



Increase of aerosol scattering by hygroscopic growth: Observation, modeling, and implications on visibility



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ABSTRACT

In situ measurements of size-resolved aerosol chemical compositions and its optical properties were concurrently carried out at an urban site in mega-city Beijing from October 24 to November 9, 2007. The main objective was to quantitatively study the relationship between aerosol chemical compositions and its hygroscopic properties, to estimate the influence of relative humidity (RH) on aerosol scattering coefficient and to quantitatively investigate visibility impairment due to particle hygroscopic growth. The hygroscopic factor of aerosol scattering coefficient ($f(RH)$), which is defined as the ratio of aerosol scattering coefficient at wet condition to that at dry condition ($RH \leq 30\%$), was calculated with the measured aerosol optical properties at dry and ambient conditions. The relationship between $f(RH)$ and RH was fitted by empirical equation and the fitting parameters were calculated. Meanwhile, $f(RH)$ for externally mixed aerosols or internally mixed aerosols was modeled based on size segregated particulate chemical composition. The modeled $f(RH)$ agreed well with the measured $f(RH)$. Empirical formula for atmospheric visibility based on mass concentration of $PM_{2.5}$ and $f(RH)$ was proposed. The result of this study is not only proven that RH, in addition to the mass concentration of $PM_{2.5}$, played an important role on visibility impairment, but also provide practical aid for air quality control to improve the visibility in the megacity region of Beijing.

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1. Introduction

Atmospheric aerosols scatter and absorb the incident light; therefore the atmospheric visibility reduces with the increase of their mass concentrations. Furthermore, the visibility would sharply decrease when the ambient relative humidity (RH) was high at the same level of aerosol mass concentration (Hodkinson, 1966; Malm et al., 2000). The main reason for this is that the mass scattering efficiency of aerosols would increase when water soluble aerosols grow larger in diameter (Deng et al., 2011). Aerosols can be divided

into hydrophilic and hydrophobic species according to its hygroscopicity. Diameter of hydrophilic salts (e.g. ammonium sulfate, ammonium nitrate) will grow many times than that at dry condition and scatter more light, but, the diameter of hydrophobic aerosols (e.g. black carbon) does not change even at high RH. Aerosol hygroscopic response is often described by the hygroscopic factor for aerosol scattering $f(RH)$, the ratio of the aerosol scattering coefficient at a given RH to that at a low reference RH (nominally $\leq 30\%$). For example, the value of $f(RH)$ for ammonium sulfate aerosols at 90% RH is about five times of that at dry conditions (Malm and Day, 2001), which implies that the ambient RH would play an important role on visibility impairment as long as there are sufficient water vapor in air as well as higher mass fraction of hydrophilic salts in aerosols.

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Many researchers had investigated the relationship among visibility, aerosol chemical species, RH, and other gaseous pollutants (Malm et al., 2000; Malm and Day, 2001; Nessler et al., 2005; Cheng et al., 2008b; Eichler et al., 2008; Liu et al., 2008; Pan et al., 2009; Liu et al., 2012). Water uptake of particles can contribute 50–60% to visibility degradation at 90% RH at rural site in Guangzhou of China (Cheng et al., 2008b). Liu et al. (2012) figured out that atmospheric visibility decreased about 35% under ambient RH ($74 \pm 13\%$) compared to dry condition ($<40\%$) in Guangzhou urban site. So, the impact of RH on light extinction could not be ignored when apportioning visibility based on chemical species. An empirical function among visibility, RH and chemical species was proposed (Malm and Day, 2001) by the IMPROVE (Interagency monitoring of protected visual environments) project. Although the chemical species of aerosols were scarcely monitored unless during the intensive campaign (Zhang et al., 2008), the mass concentration of particulate matter (PM) and RH was regularly measured by normal environmental monitoring station as ordinary course of business. Meanwhile, the monitoring of the atmospheric visibility is scarce, because visibility is now not incorporated into environmental assessment indicators. Thus if the empirical equation among visibility, PM concentration and RH is established, the visibility value from these basic parameters can be obtained. It would be meaningful for practical application to estimate the level of visibility based on the mass concentration of PM and RH.

On the other hand, public awareness of environmental protection and demands of improving atmospheric quality are increasing. The air quality understood intuitively by the people and reported by the government is often inconsistent in China. One possible reason may be that RH is ignored although RH played an important role on visibility impairment. The mass concentration of PM is measured with moisture removed, while, people's intuitive on air quality is directly to atmosphere at ambient conditions. So, investigating the relation of PM mass concentration, RH and atmospheric visibility is significant for government which draws up controlling strategies on PM for improving visibility under the direction of economic cost-effective.

2. Experiment

2.1. Experimental site

Beijing, the capital of the People's Republic of China, is the center for national politics, culture, and international exchange. The city's permanent population is 17 million and its population density is 1033 persons per square kilometer by the end of 2007. The gross domestic product in 2007 exceeded 130 billion dollars with the annual growth rate of 12.3% (<http://www.bjstats.gov.cn/nj/main/2007-tjnj/index.htm>). High population density and economic level had resulted in poor air quality in Beijing. For example, the hazy days (visibility < 10 km and RH $< 90\%$) in Beijing urban area was 17 days in 1971, sharply grew to 223 days in 1982, and keep this level (~ 200 days) until 1998 (Yang, 2008). Beijing had suffered from long-term poor air quality problems with aerosols as the major pollutants. Since 1998, Beijing had vigorously implemented a number of measures focusing on the management of coal, motor vehicles, industrial and dust pollution to improve air quality, as a result,

the hazy day quickly decreased to 73 days in 2005. Accordingly, the “blue sky”, which indicated the visibility is equal to or larger than 10 km, had increased by 100 days in 1998 and steadily grew to 246 days in 2007 (Yang, 2008).

The incidence of hazy day is high in autumn in Beijing. Hence, the measurements were carried out at an urban site in Beijing from October 24 to November 9, 2007. The measurements were carried out on the campus of Peking University (39.98°N , 116.35°E), which lies in the north-western part of Beijing. The observation point is located on the roof of a 6-floor building (~ 20 m above ground level), which is ~ 600 m to the north of the 4th ring road that acts as the main traffic line of the city. At Beijing urban site, possible air pollutant sources, which determined their chemical and hygroscopic properties, were local vehicular traffic, combustion of fuels for cooking, and some of transported pollutants.

2.2. Instrumentation and measurements

During the measurement, atmospheric visibility, aerosol scattering coefficient σ_{sp} in dry conditions, aerosol absorption coefficient σ_{ap} , NO_2 concentration and RH were measured by an online forward-scatter visibility meter (Model FD12, Vaisala, Finland), integrating nephelometer (Model M9003, Ecotech, Australia), multi-angle absorption photometer (MAAP, Model 5012, Thermo Fisher Scientific), $\text{NO}-\text{NO}_2-\text{NO}_x$ analyzer (Model 9841, Ecotech), and automatic meteorological station, respectively. The visibility meter and meteorological station are installed outdoor and nephelometer, MAAP as well as $\text{NO}-\text{NO}_2-\text{NO}_x$ analyzer are installed in the air-conditioned room.

The visibility meter consisted of a transmitter, a receiver, and a controller. The receiver detects the light scattered at 33° scattering angle, where the differences among the scattering phase functions of particles at different sizes are minimized (Chen et al., 2012). The detected signal by the receiver is converted to visibility by proprietary algorithms based on extensive calibration against transmissometer (Wauben, 2011). Visibility sensor outputs the visibility (vis) at 875 nm in unit of meter. Atmospheric extinction coefficient $\sigma_{\text{ext}}(\text{RH})$ at 550 nm (unit is Mm^{-1}) could be retrieved by Eq. (1) (Koschmieder, 1924; Seinfeld and Pandis, 2006).

$$\sigma_{\text{ext}}(\text{RH}) = \frac{3.912 \times 10^6}{\text{Vis}} \times \left(\frac{550}{\lambda} \right). \quad (1)$$

An integrating nephelometer continuously measured the light scattering coefficient by dry particles $\sigma_{sp}(\text{dry})$ at a wavelength of 532 nm. A processor-controlled heating system automatically maintained the RH of the pumped and the RH had a mean value of 24% with a standard deviation of 11% during the field study. The nephelometer was routinely calibrated by zero and span check. The nephelometer correction factor for truncation error had been done as in Liu et al. (2008).

The mass concentration of black carbon (BC) was recorded with a single-wavelength (670 nm) MAAP (Petzold and Schonlinner, 2004) and the product of mass concentration of BC by specific absorption coefficient ($6.6 \text{ m}^2 \text{ g}^{-1}$) which is from the manufacture guide specified the directly measured aerosol absorption coefficient σ_{ap} . Light absorption by gases σ_{ag}

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