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Aerosol optical properties and chemical composition apportionment in Sichuan Basin, China

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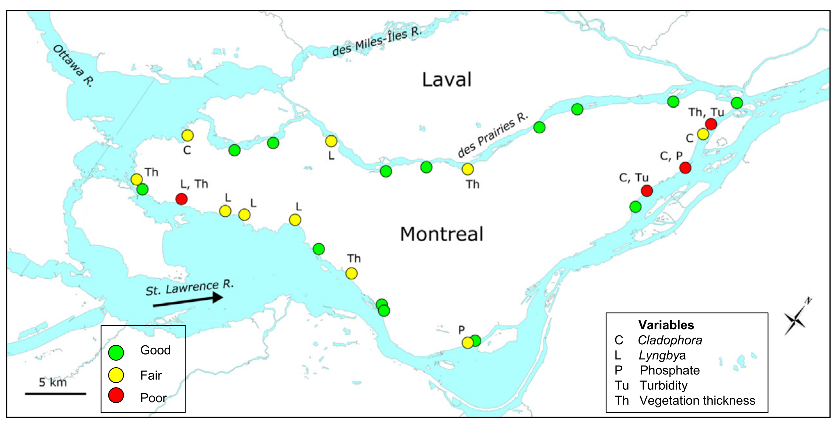
HIGHLIGHTS

- Local sources were largely responsible for the heavy aerosol pollution in Sichuan Basin.
- High relative humidity enhanced b_{ext} by up to 1.6 times.
- In winter, NH_4NO_3 and OM dominated b_{ext} in Chengdu and Chongqing, respectively.

GRAPHICAL ABSTRACT



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ABSTRACT

$PM_{2.5}$ and its major chemical components, and light scattering (σ_{scat}) and absorption (σ_{abs}) coefficients were measured in Chengdu (CD) and Chongqing (CQ) in Sichuan Basin, from October 2014 to July 2015. Annual mean $PM_{2.5}$, σ_{scat} and σ_{abs} were 67.0 ± 43.4 μg m^{-3}, 421.4 ± 290.1 Mm^{-1} and 36.7 ± 26.4 Mm^{-1}, respectively, in CD, and annual mean $PM_{2.5}$ and σ_{abs} were 70.9 ± 41.4 μg m^{-3} and 45.4 ± 24.5 Mm^{-1}, respectively, in CQ. $PM_{2.5}$, σ_{scat} and σ_{abs} were all evidently higher in winter than in other seasons mainly due to the unfavorable meteorological conditions for dispersion of local pollutants. Diurnal patterns of σ_{scat} and σ_{abs} exhibited a peak value around 7:00–8:00 LT and a valley value around 17:00–18:00 LT. High levels of $PM_{2.5}$ accompanied with low wind speed and high relative humidity conditions were the major causes of visibility impairment in Sichuan Basin. Both σ_{scat} and σ_{abs} were remarkably higher under calm wind condition, indicating that local emissions were largely responsible for the aerosol pollutions in this region. High relative humidity enhanced extinction coefficient (b_{ext}) by up to around 1.6 and 1.4 times in CD and CQ, respectively, due to the hygroscopic growth of water soluble components. On annual basis, $(NH_4)_2SO_4$ contributed the most to b_{ext}, accounting for 34.4% and 31.5% in CD and CQ, respectively, followed by NH_4NO_3 and organic matter, 28.1% and 17.5%, respectively, in

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CD, and 20.1% and 26.8%, respectively, in CQ. EC contributed about 10% and the rest contributed to <12% at both urban sites. Therefore, reducing emissions of the precursor gases such as SO_2, NO_x, NH_3 and VOCs systemically may be efficient to improve the air quality and visibility simultaneously in Sichuan Basin.

1. Introduction

With rapid economic growth and increasing anthropogenic emissions across China in the past several decades, air quality has been deteriorating and visibility impairment has occurred extensively (Chen and Xie, 2014). One of the key air pollutants is $PM_{2.5}$ (particles with an aerodynamic diameter smaller than 2.5 μm), which plays important roles in urban air quality, global climate change, and human exposure (Chang et al., 2009; de Kok et al., 2006; Mahowald, 2011; Tao et al., 2014a). Furthermore, $PM_{2.5}$ is the major cause of visibility impairment due to its strong light scattering and absorption abilities (Watson, 2002). Light extinction coefficient (b_{ext}) - the sum of the scattering (σ_{scat}) and absorption (σ_{abs}) coefficients has been used to characterize visibility. These optical parameters can be measured using commercial in-situ optical instruments, e.g., Integrating Nephelometer and Aethalometer or MultiAngle Absorption Photometer for measurement of σ_{scat} and σ_{abs}, respectively.

Aerosol optical properties are mainly decided by chemical composition and size distribution of aerosol particles, especially $PM_{2.5}$ (Bergin et al., 2001). Three typical methods have been used to estimate b_{ext} from aerosol chemical composition, including Mie method (Cheng et al., 2008), Interagency Monitoring of Protected Visual Environments (IMPROVE) algorithm (original and revised) (Pitchford et al., 2007), and multilinear regression method (Tao et al., 2014b). The Mie model has not been widely adopted in China due to its input requirement of chemical composition with continuous particle size distribution, which is difficult to obtain (Cheng et al., 2008). Both the original and revised IMPROVE algorithm use chemical species-dependent mass scattering or absorption efficiency (MSE/MAE). The fixed MSE/MAE were originally generated from data observed at rural/remote areas in the United States in the original IMPROVE algorithm. The applicability of this algorithm in China is questionable due to different chemical compositions and particle size distributions under different polluted conditions. To overcome this limitation, many studies attempted to obtain site-specific MSE and MAE values through multiple linear regressions (Tao et al., 2015; Tao et al., 2014c).

Unlike the original IMPROVE algorithm which used a constant mass extinction coefficient, a split component approach was developed in the revised IMRPOVE algorithm to account for the increase in mass extinction efficiencies (small and large particle size modes) (Pitchford et al., 2007). Other improvements in the revised IMRPOVE algorithm include calculating MSE/MAE for each chemical species using the Mie theory at a wavelength of 550 nm with assumed log-normal mass size distribution. The revised IMPROVE algorithm has been widely used in China to estimate the b_{ext} based on major chemical components (Cao et al., 2012; Cui et al., 2016; Wu et al., 2015; Yang et al., 2012).

Dominant contributors for visibility impairment include $(NH_4)_2SO_4$, NH_4NO_3, organic matter (OM), elemental carbon (EC), sea salt, fine soil (FS) and coarse particles (Pitchford et al., 2007). Their relative contributions to b_{ext} varied with location and season due to the spatiotemporal variations of their respective concentrations. Existing studies in China showed that $(NH_4)_2SO_4$ was the largest contributor to b_{ext} in Beijing, Chengdu, Jinan, Guangzhou, Xi'an and Nanjing, accounting for 40–50% of b_{ext} (Cao et al., 2012; Cui et al., 2016; Tao et al., 2012; Tao et al., 2014b; Yang et al., 2007; Zhang et al., 2015). OM is another major contributor to b_{ext} as was observed in Nanjing, Xiamen and Baoji in China, accounting for 34–40% of b_{ext} (Xiao et al., 2014; Yu et al., 2016; Zhang et al., 2012). Sea salt might be an important contributor at coastal cities, e.g., nearly 13% contribution to b_{ext} in Xiamen (Deng et al., 2016).

Although many studies have been conducted to investigate aerosol optical characteristics in the developed regions of China, such as Beijing (He et al., 2009; Jing et al., 2015; Tao et al., 2015; Tian et al., 2015), Pearl River Delta (Fu et al., 2016; Tao et al., 2012), and Yangtze River Delta (Cui et al., 2016; Han et al., 2015), studies in Sichuan Basin are very limited (Tao et al., 2014b) despite the region's special terrain and serious air pollution (Chen and Xie, 2012). Chengdu and Chongqing are two biggest cities in Sichuan Basin, located in its plat west and mountainous east, respectively. As one of the four great basins in China and surrounded by 1000–3000 m height mountains, Sichuan basin covers 260,000 km^2 with a population of around 100 million in southwest China. Visibility deterioration has been recorded in this region since 1970s, with the average visibility (6.2 km) during 2006 to 2010 being only half of that in 1960s (Chen and Xie, 2013). The wind speeds are extremely low, with annual mean of 1.3 and 1.5 m s^{-1} in Chengdu and Chongqing, respectively, and the relative humidity are high (around 80%), which play important role on the low visibility in this region. To gain a thorough knowledge of aerosol optical properties, quantitative contributions from major chemical components, and their relationship with haze formation, measurements were conducted in each selected month in four consecutive seasons at each urban site in Chengdu and Chongqing. Seasonal and diurnal variations of σ_{scat} and σ_{abs} were identified. Aerosol extinction coefficients were reconstructed based on the IMPROVE algorithm, and its applicability in Sichuan Basin was evaluated. Threshold values of $PM_{2.5}$ for preventing haze events were extracted. This information has implication for better understanding the causes of visibility deterioration and making future emission control policies to improve air quality and visibility simultaneously in this unique basin region.

2. Methods

2.1. Sampling sites

The geographical locations of the two sampling sites are shown in Fig. 1. The sampling site in Chengdu (CD) was on the roof of a sixth-floor building inside the Sichuan Academy of Environmental Science (104° 4′ E, 30° 37′ N), approximately 24 m above the ground. This sampling site is surrounded by commercial and residential districts, and there are no significant industrial sources nearby although it is close to a main road (Renmin South road of Chengdu) with high traffic density. Therefore, the sampling site could be regarded as a representative urban area in Chengdu.

The sampling site in urban Chongqing (CQ) was on the rooftop of Chongqing Monitoring Center (106° 30′ E, 29° 37′ N), 35 m above the ground. The sampling site is a typical residential area with high traffics (highway within 200 m) but without major industrial emissions.

2.2. Sample collection

The monitoring periods covered 23 October to 18 November 2014 (representative of autumn), 6 January to 2 February 2015 (winter), 2 to 29 April 2015 (spring), and 2 July to 30 July 2015 (summer). $PM_{2.5}$ samples were collected on a 47 mm Teflon filter and quartz filter (Whatman corp., USA) in parallel, with the former for mass and elemental analysis and the latter for water-soluble inorganic ions and carbonaceous components analysis. The Teflon filters were weighted before and after sampling at a constant temperature (20 °C) and relative humidity

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