Exploring the Sensitivity of Visibility to PM$_{2.5}$ Mass Concentration and Relative Humidity for Different Aerosol Types

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1. Introduction

Atmospheric horizontal visibility is defined as the distance at which the contrast of a black object with respect to its background is equal to the contrast threshold of human eyesight and is an indicator of atmospheric transparency [1]. Good visibility is a desirable feature of any geographical location, and its importance should be considered [2]. However, many megacities have suffered from air pollution incidents accompanied by a decline in visibility during the past several decades [3–5]. Poor visibility severely affects tourism, transportation, and mental health [6–8].

Visibility degradation is mainly attributed to light scattering and absorption by atmospheric fine particles [9,10]. There is an obvious, negative correlation between visibility and PM$_{2.5}$ concentration [11–14]. A form of power function or exponential function between visibility and PM$_{2.5}$ concentration was observed in many cities [11,15,16]. Notably, when the PM$_{2.5}$ concentration was lower than a certain threshold, visibility increased quickly as the PM$_{2.5}$ concentration declined [11,16–18]. The threshold values of PM$_{2.5}$ vary by city. For example, the threshold corresponding to visibility < 10 km in Beijing, Xi’an, and Shanghai
Visibility variation shows a clear dependence on relative humidity (RH) also [20,21]. Chen et al. [22] indicated that the decrease in visibility was mainly influenced by RH when RH was >90%. Hygroscopic growth usually increases aerosol extinction coefficients by enlarging the particle size due to the uptake of liquid water. In some other cases, aerosol extinction can be decreased by lowering the refractive index because the refractive index of water is smaller than that of other aerosol components [23]. Positive or negative effects are dictated to a large degree by components [21].

There are complex, nonlinear relationships among visibility, PM\textsubscript{2.5} concentration, and RH, which are deeply affected by the component composition of particles in addition to the mixing state and size distribution [21,24–26]. The effect of particles and RH on visibility is further complicated by the differences in extinction abilities of particle components [11,27,28]. Water-soluble, inorganic salts (e.g., (NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4}, NH\textsubscript{4}NO\textsubscript{3}) and carbonaceous (e.g., organic matter (OM), elemental carbon (EC) aerosols are the major extinction components in particles with various, dry extinction efficiencies and hygroscopicities [29,30]. The high content of extinction components in particles can increase the sensitivity of visibility to particle concentration. Similarly, the percentage of hygroscopic components (e.g., (NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4}, NH\textsubscript{4}NO\textsubscript{3}) in PM\textsubscript{2.5} affects the sensitivity of visibility to RH [10,24]. The content of extinction components in PM\textsubscript{2.5} varies obviously by region and period [8,11,25]. For example, the content of major extinction components ((NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4}, NH\textsubscript{4}NO\textsubscript{3}, OM, and EC) and hygroscopic components ((NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4} and NH\textsubscript{4}NO\textsubscript{3}) in PM\textsubscript{2.5} in Shanghai (83% and 42%) was 1.7 times greater than those in Chengdu (48% and 25%, respectively) [31,32]. Thus, identifying the primary factors influencing visibility is essential.

In most cases, atmospheric visibility is affected by both particle concentration and RH; if it is dominated by particulate matter, the effect of emission reduction measures is obvious, but if it is dominated by RH, the same emission reduction measures may not achieve the expected effect. Many studies were conducted to gain insights into correlations between low-visibility events and influencing factors [13,18,30]. However, the sensitivity of visibility to PM\textsubscript{2.5}, mass concentration, and RH by aerosol type remains unclear.

In predicting and preventing low-visibility events, determining whether the main cause is PM\textsubscript{2.5} or RH is essential [19,33]. There are some studies focused on the complex, nonlinear relationships among visibility, PM\textsubscript{2.5} concentration, and RH. However, the relative contribution of the two factors to visibility degradation, especially by aerosol type, was difficult to quantify. Thus, this study attempts to build a method for identifying the dominant factors of visibility on the basis of the sensitivity of visibility to PM\textsubscript{2.5} and RH changes. A total of 4453 valid hour data records of visibility, PM\textsubscript{2.5}, and RH in Tianjin, 2015, were used to verify and evaluate the method. In addition, aerosols were classified based on the extinction component ((NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4}, NH\textsubscript{4}NO\textsubscript{3}, OC, EC, fine soil, and sea salt) measured synchronously online to analyze the influence of aerosol chemical compositions on sensitivity of visibility to PM\textsubscript{2.5} and RH changes.

2. Data and Methods

2.1. The Online Observation of PM\textsubscript{2.5} Chemical Composition and Meteorological Factors

Tianjin (39°100’ N, 117°100’ E) is in the Beijing–Tianjin-Hebei urban agglomeration. It is adjacent to Bohai Bay and has the largest comprehensive port in northern China, the Tianjin Port. The climate is dominated by continental monsoons with distinct seasonal variation and holds the semi-humid characteristic of warm, temperate zones. Sampling instruments used in this study were installed on the rooftop of the Tianjin Eco-Environmental Monitoring Center, approximately 10 m above ground level. The site was expected to reflect the pollution characteristics of a thickly settled urban district.

The mass concentration of PM\textsubscript{2.5} was sampled 1 time every 5 min using a particle monitor (TEOM 1405-F, Thermo Fisher Scientific, Waltham, MA, USA) from February to December 2015. The values measured within an hour were averaged to match with
other parameters. The monitor was composed of a filter dynamics measurement system and a tapered element oscillating microbalance mass sensor installed in an individual cabinet. The method detection limit (MDL) was up to 0.1 \( \mu g/m^3 \). The inorganic ions (Ca\(^{2+}\), Na\(^{+}\), NH\(_4\)\(^{+}\), Cl\(^{-}\), NO\(_3\)\(^{-}\), and SO\(_4^{2-}\)) of PM\(_{2.5}\) were synchronously analyzed by an ion-monitoring instrument (AIM URG9000D, Enviro Technology Services, Chapel Hill, NC, USA) with 1 h resolution. The AIM was composed of one particle collection system and two ion chromatographs for anion and cation analyses. The MDLs for Ca\(^{2+}\), Na\(^{+}\), NH\(_4\)\(^{+}\), Cl\(^{-}\), NO\(_3\)\(^{-}\), and SO\(_4^{2-}\) were 2.3, 0.6, 1.8, 0.2, 0.2, and 0.3 \( \mu g/m^3 \), respectively. The 1 h resolution concentration data of carbon components (OC and EC) in PM\(_{2.5}\) were determined by a semi-continuous OC/EC analyzer (Sunset Laboratory Inc., Portland, OR, USA). The analyzer was calibrated monthly using a blank punch of pre-heated quartz fiber filter and standard sucrose solutions (3.2 mgC/mL) for quality control. The MDLs for OC and EC were 0.45 and 0.06 \( \mu g/cm^2 \), respectively. The quartz fiber filter was changed weekly during the analysis.

Relative humidity was measured by a VAISALA WST520 automatic weather station with a time resolution of 5 s. The systematic error of relative humidity measurement was within \( \pm 3\% \). Atmospheric visibility was measured using a Belfort 6000 instrument (Belfort Instrument, Belfort, CA, USA), based on forward scattering with a time resolution of 10 min. The systematic error of atmospheric visibility measurement was less than 10%. The visibility values measured within an hour were averaged to match with other parameters. The hourly concentration data for PM\(_{2.5}\), water-soluble ions, carbon species, RH, and visibility simultaneously were designated as one sample. Instrument failure, bad weather, and other factors led to partial data loss or abnormality. Singular values (e.g., abnormally high values, abnormally low values, and values above the instrument limit or with a large relative standard deviation) were removed. Details on the quality control (QC) of observations were provided in studies [34,35]. This study obtained 4453 samples from Tianjin, 2015, after validation with QC procedures.

### 2.2. Parameterization Scheme of Atmospheric Visibility

Many studies found that there is a negative exponential relationship, as shown in Equation (1) [11,36]. Considering the influence of aerosol hygroscopic growth on visibility, RH was selected as another factor for visibility parameterization. Song et al. [36] found that there was a power relationship between RH and visibility, and the function was combined with Equation (1) to simulate visibility (Equation (2)). Another equation, Equation (3), evolved by multiplying the power function between extinction coefficient and aerosol concentration and the empirically power-exponential function between extinction coefficient and RH [22,37,38]. All variables could be easily acquired; thus, the parameterization was practical.

\[
V = f(x) = a \times \exp(b \times x) + c \quad (1)
\]

where the visibility is a function of one parameter, PM\(_{2.5}\): \( f(x) \); \( V \) and \( x \) are visibility and PM\(_{2.5}\) with units of km and \( \mu g/m^3 \), respectively; and parameters \( a, b, \) and \( c \) are the regression coefficients of the schemes.

\[
V = f(x, y) = a \times \exp(b \times x) + c \times y^d + e \quad (2)
\]

where the visibility is a function of two parameters, PM\(_{2.5}\) and RH: \( f(x,y) \); \( V, x, \) and \( y \) are visibility, PM\(_{2.5}\), and RH with units of km, \( \mu g/m^3 \), and \%, respectively; and parameters \( a, b, c, \) and \( d, e \) are the regression coefficients of the schemes.

\[
V = f(x, y) = a \times x^b \times (1 - y)^c \times y \quad (3)
\]

where the visibility is a function of two parameters, PM\(_{2.5}\) and RH: \( f(x,y) \); \( V, x, \) and \( y \) are visibility, PM\(_{2.5}\), and RH with units of km, \( \mu g/m^3 \), and \%, respectively; and parameters \( a, b, \) and \( c \) are the regression coefficients of the schemes.
2.3. Classification Method of Visibility-Sensitive Regime, Depending on the Sensitivity of Visibility to PM$_{2.5}$ Concentration and RH

To test the sensitivity of visibility to PM$_{2.5}$ concentration and RH, the normalized forward sensitivity index method [19,39,40] was used, shown as Equations (4) and (5). The normalized forward sensitivity index ($K_{V-R}$) of a variable (PM$_{2.5}$ concentration or RH) to visibility is the ratio of the relative change in the variable to the relative change in the visibility.

\[
K_{V-PM_{2.5}} = \left( \left| \frac{\Delta V_x}{V_x} \right| / \left| \Delta x_i / x_i \right| \right) = \left( |\Delta V_{x_i} / \Delta x_i| \right) \times x_i / V_{x_i} \tag{4}
\]

where $V$ is visibility with units of km; $x$ is PM$_{2.5}$ with units of $\mu g/m^3$; and $y$ is RH with units of $\%$; $x_i$ is the different level of PM$_{2.5}$ concentrations, $i = 1, 2, 3, \ldots, m$; $\Delta x_i$ is the variation of $x_i$; $V_{x_i}$ is the visibility under the $i$ level of PM$_{2.5}$ concentration; $\Delta V_{x_i}$ is the visibility variation caused by $\Delta x_i$; and $K_{V-PM_{2.5}}$ is the sensitivity index of visibility under the $i$ level of PM$_{2.5}$ concentration.

\[
K_{V-RH} = \left( \left| \frac{\Delta V_y}{V_y} \right| / \left| \Delta y_j / y_j \right| \right) = \left( |\Delta V_{y_j} / \Delta y_j| \right) \times y_j / V_{y_j} \tag{5}
\]

where $V, x, y$ are visibility, PM$_{2.5}$, and RH with units of km, $\mu g/m^3$, and $\%$, respectively. $y_j$ is the different level of RH, $j = 1, 2, 3, \ldots, n$; $\Delta y_j$ is the variation of RH at value $y_j$; $V_{y_j}$ is the visibility under the $j$ level of RH; $\Delta V_{y_j}$ is the visibility variation caused by $\Delta y_j$; and $K_{V-RH}$ is the sensitivity index of visibility under the $j$ level of RH.

When the variable is a differentiable function of the parameter, the sensitivity index may be alternatively defined using partial derivatives. There is a functional relationship among visibility, PM$_{2.5}$, and RH, shown as Equations (1)–(3) in this study. $K_{V-PM_{2.5}}$ and $K_{V-RH}$ can be approximately expressed as Equations (6) and (7):

\[
|\Delta V_x / \Delta x| \approx |f'_x (x, y)| \tag{6}
\]

\[
|\Delta V_y / \Delta y| \approx |f'_y (x, y)| \tag{7}
\]

where $V$ is visibility with units of km; $x$ is PM$_{2.5}$ with units of $\mu g/m^3$; and $y$ is RH with units of $\%$; $f(x, y)$ is the parameterization scheme of visibility, PM$_{2.5}$ concentration, and RH obtained in Section 2.2; $f'_x (x, y)$ and $f'_y (x, y)$ are partial derivatives of $f(x, y)$, respectively.

Visibility relative sensitivity index $\Omega_{PM/RH}$ is defined as Equation (8):

\[
\Omega_{PM/RH} = K_{V-PM_{2.5}} / K_{V-RH} \tag{8}
\]

where $K_{V-PM_{2.5}}$ and $K_{V-RH}$ is the sensitivity index of visibility to PM$_{2.5}$ concentration and RH, respectively; and $\Omega_{PM/RH}$ is the ratio of visibility variation per unit of PM$_{2.5}$ to RH.

When $\Omega_{PM/RH} = 1$, the effects of the concentration of PM$_{2.5}$ and RH on visibility is similar. When the ratio is greater than one, indicating that visibility is more sensitive to PM$_{2.5}$ than RH, visibility is identified as being in the PM$_{2.5}$-sensitive regime. When $\Omega_{PM/RH}$ is lower than 1, the visibility variation caused by the increase in RH is greater than that caused by particle concentration, and visibility is identified as being in the RH-sensitive regime. The classification method of visibility for different control categories was used to identify the main influencing factors by determining the threshold of aerosol mass concentration and RH.

2.4. IMPROVE Equations

In this study, the extinction contribution of the composition of fine particles to visibility degradation was investigated using the Interagency Monitoring of Protected Visual Environments (IMPROVE) equation, an extensively used method for estimating the light extinction coefficient based on aerosol chemical composition [41,42]. Equation (9) is the
revised IMPROVE algorithm in which coarse particulate, NO₂, and Rayleigh scattering were excluded because of the low contributions [42,43].

\[
B_{\text{ext}} \approx 2.2 \times f_S(RH) \times \text{[Small(}NH_4\text{)SO}_4\text{]} + 4.8 \times f_L(RH) \times \text{[Large(}NH_4\text{)SO}_4\text{]} + 2.4 \times f_S(RH) \times \text{[Small(}NH_4\text{)SO}_4\text{]} + 5.1 \times f_L(RH) \times \text{[Large(}NH_4\text{)SO}_4\text{]} + 2.8 \times \text{[Small OM]} + 6.1 \times \text{[Large OM]} + 10 \times \text{[EC]} + 1 \times \text{[Fine Soil]} + 1.7 \times f_{SS}(RH) \times \text{[Sea Salt]}
\]

where \([\text{NH}_4\text{)}_2\text{SO}_4\], \([\text{NH}_2\text{NO}_3\text{}], [\text{OM}], [\text{EC}], [\text{Fine Soil}], \) and \([\text{Sea Salt}]\) are the concentrations of \((\text{NH}_4\text{)}_2\text{SO}_4\), \(\text{NH}_4\text{NO}_3\), \(\text{OM}\) (organic matter), \(\text{EC}\), fine soil, and sea salt in units of \(\mu g/m^3\); \(B_{\text{ext}}\) is the light extinction coefficient of aerosol, \(\text{Mm}^{-1}\). The fraction of the fine particle component ((\(\text{NH}_4\text{)}_2\text{SO}_4\), \(\text{NH}_4\text{NO}_3\), or OM) that is in the large mode is estimated by dividing the total concentration of the component by 20 \(\mu g/m^3\). The coefficient of the equation is the dry mass extinction efficiency of each extinction component, which represents the light scattering or absorption extinction coefficient under the unit mass concentration. \(f_S(RH)\), \(f_L(RH)\), and \(f_{SS}(RH)\) is the hygroscopic growth factor of small- and large-mode ammonium salt and sea salt, determined according to the measured RH [42].

Most of the sulfate, nitrate, and organic carbon in the particles are in the form of \((\text{NH}_4\text{)}_2\text{SO}_4\), \(\text{NH}_4\text{NO}_3\), and OM, respectively [44]. The average equivalent concentrations of \(\text{NH}_4^+\), \(\text{SO}_4^{2-}\), \(\text{NO}_3^-\) measured in this study were 0.69, 0.13, and 0.22 \(\mu eq/m^3\), respectively. There was sufficient \(\text{NH}_4^+\) to match \(\text{SO}_4^{2-}\) and \(\text{NO}_3^-\). Therefore, the concentrations of \((\text{NH}_4\text{)}_2\text{SO}_4\) and \(\text{NH}_4\text{NO}_3\) were reconstructed based on Equations (10) and (11). OM was estimated by multiplying the OC by 1.6, which is suitable for urban aerosol (Equation (12)) [42,45]. Sea salt mass was calculated based on the concentrations of \(\text{Na}^+\) and \(\text{Cl}^-\) (Equation (13)) [44]. Fine soil mass was assumed to be 20 times of that of \(\text{Ca}^{2+}\) based on previous soil source profiles (Equation (14)) [46–48].

\[
([\text{NH}_4\text{)}_2\text{SO}_4] = 1.29 \left[\text{SO}_4^{2-}\right]
\]

(10)

where \([\text{SO}_4^{2-}]\) and \([\text{(NH}_4\text{)}_2\text{SO}_4]\) are the concentrations of \(\text{SO}_4^{2-}\) and \((\text{NH}_4\text{)}_2\text{SO}_4\) in units of \(\mu g/m^3\).

\[
[\text{NH}_4\text{NO}_3] = 1.375 [\text{NO}_3^-]
\]

(11)

where \([\text{NO}_3^-]\) and \([\text{NH}_4\text{NO}_3]\) are the concentrations of \(\text{NO}_3^-\) and \(\text{NH}_4\text{NO}_3\) in units of \(\mu g/m^3\).

\[
[\text{OM}] = 1.6 [\text{OC}]
\]

(12)

where \([\text{OC}]\) and \([\text{OM}]\) are the concentrations of \(\text{OC}\) and \(\text{OM}\) in units of \(\mu g/m^3\).

\[
[\text{Sea Salt}] = 1.47 [\text{Na}^+] + [\text{Cl}^-]
\]

(13)

where \([\text{Sea Salt}], [\text{Na}^+]\) and \([\text{Cl}^-]\) are the concentrations of \(\text{sea salt}\), \(\text{Na}^+\), and \(\text{Cl}^-\) in units of \(\mu g/m^3\).

\[
[\text{Fine Soil}] = 20 [\text{Ca}^{2+}]
\]

(14)

where \([\text{Fine Soil}]\) and \([\text{Ca}^{2+}]\) are the concentrations of \(\text{fine soil}\) and \(\text{Ca}^{2+}\) in units of \(\mu g/m^3\).

3. Results and Discussion

3.1. Application of Visibility Control Category Classification Method

3.1.1. Quantification of Relationships among Visibility, PM₂.₅ Concentration, and RH in Tianjin, 2015

Visibility varied from 0.3 to 35.0 km, with an average of 12.0 ± 9.1 km, from February to December 2015 in Tianjin. The average mass concentration of PM₂.₅ was 106.0 ± 81.2 \(\mu g/m^3\), and the average RH was 46 ± 21%. There was a strong, negative correlation among visibility and PM₂.₅ (−0.64) and RH (−0.67). There was an exponential or power function among visibility, PM₂.₅, and RH (Figure 1). The visibility decreased as PM₂.₅ increased in different RH ranges. There was a threshold for the sensitivity of visibility and PM₂.₅. Under
dry conditions (23% ≤ RH < 70%), the threshold of PM$_{2.5}$, corresponding to a visibility of 10 km, was 112 µg/m$^3$; however, under an RH of 70–80%, this threshold was lowered to 40 µg/m$^3$. Visibility was almost lower by 10 km when RH was 80–90%. A nonlinear correlation was observed between visibility and PM$_{2.5}$, which was affected by RH in most cases [49]. However, the relationship between visibility and PM$_{2.5}$ was not dependent on RH at higher concentrations and higher RH, because the black and red lines overlap at higher concentrations.

![Figure 1](image)

**Figure 1.** Variation in visibility with PM$_{2.5}$ in different RH conditions in Tianjin, 2015. Data points are colored to represent relative humidity (RH ≤ 70%, 70% < RH ≤ 80%, 80% < RH ≤ 90%); data (40 µg/m$^3$, 10 km) and (112 µg/m$^3$, 10 km) are the thresholds of PM$_{2.5}$ (40 and 112 µg/m$^3$) corresponding to the visibility of 10 km under the conditions of 70% < RH ≤ 80% and RH ≤ 70%.

The relationship among visibility, PM$_{2.5}$ concentration, and RH was investigated and quantified based on hourly data obtained from Tianjin, 2015, and 4453 valid datasets were used in the regression analysis (Equations (1)–(3)). The F-test was applied with a confidence level of 95% (α = 0.05). The regression coefficients for the three parameterization schemes are listed in Table S1 in Supplementary Materials. To test the reliability, this study compared the V calculated from the regression and the measured visibilities (Figure 2). The determination $R^2$ of Equation (3) at a confidence level of 95% was higher than that of the other two equation forms (0.72 vs. 0.55 and 0.55). The slope of measured and calculated visibility by Equation (3) (0.67) was closer to 1 than these of Equations (1) and (2) (0.55 and 0.55). This result reveals that the compound form of the power and power-exponential function can increase the accuracy of the parameterization scheme for visibility calculation (Equation (15)) in Tianjin, 2015.

$$V = f(x, y) = 166.1 \times x^{-0.56} \times (1 - y)^{0.86y}$$

(15)

where the visibility is a function of two parameters, PM$_{2.5}$ and RH: $f(x, y)$; V was the visibility, varying in the range of 0.3–34.7 km; x was the PM$_{2.5}$ concentration, varying in the range of 13–581 µg/m$^3$; and y was the RH, varying in the range of 4–90% in this study.
3.1.2. Sensitivity of Visibility to PM$_{2.5}$ Concentration and RH

The parameterization scheme of visibility in Tianjin, 2015 (Equation (15)), was incorporated into Equations (6) and (7) (Section 2.3). At the same PM$_{2.5}$ ($x_i$) and RH ($y_j$) points, $V_{x_i}$ was equal to $V_{y_j}$. The visibility relative sensitivity index ($\Omega_{PM/RH}$) in Tianjin, 2015, was calculated using Equations (16)–(18).

$$K_{V-PM_{2.5}} = (-93.01) \times x_i^{-0.56} \times (1 - y_j)^{0.86}/V_{x_i}$$  \hspace{1cm} (16)

$$K_{V-RH} = 142.84 \times x_i^{-0.56} \times y_j \times (1 - y_j)^{0.65}/[\ln(1 - y_j) - y_j/(1 - y_j)]/V_{y_j}$$ \hspace{1cm} (17)

$$\Omega_{PM/RH} = (-0.65)/[y_j \times [\ln(1 - y_j) - y_j/(1 - y_j)])$$ \hspace{1cm} (18)

where $V$, $x$, and $y$ are visibility, PM$_{2.5}$, and RH with units of km, $\mu g/m^3$, and %, respectively; $x_i$ is the different level of PM$_{2.5}$ concentrations, $i = 1, 2, 3 \ldots m$; $V_{x_i}$ is the visibility under the $i$ level of PM$_{2.5}$ concentration; $y_j$ is the different level of RH, $j = 1, 2, 3 \ldots n$; $V_{y_j}$ is the visibility under the $j$ level of RH; $K_{V-PM_{2.5}}$ and $K_{V-RH}$ is the sensitivity index of visibility under the $i$ level of PM$_{2.5}$ concentration and RH, respectively; $\Omega_{PM/RH}$ is the ratio of visibility variation per unit of PM$_{2.5}$ to RH.

As shown in Figure 3, $\Omega_{PM/RH}$ decreased as RH increased. When RH was 45%, $\Omega_{PM/RH}$ was equal to one. The influence of PM$_{2.5}$ and RH on visibility was similar. When RH was lower than 45%, especially under 23% ($\Omega_{PM/RH} = 5$), $\Omega_{PM/RH}$ increased sharply with the decrease in RH. Thus, visibility was mainly influenced by PM concentration, and visibility was defined as being in the PM$_{2.5}$-sensitive regime. At an RH above 75% ($\Omega_{PM/RH} = 0.2$), $\Omega_{PM/RH}$ was close to zero and did not change. It means that RH was gradually becoming the main factor influencing visibility, and visibility was identified as being in the RH-sensitive regime when RH was above 75%.

The values of 0.6, 1, 5, 10, and 15 km of visibility are the key points that refer to the classification of dense fog, fog, mist, haze, and good days. By combining Equations (15) and (18), the PM$_{2.5}$ and RH threshold values of visibility of different control categories for the above levels were determined respectively. When $\Omega_{PM/RH} = 1$, RH was 45% and PM$_{2.5}$ was 345, 100, and 48 $\mu g/m^3$, corresponding to visibility at 5, 10, and 15 km, respectively (Figure 4).
When visibility was in the RH-sensitive regime ($\Omega_{PM/RH} = 0.2$), the threshold of RH was 75% and PM$_{2.5}$ was 106, 31, and 15 µg/m$^3$, corresponding to visibility at 5, 10, and 15 km, respectively. When visibility was in the PM$_{2.5}$-sensitive regime ($\Omega_{PM/RH} = 5$), the threshold of RH was 75% and PM$_{2.5}$ was 475, 138, and 67 µg/m$^3$, corresponding to visibility at 5, 10, and 15 km, respectively. When RH was >75%, visibility was more sensitive to the variation in RH than to the increase in PM$_{2.5}$. The threshold values of PM$_{2.5}$ were out of the observation values measured in this study (581 µg/m$^3$) at 0.6 and 1 km level of visibility,

Figure 3. Variation characteristics of the visibility relative sensitivity index ($\Omega_{PM/RH}$) in Tianjin, 2015 (abscissa is RH, 10–90%; the ordinate is relative sensitivity index).

Figure 4. The PM$_{2.5}$ thresholds for different visibility-sensitive regimes ($\Omega_{PM/RH}$) = 0.2, 1, and 5, respectively) under different visibility levels in Tianjin, 2015.
meaning that it is hard for the contribution of particulate matter to exceed the relative humidity during dense fog and fog days.

The result shows that the classification method of visibility control categories built in this study can be used to identify the dominant factors of atmospheric visibility. The precondition was that there was a linear or nonlinear relationship among atmospheric visibility, PM$_{2.5}$, and RH, and the relationships were well fitted.

3.2. Sensitivity of Visibility to PM$_{2.5}$ Concentration and RH for Different Aerosol Types

PM$_{2.5}$ and RH were the main influencing factors on visibility, and a well-fitted parameterization scheme of visibility was provided in this study (Equation (15)). This new model could be applicable and easily transferrable to other datasets worldwide. However, there remained certain deviations between the calculated and measured visibilities ($R^2 = 0.72$; slope = 0.67) due to the difference in the aerosol extinction component.

3.2.1. Constituents and Extinction Characteristics of Chemical Composition in PM$_{2.5}$

Hourly concentrations of extinction compositions, such as (NH$_4$)$_2$SO$_4$, NH$_4$NO$_3$, OM, fine soil, and sea salt, were reconstructed based on Equations (10)–(14). The extinction contribution of the compositions to visibility degradation was investigated using the revised IMPROVE equation (Equation (9)). The correlation coefficient between the extinction coefficient calculated by the IMPROVE equations and visibility (Equation (19), Koschmieder’s law) reached 0.88. The aerosol extinction coefficient calculated by the IMPROVE equations was highest in December (1147.4 Mm$^{-1}$) and lowest in June (117.0 Mm$^{-1}$), shown in Figure 5a. The difference was by up to 10 times. The primary extinction components were different in different months (Figure 5b). The contribution of hygroscopic compositions (NH$_4$NO$_3$, (NH$_4$)$_2$SO$_4$, and sea salt) to extinction coefficient was up to 80% in November, while it was only 46% in February. The contribution of OM to extinction was highest in February (35%), followed by October (25%), June (31%), and March (30%).

$$B_{ext} = k / V \times 1000$$

where $B_{ext}$ is the light extinction coefficient of aerosol, Mm$^{-1}$; $V$ is visibility, km; and $k$ is assumed to be 1.97 when the visibility is <10 km. Otherwise, $k$ is assumed to be 3.912 [46,50].

![Figure 5.](image.png)

Figure 5. The monthly average extinction coefficients (a) and contributions (b) of main extinction components in PM$_{2.5}$ in Tianjin, 2015.
The annual average proportions of \((\text{NH}_4)_2\text{SO}_4\), \(\text{NH}_4\text{NO}_3\), OM, EC, fine soil, and sea salt in PM$_{2.5}$ were 14%, 17%, 19%, 3%, 6%, and 5%, respectively. The proportions of extinction compositions in PM$_{2.5}$ changed obviously with the seasons. The monthly average proportions of extinction compositions in PM$_{2.5}$ are shown in Figure 6a. In December, November, and July, the major components accounted for the highest proportion of PM$_{2.5}$: 104%, 77%, and 68%, respectively. The proportions of major extinction compositions in PM$_{2.5}$ were lowest in February (51%) and October (51%). The content difference in the extinction component in PM$_{2.5}$ for different seasons could reach up to 2.0 times. The hygroscopic compositions of PM$_{2.5}$ accounted for the highest proportion in December (60%), followed by November (51%) and July (40%). These results suggest that constituents of the extinction composition in PM$_{2.5}$ differed by period.

\[
P_{\text{Phyg}} + P_{\text{PCE}} + P_{\text{PEC}} + P_{\text{PFS}} + P_{\text{POM}} + P_{\text{PSS}} = 100\%
\]

\[
P_{\text{Phyg}} = \left(\text{NH}_4\text{NO}_3\right)
\]

\[
P_{\text{PCE}} = \text{(NH}_4)_2\text{SO}_4
\]

\[
P_{\text{PEC}} = \text{OM, EC, fine soil, and sea salt}
\]

\[
P_{\text{PFS}} = \text{OM, EC, fine soil, and sea salt}
\]

\[
P_{\text{POM}} = \text{OM, EC, fine soil, and sea salt}
\]

\[
P_{\text{PSS}} = \text{OM, EC, fine soil, and sea salt}
\]

Figure 6. Monthly average proportions of major extinction compositions in PM$_{2.5}$ (a) and the variation of extinction coefficient with increased RH under different aerosol types, taking chemical compositions of PM$_{2.5}$ in different months for examples (b); chemical composition of PM$_{2.5}$ was the monthly mean proportions of extinction components in PM$_{2.5}$ in Tianjin, 2015; PM$_{2.5}$ concentration was set to 100 µg/m$^3$; the extinction coefficient was calculated by IMPROVE equation.

To explore the influence of composition constituents on aerosol extinction, this study used the monthly average proportions of \((\text{NH}_4)_2\text{SO}_4\), \(\text{NH}_4\text{NO}_3\), OM, EC, fine soil, and sea salt to calculate extinction coefficients with the IMPROVE equation under the same level of PM$_{2.5}$ mass concentration (100 µg/m$^3$) and RH (Figure 6b). The extinction coefficients of aerosols in December were twice that of aerosols in June, even though the PM$_{2.5}$ and RH were same. This result was observed because particles with more extinction components were more efficient light scatterers and absorbers.

In February, the sum of major extinction components accounted for 53% of PM$_{2.5}$ and was similar to the 54% in August. However, the content of hygroscopic components in August (33%) was higher than that in February (22%). At low RH, the extinction coefficient in February was higher than that in March. With the increase in RH, the growth rate of the extinction coefficient in August was faster than that in February, and it was higher than that in February when RH was more than 85% (Figure 6b). The extinction coefficient of aerosol was more sensitive to RH when the content of hygroscopic components was higher.

In summary, the total content of the extinction components in the particles was the main factor affecting the extinction ability of aerosols, and the content of the hygroscopic components played a critical role in affecting their sensitivity to RH.
3.2.2. Classification of Aerosol Types

The relationships among visibility, PM$_{2.5}$, and RH depended on the extinction capacity of the aerosol composition to a certain extent. In this study, a new aerosol classification method was established based on the percentages of hygroscopic (NH$_4$NO$_3$, (NH$_4$)$_2$SO$_4$, and sea salt) and non-hygroscopic (OM, EC, and fine soil) components in PM$_{2.5}$, shown as Equations (20) and (21):

$$\begin{align*}
P_{\text{hyg}} &= P_{(\text{NH}_4)\text{SO}_4} + P_{\text{NH}_4\text{NO}_3} + P_{\text{SS}} \\
P_{\text{no-hyg}} &= P_{\text{OM}} + P_{\text{EC}} + P_{\text{FS}}
\end{align*}$$

where $P_{(\text{NH}_4)\text{SO}_4}$, $P_{\text{NH}_4\text{NO}_3}$, $P_{\text{SS}}$, $P_{\text{OM}}$, $P_{\text{EC}}$, $P_{\text{FS}}$, $P_{\text{hyg}}$, and $P_{\text{no-hyg}}$ are the percentages of NH$_4$NO$_3$, (NH$_4$)$_2$SO$_4$, sea salt, OM, EC, fine soil, hygroscopic, and non-hygroscopic extinction components in PM$_{2.5}$, respectively.

The percentages of hygroscopic and non-hygroscopic extinction components concentrated mainly on the range of 10–70% and 10–50%, shown in Figure 7. A total of 4008 aerosol samples (accounting for 90% of all samples) were classified into six types according to the numerical value of $P_{\text{hyg}}$ and $P_{\text{no-hyg}}$. At the same particle concentration level, the higher the $P_{\text{hyg}} + P_{\text{no-hyg}}$ was, the more sensitive the visibility was to the change in the fine particle concentration. The higher the $P_{\text{hyg}}$ was, the more sensitive the visibility was to the change in RH.

![Figure 7](image-url)

Figure 7. The percentages of hygroscopic and non-hygroscopic extinction components in PM$_{2.5}$ in Tianjin, 2015 (the numbers in brackets represent the area serial numbers for different aerosol types).

The characteristics and sample size of the six aerosol types are shown in Table S2. The sample size of aerosols with a low (types 1 and 2) and medium (types 3 and 4) content of hygroscopic components was similar, accounting for 35% and 41%, respectively. Aerosols with a high content of hygroscopic components (types 5 and 6) made up 14%. The content of non-hygroscopic extinction components (types 1, 3, and 5) in most samples (68%) was lower than 30%. Of the aerosols, 22% were in types 2, 4, and 6 with a medium content of non-hygroscopic extinction components. A good correlation between visibility and PM$_{2.5}$
was found for different aerosol types with the correlation coefficient varying from $-0.54$ to $-0.71$. The correlation coefficient between visibility and RH increased from $-0.16$ to $-0.70$ with the increase of the content of hygroscopic components.

3.2.3. Impacts of Chemical Compositions on the Sensitivity of Visibility to PM$_{2.5}$ and RH

The varied correlation among visibility, PM$_{2.5}$, and RH for different aerosol types indicated that the content of chemical compositions greatly influences the sensitivity of visibility to PM$_{2.5}$ and RH. In this study, the quantitative relationship among visibility, PM concentration, and RH in different aerosol types was fitted based on Equations (1)–(3). The compound form of the power and power-exponential function was still identified as the optimal fit among visibility, PM$_{2.5}$, and RH (Equation (3)). Coefficients and fitting effects varied by aerosol type (Table 1). The calculated visibility for types 1, 2, and 4 fitted the measured visibility less effectively; $R^2$ was 0.65, 0.66, and 0.64, respectively. The $R^2$ between the measured and calculated visibility in aerosols with high hygroscopic and low non-hygroscopic content (types 3, 5, and 6) was above 0.76. The linear slopes of correlation between calculated and measured visibility were higher (0.72–0.84) in types 3, 5, and 6 than in other types. The fitting equation for types 3, 5, and 6 increased the accuracy of the parameterization scheme for visibility calculation.

Table 1. Fitting equation and effect of visibility, PM$_{2.5}$, and RH for different aerosol types.

| Type | Fitting Equation | Relationship between Measured and Calculated Visibility | $R^2$ |
|------|------------------|--------------------------------------------------------|-------|
| 1    | $V = 447.9 \times [PM_{2.5}]^{-0.77} \times (1 - RH)^{0.67 \cdot RH}$ | $y = 0.62x + 4.66$ | 0.65  |
| 2    | $V = 159.4 \times [PM_{2.5}]^{-0.55} \times (1 - RH)^{0.42 \cdot RH}$ | $y = 0.64x + 5.83$ | 0.66  |
| 3    | $V = 496.6 \times [PM_{2.5}]^{-0.82} \times (1 - RH)^{0.99 \cdot RH}$ | $y = 0.75x + 1.93$ | 0.76  |
| 4    | $V = 176.7 \times [PM_{2.5}]^{-0.59} \times (1 - RH)^{0.72 \cdot RH}$ | $y = 0.61x + 4.58$ | 0.64  |
| 5    | $V = 491.9 \times [PM_{2.5}]^{-0.85} \times (1 - RH)^{1.08 \cdot RH}$ | $y = 0.84x + 0.65$ | 0.84  |
| 6    | $V = 132.4 \times [PM_{2.5}]^{-0.51} \times (1 - RH)^{1.04 \cdot RH}$ | $y = 0.72x + 1.90$ | 0.76  |
| annual | $V = 166.1 \times [PM_{2.5}]^{-0.56} \times (1 - RH)^{0.86 \cdot RH}$ | $y = 0.67x + 3.65$ | 0.72  |

Based on the division method of visibility control categories established in Section 2.3, the thresholds for identifying the PM$_{2.5}$-sensitive regime and RH-sensitive regime under different aerosol types were determined (Figure 8).

The PM$_{2.5}$ and RH thresholds of varied visibility-sensitive regimes differed with aerosol types. When visibility was 10 km, the PM$_{2.5}$-sensitive regime was identified by RH < 30%, 31%, 26%, 26%, 25%, and 20% and PM$_{2.5}$ > 129, 141, 106, 117, 91, and 146 µg/m$^3$ in types 1–6, respectively. The RH-sensitive regime was characterized by RH > 83%, 85%, 79%, 78%, 78%, and 70% and by PM$_{2.5}$ < 40, 45, 26, 30, 22, and 29 µg/m$^3$ in types 1–6, respectively. Other visibility control categories were classified into transition regimes dominated by PM$_{2.5}$ and RH with RH = 56%, 58%, 50%, 49%, 49%, and 41% and PM$_{2.5}$ = 95, 105, 77, 86, 65, and 103 µg/m$^3$, respectively. The threshold difference of PM$_{2.5}$ was by up to 1.6 times.
Table 1. Fitting equation and effect of visibility, PM2.5, and RH for different aerosol types.

| Type | Fitting Equation | Relationship between Measured and Calculated Visibility | $R^2$ |
|------|------------------|--------------------------------------------------------|-------|
| 1    | $V = 447.9 \times [PM2.5] - 0.77 \times (1 - RH)^{0.67} \cdot RH$ | $y = 0.62x + 4.66$ | 0.65 |
| 2    | $V = 159.4 \times [PM2.5] - 0.55 \times (1 - RH)^{0.42} \cdot RH$ | $y = 0.64x + 5.83$ | 0.66 |
| 3    | $V = 496.6 \times [PM2.5] - 0.82 \times (1 - RH)^{0.99} \cdot RH$ | $y = 0.75x + 1.93$ | 0.76 |
| 4    | $V = 176.7 \times [PM2.5] - 0.59 \times (1 - RH)^{0.72} \cdot RH$ | $y = 0.61x + 4.58$ | 0.64 |
| 5    | $V = 491.9 \times [PM2.5] - 0.85 \times (1 - RH)^{1.08} \cdot RH$ | $y = 0.84x + 0.65$ | 0.84 |
| 6    | $V = 132.4 \times [PM2.5] - 0.51 \times (1 - RH)^{1.04} \cdot RH$ | $y = 0.72x + 1.90$ | 0.76 |
| annual | $V = 166.1 \times [PM2.5] - 0.56 \times (1 - RH)^{0.86} \cdot RH$ | $y = 0.67x + 3.65$ | 0.72 |

Based on the division method of visibility control categories established in Section 2.3, the thresholds for identifying the PM2.5-sensitive regime and RH-sensitive regime under different aerosol types were determined (Figure 8).

The PM2.5 and RH thresholds of varied visibility-sensitive regimes differed with aerosol types. When visibility was 10 km, the PM2.5-sensitive regime was identified by RH < 30%, 31%, 26%, 26%, 25%, and 20% and PM2.5 > 129, 141, 106, 117, 91, and 146 µg/m³ in types 1–6, respectively. The RH-sensitive regime was characterized by RH > 83%, 85%, 79%, 78%, 78%, and 70% and by PM2.5 < 40, 45, 26, 30, 22, and 29 µg/m³ in types 1–6, respectively. Other visibility control categories were classified into transition regimes dominated by PM2.5 and RH with RH = 56%, 58%, 50%, 49%, 49%, and 41% and PM2.5 = 95, 105, 77, 86, 65, and 103 µg/m³, respectively. The threshold difference of PM2.5 was by up to 1.6 times.

Figure 8. PM2.5 (the histogram) and RH (the pink scatter) threshold values of visibility-sensitive regimes for aerosol types in Tianjin, 2015 (a–c are the values with a relative sensitivity index ($\Omega_{PM/RH}$) = 5, 1, and 0.2, respectively).

According to the threshold values of the visibility control categories, the proportion of those in different types in Tianjin, 2015, is shown in Table 2. The proportion of transition regimes dominated by PM2.5 in type 1 was up to 48%, and that of transition regimes dominated by RH was 23%. The proportion of transition regimes dominated by PM2.5 was still dominant (47%) in type 2. The proportions of the PM2.5-sensitive regime (47%) in type 2 were higher than those of the other types. The proportions of the transition regimes dominated by PM2.5 and RH were similar (31% vs. 31% and 59% vs. 54%, respectively) in types 3 and 4. The transition regime dominated by RH became prominent in types 5 and 6 (77% and 46%). The proportions of the RH-sensitive regime were higher than those of other types in type 6 (44%).

Table 2. Proportion of different visibility-sensitive regimes under varied aerosol types in Tianjin, 2015 (%).

| Aerosol Types | PM2.5-Sensitive Regime | Transition Regime Dominated by PM2.5 | Transition Regime Dominated by RH | RH-Sensitive Regime |
|---------------|------------------------|--------------------------------------|----------------------------------|---------------------|
| 1             | 27                     | 48                                   | 23                               | 2                   |
| 2             | 47                     | 47                                   | 6                                | 0                   |
| 3             | 6                      | 31                                   | 59                               | 3                   |
| 4             | 9                      | 31                                   | 54                               | 6                   |
| 5             | 1                      | 13                                   | 77                               | 9                   |
| 6             | 3                      | 8                                    | 46                               | 44                  |
| annual        | 13                     | 31                                   | 47                               | 9                   |

4. Conclusions

Many previous studies were performed to explore the complex relationships among visibility, PM2.5 concentration, and RH. However, the relative contribution of the two fac-
tors to visibility degradation, especially for different aerosol types, was difficult to quantify. The impact of chemical compositions on the sensitivity of visibility to PM$_{2.5}$ and RH was explored based on the online monitoring data of RH, visibility, PM$_{2.5}$, and its major components in this study. The normalized forward sensitivity index method for identifying the dominant factors of visibility was used based on the functional equation among visibility, RH, and PM$_{2.5}$ mass concentration given different aerosol types. Visibility was identified as being in the PM$_{2.5}$- or RH-sensitive regime according to the corresponding threshold.

The method was verified and evaluated in Tianjin. There was a compound relationship of the power and power-exponential functions among visibility, PM$_{2.5}$, and RH. The R$^2$ between the calculated and measured visibilities reached 0.72, and the slope was 0.67. Visibility was identified as being in the PM$_{2.5}$-sensitive or RH-sensitive regime. Corresponding thresholds of PM$_{2.5}$ were determined by function among visibility, PM$_{2.5}$, and RH. Chemical compositions ((NH$_4$)$_2$SO$_4$, NH$_4$NO$_3$, OC, EC, fine soil, and sea salt) influenced the sensitivity of visibility to PM$_{2.5}$ and RH due to varying degrees of dry extinction efficiency and hygroscopicity. A good correlation between visibility and PM$_{2.5}$ was found for different aerosol types with the correlation coefficient varying from −0.54 to −0.71. The correlation coefficient between visibility and RH changed from −0.16 to −0.70 with the increase of the content of hygroscopic components. The results showed that the normalized forward sensitivity index method is suitable for various aerosol types and can be applied to identify the leading factors of atmospheric visibility decline as long as synchronous, high-resolution data of visibility, PM$_{2.5}$, and RH are collected. However, for different aerosol types, the PM$_{2.5}$ thresholds under the same visibility-sensitive regimes can be several times different, while the RH threshold changes gently. The thresholds for different aerosol types obtained in this study can be used as a reference for other cities with the data of extinction component content in PM$_{2.5}$. Next, we will further evaluate the applicability of the method in other cities.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/atmos13030471/s1; Table S1: Summary of regression coefficients for the Equations (1)–(3) parameterization schemes at 95 % confidence level ($\alpha = 0.05$); Table S2: Characteristics and sample size of six aerosol types in Tianjin 2015

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