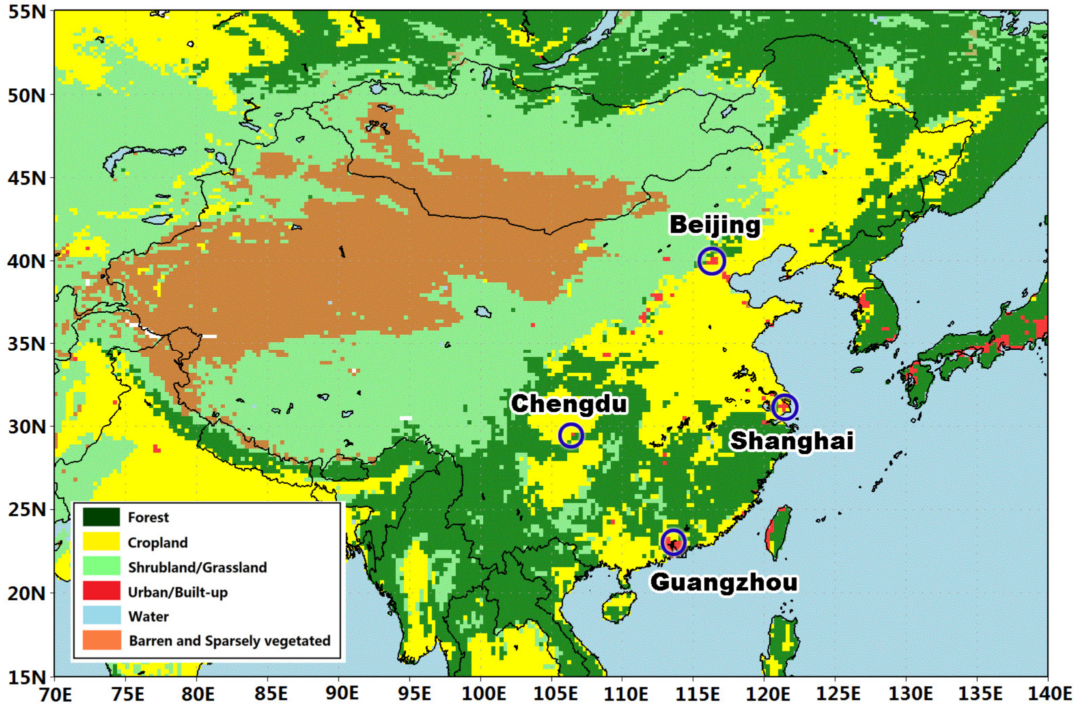

Fig. 1. Map showing the land-use pattern in China (based on 2004 MODIS products) and the locations of Beijing, Shanghai, Guangzhou and Chengdu

reflects urban pollution. The air quality monitoring stations were located in urban areas in all the cities studied. Table 1 provides a detailed description of the stations.

The visibility data used for analysis in this study was recorded by trained observer at 0200, 0800, 1400 and 2000 (local standard time) every day throughout 2005 to 2009, in according with standard CMA procedures. Other meteorological data—temperature, relative humidity (RH), precipitation and wind speed (WS)—were also recorded at these times. A daily mean was calculated for analysis in this study. Air quality data, including daily concentrations of SO_2, NO_2 and PM_{10}, was measured at the air quality monitoring stations managed and operated by the Environmental Protection Bureau (EPB), by UV fluorescence method analyzers (Thermo Electron Corporation Model 43c in Beijing, Shanghai and Guangzhou, and an API 100E in Chengdu), dual-channel chemiluminescence analyzers (Thermo Electron Corporation Model 42c in Beijing, Shanghai and Guangzhou, and an API 200E in Chengdu), and R&P TEOM Series 1400a ambient particulate monitors. The detection limit was 1 $\mu g/m^3$. The quality assurance (QA) and quality control (QC) procedures, including regular instrument calibration with Standard Reference Materials (SRM) or standards traceable to SRM, following Chinese government standard QA/QC requirements, were carried throughout the period at these stations. The data was stored as five-minute average values, from which daily average data was computed.

An overview of the general information of these four megacities is presented in Table 1. Care should be taken when comparing this data, especially visibility, with other previously reported observation results because of inherent systematic errors attributable to the limited number of stations in this study. The main focus was on the changes of visibility and air pollutant concentrations and their correlations in each individual city, rather than comparing the absolute values of visibility.

Time Series Analysis and Stepwise Regression Method

With an aim to better investigate the seasonal and inter-annual changes of visibility, a Kolmogorov-Zurbenko (KZ) filter was applied to the daily mean data set to separate the original time series into several components representing different time scale variation. A detailed description of KZ filter can be found in the literature (e.g. Rao and Zurbenko, 1994; Zurbenko *et al*., 1996; Eskridge *et al*., 1997). Briefly, the KZ filter is based on an iterative moving average that removes high-frequency variation from the data. In this study, a 15-day moving average procedure was repeated five times in order to remove all signals of less than $15\sqrt{5} \approx 33$ days: that is, the high-frequency meso- and synoptic scales were removed in the KZ (15,5) time series and allows seasonal signals to be identified (Eskridge *et al*., 1997). Seasonal variations and the signals of smaller time scales were removed by KZ filtering for a window size of 365 days and 3 iterations. The resulting KZ (365,3) time series, containing signals with time scales of greater than $365\sqrt{3} \approx 632$ days (1.7 year), and a fitted linear regression line were then used to reveal the inter-annual variation of visibility over the five-year period.

Visibility is affected by ambient air pollutant concentrations and meteorological conditions: in particular, degradation of visibility is directly proportional to the loading of airborne PM (light scattering and absorption); NO_2 also contributes due to its strong blue-light-absorbing capacity; and SO_2, which itself does not affect visibility but can oxidize to form sulfates, some of the most important atmospheric species scattering light and degrading visibility. On the

Table 1. General information, meteorological conditions and air pollutant concentrations in Beijing, Shanghai, Guangzhou and Chengdu in the period 2005–2009.

	Beijing	Shanghai	Guangzhou	Chengdu
Area (km^2)	16800	6300	7400	12400
Population (million)[a]	19.7	18.9	10.3	12.9
Visibility (km)[b]	16.9 ± 7.7	16.7 ± 5.5	12.4 ± 4.9	9.5 ± 2.7
Temperature (°C)[b]	13.6 ± 11.0	17.7 ± 9.1	23.1 ± 6.2	16.7 ± 7.5
Relative Humidity (%)[b]	51.3 ± 19.2	69.3 ± 11.5	70.1 ± 13.3	75.4 ± 9.7
Wind Speed (m/s)[b]	1.4 ± 1.0	2.4 ± 1.2	1.1 ± 0.6	0.6 ± 0.4
Total Precipitation (mm)	2316	6182	9843	4006
Air Quality Monitoring Station	Wanliu	Shanghai Normal University	Guangdong Business College	Caotang
SO_2 ($\mu g/m^3$)[b]	44 ± 50[c]	58 ± 38	48 ± 37	56 ± 32
NO_2 ($\mu g/m^3$)[b]	55 ± 26[c]	61 ± 25	54 ± 31	57 ± 20
PM_{10} ($\mu g/m^3$)[b]	141 ± 94[c]	92 ± 54	77 ± 48	120 ± 69
SO_2 Exceeding Rate[d]	5.1%	2.3%	2.4%	1.7%
NO_2 Exceeding Rate[d]	13.6%	20.4%	17.4%	11.7%
PM_{10} Exceeding Rate[d]	30.0%	12.5%	7.8%	23.8%

[a] Data in 2008.
[b] Average ± standard deviation.
[c] Data in the period of 2006–2009.
[d] Exceeding rate is defined as the percentage of episode days with daily concentration exceeding Grade II of the Chinese National Ambient Air Quality Standard (SO_2: 150 $\mu g/m^3$; NO_2: 80 $\mu g/m^3$; PM_{10}: 150 $\mu g/m^3$; GB3095-1996) within the study period.

other hand, meteorological conditions can also influence visibility by affecting air pollutant concentrations or their light-scattering capability. Specifically, aerosols grow in size when they interact with water vapor, causing increased scattering of sunlight in the atmosphere with high RH. High temperature and WS improve visibility by enhancing the dispersive capability of the atmosphere (via thermal and mechanical turbulence respectively) and reducing aerosol concentration levels.

In order to investigate the predominant factors affecting visibility and better predict visibility from air quality and meteorological data, an empirical model was developed incorporating the stepwise regression method. Generally, the stepwise procedure comprises an "enter" process and a "remove" process. The "enter" process allows each independent variable to be added into the regression equation only if it significantly increases the correlation of the equation. The independent variables are entered into the equation in descending order of their individual Pearson correlations with the dependent variable. In the present study, a significance level of 0.001 was adopted. After the new variable is entered into the regression equation, previously entered variables may lose some of their explanatory power. If the significance of a variable already in the regression equation falls below 0.005, it is removed from the equation. Daily visibility was set to be the dependent variable. Since visibility is generally associated with air quality (concentrations of SO_2, NO_2 and PM_{10}), temperature, RH and WS, these parameters were selected as the independent variables (Tsai, 2005). Note particularly that days with RH > 90% or rainfall were excluded from the regression analysis (Tsai, 2005; Chang *et al*., 2009).

RESULTS AND DISCUSSION

Characteristics of Air Pollution

Table 1 shows the "exceeding rates" of three major air pollutants (SO_2, NO_2 and PM_{10}) in the four megacities during 2005–2009. Exceeding rate was defined as the percentage of episode days with daily concentration exceeded Grade II of the Chinese National Ambient Air Quality Standard (NAAQS; GB3095-1996) within the study period. In Beijing and Chengdu, PM_{10} was the most dominant air pollutant, with exceeding rates (30% and 23.8% respectively) more than double those of the next most dominant air pollutant, NO_2. On the other hand, Shanghai and Guangzhou were mainly affected by NO_2 (with exceeding rates of 20.4% and 17.4% respectively). The severe PM_{10} pollution in Beijing and Chengdu is probably because these two cities are always affected by dust storms (e.g., Zhang *et al*., 2005, 2010; Tao, J., unpublished data). Low precipitation in these two cities may be another reason; this would prolong the lifetime of aerosols in the atmosphere (Table 1). In Chengdu the stable atmosphere reflected by low WS also favors the accumulation of particulate matter (Table 1). (Similar meteorological conditions and their effect on air pollution have also been found in Chongqing, another basin city near Chengdu (Yang *et al.*, 2008).) The NO_2 pollution in Shanghai and Guangzhou highlights the potential role of photochemical pollution in these two cities. This result agrees well with previous satellite studies that showed that the YRD, PRD and BTT regions were the major NO_2-polluted areas in China (van der A *et al*., 2006).

Recent Changes in Visibility and Air Quality

Fig. 2 shows the visibility time series of average daily KZ (15,5) and KZ (365,3) values, as well as a linear regression line fit to the KZ (365,3) data. Generally, visibility in Shanghai, Guangzhou and Chengdu presented more or less similar seasonal patterns, with maximums in summer and minimums in winter negatively related to those of air pollutant concentrations (not shown). Such seasonal cycles are widely observed in China and can be explained by the seasonal variation of meteorological conditions dominated by the East Asian monsoon (e.g., Chang *et al*., 2009; Wan *et al*., 2011). However, visibility in Beijing presented a different seasonal pattern, with a maximum in winter and a minimum in summer. Similar seasonal patterns can be found after excluding precipitation, and therefore we do not believe that a higher rainfall in summer (not shown) to be one of the important causes. In fact, the degradation of visibility in Beijing in summer has been observed in other studies, and was due to the transport of air pollutants contributed by industrial emissions, vehicular fossil fuel combustion and agricultural biomass burning from upwind region such as Hebei and Shandong Provinces (Li and Shao, 2009; Li *et al*., 2010). As illustrated by Wang *et al*. (2010), this unique seasonal pattern may also be partly owing to the high RH in summer (approximately 30% higher than that in winter; data not shown), while the RH in the other three cities were more or less consistent throughout the year.

Visibility in Beijing improved from 2005 to 2009 at a growth rate of 0.58 km/year (R^2 = 0.497). Although Chang *et al*. (2009) suggested that visibility in Beijing was decreasing with a rate of –0.78 km/decade between 1973 and 2007, the increasing visibility observed here is identical to the result of Che *et al*. (2009), who found that visibility in Beijing began to improve after 1995 due to the stricter environmental control measure implemented. Visibility in Shanghai has been improving since 1990 (Chang *et al*., 2009; Che *et al*., 2009) but, in recent years, the annual variation of visibility in Shanghai was relatively small and did not show a significant increasing trend (R^2 = 0.008) during 2005–2009. Although visibility in Guangzhou was dramatically degrading in the 1970s, it was maintained at a relatively steady level and seemed to begin improving since 2005 (Deng *et al*., 2008; Chang *et al*., 2009). In fact, with a notable growth rate of 0.83 km/year (R^2 = 0.825), the visibility in Guangzhou did improve from 2005 to 2009, which was the most pronounced increase in any of these four cities. Visibility in Chengdu has been degrading in the past three decades (Chang *et al*., 2009; Che *et al*., 2009). In contrast to the cases of Beijing, Shanghai and Guangzhou, visibility in Chengdu in the past five years did not stabilize or improve, but continued to decrease (–0.21 km/year, R^2 = 0.885).

Table 2 summarizes the annual averages of the meteorological parameters and major air pollutant concentrations in the four megacities. Most of meteorological parameters did not show significant trends except the decreases of RH in Guangzhou and Chengdu as well as WS in Shanghai and Chengdu. Such decreases are probably due to rapid urbanization, which would result in an urban heat-island effect (Brazel and Balling, 1986) and increased surface roughness (Vautard *et al*., 2010), although longer data records are still needed for further investigation. These results imply that the observed visibility changes are more related to the changes of air quality.

In fact, significantly decreasing trends were observed in all air pollutant concentrations (Table 2), suggesting that air quality was improving throughout the five-year period. Concentrations of SO_2 in Beijing continued to decrease from 2006 to 2009, while those in Shanghai, Guangzhou and Chengdu decreased sharply following increases in 2005–

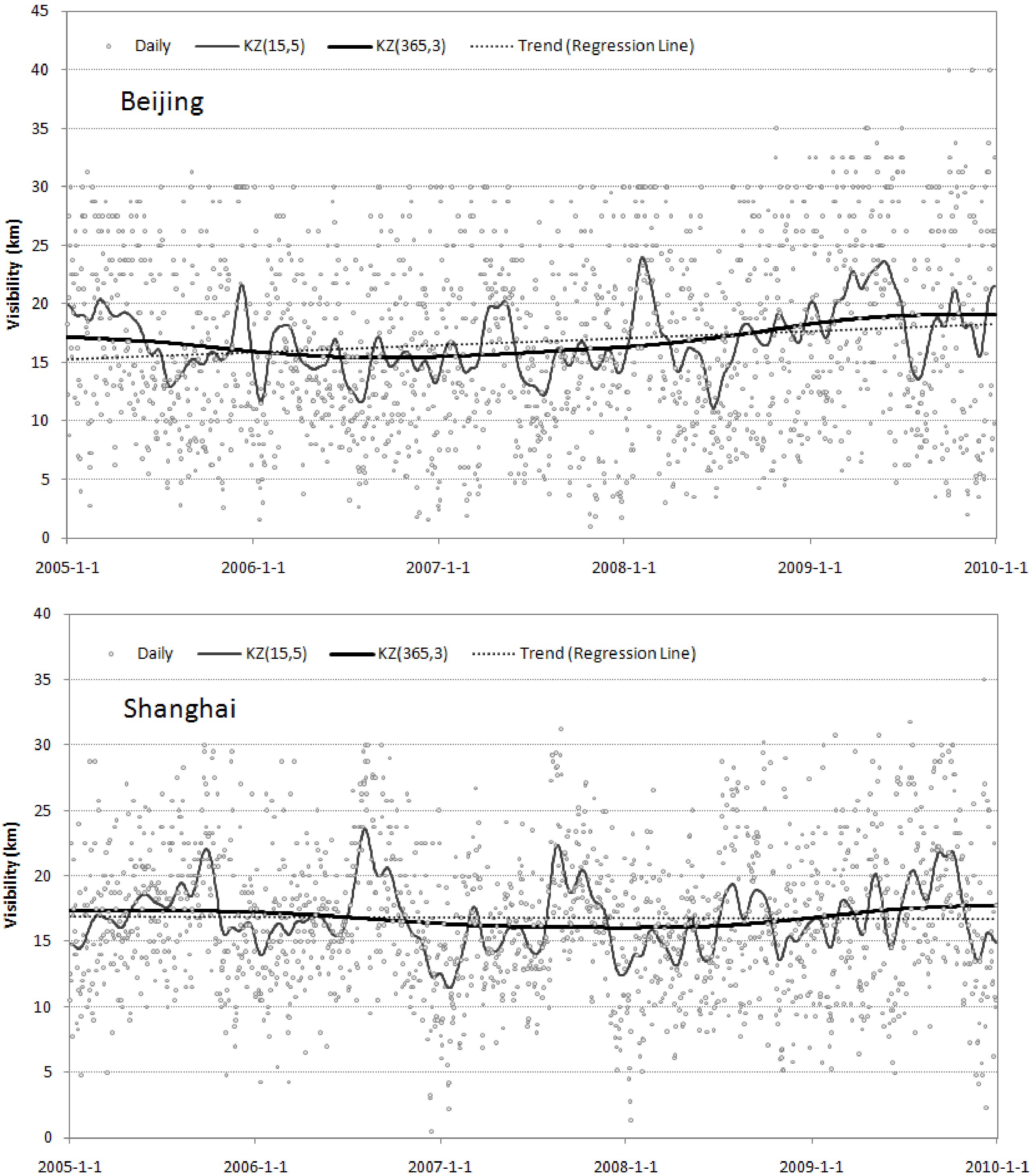

Fig. 2. Visibility time series of daily mean (circle), KZ(15,5) (thin line), KZ(365,3) (thick line) and a linear regression line fit to KZ(365,3) (dot line) from 2005-1-1 to 2009-12-31 in Beijing, Shanghai, Guangzhou and Chengdu.

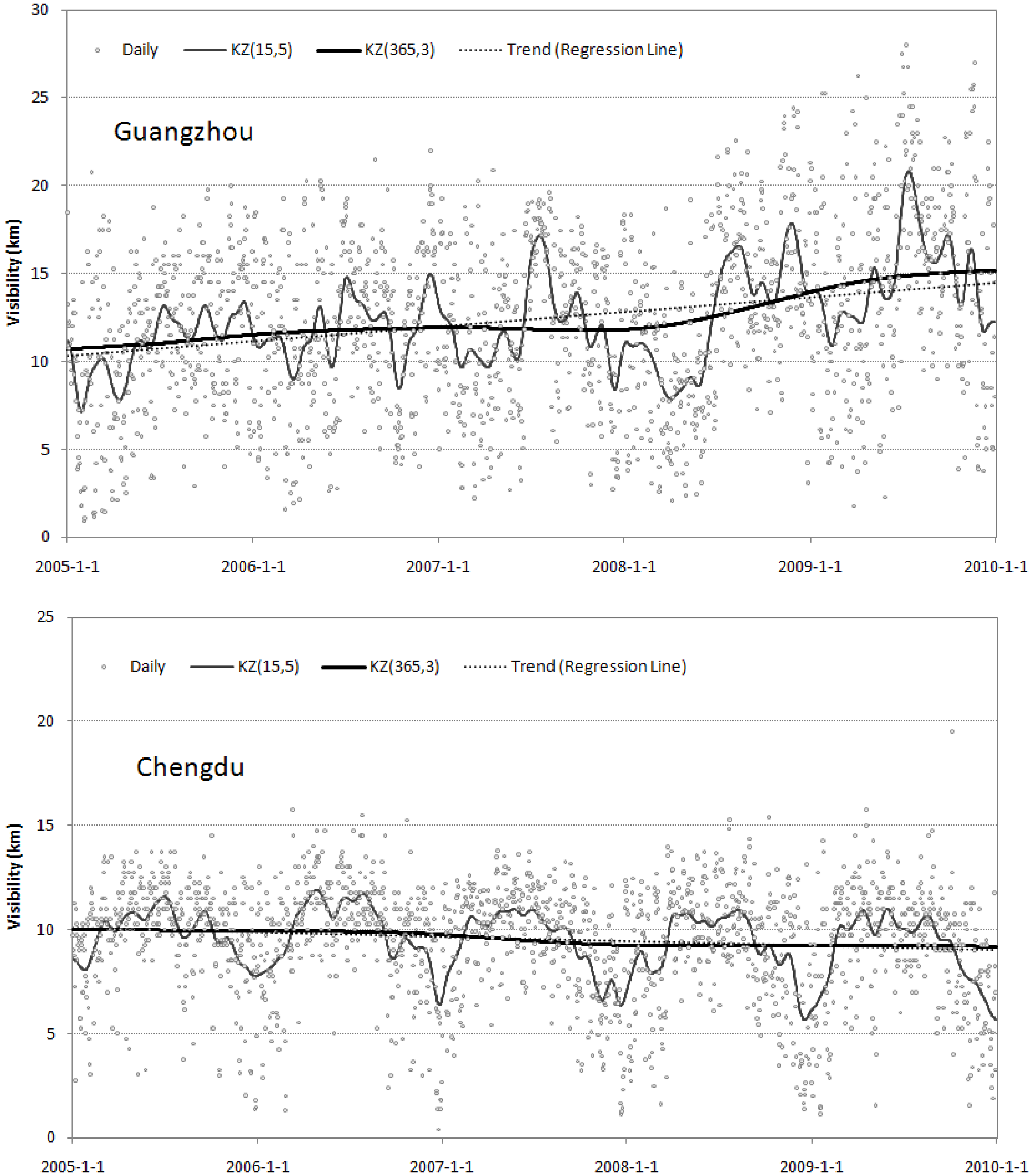

Fig. 2. *(continued).*

2007. Such decreasing trends, especially for sharp decreases in 2007–2008, were probably due to the implementation of gas-desulfurization in power plants (Lu *et al*., 2010). Slight decreases in NO_2 concentration were also observed in all four megacities, in contrast to the increasing trend from 1997 to 2006 based on satellite measurements (Zhang *et al*., 2007). Marked decreasing trends of particulate pollution were attributed to the strong efforts of the Chinese government to control particulate emissions from various industrial sources (http://www.mep.gov.cn/). Recently, a similarly notable decreasing trend in PM_{10} concentration was also observed in Foshan, an industrial city in the PRD (Wan *et al*., 2011). However, it is worthwhile pointing out that the concentrations of air pollutants, especially SO_2 and PM_{10}, were still remarkably higher than those in most developed countries (Wan *et al*., 2011).

Table 2. Annual means and inter-annual trends of air pollutant concentration and meteorological parameters in four megacities.

Air Pollutant	City	2005	2006	2007	2008	2009	Trend
SO_2 (μg/m^3)	Beijing	n.a.	56	44	41	35	–6.5 (R^2 = 0.93)
	Shanghai	68	69	72	48	35	–8.7 (R^2 = 0.73)
	Guangzhou	49	53	56	42	38	–3.5 (R^2 = 0.50)
	Chengdu	65	62	72	48	39	–6.9 (R^2 = 0.60)
NO_2 (μg/m^3)	Beijing	n.a.	60	54	51	55	–1.9 (R^2 = 0.44)
	Shanghai	63	67	65	59	51	–3.0 (R^2 = 0.64)
	Guangzhou	57	59	54	51	49	–2.4 (R^2 = 0.81)
	Chengdu	59	58	57	55	55	–1.2 (R^2 = 0.92)
PM_{10} (μg/m^3)	Beijing	n.a.	168	142	127	127	–13.8 (R^2 = 0.85)
	Shanghai	96	103	90	88	86	–3.6 (R^2 = 0.64)
	Guangzhou	102	79	69	72	62	–8.8 (R^2 = 0.81)
	Chengdu	130	133	108	115	112	–5.4 (R^2 = 0.58)
Temperature (°C)	Beijing	13.4	13.6	14.1	13.5	13.5	0.0 (R^2 = 0.00)
	Shanghai	17.2	18.0	18.3	17.3	17.6	0.0 (R^2 = 0.00)
	Guangzhou	22.9	23.3	23.3	22.6	23.1	0.0 (R^2 = 0.00)
	Chengdu	16.4	17.1	17.0	16.4	16.9	0.0 (R^2 = 0.00)
Relative humidity (%)	Beijing	48.5	52.1	53.6	51.8	50.5	0.0 (R^2 = 0.09)
	Shanghai	69.6	69.3	68.2	69.6	69.7	0.0 (R^2 = 0.01)
	Guangzhou	70.3	70.8	70.0	70.2	69.1	–0.3 (R^2 = 0.59)
	Chengdu	77.2	75.9	75.8	74.7	73.2	–0.9 (R^2 = 0.94)
Wind speed (m/s)	Beijing	1.54	1.33	1.31	1.36	1.42	–0.02 (R^2 = 0.11)
	Shanghai	2.75	2.50	2.37	2.27	2.24	–0.13 (R^2 = 0.91)
	Guangzhou	1.14	1.08	1.10	1.13	1.09	–0.01 (R^2 = 0.14)
	Chengdu	0.75	0.73	0.66	0.56	0.53	–0.06 (R^2 = 0.95)

These results emphasize the important role of the Chinese government's efforts in air quality improvement. Specifically, the hosting of international events, such as the 2008 Olympic Games in Beijing, the 2010 World EXPO in Shanghai and the 2010 Asian Games in Guangzhou, may have presented a precious opportunity for China to push and enforce the implementation of emission control measures to improve air quality. The global financial crisis and China's economic recession may also have exerted certain impacts on air quality improvement. However, the discrepancy between visibility and air quality changes in Shanghai and Chengdu highlight their complex relationship, which is discussed below.

Dependence of Visibility on Air Quality and Meteorological Conditions

As discussed, variation of visibility usually results from changes in both air quality and meteorological conditions. Fig. 3 presents the average visibility of the four megacities in various cases. "Episode day" was defined as those days on which the concentration of air pollutant (SO_2, NO_2 or PM_{10}) exceeded the recommended NAAQS II level. Other days were defined as "normal" days. For each of the meteorological parameters temperature, RH and WS, "high" days were defined as days on which the daily mean exceeded the 2005–2009 average, while the others were characterized as "low" days.

Fig. 3 shows that visibility on SO_2, NO_2 and PM_{10} episode days was noticeably reduced compared to normal days, emphasizing the effects of air pollution on visibility degradation mentioned above. The average visibility in various meteorological conditions is also presented in Fig. 3. The lower visibility in high RH cases highlights the impact of RH on the light-extinction capacity of aerosols. As discussed, high WS and temperature enhance the dispersion of air pollutant and thus improve visibility, which is also supported by the present results. Note the exception of Beijing, which experiences lower visibility on high-temperature days. This is probably due to the fact that visibility in Beijing has always been degraded in summer, as illustrated previously, since it is related to air pollutant transport and high humidity (Li & Shao, 2009; Li *et al*., 2010; Wang *et al*., 2010).

To better understand the dependence of visibility on air quality and meteorological conditions, it was necessary to calculate the correlation between them. In fact, the Pearson correlations between visibility and temperature, RH, WS, SO_2, NO_2 and PM_{10} concentrations in the four megacities were all significant at the $p = 0.001$ level. Therefore, a stepwise multiple regression method was applied to further examine their underlying relationships, and which would consider their mutual impact. A model using this approach was developed to simulate daily visibility based on air quality and meteorological parameter data. To enable the validity of the model to be evaluated, only data from 2005 to 2008 was used in its development (Table 3); the model was then interrogated by the newly recorded 2009 data (Fig. 4). Overall, with the exception of Beijing, the model tended to overestimate or underestimate the visibility in many cases (Fig. 4), suggesting that it was too ambitious to expect a single regression model to be able to predict the visibility

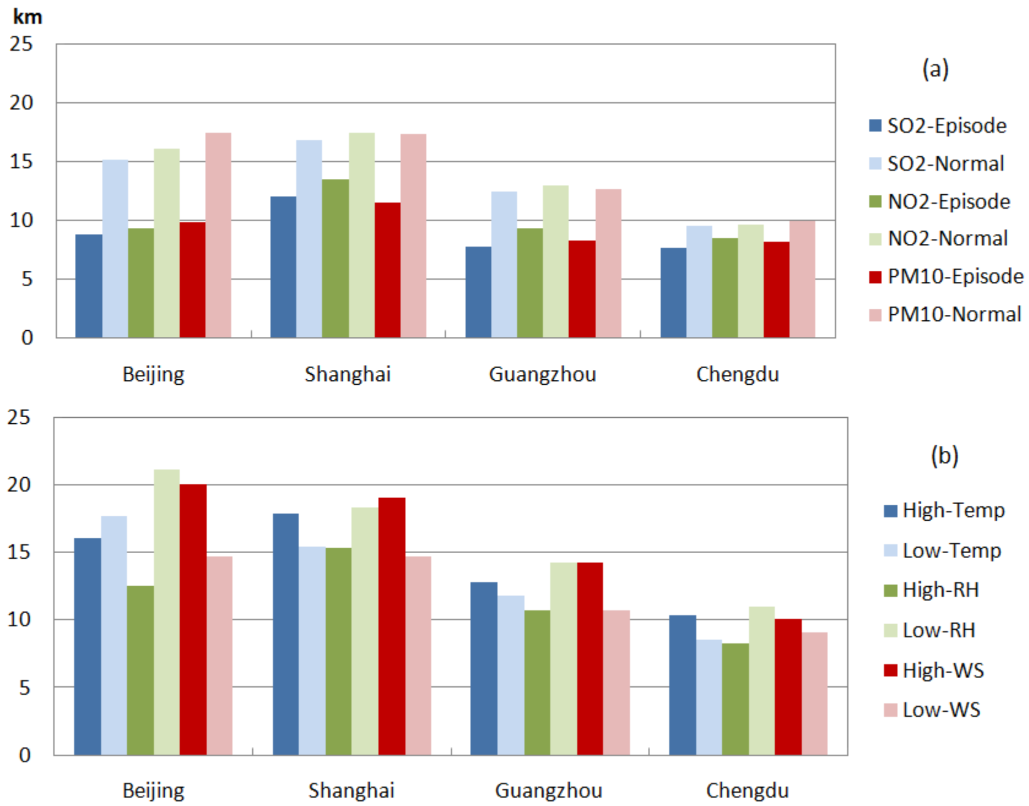

Fig. 3. Visibility means in (a) air pollutants (SO_2, NO_2 or PM_{10}) episode or normal days and (b) days with various meteorological conditions. ("Episode" or "Normal" stands for the days with air pollutant concentration exceeding or below the daily NAQQS II respectively; "High" or "Low" stands for the days with daily mean of meteorological parameter higher or lower than the average of 2005–2009 respectively; "Temp", "RH" and "WS" stand for temperature, relative humidity and wind speed respectively.)

in such a wide range of situations. Accordingly, an optimal empirical regression model was proposed and developed to produce a formula set containing two stepwise regression models, as described below.

Optimal Empirical Regression Models for Visibility

The optimal empirical regression models for each megacity are shown in Table 3. The original data for each megacity was grouped into two categories and the detailed grouping procedure appears in the Appendix. That is, each optimal model is a formula set containing two stepwise regression models as foreshadowed above. Overall, as revealed by the higher determination coefficients R^2 (where $R^2 \times 100\%$ of data can be explained by the model), the optimal regression models provide better prediction capacity than the original models did (Table 3).

To further verify the validity of the optimal model, daily observed visibility data for 2009 were examined by linear regression, which regards simulated visibility as an independent variable and observed visibility as the dependent variable. The results confirm that the newly developed optimal empirical regression model simulated the observed visibility data more accurately than the original stepwise regression model did, giving a higher R^2 value, a slope approaching 1 and an intercept approaching 0 (Fig. 4). As examples, in some cases for Beijing (e.g. 2009-2-9 and 2009-12-24) which the original model had underestimated, the optimal empirical model now gave improved results. For Shanghai, in some cases (e.g. 2009-6-18), the optimal empirical model performed better than the original model had done, although it was still far from exactly accurate. For Guangzhou, the optimal empirical model simulated visibility more accurately in summer. In Chengdu, although R^2 in the correlation between simulated and observed visibility was slightly less than that in the original model, both the slope and intercept were remarkably smaller. The original model had tended to overestimate the visibility in several cases (e.g. 2009-1-18), but the newly developed model did not.

Possible Causes of Visibility Changes

As well as the ability to better simulate daily visibility, the new optimal empirical stepwise regression model developed in this study also provided clues to answering the question of the extent to which air quality and meteorological parameters affect visibility.

In Beijing, visibility was affected by concentrations of PM_{10} and NO_2 as well as RH, when RH was above the five-year average. In other cases, visibility was influenced only by PM_{10} concentration and RH. Such results indicated that PM_{10} concentration was the common factor in visibility degradation in Beijing. Considering that the increasing trend

Table 3a. Summary of original stepwise regression models.[a]

	Stepwise regression model	R^2
Beijing	[Vis] = −0.22 × [RH] − 0.03 × [PM_{10}] − 0.06 × [NO_2] + 34.83	0.76
Shanghai	[Vis] = −0.05 × [PM_{10}] − 0.16 × [RH] + 0.77 × [WS] + 0.07 × [Temp] + 29.79	0.56
Guangzhou	[Vis] = −0.04 × [PM_{10}] − 0.19 × [RH] + 0.09 × [Temp] + 26.33	0.50
Chengdu	[Vis] = −0.14 × [RH]− 0.01 × [PM_{10}] + 0.07 × [Temp] + 0.68 × [WS] + 20.10	0.56

Table 3b. Summary of optimal empirical stepwise regression models.

	Optimal empirical stepwise regression model	R^2
Beijing	If [RH] < 51%, [Vis] = −4.75 × ln[PM_{10}] − 0.07 × [NO_2] − 0.15 × [RH] + 51.52 Otherwise, [Vis] = −7.51 × ln[PM_{10}] − 0.27 × [RH] + 67.31	0.80
Shanghai	If [WS] < 3 m/s, [Vis] = −7.05 × ln[PM_{10}] − 0.18 × [RH] + 0.05 × [Temp] + 59.27 Otherwise, [Vis] = −7.12 × ln[PM_{10}] − 0.23 × [RH] + 0.13 × [Temp] + 62.08	0.61
Guangzhou	If [Temp] < 23°C, [Vis]= −0.18 × [RH] − 2.63 × ln[PM_{10}] − 0.02 × [SO_2] + 35.86 Otherwise, [Vis]= −5.17 × ln[PM_{10}] + 0.52 × [Temp] − 0.15 × [RH] + 30.44	0.57
Chengdu	If [RH] > 75% and [PM_{10}] > 100 μg/m^3, [Vis] = −0.30 × [RH] + 0.08 × [Temp] + 2.01 × [WS] − 0.01 × [PM_{10}] + 30.01 Otherwise, [Vis] = −0.12 × [RH] − 0.01 × [PM_{10}] + 0.05 × [Temp] + 0.56 × [WS] + 18.36	0.58

[a] [Vis], [Temp], [RH], [WS], [SO_2], [NO_2] and [PM_{10}] stand for visibility (unit: km), temperature (unit: °C), relative humidity (unit: %), wind speed (unit: m/s), concentrations of SO_2, NO_2 and PM_{10} (unit: μg/m^3) respectively.

of improved visibility (0.58 km per year) coincided with a decreasing trend in PM_{10} concentration (13.8 μg/m^3 per year), it is reasonable to conclude that the reduction of PM_{10} has been the main reason for visibility improvement.

In Shanghai, visibility was related to PM_{10} concentration, RH and temperature. Although a deceasing trend was observed in PM_{10} concentration, visibility did not show any significant improvement. In a recent study, Lin *et al.* (2010) suggested that PM_{10} emission control policy had not been successful in reducing concentrations of fine particles, which are more effective in scattering light and degrading visibility, due to the formation of secondary fine particles from precursor species. Therefore, the steady level of visibility was probably due to the similarly steady level of $PM_{2.5}$ concentration, although the concentration of coarse particles decreased significantly; unfortunately, long-term observation data of $PM_{2.5}$ concentration was not available for the present study, and this hypothesis could not therefore be tested. Our results imply that long-term observation of $PM_{2.5}$ concentration and composition is of importance for improved studies of the causes of long-term visibility change. Note particularly that if the entrance and removal significant levels in stepwise regression method are set to be 0.05 and 0.10 respectively, WS is one of the significant independent variables included in the model of "Low WS" cases (with a coefficient of 0.639 km per m/s, not shown), highlighting the potential impact of WS on visibility: in other words, the decreasing trend of WS could be a possible factor in maintaining a steady visibility level. Nevertheless, the exact cause of visibility change in Shanghai was difficult to explain currently and needs to be further investigated.

In Guangzhou, visibility was affected by RH and concentrations of PM_{10} and SO_2 when temperatures were below the five-year average. Otherwise, temperature exerted a positive impact on visibility, while no significant correlation was found between visibility and SO_2 concentration. This result suggested that high temperatures were of importance to the dispersive capability and of atmospheric visibility (usually in summer, average 29.0°C), but had negligible effect in other cases when the SO_2 concentration exerted a negative impact on visibility and the influence of PM_{10} concentration was smaller. As mentioned before, SO_2 impairs visibility through the formation of secondary fine sulfate particles which scatter light efficiently, and therefore the observed significant impact of SO_2 on visibility degradation at low temperatures implied that the oxidation of SO_2 and formation of sulfates were probably enhanced in winter (average temperature 15.6°C). It is worthwhile noting that, from the $PM_{2.5}$ chemical speciation data observed in the period mid-2009 to early 2010, elevated concentrations of sulfate and high ratios of sulfate: (SO_2 + sulfate) were detected in autumn and winter (J. Tao, unpublished data), which supported this hypothesis and highlighted the crucial role of secondary sulfate formation on visibility degradation at low temperatures. This result also agrees with the fact that SO_2 dissolves more readily in water and oxidizes into secondary sulfates via heterogeneous reactions at low temperatures (Ota and Richmond, 2011). A similar relationship between

SO_2 and visibility impairment was also found in Hong Kong, another megacity in the PRD (Lin *et al.*, manuscript in preparation). Overall, the observed improvement in visibility may reasonably be attributed to prominent decreases of PM_{10} concentration. Significant decreases of SO_2 concentration in winter (from 64 µg/m^3 in 2005 to 37 µg/m^3 in 2009; data not shown) also exerted a positive impact on the improvement of visibility.

In Chengdu, visibility was affected by PM_{10} concentration, RH, temperature and WS. In particular, the negative effect of RH and positive influence of WS on visibility were much more vital when RH and PM_{10} concentrations were high. Although PM_{10} concentration and RH both decreased in the past five years (from 130 µg/m^3 and 77.2% in 2005 to 112 µg/m^3 and 73.1% in 2009, respectively), visibility in Chengdu did not improve but continued to deteriorate. The observed

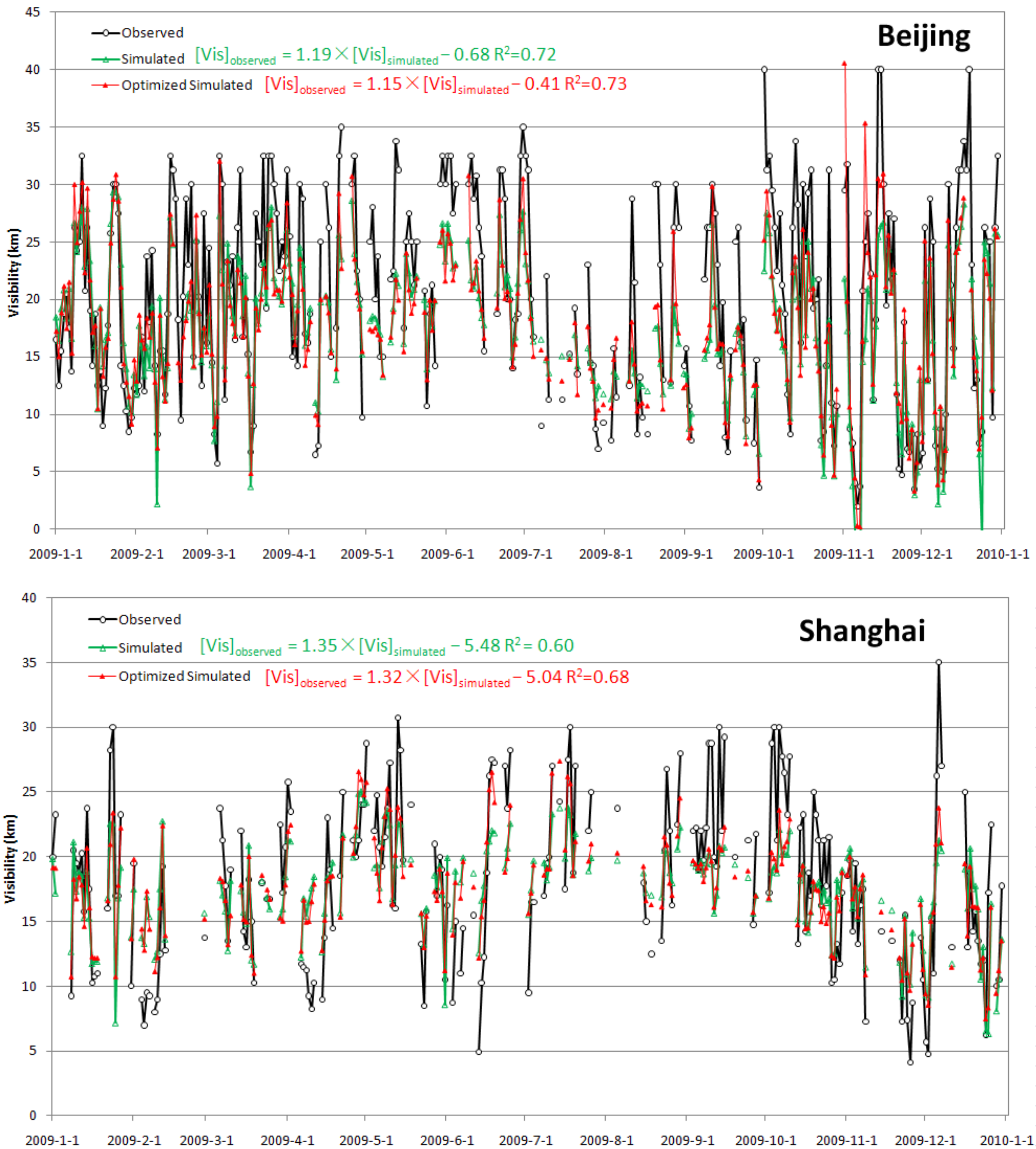

Fig. 4. Time series of daily observed visibility (black) and daily visibility simulated by original stepwise regression model (green) and optimal empirical regression model (red) in 2009 in Beijing, Shanghai, Guangzhou and Chengdu.

Fig. 4. *(continued).*

decreases in visibility were probably due to the reduction of WS (0.75 m/s in 2005 to 0.53 m/s in 2009), although the coefficient was just 2.01 km per m/s, too low to trigger the notably decreasing trend in visibility. This indicates that visibility degradation in Chengdu was still affected by other unknown factors. As mentioned previously, visibility is much more related to fine particle concentration than coarse particle concentration due to their different scattering effects. The decreasing trend of visibility in Chengdu was thus probably caused increased $PM_{2.5}$ concentration as well—which, however, could not be investigated in this study. Note particularly that, as mentioned above, the air quality monitoring station in Chengdu was located about 10 km from the meteorology observation station (located in the suburban area), which in itself might suggest another reason for the observed discrepancy.

CONCLUSION

Visibility degradation has been widely observed in China. In this study, the recent changes of visibility in four major megacities were investigated: Beijing, Shanghai, Guangzhou and Chengdu. Optimal empirical regression models were developed to investigate the underlying causal relationships between visibility, air quality and meteorological conditions. The models also provide a simple tool for predicting daily visibility with regard to air pollutants (SO_2, NO_2 and PM_{10}) and meteorological data (temperature, RH and WS) in the future.

Because of the stricter air pollutant emission control regulations imposed by the government, air quality in terms of three major air pollutant concentrations was observed to have improved in these four megacities during the five years 2005–2009, although pollution levels were still higher than in other megacities in developed countries. Similar significant improvements in atmospheric visibility were observed in Beijing (0.58 km/year) and Guangzhou (0.83 km/year). However, visibility was slightly degraded in Chengdu (–0.21 km/year). In Shanghai, visibility decreased slightly before 2007, then increased somewhat after 2007. Results of regression analysis have suggested that the improvements of visibility in Beijing and Guangzhou were mainly due to decreases in PM_{10} concentrations. Moreover, reduction of SO_2 concentration in Guangzhou also exerted a positive effect on visibility improvement there. On the other hand, the exact reasons for the lack of visibility improvement in Shanghai and Chengdu are not yet clear. The decreasing trend of wind speed, and therefore a weaker atmospheric dispersive capability, is probably one reason. Changes of $PM_{2.5}$ concentrations and compositions might also affect the visibility change in all of these cities, especially in Shanghai and Chengdu. These results highlight that policy measures which focus upon PM_{10} reduction as the air quality metric may not achieve corresponding improvements in visibility. A longer time data set will help to more clearly identify the trends.

ACKNOWLEDGEMENTS

This study was supported by the Special Scientific Research Funds for Environment Protection Commonweal Section (No. 200809143 & No. 201009001), the Fundamental Research Funds for the Central University (No. 2010380003161540), the National Natural Science Foundation of China (No. 40875075), a joint fund of the National Natural Science Foundation of China and Natural Science Foundation of Guangdong Province, China (No. U0833001), the Fundamental Research Funds for the Central Universities and the Trans-Century Training Programme Foundation for the Talents offered by the Ministry of Education of China.

SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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Received for review, November 29, 2011
Accepted, February 19, 2012

Appendix:

In this study, we tried several possible ways of grouping the data into two categories for each megacity, and various regression models in terms of a formula set containing two stepwise regression models were derived. The models with best simulation results were regarded as the optimal empirical regression models (while the others were not shown). The grouping methods of optimal empirical regression models in each city and their reasons are summarized below.

Relative humidity in Beijing shows a large variation compared to other three cities (Fig. a). Given that the relationship between particle scattering capability and relative humidity is nonlinear, the impacts of relative humidity on visibility in high or low relative humidity environments are different (Malm & Day, 2001, Wang et al., 2010). Therefore, we defined the cases with relative humidity > 51% (mean relative humidity in Beijing) as "High RH" cases; otherwise they were classified as "Low RH" cases.

In Shanghai, hazy days with heavy aerosol pollution always occurred on days with low wind speed (Fu et al., 2008). In this study we found that the impact of wind speed variation on visibility change was different on days when wind speed was greater or less than 3 m/s. In particular, when wind speed was < 3 m/s, the correlations between wind speed and visibility (R^2=0.140) and the slope of linear regression (2.45 km per m/s) were both stronger than those when wind speed was > 3 m/s (R^2=0.057, 1.78 km per m/s respectively) (Fig. b). Therefore, "High WS" and "Low WS" cases with wind speed greater and less than 3 m/s respectively were grouped.

In Guangzhou, negative regression slope (-0.451) between temperature and visibility was found when temperature was < 23°C (mean temperature in Guangzhou). In contrast, the slope was positive (0.770) when temperature was > 23°C (Fig. c). This result suggested that the dispersive effect of high temperature on visibility improvement was observed only on days with temperature > 23°C. As a result, we grouped the dataset into two groups (i.e., "High TEMP" cases with temperature higher than 23°C and "Low TEMP" cases otherwise).

Furthermore, Tsai (2005) found that the parameter of ln[PM_{10}] could better predict the impacts of PM_{10} on visibility than [PM_{10}] (concentration of PM_{10}). In this study, we also found that correlation between [PM_{10}] and visibility in Beijing was –0.647, while that between ln[PM_{10}] and visibility could be raised to 0.740. Such a phenomenon was also observed in the data of Shanghai and Guangzhou, where the correlations between ln[PM_{10}] and visibility (0.688 and 0.559 respectively) were slightly larger than that between [PM_{10}] and visibility (–0.657 and –0.532 respectively). As a result, we used ln[PM_{10}] rather than [PM_{10}] as an independent variable in the optimal empirical regression model for Beijing, Shanghai and Guangzhou.

In Chengdu, the original model tended to underestimate visibility when both PM_{10} concentration and relative humidity were high (Fig. d), which was probably because of the nonlinear growth of particle scatting capability in high relative humidity environment (Malm & Day, 2001). We thus defined "episode day" as a day with relative humidity > 75% (mean relative humidity in Chengdu) and PM_{10} concentration > 100 $\mu g/m^3$ (annual average of NAAQS II), then separated the data into two groups (i.e., "episode day" and "non-episode day").

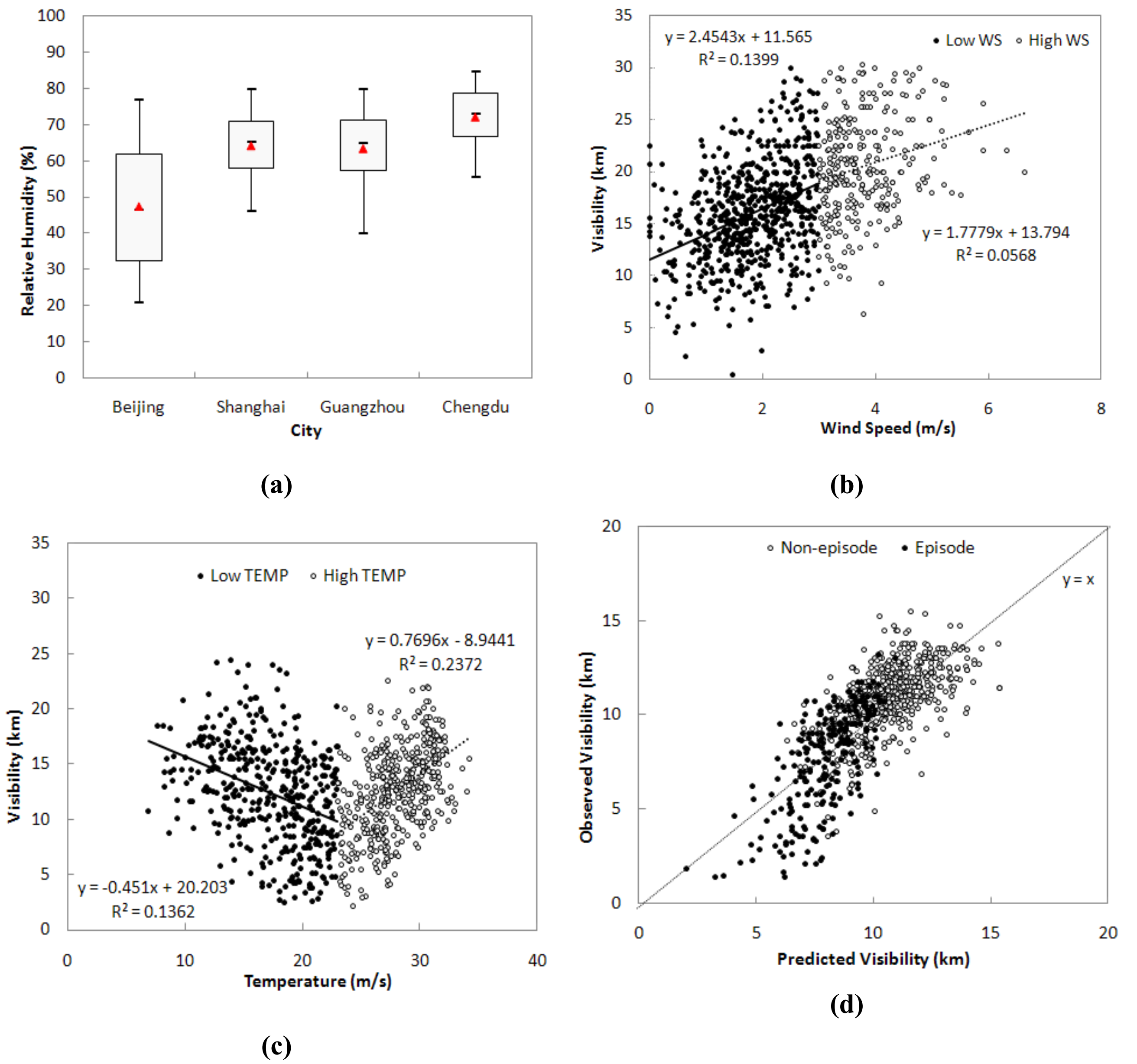

Fig. (a) Different percentiles (box & whisker) and average (triangle) of relative humidity in Beijing, Shanghai, Guangzhou and Chengdu; (b) Scatter plot of visibility and wind speed in Shanghai; (c) Scatter plot of visibility and temperature in Guangzhou; (d) Scatter plot of observed and predicted visibility in Chengdu.