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Source apportionment of $PM_{2.5}$ light extinction in an urban atmosphere in China

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ABSTRACT

Haze in China is primarily caused by high pollution of atmospheric fine particulates ($PM_{2.5}$). However, the detailed source structures of $PM_{2.5}$ light extinction have not been well established, especially for the roles of various organic aerosols, which makes haze management lack specified targets. This study obtained the mass concentrations of the chemical compositions and the light extinction coefficients of fine particles in the winter in Dongguan, Guangdong Province, using high time resolution aerosol observation instruments. We combined the positive matrix factor (PMF) analysis model of organic aerosols and the multiple linear regression method to establish a quantitative relationship model between the main chemical components, in particular the different sources of organic aerosols and the extinction coefficients of fine particles with a high goodness of fit (R^2 = 0.953). The results show that the contribution rates of ammonium sulphate, ammonium nitrate, biomass burning organic aerosol (BBOA), secondary organic aerosol (SOA) and black carbon (BC) were 48.1%, 20.7%, 15.0%, 10.6%, and 5.6%, respectively. It can be seen that the contribution of the secondary aerosols is much higher than that of the primary aerosols (79.4% versus 20.6%) and are a major factor in the visibility decline. BBOA is found to have a high visibility destroying potential, with a high mass extinction coefficient, and was the largest contributor during some high pollution periods. A more detailed analysis indicates that the contribution of the enhanced absorption caused by BC mixing state was approximately 37.7% of the total particle absorption and should not be neglected.

Introduction

Visibility is an important indicator of the urban air quality, which depends on the extinction of the atmosphere. The atmospheric extinction effect includes the extinction of gases and particles, in which the extinction of particles, *i.e.*, the scattering and absorption of sunlight by the atmospheric particles, is the primary factor (Watson, 2002). The optical properties of particles depend on the particle size, morphology, chemical composition, mixed state and the hygroscopic properties of the particles (Meier et al., 2009; Ma et al., 2012; Liu et al., 2014; Cao et al., 2012). Therefore, the study of the

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physical and chemical properties of particulate matter is key to the quantitative study of solar radiation and particle environmental effects.

Early studies of the extinction of particulate matter have primarily focused on the relationship between the mass concentration of particles and the extinction coefficient and have found a significant positive correlation between the two (Chan et al., 1999; Wang et al., 2006); however, due to the complex chemical composition of particles and large differences in particle size, changes in the extinction coefficient cannot be well characterised by only the concentration of the particles. In addition, some studies have used the Mie model to simulate the mixed state of particulate matter (Cheng et al., 2008) to explore particle extinction properties; however, the Mie model, as a theoretical model, cannot quantitatively represent the impact of particles on the visibility in a satisfactory manner. Using the multiple regression analysis method, the United States IMPROVE project (Interagency Monitoring of Protected Visual Environments) constructed the IMPROVE formula to calculate the extinction coefficient with multiple chemical species mass concentrations and extinction efficiencies (Sisler and Malm, 2000). This formula has been widely used in related studies involving the extinction of particulate matter (Wang et al., 2016; Zhou et al., 2016; Yu et al., 2016). Note that the IMPROVE formula is primarily designed for the chemical compositions and optical properties of $PM_{2.5}$ and cannot be used for PM_1, which is more closely related to atmospheric extinction. In addition, most studies have calculated the extinction contribution of organic aerosols as a whole (Wang et al., 2016; Zhou et al., 2016; Yu et al., 2016); however, the physical and chemical properties of organic aerosols are very complex and the optical properties of different types of organic aerosols from different sources are very different. Therefore, it is necessary to explore the effect of different types of organic aerosols on atmospheric extinction.

Traditional filter-based sampling methods have a shading effect and a multiple scattering effect (Weingartner et al., 2003; Bond and Bergstrom, 2006); in particular, the BC mass concentration and extinction coefficient is greatly influenced, which has a direct impact on the regression fit. In addition, off-line sampling has lower time resolution and its sample numbers and accuracy cannot satisfy our demands. In this study, we use high time resolution aerosol observation equipment to obtain *in situ* measurement data to establish the relationship between the chemical composition and extinction coefficient of atmospheric fine particles using the multiple linear regression method. In particular, the influence of different types of organic aerosols on the extinction of particulate matter is quantified and the influence of each component on the extinction coefficient is discussed to provide a scientific basis for haze management in China.

1. Material and method

1.1. Sampling site and period

The measurements were conducted in Dongguan City in the wintertime, from 11 December 2013, to 10 January 2014, during the polluted dry season in PRD with prevailing wind from the mainland (Huang et al., 2014). The Dongguan (DG) site is an urban site (23.0°N 113.7°E) in the middle of PRD with high urbanisation and industrialisation. The site was within a building beside a sports stadium located in the downtown area of Dongguan.

1.2. Instruments

The instruments were placed in a temperature-controlled room and the air was induced through a $PM_{2.5}$ cyclone inlet placed on the rooftop and then dried before entering the inlets of the instruments. A high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) (Aerodyne Research, MA, USA) was used to measure non-refractory species of $PM_{1,}$ including the organic carbon, sulphate, nitrate, ammonium ions and chloride ions. A detailed description of the instrument is given by DeCarlo et al. (2006), and the calibration followed standard protocols (Jayne et al., 2000; Jimenez et al., 2003; Drewnick et al., 2005). Additional details concerning the HR-ToF-AMS operation can be found in He et al. (2011) and Huang et al. (2013).

A single particle soot photometer (SP2) (Droplet Measurement Technologies, CO, USA) was used to measure the black carbon (BC) mass concentration and size distribution. The technical details are described in Schwarz et al. (2006, 2008). The calibration of the SP2 was conducted with fullerene soot (Alpha Aesar, Inc., Ward Hill, MA) selected by size using a differential mobility analyser upstream of the SP2. Additional details concerning the SP2 operation are provided in Huang et al. (2012). The detection limit of the BC particles in this study was approximately 0.07 μg in volume equivalent diameter.

A three-wavelength Photo-acoustic Soot Spectrometer (PASS-3) (Droplet Measurement Technologies, CO, USA) was simultaneously used to obtain the light absorption b_{ap} and scattering b_{sp} coefficients at 532 nm. The principles and technical details of the PASS-3 are described by Arnott et al. (1999), and additional details concerning the calibration and operation can be found in Yuan et al. (2016). The aerosols were dried prior to the inlet; therefore, it is the dry aerosol extinction coefficients measured by this instrument that are later referred to.

A Nitrogen Oxide Analyser (EC9841) was used to measure the nitrogen dioxide, and the time resolution was set to 1 min. The absorption of solar visible wavelengths by nitrogen dioxide is the most significant for a gas; therefore, the gas absorption coefficient b_{ag} was calculated from the measured concentration of nitrogen dioxide. Because the concentration of gas in the atmosphere is relatively stable, the scattering effects of other gas molecules are generally constant, and generally the value of the atmospheric gas scattering coefficient b_{sg} is taken to be 13 Mm^{-1} (Cohen, 1975).

The atmospheric total extinction coefficient, b_{ext}, was obtained by summing the gas absorption coefficient b_{ag}, the gas scattering coefficient b_{sg}, the particle absorption coefficient b_{ap} and the particle scattering coefficient b_{sp}.

2. Results and discussion

2.1. Atmospheric extinction

Fig. 1 shows a time series of the atmospheric extinction in the Dongguan area during the campaign. The average extinction

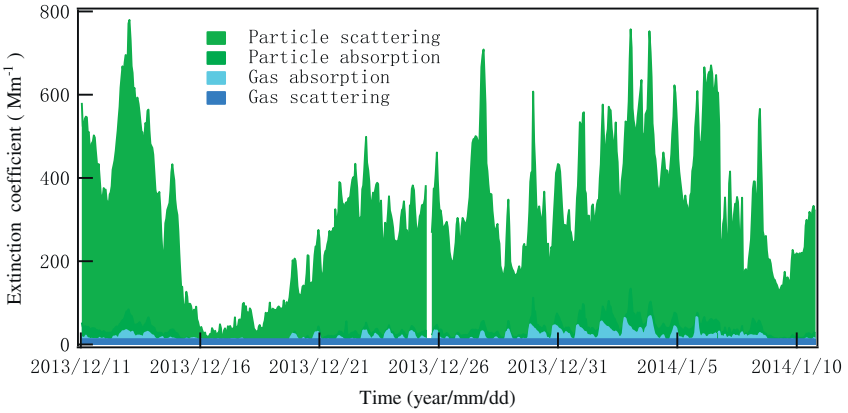

Fig. 1 – Time series of the light extinction coefficient.

coefficient of the atmosphere is 389 ± 192 Mm^{-1}. In the total extinction coefficient, the average particle scattering is 83.2%, the average particle absorption is 8.7%, the average gas scattering is 3.3% and the average gas absorption is 4.8%. The total extinction contribution of the particulate matter was 91.9%, and the study result is consistent with that of Hangzhou (Wang et al., 2016), Shanghai (Zhou et al., 2016) and Nanjing (Yu et al., 2016), indicating that the extinction of particulate matter is the primary cause of the decline in visibility. When including the effect of the relative humidity on the extinction of particulate matter, the actual extinction of the particulate matter in the total extinction coefficient will be more significant.

2.2. Submicron particle chemical composition

2.2.1. PM_1 chemical composition

Fig. 2 shows a time series and average mass percentage of each chemical component in PM_1. The mass concentration of PM_1 fluctuates in the range of 2.9–191 μg/m^3 during the observation period, with an average value of 56.8 ± 30.9 μg/m^3. It can be seen that the concentrations of the pollutants vary greatly and show obvious characteristics of urban pollution, and there were two obvious pollution processes, *i.e.*, Dec.11–14, 2013 and Dec.29, 2013–Jan. 5, 2014. The former process had higher concentrations of sulphate, while the later process had far higher concentrations of organic aerosol. It is interesting to note that the two pollution processes had similar light extinction coefficients in Fig. 1, despite of their different aerosol pollution levels, implying that the light extinction were strongly related with aerosol composition. During the entire observation period, the organic matter contributed the most to the average mass concentration, accounting for 41.0% of the total mass concentration of PM_1, followed by sulphate, accounting for 23.2% and nitrate, ammonium ion, BC and chloride ion accounting for 14.0%, 11.4%, 8.2% and 2.2% respectively.

2.2.2. Positive matrix factor (PMF) analysis of organic aerosol

During the observation period, the organic matter accounted for the largest proportion of PM_1; therefore, the effects of organic aerosols on the atmospheric extinction cannot be ignored. To determine the different effects of the different sources of organic aerosols (OA) on the atmospheric extinction in this study, a factorisation analysis of the organic aerosol from high-resolution mass spectrometry data obtained from AMS

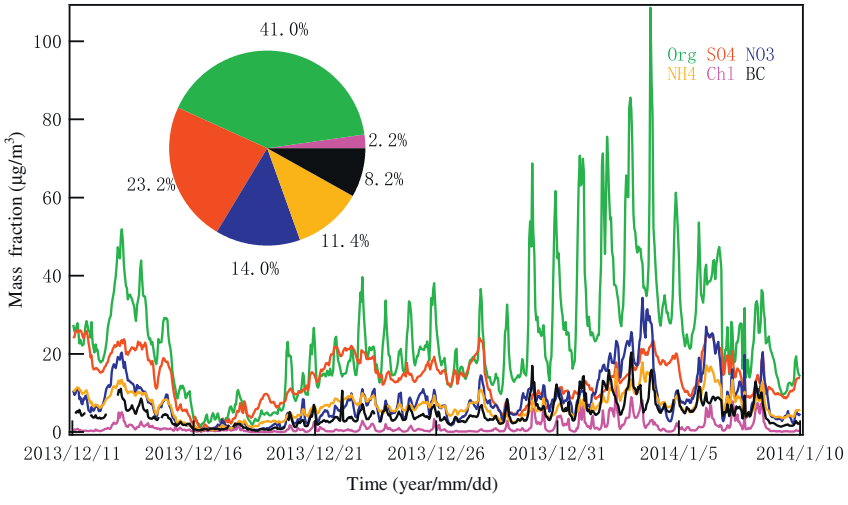

Fig. 2 – Time series and average mass fractions of the PM_1 chemical components.

during the observation period was performed using the PMF analysis model; for the specific methods see Ulbrich et al. (2009). In this study, five factors were determined: hydrocarbon-like OA (HOA), cooking OA (COA), more oxidised oxygenated OA (MO-OOA), less oxidised oxygenated OA (LO-OOA) and biomass burning OA (BBOA). The mass spectra of each OA component resolved by PMF are provided in the Supplementary Materials. HOA, COA and BBOA belong to primary OA (POA); MO-OOA and LO-OOA are secondary OA (SOA) (Mohr et al., 2009; Canagaratna et al., 2004).

The time series and average mass percentages during the observation period of the different types of OA are shown in Fig. 3. The average mass percentages of MO-OOA, COA, HOA, LO-OOA and BBOA were 40.6%, 20.0%, 13.6%, 13.4% and 12.4%, respectively. SOA accounts for 54.0% of the total organic aerosol and 22.3% of the mass concentration of PM_1 during the observation period, which indicates that SOA is a very important pollution component of atmospheric submicron particles in the winter in Dongguan and may have a relatively large impact on the reduction of visibility.

2.3. Model of the quantitative relationship between the extinction coefficient and the PM_1 chemical composition

Because the particle size (0.1–1 μm) of the submicron particles in the atmosphere is close to the wavelength of the visible solar band (0.3–0.8 μm), the extinction effect of submicron particles is more pronounced than that of larger particle sizes. In the previous analysis, the extinction coefficient of particulate matter accounted for 91.9% of the total extinction coefficient of the atmosphere. Based on these two points, a multiple regression was analysed between the absorption and scattering coefficients of the particulate matter at 532 nm as measured by PASS-3 and the chemical composition of PM_1 obtained from HR-ToF-AMS and SP2 during the same period using the SPSS (V18) software. This study considers the possibility that the extinction effect of different types of OA may be different. Therefore, in the following model construction, the source analysis results of the OA are used to replace the organic matter in the fitting.

In this study, it is assumed that all particulates are in an external mixed state and that the scattering effect of each chemical species is relatively independent (Lowenthal et al., 1995). In addition, because the contribution of minerals, dust and metals to the PM_1 mass concentration is very small and has a low extinction efficiency (Watson, 2002; Tao et al., 2014), these factors are not considered in the fitting process. Taking into account the simplicity of the fitting formula, the different degrees of ageing of MO-OOA and LO-OOA are merged into SOA and the fit formula does not include any constant items.

2.3.1. Fitting of the particle scattering coefficient

A large number of studies have shown that chemical substances such as ammonium sulphate, ammonium nitrate, ammonium chloride and organic matter in particulate matter are the main components that scatter sunlight (Watson, 2002). In addition, the scattering effect is enhanced by the hygroscopicity of the sulphates, nitrates and ammonium salts (Liu et al., 2014). However, because the scattering coefficient of the particulate matter in this study is directly measured by PASS-3 through the drying tube, the relative humidity is not higher than 35% and the moisture absorption of the particulate matter is not obvious; therefore, the impact of the humidity on the scattering of the particles is not taken into account.

To facilitate the fitting, it is assumed that the sulphate, nitrate and chloride salts in the particulate matter are in the form of an ammonium salt; the mass concentration of ammonium sulphate, ammonium nitrate and ammonium chloride can be determined by:

$$[(NH_4)_2SO_4] = 1.375\,[SO_4^{2-}],$$
$$[NH_4NO_3] = 1.29\,[NO_3^-]$$
$$[NH_4Cl] = 1.507\,[Cl^-].$$

We include $(NH_4)_2SO_4$, NH_4NO_3, NH_4Cl, SOA, COA, HOA and BBOA in the fitting of the particle scattering coefficient so that the standard equation is:

$$b_{sp} = a[(NH_4)_2SO_4] + b[NH_4NO_3] + c[NH_4Cl] + d[SOA] + e[HOA] + f[COA] + g[BBOA].$$

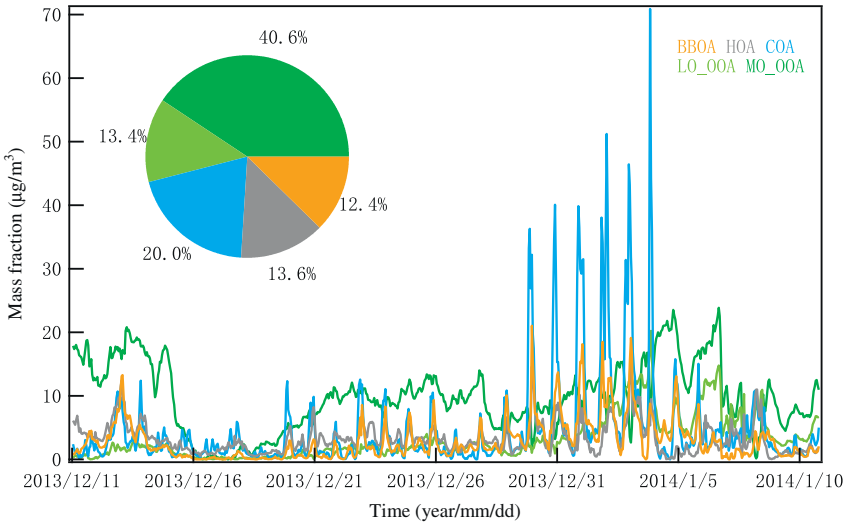

Fig. 3 – Time series and average mass fractions of different organic aerosols.

Then, using a gradual multiple regression to remove the factors that are negative and have low efficiency, we obtain the final relationship as follows:

$$b_{sp} = 9.221 \times [(NH_4)_2SO_4] + 6.780 \times [NH_4NO_3] + 2.809 \times [SOA] + 16.725 \times [BBOA]. \quad (1)$$

The regression equation and the regression coefficient are determined by the significance test (Sig.F = 0.000, Sig.T = 0.000). The coefficient of determination R^2 = 0.994 and the normalised residual have normal distributions. It can be seen that SOA and BBOA have significant impacts on the light scattering as two different types of organic matter; however, the contributions of HOA and COA can be ignored.

2.3.2. Fitting of the particle absorption coefficient

In general, BC is assumed to be the only atmospheric particle that absorbs the full spectrum of sunlight; however, studies have shown that organic compounds in aerosols including soil humus, humus-like substances and bio-aerosols have an absorption effect on sunlight (Andreae and Gelencser, 2006). When fitting the relationship between the winter atmospheric extinction coefficient in Shenzhen and the chemical composition of fine particles, Yao et al. (2010) found that even though the organic light absorption efficiency is much lower than that of BC, the mass concentration of organic matter is several times higher than that of BC. These results show that the average contribution of BC to the absorption coefficient of particulate matter is 73% and the contribution of organic matter is 21%. In addition, many studies have indicated that the BC mixing state is one of the important influencing factors enhancing the light absorption due to the 'lens effect' when a BC core is coated by other chemical components, especially sulphate and nitrate (Jacobson, 2001; Chung and Seinfeld, 2005; Lan et al., 2013). Therefore, in the fitting of the absorption coefficient, we take BC, organic matter, sulphate and nitrate into account.

Considering the enhanced effect of inorganic salts and organic compounds on the light absorption (Jacobson, 2001), as discussed later, we take $(NH_4)_2SO_4$, NH_4NO_3, BC, SOA, HOA, COA and BBOA together into the fitting of the particle absorption coefficient; however, ammonium chloride is not considered due to its lack of quality. Similar to the scattering fitting process, the final relationship is obtained as follows:

$$b_{ap} = 4.194 \times [BC] + 0.273 \times [(NH_4)_2SO_4] + 0.260 \times [NH_4NO_3] + 0.110 \times [SOA] + 1.480 \times [BBOA]. \quad (2)$$

The regression equation and the regression coefficient are determined by the significance test (Sig.F = 0.000, Sig.T = 0.000). The coefficient R^2 = 0.998 and the normalised residual have normal distributions. It can be seen that SOA and BBOA are still the two primary components of the organic matter that have a significant influence on the light absorption.

In Eq. (2), the BC coefficient of 4.2 m^2/g can be taken to be the mass absorption efficiency of pure BC, which is within the range of 4.0–15.9 m^2/g suggested by Bond and Bergstrom (2005). Our result of 4.2 m^2/g is relatively low in this range; this may be because China's BC particle size is relatively large (Huang et al., 2006), which would lead to low mass absorption efficiency; in addition, the differences between the instruments used in the study may lead to large differences in the results. In this paper, we use all *in situ* measurements, including PASS-3 and SP2, which may be more reliable compared to the filter-based measurements used in previous studies.

In addition, the absorption coefficients of $(NH_4)_2SO_4$, NH_4NO_3 and organic matter can be taken as the enhanced absorption contribution *via* the chemical components coating BC. Then, we can calculate the average contributions of ammonium sulphate, ammonium nitrate and organic matter to be 15.1%, 8.2% and 17.3%, respectively. Meanwhile the absorption contribution of OA may also be due to their own absorption from brown carbon. To further clarify the specific contribution of the organic matter, we can calculate the absorption of brown carbon (BrC) using the Absorption Angstrom Exponent (AAE). The PASS-3 simultaneously measured light absorption at 405 nm, 532 nm and 781 nm; we applied an $AAE_{532-781\ nm}$ of 7.0, which is a more reasonable pure BC AAE value measured in winter in an urban city near Dongguan by Yuan et al. (2015). The details of the calculation of AAE and the light absorption of BrC are described in Yuan et al. (2015). Then, we calculated that the average BrC

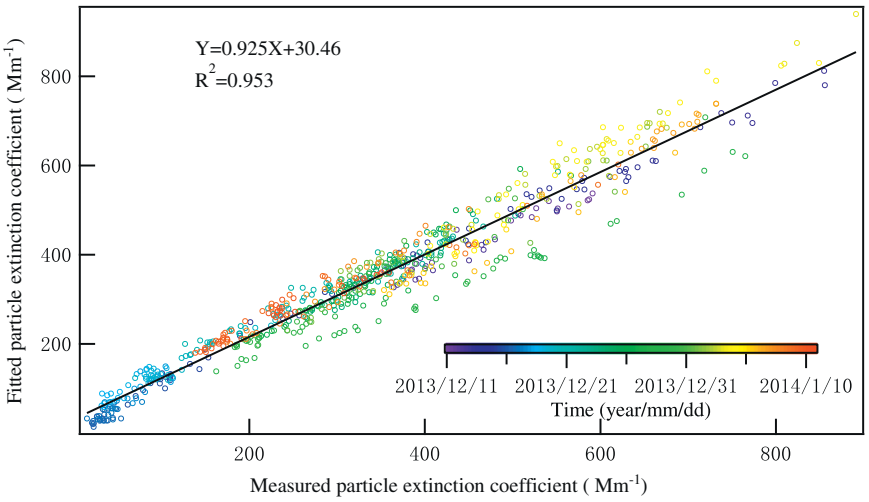

Fig. 4 – Linear relationship between the fitted and the measured values of the particle extinction coefficient.

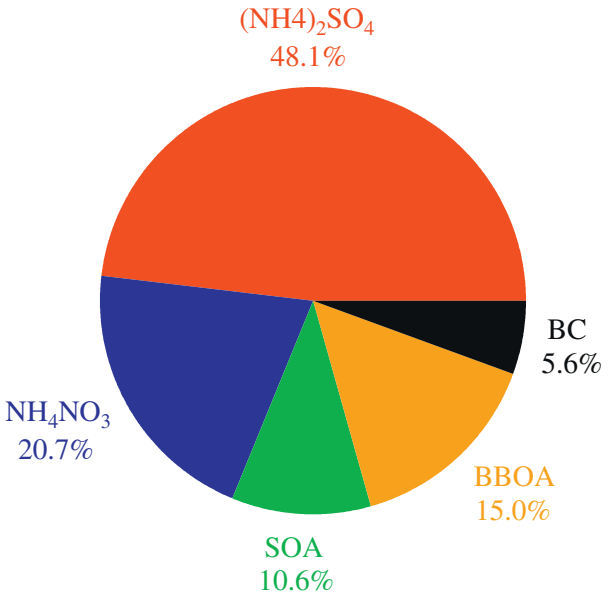

Fig. 5 – Average extinction contribution of the main chemical components of PM_1.

light absorption contribution at 532 nm was 2.9%. The coating effect contribution of the organic matter can be calculated by subtracting the BrC absorption from the total organic matter absorption. Therefore, the average contribution of the organic coating effect was 14.4%. Adding the absorption contribution of ammonium sulphate and ammonium nitrate, the total absorption contribution enhanced by the BC mixing state is 37.7%.

2.3.3. Establishment of the extinction relationship of particulate matter

The total extinction coefficient of particles in the winter in Dongguan can be obtained from the absorption fitting formula, Eq. (1), added to the scattering fitting formula, Eq. (2):

$$b_{ext,p} = 4.2 \times [BC] + 9.5 \times [(NH_4)_2SO_4] + 7.0 \times [NH_4NO_3] + 2.9 \times [SOA] + 18.2 \times [BBOA]. \quad (3)$$

Fig. 4 shows a comparison between the fitting value of the total extinction coefficient of the particles and the measured values. The determination coefficient $R^2 = 0.953$ shows that the model can predict the extinction of the particles with the PM_1 chemical composition data. In the fitting formula, Eq. (3), it can be seen that BBOA has the highest extinction efficiency of 18.2 m^2/g, indicating that the effect of biomass combustion emissions on atmospheric visibility is very important; the extinction efficiencies of ammonium nitrate and ammonium sulphate are the second highest. Hand and Malm (2007) summarised 48 reports on the relationship between $PM_{2.5}$ components and extinction coefficients using multivariate linear regression methods, and the average extinction efficiencies of ammonium sulphate and ammonium nitrate in dry aerosols were 2.8 ± 0.5 m^2/g and 2.8 ± 0.5 m^2/g, respectively. Considering that the ratio of sulphates, nitrates and ammonium salts in PM_1 account for approximately 60%–70% of those in $PM_{2.5}$ (Zhang et al., 2013) and that PM_1 has a particle size closer to the wavelength of visible light (a stronger scattering capacity), the coefficients of ammonium sulphate and ammonium nitrate obtained in this paper are reasonable.

2.4. Extinction contribution of the major PM_1 chemical composition

According to the fitting formula, Eq. (3), we can obtain the average extinction contribution of the PM_1 chemical components and time series, as shown in Figs. 5 and 6, respectively. During the entire observation period, ammonium sulphate contributed most to the extinction with an average rate of 48.1%. The contribution rate of ammonium nitrate, SOA, BBOA and BC was 20.7%, 10.6%, 15.0% and 5.6%, respectively. It can be seen that in different sources of organic matter, HOA and cooking source are far less important than secondary organic matter and biomass combustion for visibility; these two sources are also the most important sources of carbonaceous aerosols worldwide (Bond et al., 2004). However, secondary species, including ammonium sulphate, ammonium nitrate and SOA, dominate the particulate matter extinction with a total contribution of 79.4%, while the contribution of the primary source accounts for a total contribution of 20.6%.

In this study, the extinction contribution of ammonium sulphate was the highest; similar studies have found that organic matter has the highest contribution to the extinction in Shenzhen (Yao et al., 2010); in Xi'an, the highest contribution is from ammonium nitrate (Wang et al., 2014); and in Beijing, the highest contribution is organic or ammonium

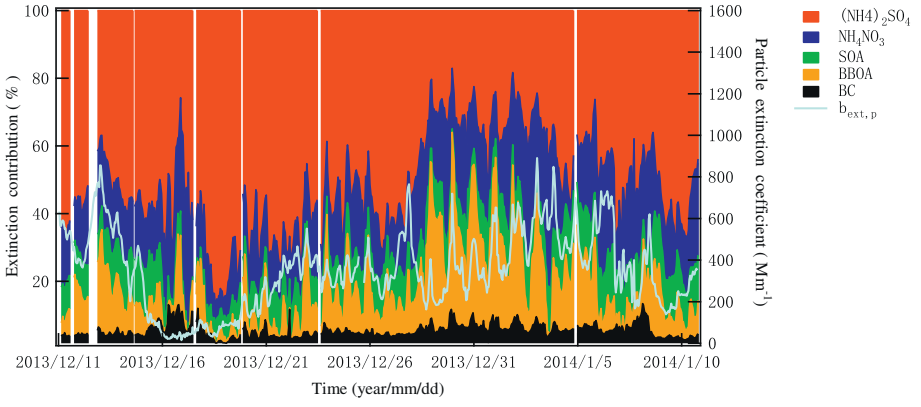

Fig. 6 – A time series of the extinction contribution of the main chemical components.

nitrate (Zhang et al., 2016; Han et al., 2015; Xu et al., 2016; Wang et al., 2015). Therefore, it is clear that different regions need to use different pollution control strategies to more effectively improve atmospheric visibility.

From Fig. 6 we can see that, in the case of a particular pollution episode (29 December 2013, to 3 January 2014), the BBOA extinction contribution is more prominent than it is on normal days and the contribution rate can reach as high as 54.0%. This indicates that BBOA may be a major factor leading to a visibility decline in special high pollution periods.

3. Conclusions

Based on high time resolution aerosol observation, we combined the PMF analysis model and the multiple linear regression method to establish a relationship model between the detailed aerosol composition and light extinction in winter in Dongguan, China. The major findings are as below.

(1) During the observation period, the atmospheric extinction coefficient in Dongguan was 389 ± 192 Mm^{-1} and the extinction coefficient of the particulate matter was 91.9% of the total extinction coefficient, which indicates that the extinction of particulate matter is the main reason for the decrease in visibility in the Dongguan area.
(2) Organic matter contributes most to the mass concentration of PM_1, accounting for 41.0%, followed by sulphate, nitrate, ammonium ion, BC and chloride accounting for 23.2%, 14.0%, 11.4%, 8.2% and 2.2%, respectively. Using the PMF model to analyse the types of OA in PM_1, the contributions of HOA, BBOA, MO-OOA, LO-OOA and COA were 13.6%, 12.4%, 40.6%, 13.4% and 20.0%, respectively.
(3) A quantitative relationship between the extinction coefficients and the chemical composition of the fine particles was established using a multiple linear regression method, as below:

$$b_{ext,p} = 4.2 \times [BC] + 9.5 \times [(NH_4)_2SO_4] + 7.0 \times [NH_4NO_3] + 2.9 \times [SOA] + 18.2 \times [BBOA].$$

According to the model calculation results, ammonium sulphate makes the greatest contribution to the extinction coefficient of particulate matter with a contribution rate of 48.1%; this is followed by ammonium nitrate with a contribution rate of 20.7%. The extinction rates of SOA, BBOA and BC were 10.6%, 15.0% and 5.6%, respectively.
(4) BBOA has the greatest contribution in the high pollution period and is the primary factor leading to low visibility. Note that the absorption enhanced by the BC mixing state contributes to 37.7% of the total particle absorption.

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jes.2017.07.016.

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