Research Article

Twelve-Year Trends of PM₁₀ and Visibility in the Hefei Metropolitan Area of China

Lin Huang, Mindong Chen, and Jianlin Hu

*Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control,
Jiangsu Engineering Technology Research Center of Environmental Cleaning Materials,
Collaborative Innovation Center of Atmospheric Environment and Equipment Technology,
School of Environmental Science and Engineering, Nanjing University of Information Science and Technology,
219 Ningliu Road, Nanjing, Jiangsu 210044, China*

Correspondence should be addressed to Jianlin Hu; hu_jianlin@126.com

Received 12 November 2015; Accepted 18 January 2016

Academic Editor: Renate Forkel

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China has been experiencing severe air pollution and previous studies have mostly focused on megacities and a few hot spot regions. Hefei, the provincial capital city of Anhui province, has a population of near 5 million in its metropolitan area, but its air quality has not been reported in literature. In this study, daily PM₁₀ and visibility data in 2001–2012 were analyzed to investigate the air quality status as well as the twelve-year pollution trends in Hefei. The results reveal that Hefei has been suffering high PM₁₀ pollution and low visibility during the study period. The annual average PM₁₀ concentrations are 2–3 times of the Chinese Ambient Air Quality Standard. PM₁₀ shows fluctuating variation in 2001–2007 and has a slightly decreasing trend after 2008. The annual average visibility range is generally lower than 7 km and shows a worsening trend from 2001 to 2006 followed by an improving trend from 2007 to 2012. Wind speed, precipitation, and relative humidity have negative effects on PM₁₀ concentrations in Hefei, while temperature could positively or negatively affect PM₁₀. The results provide a general understanding of the status and long-term trends of PM₁₀ pollution and visibility in a typical second-tier city in China.

1. Introduction

Particulate matter (PM) is a pollutant of great environmental concern due to its tight correlation with health conditions of people [1–5] and the climate of the earth [6–8]. PM is actually a complex mixture of extremely small particles and liquid droplets, with chemical components of sulfate, nitrate, ammonium, elemental carbon, organic chemicals, metals, and so forth [9], the emissions of which originate from both anthropogenic and natural sources [6, 10, 11]. Based on the aerodynamic diameter of particles, PM is classified into PM_{2.5} (PM with an aerodynamic diameter $\leq 2.5 \mu\text{m}$), PM₁₀ (PM with an aerodynamic diameter $\leq 10 \mu\text{m}$), and TSP (total suspended particles, PM with an aerodynamic diameter $\leq 100 \mu\text{m}$) [12].

Visibility usually refers to the horizontal distance, at which the contrast between a target and its sky background equals the threshold of human eyes [13]. Visibility can be significantly associated with air quality as it could be

decreased by air pollutants via light scattering and absorption by fine particles in the atmosphere [13–15]. Thus, visibility can be taken as a highly relevant visual indicator of air pollution level [16]. China has been experiencing severe air pollution in recent decades resulting from the rapid industrial development and urbanization. Many studies have reported the high PM pollution levels and the degradation of visibility due to the worsening air quality in China [13, 16–18].

Analysis on long-term changing trends of PM pollution and visibility is an important approach to evaluate the overall impact on air quality at local, regional, continental, and global scales [13, 19]. In China, the relevant studies have been focused on the megacities [20–24] and some hot spot regions such as the North China Plain [14, 23, 25–27], the Yangtze River Delta region [17, 18, 28, 29], and the Pearl River Delta region [16, 30]. Few studies on PM pollution and visibility over a specific second-tier city, such as Hefei, have been carried out, and therefore long-term changing trends of PM₁₀ pollution and visibility remain unknown in these areas.

FIGURE 1: Location of Hefei city, as marked by the red rectangle.

The government of China started routinely monitoring PM_{10} , sulfur dioxide (SO_2), and nitrogen dioxide (NO_2) in Hefei in June 2000. Routine measurement of $PM_{2.5}$ was introduced until January 2013, and no routine measurement of TSP was available in Hefei. Therefore, long-term trends analysis is only feasible for PM_{10} in Hefei. In this study, analysis of twelve-year trends of PM_{10} and visibility in Hefei during the period of 2001–2012 was performed. The study investigated the general patterns of PM_{10} and visibility trends in Hefei over the recent twelve-year period by analyzing the seasonal and annual behavior of PM_{10} and visibility. In addition, the meteorological factors that may affect PM_{10} pollution and visibility were also examined. The results provide a general understanding of the long-term PM_{10} pollution and visibility characteristics, which can be served as a basic data support for local government to further mitigate the environment pollution and to improve air quality control plans for the city.

2. Materials and Methods

2.1. Study Areas. Hefei (center: $117.19'37''$, $31.44'55''$) is the capital city of Anhui province, located in the east center of China, about 1000 km south to the national capital city of Beijing and 450 km west to the city of Shanghai (Figure 1). Hefei is the largest city and the political, economic, and cultural center of Anhui province. Hefei has a total population of about 8 million and near 5 million inhabitants reside in its metropolitan area in 2012 [31]. Hefei has a GDP of 416 billion Chinese currency, ranked number 1 in Anhui province and number 31 in China in 2012 [31]. The main industry includes machinery, electronics, chemistry, steel, textile, and cigarette industries. Hefei has been identified by The Economist in the December 2012 as world's number 1 fastest growing metropolitan economy (<http://www.economist.com/node/21567579>).

2.2. Data Source. Daily air pollution index (API) and the major pollutant data during June 2000 to January 2013 in Hefei were downloaded from the website of the Ministry of

TABLE 1: The bottom and upper limit of API values and corresponding PM_{10} concentrations.

API		PM_{10} concentrations ($\mu g m^{-3}$)	
I_{bottom}	I_{upper}	C_{bottom}	C_{upper}
0	50	0	50
50	100	50	150
100	200	150	350
200	300	350	420
300	400	420	500
400	500	500	600

Environmental Protection of the People's Republic of China Data Center (<http://datacenter.mep.gov.cn/>). Multiple sites have been built up in Hefei (4 sites in 2000 and then gradually expanded to 9 sites with the expansion of the city by 2010). The monitoring sites are all located in the urban area. Even though ambient concentrations of PM_{10} , NO_2 , and SO_2 have been routinely monitored at multiple sites, the data website only publishes city-average API values (averaged over all sites). The complete data in 2001–2012 were therefore analyzed in this study. Daily API values were estimated based on the ambient concentrations of PM_{10} , NO_2 , and SO_2 . First, sub-API (i.e., API of one specific pollutant) values were estimated for PM_{10} , NO_2 , and SO_2 , respectively, and then the maximum sub-API value was determined as the daily API and the associated pollutant was determined as the major pollutant. In Hefei, PM_{10} was the major pollutants on all the pollution days in 2001–2012 (http://datacenter.mep.gov.cn/report/air_daily/air_dairy_aqi.jsp). Therefore, daily PM_{10} mass concentrations were converted back from API values using the following:

$$C = (I - I_{bottom}) \times \frac{C_{upper} - C_{bottom}}{I_{upper} - I_{bottom}} + C_{bottom}, \quad (1)$$

where C is the PM_{10} concentration and I is the API value. I_{upper} and I_{bottom} are the upper and bottom limit values for I (i.e., $I_{bottom} \leq I < I_{upper}$). C_{upper} and C_{bottom} are the upper and bottom limit concentrations corresponding to I_{upper} and I_{bottom} . Table 1 shows the values of I_{bottom} , I_{upper} , C_{bottom} , and C_{upper} .

The meteorological observation data in 2001–2012 were obtained from the National Climate Data Center (NCDC) (<ftp://ftp.ncdc.noaa.gov/pub/data/noaa/>). The meteorological data includes hourly surface observations of air temperature, dew point temperature, wind speed, wind direction, precipitation. Relative humidity was estimated using the temperature and dew point temperature. Hourly meteorological parameters then were averaged to daily values to investigate their correlations to daily PM_{10} and visibility.

3. Results and Discussion

3.1. PM_{10} Trends. Figure 2(a) displays the long-term trends of the PM_{10} concentrations in Hefei during the period from 2001 to 2012. The statistical results for each year were computed

FIGURE 2: (a) Statistical box chart of long-term trends for the PM₁₀ concentration in Hefei during the period from 2001 to 2012. The range of the box represents the interquartile range of the data. (b) The worst 20% and 50% and best 20% region-average PM₁₀ concentration trends in Hefei during the period from 2001 to 2012. Dotted lines are the linear regression curves of the corresponding trend lines.

using the individual daily data. The annual average concentrations of PM₁₀ show fluctuating variation in 2001–2007 and slightly decrease after 2008, with an overall increasing trend of $0.24 \mu\text{g m}^{-3}$ per year (Figure 2(b)). The minimum PM₁₀ annual average concentration is $93 \pm 41 \mu\text{g m}^{-3}$ observed in 2005 and the maximum is $126 \pm 55 \mu\text{g m}^{-3}$ observed in 2008. The annual average PM₁₀ concentrations are 2–3 times of Grade I Chinese Ambient Air Quality Standard for annual average PM₁₀ ($40 \mu\text{g m}^{-3}$) [32], underscoring serious air pollution status in Hefei. The minimum PM₁₀ concentration values for the twelve years ranged from 9 to $23 \mu\text{g m}^{-3}$ with a low deviation magnitude; however, the maximum values revealed a significant annual variation with no clear trend during this period. The maximum PM₁₀ concentration was $248 \mu\text{g m}^{-3}$ in 2001 and then concentration value drastically increased to $470 \mu\text{g m}^{-3}$ in 2002, followed by a continuous decrease from 2004 to 2007. The concentration went back to $506 \mu\text{g m}^{-3}$ in 2008 and showed a zigzag behavior after that. The maximum PM₁₀ in Hefei is $230 \mu\text{g m}^{-3}$ in 2012, which is the lowest value during the twelve years but is 4 times of Grade I Chinese Ambient Air Quality Standard for 24-hour average PM₁₀ ($50 \mu\text{g m}^{-3}$) [32].

Figure 2(b) shows the worst 20% and 50% and best 20% average PM₁₀ concentration trends in Hefei during the period from 2001 to 2012. Dotted lines are the linear regression curves of the corresponding trend lines. An increasing trend is found for the best 20% PM₁₀ concentrations at the rate of $0.52 \mu\text{g m}^{-3}$ per year. A similar but relatively smaller trend is

found for the average PM₁₀ concentrations of $0.24 \mu\text{g m}^{-3}$ per year. However, the worst 20% PM₁₀ concentration exhibited a declining trend of $1.18 \mu\text{g m}^{-3}$ per year. It should be noted that the trends are not statistically significant, as the correlation coefficients (R^2) are all below 0.15 for the three trends. Therefore, overall the PM₁₀ pollution situation has not been significantly improved during these years in Hefei. Considering the high PM₁₀ levels, further legislation efforts will be expected to inhibit PM pollution and improve air quality in Hefei.

Figure 3 shows the frequency distribution of daily PM₁₀ concentrations in Hefei in each year during 2001–2012 in five concentration ranges: 0–50, 50–100, 100–150, 150–200, and $>200 \mu\text{g m}^{-3}$. PM₁₀ concentrations in the range of 50–100 and 100–150 $\mu\text{g m}^{-3}$ dominate most days of a year, accounting for approximately 70% of all days in a year. The number of days with PM₁₀ concentration less than $100 \mu\text{g m}^{-3}$ shows an increasing trend during 2008 to 2012, indicating a slight improvement in air quality since 2008. The days with the PM₁₀ concentration over $200 \mu\text{g m}^{-3}$ only account for less than 10% of the total days over all the years, except 2002 and 2008, and show no clear trend.

To learn more about the temporal variations of PM₁₀, the data are divided into four seasonal groups. Each season is defined as spring (March to May), summer (June to August), fall (September to November), and winter (December to February). Figure 4 presented the seasonal variations for PM₁₀ concentration in Hefei during 2001–2012. Of all the twelve years, summer has the lowest PM₁₀ concentrations

FIGURE 3: Frequency distribution of daily PM_{10} concentration in Hefei during the period from 2001 to 2012. Note: PM_{10} concentration unit: $\mu g m^{-3}$.

among all seasons with the lowest standard deviation, varied in the range of 71 ± 18 (2005) to 104 ± 45 (2008) $\mu g m^{-3}$. The other three seasons have similar PM_{10} concentrations, with the 12-year mean values of 114 ± 14 (spring), 114 ± 11 (fall) and 112 ± 11 (winter) $\mu g m^{-3}$, respectively. Higher dilution due to warmer temperature and mixing layer and higher removal due to more precipitation in summer are the main driver of relatively clean PM_{10} status in summer. In addition to the effects of meteorological conditions, additional emissions in other seasons (such as agricultural biomass burning in the spring and fall [8], residential house heating, and fireworks during the Chinese New Year [33]) also contribute to the difference among the seasons. Even though the summer concentrations are the lowest, still the concentrations are several times higher than Grade I Chinese Ambient Air Quality Standard. The relatively weak seasonal variation, especially among spring, fall, and winter, is not consistent from the seasonal pattern in other cities [34], suggesting the pollution sources and processes are different in Hefei.

3.2. Visibility Trends. Long-term trend of visibility in Hefei has been analyzed for the period of 2001–2012, and the annual statistical results are shown in Figure 5(a). The annual average visibility is less than 7 km in all the years except 2001 and 2004. The best visibility is found in 2001 with the highest value of 23.2 km and the average value of 8.6 km. The visibility shows a worsening trend from 2001 to 2007. In 2007, the highest visibility is 11.7 km, and the lowest value is only 0.7 km, with an average one of 5.6 km. The mean visibility presents a slightly increasing trend by 9.5% on average since 2007. In 2012, the average visibility can reach 6.1 ± 2.5 km, and the lowest value is 1.2 km. The highest visibility shows a clear degradation trends from 2001 (23.2 km) to 2012 (12.0 km).

FIGURE 4: Seasonal variations of PM_{10} concentration in Hefei during 2001–2012. The error bars indicated the standard deviation for the average PM_{10} concentration value in each season every year.

The general visibility situation is very serious and continuous and effective pollution control strategy is necessary to counteract the degradation of atmospheric visibility.

The worst 20% and 50% and best 20% region-average visibility trends in Hefei during 2001–2012 are shown in Figure 5(b). Dashed lines are the linear regression curves of the corresponding trend lines. Similar decreasing trends are found over the twelve years, with the rates of 0.01, 0.08, and 0.13 km per year which were observed for the worst 20% and 50% and best 20% average visibility, respectively. However, it is encouraging to notice slight increasing trends in general since 2006 or 2007 for the three groups of data.

The trends of annual percentages (%) of visibility for lower than 5 km and higher than 10 km in Hefei during 2001–2012 are shown in Figure 6. The percentage of visibility lower than 5 km is in the range of 21.9–42.7%, with an overall increasing rate of 1.14% per year, while that of visibility higher than 10 km is in the range of 3.8–35.3%, with an overall decreasing rate of 1.51% per year. However, it is worthy to notice that there is a breaking point for both trends in 2006, similar to the visibility trends in Figure 5. The percentage lower than 5 km visibility before 2006 exhibits a significantly increasing trend with the rate of 3.04% per year, while, after 2006, a decreasing trend is observed with a declining rate of 1.09% per year. The percentage higher than 10 km visibility before 2006 exhibits a fluctuating behavior with a decreasing rate of 3.77% per year, and in contrast, after 2006, a steady increasing trend with a high R^2 value of 0.917 and a changing rate of 0.91% per year is obtained. The decreasing trend in PM_{10} after 2008 and the increasing trend in visibility after 2006 are consistent with the air control programs that have been implemented during the 11th five-year plan (2006–2010), as suggested by other studies [13, 15].

FIGURE 5: (a) Statistical box chart of long-term trends for the visibility in Hefei during the period from 2001 to 2012. The range of the box represents the interquartile range of the data. (b) The worst 20% and 50% and best 20% region-average visibility trends in Hefei during the period from 2001 to 2012. Dashed lines are the linear regression curves of the corresponding trend lines.

FIGURE 6: Annual region-average percentages (%) of visibility lower than 5 km (left y-axis) and higher than 10 km (right y-axis) in Hefei during 2001–2012. Black dashed lines and blue dotted lines are the linear regression curves of the corresponding trend lines, splitted by year 2006 (the red short dashed line).

However, more effective control programs are further needed to keep improving the air quality and visibility in Hefei.

3.3. PM_{10} and Visibility versus Meteorological Factors. The meteorological conditions are key factors determining the status of the PM pollution and visibility. Figure 7 displays the PM_{10} concentrations and visibility values in different categories of meteorological parameters of air temperature, wind speed, wind direction, precipitation, and relative humidity in Hefei from 2001 through 2012. The daily averages of the five meteorological parameters are first divided into eight categories, and PM_{10} concentrations and visibility values are sorted into the eight categories correspondingly and then the mean and standard deviation values are calculated for PM_{10} and visibility in each category. The PM_{10} follows a clear “n” shape with the temperature, with the PM_{10} concentration increasing with temperature when temperature is lower than 12°C , and PM_{10} concentration reaches a peak point of approximately $120 \mu\text{g m}^{-3}$ at around 12°C ; then PM_{10} concentration decreases with temperature. The visibility shows somewhat an inverse correlation with temperature with a decreasing trend, indicating the degradation effect of particles on visibility. The effects of temperature on PM_{10} concentrations are complicated. On one hand, vertical mixing, which dilutes particles in the boundary layer, is affected by the change in temperature and wind with height. In general vertical mixing is stronger in hot seasons when temperature is also higher [35]. On the other hand, temperature affects the chemical formation of particles. Higher temperature accelerates the reactions to form more products that can partition into particulate phase (positive), but meanwhile the products tend to remain in their gaseous phase when temperature

FIGURE 7: Effects of meteorological factors on PM_{10} concentration and visibility in Hefei during 2001–2012: (a) air temperature, (b) wind speed, (c) wind direction, (d) precipitation, and (e) relative humidity.

TABLE 2: Multiple regression statistics for visibility to temperature, relative humidity, and PM₁₀ concentrations.

(a)

Regression statistics	
Multiple R	0.69
R^2	0.47
Adjusted R^2	0.47
Std. error	1.36
Observations	4586

(b)

	Coefficients	Std. error	t stat.	P value	Lower 95%	Upper 95%
Intercept	11.46	0.14	80.07	0.00	11.18	11.74
Temperature	0.04	0.00	18.20	0.00	0.03	0.04
RH	−0.09	0.00	−57.37	0.00	−0.09	−0.08
PM ₁₀	−0.01	0.00	−34.26	0.00	−0.02	−0.01

is too high (negative) [36]. Therefore, the results in Hefei indicate that the positive effect of temperature on particulate dominates when temperature is lower than 12°C, and the negative effects become more important when temperature is higher than 12°C.

PM₁₀ concentration decreases with the wind speed, and visibility increases with the wind speed. Calm winds prohibit the transport of pollutants and favor the accumulation of PM, leading to higher PM₁₀ concentrations and worse visibility [15]. In terms of wind direction effect, the PM₁₀ concentration is the highest under the southeast winds, followed by the east winds. The PM₁₀ concentrations are similar under the rest of wind directions. This phenomenon is likely due to the more industries located in cities east to Hefei, such as Ma'anshan and Nanjing. In winter the prevailing wind direction is north (northwest, north, and northeast winds), accounting for 55% of total winds, and the prevailing wind direction in summer is east (northeast, east, and southeast winds), accounting for 59% of total winds. The fact that the prevailing winds in summer are associated with higher PM₁₀ concentrations while the high temperatures in summer lead to decrease in PM₁₀ concentrations (Figure 7(a)) confirms the negative effect of temperature in summer that is discussed previously. The visibility trend indicates that visibility is generally the best when the wind is from the southwest and is the worst when the wind is from the north.

PM₁₀ decrease when the precipitation and the relative humidity increase. Washout effect by precipitation is the main removal process of PM₁₀ [6, 37]. Stronger precipitation leads to more washout of PM and thus lower ambient PM concentrations. High relative humidity can significantly alter the particle properties. Some particles, such as sulfates, accumulate water and grow to larger sizes at which they are more efficient to deposit [15]. Previous study in Shanghai also found similar correlation between PM₁₀ and relative humidity [38]. The visibility also shows decreasing trends with increasing precipitation rate and relative humidity, which is consistent with other studies [39].

PM₁₀ can directly affect visibility by scattering and absorption light. Multiple linear regression analysis was

conducted for visibility to PM₁₀ and the meteorological parameters. When excluding the wind speed, wind direction, and precipitation in the analysis, the multiple regression correlation coefficient (multiple R) is the greatest, and the statistical results are shown in Table 2. In Hefei, visibility is negatively correlated to both RH and PM₁₀. The fact that visibility is negatively correlated to PM₁₀ but meanwhile visibility and PM₁₀ are both negatively correlated with RH is presumably due to the synthesized effects by other factors, such as particle sources, hygroscopicity, and hygroscopic growth rate of particles versus loss by deposition. The analysis of PM₁₀ and visibility to the meteorological parameters in Figure 7 is rather qualitative. More studies are needed to quantify the relationships between PM₁₀ pollution and visibility and meteorological conditions in Hefei.

4. Conclusions

Twelve-year daily PM₁₀ and visibility data in 2001–2012 in Hefei were analyzed. Overall Hefei has been suffering serious PM₁₀ pollution during these years. The annual average PM₁₀ concentrations are 2–3 times of the Chinese Ambient Air Quality Standard, and the extreme daily PM₁₀ concentrations are over 4 times of the standard. An overall worsening trend is revealed for annual average and the best 20% PM₁₀ concentrations, even though the worst 20% PM₁₀ (i.e., the extreme PM₁₀ pollution) shows a decreasing trend. Unlike other cities, PM₁₀ in Hefei has a relatively weak seasonal variation among spring, fall, and winter but has lower concentrations in summer. The annual average visibility range is generally lower than 7 km, indicating serious visibility degradation in Hefei. Moreover, visibility exhibits an overall worsening pattern through the period. Detailed analysis reveals that the annual average concentrations of PM₁₀ show fluctuating variation in 2001–2007 and slightly decrease after 2008, and the visibility shows a worsening trend from 2001 to 2006 followed by a steadily improved trend from 2007 to 2012, indicating a gradually improved trend after the 11th five-year plan. However, it is necessary to implement more effective control programs

in order to keep improving the air quality and visibility in Hefei. Wind speed, precipitation, and relative humidity have a negative effect on PM_{10} concentrations, while temperature has a negative effect on PM_{10} when it is above $12^{\circ}C$ and has a positive effect when it is lower than $12^{\circ}C$. Precipitation and relative humidity also have a negative effect on visibility.

This study reports the long-term trends of PM_{10} and visibility for the first time in the Hefei metropolitan area and provides a general understanding of the status and trends of PM_{10} pollution and visibility in a typical second-tier city in China. More studies are still needed in this area, such as field measurement and modeling studies to investigate the chemical and physical properties of particles during PM pollution events, source apportionment studies of PM and visibility to reveal the contributions of different sources to PM and visibility, modeling studies of regional transport of PM and its precursors, and effects of meteorological condition change on the year-to-year variability of PM concentrations. These studies will provide necessary scientific support for developing local and regional emission mitigation programs around the Hefei metropolitan area.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

This work is financially supported by the Startup Fund for Talent at Nanjing University of Information Science and Technology (NUIST) under Grant no. 2243141501009, the Natural Science Foundation of China (21577065), International ST Cooperation Program of China (2014DFA90780), a Significant Research Plan of National Natural Science Foundation of China (91543115), and the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD).

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