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Quantifying the Influences of PM_{2.5} and Relative Humidity on Change of Atmospheric Visibility over Recent Winters in an Urban Area of East China

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Abstract: Fine particulate matters (PM_{2.5}) and relative humidity (RH) in the ambient atmosphere are the leading anthropogenic and natural factors changing atmospheric horizontal visibility. Based on the analysis of environmental and meteorological data observed over 2013–2019 in Nanjing, an urban area in East China, this study investigated the influences of PM_{2.5} and RH on atmospheric visibility changes over recent years. The visibility had significantly negative correlations with the PM_{2.5} concentrations and RH changes. The nonlinear relationships existed between PM_{2.5} concentrations and visibility, as well as between RH and visibility, with the inflection points in the atmospheric visibility changes. The PM_{2.5} inflection concentrations were 81.0 $\mu\text{g m}^{-3}$, 76.0 $\mu\text{g m}^{-3}$, 49.0 $\mu\text{g m}^{-3}$, and 33.0 $\mu\text{g m}^{-3}$, respectively, for the RH ranges of $\text{RH} < 60\%$, $60\% \leq \text{RH} < 80\%$, $80\% \leq \text{RH} < 90\%$, and $\text{RH} \geq 90\%$, indicating that the improvement of visibility with reducing PM_{2.5} concentrations could be more difficult under the humid meteorological condition. The visibility changes were most sensitive to PM_{2.5} concentrations in the RH range of 60–80% in this urban area of East China. The relative contributions of natural factor RH and anthropogenic factor PM_{2.5} to variations of wintertime atmospheric visibility were quantified with 54.3% and 45.7%, respectively, revealing an important role of natural factor RH in the change of atmospheric visibility in the urban area of East Asian monsoon region.

Keywords: atmospheric visibility; PM_{2.5}; relative humidity; Nanjing

1. Introduction

Atmospheric horizontal visibility (hereinafter, visibility) is meteorologically defined as the distance at which a normal observer can perceive a black object viewed against the horizon [1]. For the air environment, visibility can express the ambient atmospheric opacity reflecting the levels of air pollution, especially haze pollution resulting from high aerosol concentrations, which could deteriorate atmospheric visual range [1,2]. With the adverse influence of low visibility on traffic, human health, climate change, and other significant aspects, the spatial and temporal changes of visibility have attracted considerable attention worldwide as part of scientific studies of atmospheric

environment [3–8]. The changes in atmospheric visibility in long-term data of observation is of major interest in air pollution studies with climatology of atmospheric environment [9–12].

Visibility is determined by the light extinctions of aerosol particles [13,14] and gaseous pollutants (NO_x, SO₂ and volatile organic compounds (VOCs)) [5,15,16] in the ambient atmosphere. Compared to aerosol particles, the gaseous species have much weaker light extinction for less influence on visibility [17]. The size distribution, chemical composition, and mass concentrations of aerosol particles can heavily influence the extinction of aerosols changing atmospheric visibility [18–20]. The extinction efficiency of aerosol particles depends not only on the particle size, but also on the chemical compositions [21]. The chemical composition of PM_{2.5} (particulate matter with an aerodynamical diameter equal to or less than 2.5 μm) is quite complicated, including primary particulate matter, secondary inorganic and organic particles, carbonaceous species (organic carbon and elemental carbon), crustal elements, and water [22]. Visibility is mainly deteriorated to a large extent by enhancing concentrations of secondary aerosols [23,24]. Most secondary aerosol species (e.g., (NH₄)₂SO₄, NH₄NO₃) can strengthen the light scattering [25], while other primary aerosol species, such as elemental carbon and soil dust, can cause a strong light absorption but reflect less visible light [21,26,27].

Relative humidity (RH) in the ambient atmosphere is another important factor that impacts atmospheric visibility by affecting the hygroscopic property of particles, and the humidification increases the particle scattering in the atmosphere [28,29]. The aerosol chemical compounds contain a large amount of hydrophilic aerosol particles (e.g., (NH₄)₂SO₄, NH₄NO₃), which have a strong hygroscopic property [21,30]. Hygroscopic aerosol particles take up water as RH increases [31], and aerosol water can affect both the size and refractive indices of atmospheric aerosols, thereby influencing the mass concentrations, size distribution, and optical properties, increasing the scattering and absorption capacities to the visible light, resulting in impairment of visibility [32–37]. When the RH values reach 70–80%, water content of aerosols, such as sulfate and nitrate, can generally contribute 50% or more of PM_{2.5} to visibility changes, becoming a controlling factor in enlarging light scattering of aerosol optical properties [17,38,39].

In China, there was a notable decrease in visibility of about 2.1 km per decade from 1990 to 2005 [10], and air pollution of high PM_{2.5} concentrations is becoming a serious environmental problem [40–44]. Extreme low visibility events resulted from high PM_{2.5} levels are commonly accompanied by high RH weather [45–47]. In some cases, the variations on visibility are mainly determined by PM_{2.5} concentrations, so visibility can very well represent the level of air quality. In contrast, in the cases where RH dominates the variations in visibility, the visibility may not be a good proxy of air quality [48]. Visibility can be generally influenced by anthropogenic and natural factors. PM_{2.5} and RH, as the leading anthropogenic and natural factors with their relative contributions to changing visibility, have been poorly understood in the change of atmospheric environment.

Nanjing, a major urban area in East China, has confronted the environmental problem in atmosphere associated with high concentrations of PM_{2.5} and low visual range over recent years [49,50]. In this study, we selected Nanjing as an urban area with seven-year (2013–2019) data of meteorological and environmental observations to understand the anthropogenic and natural influences on the change of atmospheric visibility over recent winters. The main objectives of this study were to recognize the influences of anthropogenic factor PM_{2.5} and natural factor RH on atmospheric visibility change and to quantify the relative contributions of PM_{2.5} and RH to visibility change over recent years in an urban area of East China.

2. Data and Methods

2.1. Data

The seven-year (2013–2019) data of hourly PM_{2.5} concentrations were used in this study, including nine observational sites in Nanjing with eight urban sites in residential and industrial

zones, as well as one suburban site over the national air quality monitoring network in China (Figure 1). The $PM_{2.5}$ concentrations were averaged over nine observational sites in Nanjing to characterize the variations of fine aerosol particles over this urban area. The meteorological data of surface observation in Nanjing were obtained from the Meteorological Data Sharing Network of China Meteorological Administration [51]. The meteorological data of surface observation included visibility and RH with temporal resolutions of 3 h in order to analyze the meteorological variations and the influence of RH on visibility for this study. Daily mean visibility, RH, and $PM_{2.5}$ concentrations were computed from the hourly observation data of meteorology and environment.

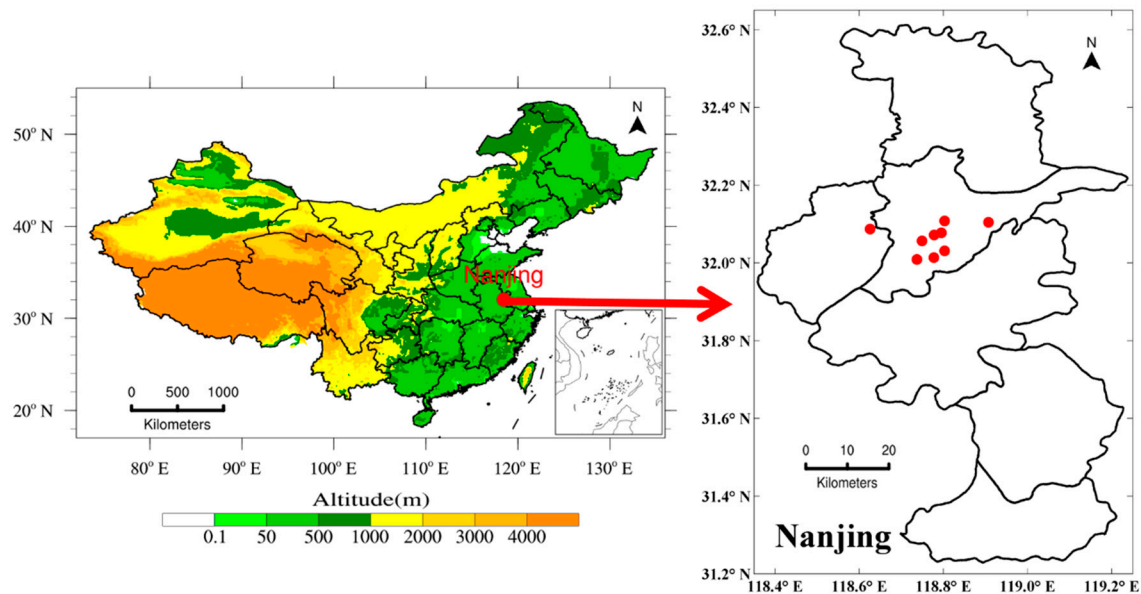


Figure 1. Topographical height (m, a. s. l.) in China with the locations of Nanjing (left panel) and nine sites of air quality measurement in Nanjing, an urban area in East China (right panel).

2.2. Methods

2.2.1. Categorizations of Atmospheric Visibility and RH

In this study, we categorized daily visibility into the good, low, and poor visibility levels, respectively, with the lower limit of 10 km and upper limits of 10 km and 5 km [35,52]. To more quantitatively analyze the variation of visibility, the visibility values were divided into three levels: > 10.0 km, $5.0\text{--}10.0$ km, and < 5.0 km. Aerosol light scattering enhancement factor grows smoothly with $RH < 60\%$ and exhibits a rapid enhancement at high $RH > 60\%$ [45]. The aerosol hygroscopic effect becomes significant when RH exceeds 90% [45]. Thus, we divided RH into four ranges: $RH < 60\%$, $60\% \leq RH < 80\%$, $80\% \leq RH < 90\%$, and $RH \geq 90\%$.

2.2.2. A Measure of Relative Importance in Multiple Regression

Multiple regression analysis has two distinct applications: Prediction and explanation [53]. When multiple regression is used for explanatory purpose, we are interested in the extent of each variable contributing to the response variable [54]. Standardized regression coefficients are the most common measure of relative importance when multiple regression is used [55,56].

We constructed a stepwise multiple linear regression model to quantify the effect of RH and $PM_{2.5}$ on visibility variability. The model fitted the daily visibility with daily RH and $PM_{2.5}$. The daily values of visibility, $PM_{2.5}$ and RH satisfied approximately normal distribution. The fit has the form:

$$Y(t) = \sum_{k=1}^n \beta_k X_k(t) \quad (1)$$

where $Y(t)$ is the standardized time series of daily visibility for all winters over 2013–2018 in Nanjing, and $X_k(t)$ is the corresponding time series for the standardized affecting factors (PM_{2.5} and RH) $k \in [1, n]$. β_k is the standardized regression coefficient. The contribution (C_{x_k}) of a given predictor (x_k) to response variable (Y) is:

$$C_{x_k} = \frac{|\beta_k|}{\sum_{k=1}^n |\beta_k|} \quad (2)$$

3. Results and Discussion

3.1. Variations in Atmospheric Visibility

Figure 2 showed the monthly variations of visibility in Nanjing averaged over 2013–2019, with the error bars indicating the standard deviations for the mean visibility based on the daily values. Similar to most urban areas in China, visibility in Nanjing varied seasonally from the high values in summer (July and August) to the low values in winter (December, January, and February) during the past seven years, which could be in association with a combination of air pollutant emissions and seasonal variance in meteorology [57]. Hence, we focused this study on the wintertime visibility variations and their relationship with wintertime PM_{2.5} and RH.

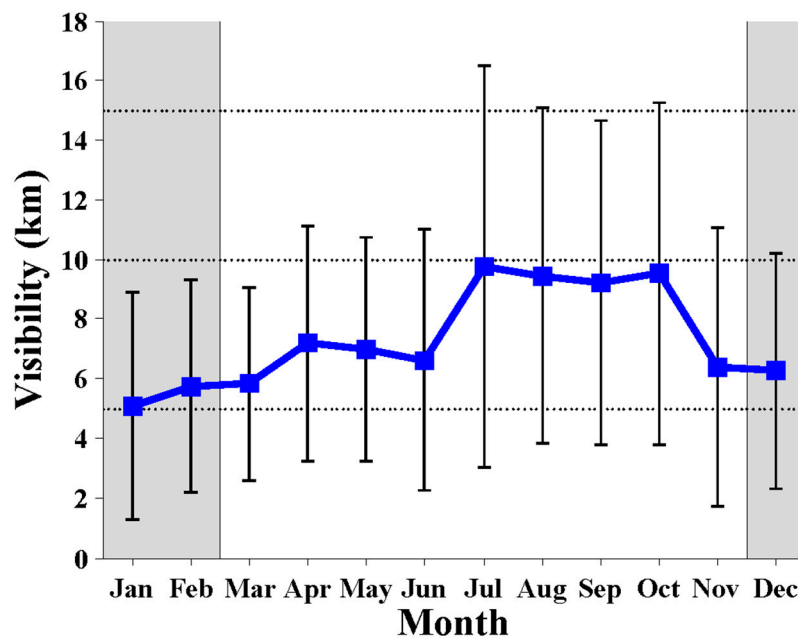


Figure 2. Monthly variations of atmospheric visibility in Nanjing averaged over 2013–2019. Error bars denote the standard deviations of daily visibility.

The occurrence frequencies of good, low, and poor visibility levels were calculated as a percentage of the number of daily visibility values in each level during winter. The interannual variations of wintertime mean visibility, RH, PM_{2.5} concentrations, and occurrence frequencies of good, low, and poor visibility levels in winter over 2013–2018 are shown in Figure 3. The interannual variations of PM_{2.5} concentrations presented a significantly decreasing trend over 2013–2018, and the values of visibility enhanced after 2013 (Figure 3a), which could present the effect of China’s emission control strategies introduced in 2013 [58,59]. Differently, the interannual variations of wintertime RH presented a weak humid trend in the urban environment from 2014 to 2018, reflecting the change of natural factor RH in the East Asian monsoon region [60,61]. The slight increase in RH during these winter periods could intensify the hygroscopic growth of aerosol particles, thus increasing light extinctions and reducing visibility. Although the number of days with the level of “good visibility” had an obvious

enhancement over the past winters, especially since 2016, the level of “poor visibility” dominated the wintertime ambient air with 42–63% days, with a weak interannual change over 2013–2018 in Nanjing (Figure 3b). All of the mean visibility values averaged over each winter ranged between 5–10 km in the level of “low visibility” (Figure 3a).

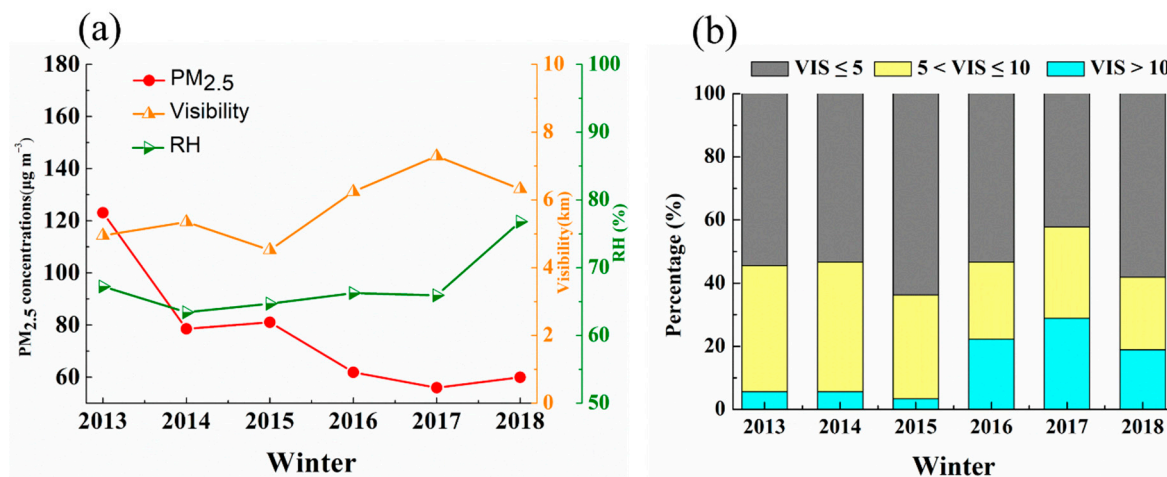


Figure 3. Interannual variations of wintertime (a) Fine particulate matter (PM_{2.5}) concentrations, visibility, relative humidity (RH) and (b) the percentages (%) of days with good (VIS > 10 km), low (5 km < VIS ≤ 10 km), and poor (VIS ≤ 5 km) visibility in winter observed in Nanjing from 2013 to 2018.

The correlation coefficients of daily visibility with PM_{2.5} and RH during wintertime of 2013–2018 were given in Table 1. The visibility had significantly negative correlations with the PM_{2.5} concentrations and RH. The light extinction efficiency of the coarse particles was substantially lower than that of fine particles, and aerosols with diameters of 0.5–2 µm were most efficient for scattering visible light [49,62]. In terms of particles, we could expect that the visibility degradation was mainly caused by the increment of the PM_{2.5} concentrations during winter in Nanjing. However, the better correlation between visibility and RH was found than that between visibility and PM_{2.5} concentrations for all winters (Table 1), which could indicate that the natural factor RH might play the more important role in visibility change compared with the anthropogenic factor PM_{2.5} concentrations in this urban region. The impact of RH on the size and chemical composition of aerosol particles could alter aerosol optical property for changing visibility in the ambient atmosphere [32,35].

Table 1. Correlation coefficients of daily visibility with PM_{2.5} concentrations and RH in Nanjing during wintertime of 2013–2018 (passing the confidence level of 99%).

Winter	Visibility vs. PM _{2.5}	Visibility vs. RH
2013	−0.52	−0.63
2014	−0.56	−0.73
2015	−0.70	−0.79
2016	−0.56	−0.62
2017	−0.60	−0.72
2018	−0.54	−0.59

3.2. Connection of Visibility with PM_{2.5} and RH

Figure 4 exhibited the changes in atmospheric visibility with respect to PM_{2.5} concentrations and RH in ambient air. The nonlinear relationships of visibility with PM_{2.5} concentrations and RH could be fitted as power functions with the goodness of fitting respectively at 0.30 and 0.37, passing the confidence level of 95%. Reduction of PM_{2.5} over the high levels could improve atmospheric visibility insignificantly, and as PM_{2.5} concentrations were reduced to the low levels reaching the inflection point

in changing visibility, further decreasing $PM_{2.5}$ concentrations could enhance the visibility sharply (Figure 4a). Similarly, an inflection point existed in the nonlinear relationship between atmospheric visibility and RH changes (Figure 4b). In humid air environment with high RH, the visibility dropped slowly with increasing RH, while RH changed visibility significantly in dry air with low RH.

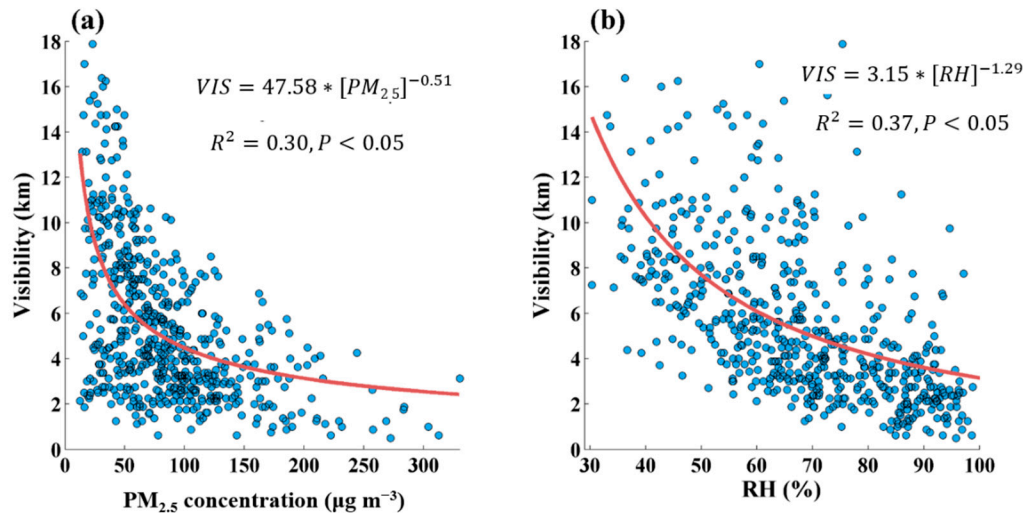


Figure 4. Scatterplots of (a) visibility and $PM_{2.5}$ as well as (b) visibility and RH in Nanjing during winter over 2013–2018 with the curves of the power function fitting equations.

Based on the observation analysis, it was found that the nonlinear relationship between $PM_{2.5}$ concentrations and visibility depended on different RH values in the ambient atmosphere. With the same $PM_{2.5}$ concentrations, the higher the RH was, the lower the visibility was (Figure 5). The increases of RH can promote the moisture absorption of the particles and strengthen the scattering and absorption capacities to the visible light, leading to the impairment of atmospheric visibility [32,35]. Power functions were used to fit the nonlinear relationships between visibility and $PM_{2.5}$ concentrations in different RH ranges (Table 2). The visibility changes in this urban area of East China were most sensitive to $PM_{2.5}$ concentrations in the RH range of 60–80% with a power exponent of -0.77 , and the visibility changes showed the least sensitivity to $PM_{2.5}$ concentrations when $RH \geq 90\%$ with a power exponent of -0.55 (Table 2). Under the high humid condition ($RH \geq 90\%$), the visibility values were mostly lower than 5 km for the “poor visibility,” even with low $PM_{2.5}$ concentrations (Figure 5), which could have resulted from the formation of fog droplets in ambient air with water vapor saturation. Under the dry air condition ($RH < 60\%$), a large part of the observed visibility was longer than 5 km in the good and low visibility levels with the enhancement of $PM_{2.5}$ concentrations (Figure 5), which might have been caused by the low aerosol water contents and weak water vapor uptake by fine aerosol matters [49].

Figure 5 also showed the inflection points on the fitting curves of visibility and $PM_{2.5}$ changes. The inflection point of $PM_{2.5}$ concentrations was a turning point in the change of visibility induced by $PM_{2.5}$ concentrations. Atmospheric visibility showed low and high sensitivity with $PM_{2.5}$ concentration changes, respectively, above and below the inflection points (Figure 5). Under four RH ranges of $RH < 60\%$, $60\% \leq RH < 80\%$, $80\% \leq RH < 90\%$, and $RH \geq 90\%$, the inflection points of $PM_{2.5}$ concentrations in changing visibility reduced to $81.0 \mu\text{g m}^{-3}$, $76.0 \mu\text{g m}^{-3}$, $49.0 \mu\text{g m}^{-3}$, and $33.0 \mu\text{g m}^{-3}$ (Figure 5, Table 3), which indicated that improving atmospheric visibility with reducing $PM_{2.5}$ concentrations could be more difficult in more humid air environment. An important task of atmospheric environment control is reducing the $PM_{2.5}$ concentrations below the inflection points in order to achieve a significant improvement in visibility [36].

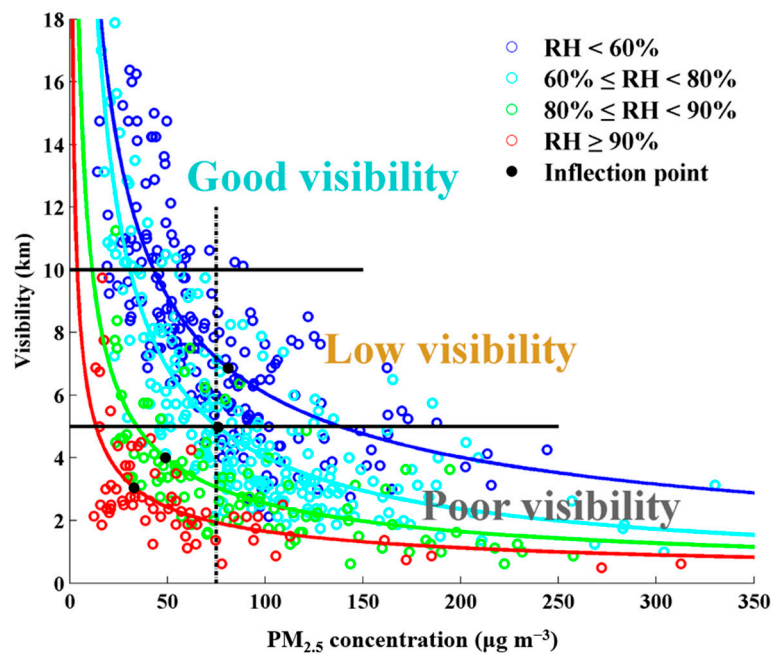


Figure 5. Scatterplot of wintertime visibility and PM_{2.5} concentrations in different RH ranges in Nanjing over 2013–2018 with circles colored in different RH. The curves and black dots present the fitting curves and their inflection points; black lines of 5 km and 10 km visibility indicate the poor, low, and good visibility levels; and the dashed line represents the light PM_{2.5} pollution level of 75 µg m⁻³.

Table 2. Nonlinear fitting equations of visibility and PM_{2.5} in different RH ranges and adjusted coefficients of determination (Adj R²) passing the confidence level of 99%.

	Nonlinear Fitting Curves	Adj R ²
RH < 60%	VIS = 93.08 × [PM _{2.5}] ^{-0.59}	0.58
60% ≤ RH < 80%	VIS = 139.40 × [PM _{2.5}] ^{-0.77}	0.70
80% ≤ RH < 90%	VIS = 46.44 × [PM _{2.5}] ^{-0.63}	0.49
RH ≥ 90%	VIS = 20.94 × [PM _{2.5}] ^{-0.55}	0.39

Table 3. The critical PM_{2.5} concentrations on the fitting curves at visibility of 10 km, and the inflection point PM_{2.5} concentrations and the critical visibility values on the fitting curves at PM_{2.5} concentrations of 75 µg m⁻³ in different RH ranges.

	Critical PM _{2.5} Concentrations (µg m ⁻³) at Visibility of 10 km	Inflection Point PM _{2.5} Concentrations (µg m ⁻³)	Critical Visibility (km) at PM _{2.5} Concentrations of 75 µg m ⁻³
RH < 60%	43.0	81.0	7.2
60% ≤ RH < 80%	30.7	76.0	5.0
80% ≤ RH < 90%	11.4	49.0	3.0
RH ≥ 90%	3.8	33.0	1.9

According to the National Ambient Air Quality Standards of China released by the Ministry of Ecology and Environment of China, light air pollution level of PM_{2.5} is categorized by the daily average PM_{2.5} concentration exceeding 75 µg m⁻³ in ambient air. There is a widely used haze definition with visibility of less than 10 km and RH of less than 90% [63,64]. Based on the fitting curves of nonlinear relationships between visibility and PM_{2.5} concentrations in four RH ranges of RH < 60%, 60% ≤ RH < 80%, 80% ≤ RH < 90%, and RH ≥ 90%, Table 3 presented the critical values of PM_{2.5} concentrations on the fitting curves at visibility of 10km in low visibility level. For the visibility at 10 km, the critical

PM_{2.5} concentrations varied significantly between 43.0 µg m⁻³, 30.7 µg m⁻³, 11.4 µg m⁻³, and 3.8 µg m⁻³ in dry and humid air with four RH ranges of RH < 60%, 60% ≤ RH < 80%, 80% ≤ RH < 90%, and RH ≥ 90% (Table 3), and all the critical PM_{2.5} concentrations with much lower than the light PM_{2.5} pollution level at 75 µg m⁻³ implied that the standard of clean air environment with PM_{2.5} < 75 µg m⁻³ had apparent inconformity with the threshold of visibility at 10 km for haze pollution, reflecting the different influences of anthropogenic and natural factors on atmospheric environment changes.

Table 3 also gave the critical values of visibility on the fitting curves at PM_{2.5} concentrations of 75 µg m⁻³ in light air pollution level. The critical visibility values at PM_{2.5} of 75 µg m⁻³ were 7.2 km, 5.0 km, 3.0 km, and 1.9 km, respectively, with different RH ranges (Table 3). The increases of RH could significantly deteriorate atmospheric visibility, providing a larger contribution to the visibility change. The critical values of visibility were below the visibility threshold of 10 km for haze pollution under all RH conditions, indicating a discrepancy of haze pollution defined by visual ranges and PM_{2.5} concentrations. PM_{2.5} concentrations and RH, as the leading anthropogenic and natural factors changing atmospheric visibility, exerted an integrate influence on wintertime visibility impairment in this urban area of East China.

3.3. Relative Contribution of RH and PM_{2.5} to Wintertime Visibility Variations

The impairment of wintertime visibility is generally linked with rapid increases in anthropogenic pollutant emissions in conjunction with unfavorable meteorological conditions for air pollutant dispersion [65,66]. This has led to a hot debate concerning whether the notable changes in visibility were mainly attributed to the meteorological conditions and anthropogenic emission control. Based on the discussion above, the anthropogenic factor PM_{2.5} led to the visibility impairment by light extinction, and natural factor RH promoted aerosols' hygroscopic growth to a certain extent. We quantified their relative contribution of the leading anthropogenic and natural factors, PM_{2.5} and RH, to the change of wintertime visibility in Nanjing over 2013–2018 with the multiple linear regression analysis of visibility, RH, and PM_{2.5} concentrations. The regression equation was:

$$[\text{VIS}] = -0.57[\text{RH}] - 0.48[\text{PM}_{2.5}], R^2 = 0.58, p < 0.01, \quad (3)$$

The goodness of fitting was 0.58, passing the confidence level of 99%, which could mean the inclusion of PM_{2.5} and RH accounted for the major contribution to wintertime visibility variations from 2013 to 2018 in Nanjing.

We estimated the relative importance of RH and PM_{2.5} by the method of comparing the standardized regression coefficients mentioned in Section 2.2.2. The daily variations of wintertime visibility represented by natural factor RH was 54.3%, indicating a higher contribution as compared with the anthropogenic factor PM_{2.5} contribution of 45.7%. That is, in spite of the effective PM_{2.5} reduction in this urban area, natural factor RH was a dominant driver for the changes of wintertime visibility over recent years. The change of natural factor RH was closely connected with climate change in this East Asian monsoon region, implicating an importance of the regional climate change for variation of atmospheric visibility and environment.

4. Conclusions

Anthropogenic and natural factors affecting visibility change were investigated based on data of environmental and meteorological observations over 2013–2019 in Nanjing, an urban area of East China. We recognized PM_{2.5} and RH changing atmospheric visibility and quantified the relative contribution of leading anthropogenic and natural factors to the variations of atmospheric visibility over recent winters in this urban area of East China. The application of these results will improve the representativeness and reliability of the use of visibility in the field of air pollution attribution and the determination of its climatic effects.

In spite of the effective PM_{2.5} reductions after 2013, the level of “poor visibility” dominated the ambient atmosphere over winter in this urban area of East China. Although the visibility had significantly negative correlations with the PM_{2.5} concentrations and RH, the inflection points existed in the nonlinear relationships between visibility and PM_{2.5} concentrations, as well as between visibility and RH. Under the RH ranges of RH < 60%, 60% ≤ RH < 80%, 80% ≤ RH < 90%, and RH ≥ 90%, the inflection points of PM_{2.5} concentrations were 81.0 μg m⁻³, 76.0 μg m⁻³, 49.0 μg m⁻³, and 33.0 μg m⁻³, respectively, reflecting that reducing the PM_{2.5} concentrations to the inflection points for improving the visibility would be more difficult in more humid air environment in context of climate change. The visibility changes were most and least sensitive to PM_{2.5} concentrations, respectively, in the RH range of 60–80% and RH ≥ 90% in this urban area of East China. The relative contributions of natural factor RH and anthropogenic factor PM_{2.5} to the changes of visibility were quantified with 54.3% and 45.7%, respectively, in this urban area, indicating an important role of natural factor RH in the changes of atmospheric visibility in the urban area of East Asian monsoon region.

This study revealed the influences of PM_{2.5} and RH on changing wintertime visibility with the seven-year data of environmental and meteorological observations over an urban area in East China. It should be emphasized that the particle size distribution, optical properties, and the chemical composition of aerosols exert the impacts on atmospheric visibility. The influences of atmospheric particles and meteorological conditions to atmospheric visibility are more complicated, which could be further investigated based on the long-term observations on physical and chemical properties of particle and fine meteorology. The influences of long-range transport of PM_{2.5}, weather patterns, and human activities on visibility change could be a further study with a more comprehensive numerical model of air quality and fine observations of meteorology and environment.

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