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Comparison of parameterizations for the atmospheric extinction coefficient in Lin'an, China

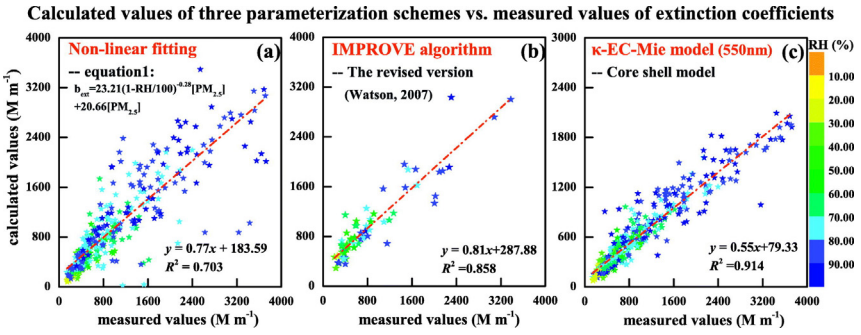
Lin Jiang [a], Zefeng Zhang [a], Bin Zhu [a,*], Yan Shen [b], Honglei Wang [a], Shuangshuang Shi [a], Dandan Sha [a]

[a] Collaborative Innovation Centre of Forecasting and Evaluation of Meteorological Disasters, Key Laboratory for Aerosol-Cloud-Precipitation, China Meteorological Administration, Nanjing University of Information Science & Technology, Nanjing 210044, China
[b] Enches Meteorological Bureau of Zhejiang Province, Fenghua 315500, China

HIGHLIGHTS

- Three parameterization schemes for studying the atmospheric extinction coefficient are evaluated.
- The uncertainties of the IMPROVE algorithm are influenced by the chemical composition of PM.
- The κ-EC-Mie model can reflect the visibility more objectively.

GRAPHICAL ABSTRACT



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ABSTRACT

A more precise atmospheric extinction coefficient would be advantageous for improving air quality (visibility) forecasting. In this study, the size distribution, chemical composition and relative humidity were measured in Lin'an from January 9 to 31, 2015. The merits and weaknesses of three parameterization schemes are discussed in this paper, including the non-linear fitting scheme, the IMPROVE (Interagency Monitoring of Protected Visual Environment) algorithm and the κ-elemental carbon (EC)-Mie model. Comparing the three schemes mentioned above, we find that the non-linear fitting equation requires the least amount of data, and its calculation process is the simplest. However, its calculated values are significantly influenced by specific data and fitting formulas. The uncertainty of the variable coefficients makes it difficult to directly implement this method for other datasets. The calculated values of the three versions of the IMPROVE algorithm strongly correlate with the measured values, with slopes near 1.0 and statistical indexes (R^2) of 0.848, 0.858 and 0.866. However, this method is affected by the chemical compositions of the particles in different regions; for example, when the quality of $PM_{2.5}$ is reconstructed from the measured data, the coefficients of different components change, thus affecting the final results. The κ-EC-Mie model objectively reflects the changes in the law of visibility, as the model considers both the chemical composition and size distribution of particles, and its predominant merit is derived from the fact that it is calculated without an empirical formula, which may eliminate the computational errors caused by the uncertainties of coefficients.

* Corresponding author.
E-mail address: binzhu@nuist.edu.cn (B. Zhu).

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1. Introduction

Atmospheric visibility degradation is an important environmental issue due to its demonstrated association with air pollution. Recently, visibility in clear skies has been found to have decreased over land globally from 1973 to 2007 (Wang et al., 2009). The reduction in visibility is not only an aesthetic problem but also an important indicator of deteriorating ambient air quality in urban areas (Baumer et al., 2008; Watson, 2002; Ying et al., 2007). Aside from meteorological elements, the reduction in visibility is mainly due to airborne particulate matter (PM), which can absorb or scatter light away from the initial path of transfer (Appel et al., 1985; Chan et al., 1999; Elias et al., 2009).

The atmospheric extinction coefficient represents the relative attenuation rate of light in the air when it propagates a unit distance. Observational studies of aerosol optical properties have been performed to reduce the uncertainties in estimating and forecasting visibility (Bellouin et al., 2003; Change, 2001; Liao and Seinfeld, 2005; Wu et al., 2012; Yu et al., 2016). Large uncertainties still exist in estimating aerosol optical properties; these uncertainties are related to the aerosol loadings, profiles, compositions, mixing states and atmospheric humidity (Höller et al., 2003). Research in Nanjing reveals that scattering aerosols are still the key control on atmospheric visibility. High relative humidity (RH) leads to a considerably higher scattering coefficient and a reduction in scattering aerosols (Zhuang et al., 2016). To systematically evaluate and predict atmospheric visibility for implementation in the early warning and forecasting business, the physical meaning of the extinction coefficient is considered. Many studies have attempted to establish a parametric model between the physical and chemical properties of aerosols and extinction coefficients.

Koschmieder found that atmospheric visibility was inversely proportional to atmospheric light transmittance in 1924 (Koschmeider, 1924), thus establishing an equation relating visibility and an extinction coefficient: $bext = \ln\varepsilon / Vis$. After that, many scientific groups have provided different fitting schemes to quantitatively describe the relationship between the extinction coefficient and its influencing factors. Of these schemes, the most widely used methods are as follows. The first method establishes a statistical relationship between the extinction coefficient and its influencing factors, including the concentrations of PM, such as PM_{10} and $PM_{2.5}$, and their chemical components, ozone gaseous pollutants, such as sulfur dioxide and nitrogen dioxide, and meteorological conditions, such as wind speed and direction, RH, and temperature. The second method uses an empirical formula established by IMPROVE (Interagency Monitoring of Protected Visual Environment) to calculate the aerosol extinction coefficient. This coefficient accounts for the chemical components of $PM_{2.5}$, and the total extinction is apportioned to individual major aerosol components: sulfate, nitrate, organic carbon (OC), black carbon (BC), soil and coarse mass (Han et al., 2012; Jun et al., 2009; Jung et al., 2009). The final method uses the Mie scattering model (Mie, 1908) to calculate the main optical parameters of spherical particles; in addition to considering the chemical composition, the particle size distributions of ions are also calculated. Based on the assumption that all aerosol samples are spherical particles, this distribution is combined with the aerosol concentration spectrum distribution. After confirming the hygroscopic growth regularity of particle size, the particle size distribution under different RHs is obtained. Afterwards, based on the Mie theory, the extinction properties of the particles at different levels of RH are calculated (Chen et al., 2011; Hulst and Twersky, 1957; Zhang et al., 2017). In the following section, we evaluate the accuracies of the values calculated by these schemes and their applicability to different datasets (which can be directly applied to the calculation in different regions).

The Yangtze River Delta (YRD) is one of the most rapidly developing regions in China both economically and socially (Cheung and Wang, 2001; Feng et al., 2015; Pu et al., 2014). To explore the air quality of a region, the measurement of pollutants in locations reflecting the "background" atmospheric conditions of the region is essential (Zheng et al., 2005). The Lin'an background air monitoring station is a site with negligible local anthropogenic emissions, but it is influenced by regionally transported pollutants. It is the only station that monitors changes in the atmospheric background composition of the YRD. Several studies have previously been conducted in Lin'an, and these studies confirm that Lin'an station is representative of the regional background of the YRD (Meng, 2014; Pu et al., 2014; Xu et al., 2002).

To the best of our knowledge, there have been few studies comparing different parameterization schemes. To determine whether these schemes have a wide range of applicability, the merits of their fit results were analysed in the context of meteorological data, PM concentrations, and chemical composition data. We intend to determine a method that can be applied in different regions to calculate aerosol extinction characteristics with high accuracy. If we can confirm its validity, this method may simplify the parameterization for atmospheric extinction, allowing for greater efficiency in future research.

2. Instruments and experiments

2.1. Observation stations and time

The Lin'an Background Air Monitoring Station (30°18′N, 119°44′E; located on top of a mountain 138.6 m above sea level), located in Zhejiang province, is one of the Global Atmospheric Watch background stations of the World Meteorological Organization in China, and it serves as a regional background site for the YRD region. The Lin'an station is located in a hilly agricultural/forested area with heavy vegetation coverage. There is no large village within 3 km. It is approximately 50 km east of Hangzhou, the capital city of Zhejiang province, and approximately 210 km southwest of Shanghai, the mega-city in the YRD region. To the west of Lin'an is the mountainous area of Zhejiang and Anhui provinces. It is a site with negligible local anthropogenic emissions, but it is influenced by regionally transported pollutants.

The sampling period for this study occurred from January 9 to 31, 2015. Winter is a period with a stable atmospheric stratum in the YRD. During this period, the near-surface wind speed is relatively low and pollutants are easy to accumulate and difficult to spread. Low visibility events occur frequently.

2.2. Instruments

The KC-120H flow sampler is used to collect aerosol samples at a sampling flow rate of 100 $L \cdot min^{-1}$; all samples were collected every 6 h. The particle number concentrations in the size range of 10 nm–10 μm were measured using a wide-range particle spectrometer (WPS) produced by USA MSP. The WPS combines the principles of differential mobility analyses (DMA), condensation particle counting (CPC), and laser light scattering (LPS). One complete scan of the entire size range with a 3 s scanning period of each channel takes approximately 5 min. Since the ambient humidity of the WPS is 0% to 90%, i.e., non-condensing, some invalid data were removed after data processing according to their observed implementation. A detailed description of this process was reported by Gao et al. (Gao et al., 2009) and Kang et al. (Kang et al., 2013).

The following water-soluble ions were measured by an 850 professional ion chromatograph (IC) (Metrohm model 850 professional IC, Switzerland): NH_4^+, Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, NO_3^-, SO_4^{2-}, F^- and NO_2^-. The filters were weighted after being balanced at a constant temperature (25 °C) and RH (50%) for 48 h before and after sampling. Half of each Teflon filter was cut, placed into a 20 mL PET vial, ultrasonically extracted for 0.5 h, and mechanically oscillated for 1 h with 10 mL of deionized water (18.2 MU). PTFE filters were used to remove the insoluble particles and filter chips in the extraction prior to the IC analysis. The chromatograph includes a column oven, a conductivity detector, an 858 auto-injector and a MagIC net chromatography workstation (Metrohm, Switzerland). The column oven consists of a Metrosep