



## Aerosol scattering coefficients and major chemical compositions of fine particles observed at a rural site in the central Pearl River Delta, South China

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### Abstract

During November–December 2010 aerosol scattering coefficients were monitored using a single-waved (525 nm) Nephelometer at a regional monitoring station in the central Pearl River Delta region and 24-hr fine particle (PM<sub>2.5</sub>) samples were also collected during the period using quartz filters for the analysis of major chemical components including organic carbon (OC), elemental carbon (EC), sulfate, nitrate and ammonium. In average, these five components accounted for about 85% of PM<sub>2.5</sub> mass and contributed 42% (OC), 19% (SO<sub>4</sub><sup>2-</sup>), 12% (NO<sub>3</sub><sup>-</sup>), 8.4% (NH<sub>4</sub><sup>+</sup>) and 3.7% (EC), to PM<sub>2.5</sub> mass. A relatively higher mass scattering efficiency of 5.3 m<sup>2</sup>/g was obtained for fine particles based on the linear regression between scattering coefficients and PM<sub>2.5</sub> mass concentrations. Chemical extinction budget based on IMPROVE approach revealed that ammonium sulfate, particulate organic matter, ammonium nitrate and EC in average contributed about 32%, 28%, 20% and 6% to the light extinction coefficients, respectively.

**Key words:** scattering coefficients; visibility; fine particle (PM<sub>2.5</sub>); chemical compositions

**DOI:** 10.1016/S1001-0742(11)60730-4

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### Introduction

Aerosols in the atmosphere can scatter and absorb solar radiation and therefore play important roles in climate forcing and visibility degradation. Uncertainties in aerosols' optical properties contribute substantially to uncertainties in climate forcing (IPCC, 2007) and in regulatory efforts to protect visibility. Visibility degradation is probably the most readily perceived impact of air pollution (Seinfeld and Pandis, 2006). Although visibility is reduced by the absorption and scattering of light by both gases and particles, light scattering by particles of sizes comparable to the wavelength of visible light (the Mie scattering range) is mostly responsible for the visibility reduction in the atmosphere, and scattering by air molecules, as well as absorption of certain wavelength of light by gas molecules and particles, usually has a minor influence on urban visibility. As the main constituent of the pervasive haze that impairs visibility, fine particles are mostly anthropogenic pollutants, either emitted as particles or formed in the atmosphere by gas-to-particle conversion. The geographic areas affected by haze have increased on the global or regional scale, and haze has become a common landscape

feature in many densely populated parts with population growth and the spread of industrialization (Che et al., 2007; Chang et al., 2009; Wang et al., 2009).

The Pearl River Delta (PRD) in South China is one of the most urbanized and industrialized regions in China and its rapid economical growth in recent decades has resulted in deteriorated air quality (Chan and Yao, 2008), and visibility degradation is becoming an air pollution problem of wide concern in the region (Wu et al., 2005). A 52-year record between 1954–2006 revealed that annual haze days in Guangzhou, a central city in PRD, increased rapidly since 1972 and remained fairly constant to be 150 days since 1980 (Deng et al., 2008). The visibility impairment due largely to fine particles impacts greatly on public health, as can be seen from good statistical evidence for a relationship between the degradation of air quality based on aerosol extinction coefficients and the mortality associated with lung cancers during 1952–2006 in Guangzhou (Tie et al., 2009), or from elevated DNA damage potential of extractable organic matters during haze days in Guangzhou (Xu et al., 2008). Visibility is very sensitive to aerosol concentrations in the PRD (Deng et al., 2008), and the frequent haze episodes in the region are in fact a result of fine/ultra-fine particle pollution (Tan et al., 2009). During winter 2002, PM<sub>2.5</sub>

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and  $PM_{10}$  levels in PRD averaged 72.6 and 111.5  $\mu\text{g}/\text{m}^3$  with  $PM_{2.5}$  constituting 70.4% of the  $PM_{10}$  mass (Cao et al., 2003).  $PM_{2.5}$  concentration in urban Guangzhou was up to 151  $\mu\text{g}/\text{m}^3$  in the fall of 2004 (Andreae et al., 2008) and 162  $\mu\text{g}/\text{m}^3$  in the fall of 2007 (Xu et al., 2008). For effective control measures to improve visibility in the PRD region, it is important to know the contribution of major aerosol chemical species to the overall light extinction. Using the multi-linear regression equation developed by the Interagency Monitoring of Protected Visual Environments (IMPROVE) project (Malm et al., 2000), Cheung et al. (2005) concluded that ammonium sulfate and organic matter (OM) contributed 52% and 38% to the light extinction, respectively, in Hong Kong. Tao et al. (2009) found that ammonium sulfate was the dominant species affecting light scattering in urban Guangzhou. No doubt that more data from ground-based measurements are needed to explore the role of aerosols in visibility impairment considering their chemical compositions and size distributions.

In the present study, visibility and aerosol scattering coefficients were measured continuously at a regional monitoring station in the central PRD region during November–December 2010, and filter-based measurements of major chemical components of fine particles were also conducted. The purpose was to estimate mass scattering efficiency of fine particles and the contribution of major components to the light extinction or visibility impairment.

## 1 Materials and methods

### 1.1 Field sampling

Field sampling and measurements were conducted on the rooftop, about 30 m above ground, of a seven-floor building in a high school at a rural town Wangqingsha (WQS, 22°42'N, 113°32'E) from November 2 to December 25, 2010. As shown in Fig. 1, WQS is located in the central Pearl River Delta region, with the Pearl River estuary in the south and surrounding city clusters approximately 50 km away. Also the surrounding terrain is flat with large farmlands nearby and rare traffic, so this site can serve as



**Fig. 1** Sampling site Wangqingsha (WQS) surrounded by city clusters in the Pearl River Delta (PRD).

an ideal location to monitor regional air pollution. The 24-hr  $PM_{2.5}$  samples were collected onto  $8 \times 10$  inch quartz filters using a high volume sampler (Tisch Environmental Inc., Ohio, USA) at a rate of 1.1  $\text{m}^3/\text{min}$ . Aerosol scattering coefficients ( $b_{\text{sp}}$ ) were measured with a single-waved (525 nm) integrating Nephelometer (Aurora-1000, Ecotech, Australia). To eliminate the effects of humidity, the internal sample heater was enabled to keep relative humidity (RH) < 40% and temperature ( $25 \pm 2$ )°C. Measured  $b_{\text{sp}}$ , as well as sample air temperature, chassis temperature, relative humidity and sample pressure were recorded every 60 sec. The lower detection limit for  $b_{\text{sp}}$  is less than 0.3  $\text{Mm}^{-1}$  (60 sec averaged data). The meteorological parameters were measured by a mini weather station (Vantage Pro2TM, Davis Instruments Corp., USA) with wind speed/direction, RH, temperature and global solar radiation recorded every 1 min. Visibility was detected by a Belfort Model 6000 visibility sensor (Belfort Instrument, USA).

### 1.2 Laboratory analysis

$PM_{2.5}$  mass concentrations were obtained by weighing the filters in the lab before and after field sampling under same conditions. To determine organic carbon (OC) and elemental carbon (EC) contents, a punch (1.5 cm  $\times$  1.0 cm) of each filter was taken for OC/EC analysis using the thermo-optical transmittance (TOT) method (NIOSH, 1999) by an OC/EC analyzer (Sunset Laboratory Inc., USA). An additional punch was taken from each filter and extracted twice in 10 mL of 18-MO $\Omega$  milliQ water and sonicated for 15 min in an ice-water bath. After filtered through a 0.22  $\mu\text{m}$ -pore size filter, the total extract (20 mL) was analyzed for ions ( $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ) and anions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{NO}_2^-$  and  $\text{PO}_4^{3-}$ ) with an Ion Chromatography (Metrohm 883 Basic IC plus, Switzerland).

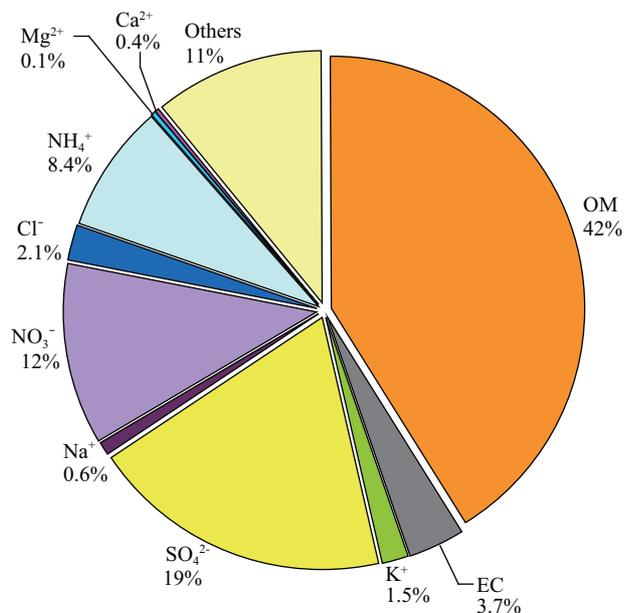
### 1.3 Quality control and quality assurance (QC/QA)

Prior to field sampling, quartz filters were baked at 450°C for 4 hr to remove organic impurities, wrapped in aluminum foils, and zipped in Teflon bags. After sampling, the filters were again wrapped in aluminum foils, zipped in Teflon bags, and stored in a freezer at  $-20^\circ\text{C}$  until analysis. Each week there was a clean filter exposed to the atmosphere during the sampling period and processed as a field blank. All the OC/EC and ions/anions data were corrected using the field blanks.

## 2 Results and discussion

### 2.1 $PM_{2.5}$ mass concentrations and chemical compositions

During the sampling period observed  $PM_{2.5}$  mass concentrations ranged 13.4–155.8  $\mu\text{g}/\text{m}^3$  with an average of ( $70.8 \pm 27.8$ )  $\mu\text{g}/\text{m}^3$ , about 37% lower than that of ( $113 \pm 23.6$ )  $\mu\text{g}/\text{m}^3$  observed at the same site in the same period in 2007 (Ding et al., 2011). Figure 2 shows the relative contributions of the carbonaceous and inorganic components to  $PM_{2.5}$ . The dominant inorganic species  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$



**Fig. 2** Average chemical composition of fine particles at WQS during the sampling period.

and  $\text{NH}_4^+$  altogether contributed nearly 40% of the  $\text{PM}_{2.5}$  mass, while carbonaceous aerosols contributed about 46% of the  $\text{PM}_{2.5}$  mass. Organic matter is the dominant fraction which alone contributed 42% of the  $\text{PM}_{2.5}$ , quite similar to the 43.7% of POM in  $\text{PM}_{2.5}$  observed at urban Guangzhou in 2004 (Andreae et al., 2008). Sulfate as a regional pollutant showed a less share (19%) if compared to a share of 21% at the same site in 2007 (Ding et al., 2011) or a share of 27% observed at urban Guangzhou in 2004 (Andreae et al., 2008).

Compared to results observed at the same site in 2007 (Ding et al., 2011), average sulfate decreased from  $24.2 \mu\text{g}/\text{m}^3$  in 2007 to  $13.6 \mu\text{g}/\text{m}^3$  in 2010, while average nitrate were only about  $1 \mu\text{g}/\text{m}^3$  lower from  $9.5 \mu\text{g}/\text{m}^3$  in 2007 to  $8.4 \mu\text{g}/\text{m}^3$  in 2010. For carbonaceous aerosols, the ratios of OC/EC in 2007 and 2010 were the same (5.7), indicating a larger fraction of secondary organic aerosols in OC if a ratio of 2.0 was taken for the primary sources. OC was about  $5 \mu\text{g}/\text{m}^3$  lower from  $19.3 \mu\text{g}/\text{m}^3$  in 2007 to  $14.5 \mu\text{g}/\text{m}^3$  in 2010, resulting in about  $10 \mu\text{g}/\text{m}^3$  decrease of particulate organic matter (POM) if a factor of 2.0 was used to calculate POM from measured OC (Bae et al., 2006; Chen and Yu, 2007); EC was only about  $1 \mu\text{g}/\text{m}^3$  lower from  $3.6 \mu\text{g}/\text{m}^3$  in 2007 to  $2.6 \mu\text{g}/\text{m}^3$ . These EC values in  $\text{PM}_{2.5}$  were lower than those measured earlier in both fall and summer seasons in the PRD region (Cao et al., 2003; 2004), but was significantly higher than those reported in the other places of the world (Castro et al., 1999; Malm et al., 2000; Zheng et al., 2006; Sciare et al., 2008). The mass ratio of nitrate to sulfate in average was 0.62 (molar ratio 0.92) in 2010, while that was 0.39 (molar ratio 0.62) in 2007. This drop of the sulfate levels and its contribution to  $\text{PM}_{2.5}$ , as well as the increase nitrate/sulfate ratios at the WQS regional monitoring site, might be indicative of the effective  $\text{SO}_2$  emission control by local Guangdong and Hong Kong governments in recent years.

## 2.2 Mass scattering efficiencies

Figure 3 shows the time series of measured aerosol visibility, scattering coefficients and meteorological parameters. One-hour visibility during the sampling period averaged 12.7 km with the lowest less than 1 km and the highest near 80 km. As shown in Fig. 3, visibility impairment episodes and distinctively elevated aerosol scattering coefficients typically occurred when wind speeds were relatively low and therefore fine particles accumulated to high levels. The extremely high visibility and low scattering coefficients occurred when a cold air mass intruded on December 17. To derive mass scattering efficiencies ( $\alpha_{\text{sp}}$ ), the simplest method is to divide the average scattering coefficient by the average mass concentration for a given sampling period, or the slope from a linear regression of scattering coefficients and mass concentrations can be interpreted as the specific mass scattering efficiency (Hand and Malm, 2007). As shown in Fig. 4, scattering coefficients were significantly correlated with  $\text{PM}_{2.5}$  mass concentrations ( $p < 0.001$ ) and the slope was  $5.3 \text{ m}^2/\text{g}$ , which could be interpreted as mass scattering efficiency of fine particles at WQS during the November–December in 2010. This fine mode specific mass scattering efficiency was much higher compared to average values from previous studies (Hand and Malm, 2007; and references therein). Much higher values ( $> 5 \text{ m}^2/\text{g}$ ) similar to those from this study were also reported in Mexico City, Mexico (Chow et al., 2002) and Fort Meade, USA (Chen et al., 2003). The higher values during polluted periods or in urban areas were probably attributed to more efficient light scattering of fine mode particles due to their size distribution and/or composition (Tan et al., 2009). The fine mode aerosol mass scattering efficiency in the present study was also higher than the value ( $3.0 \pm 0.6$ )  $\text{m}^2/\text{g}$  reported in June 1999 in Beijing, China (Bergin et al., 2001).

## 2.3 Measured and calculated visibility light extinction coefficients

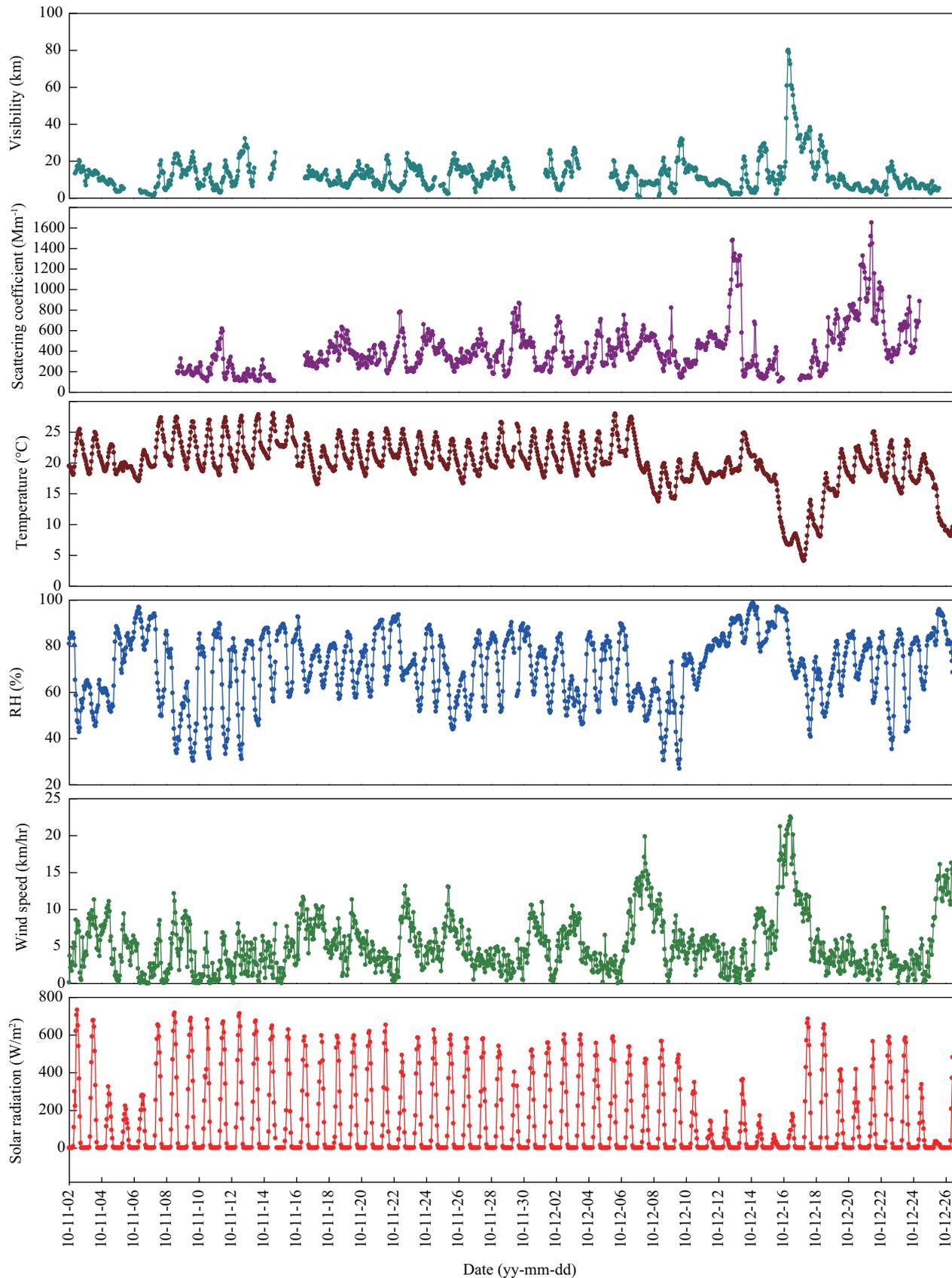
Light extinction coefficient  $b_{\text{ext}}$  can be deduced from measured visibility (Vis) from Koschmeider equation (Seinfeld and Pandis, 2006):

$$b_{\text{ext}} = 3.912/\text{Vis}$$

Daily mean  $b_{\text{ext}}$  at WQS observed during the sampling period ranged  $97\text{--}1469 \text{ Mm}^{-1}$  with an average of  $408 \text{ Mm}^{-1}$ . According to a chemical extinction budget from IMPROVE study (Malm et al., 2000),  $b_{\text{ext}}$  can be calculated as:

$$b_{\text{ext}} \approx 3 \times f(\text{RH}) \times [\text{AS}] + 3 \times f(\text{RH}) \times [\text{AN}] + 4 \times [\text{POM}] + 10 \times [\text{LAC}] + 1.0 \times [\text{Soil}] + 0.6 \times [\text{CM}] + 10$$

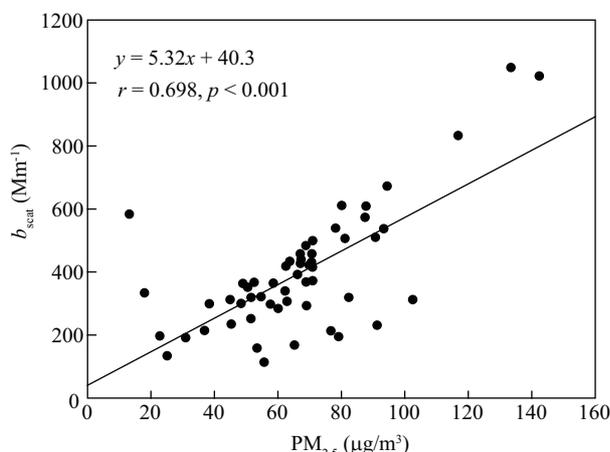
assuming an externally mixed aerosol model consisting of fine ammonium sulfate [AS], ammonium nitrate [AN], particulate organic material [POM], fine soil [Soil], coarse mass [CM] and light absorbing carbon [LAC]. The scattering enhancement factor  $f(\text{RH})$  takes into account RH effects for each hygroscopic species (Malm and Day, 2001). Since that coarse matter, soils and sea salts had minor contribution to aerosol mass (Andreae et al., 2008;



**Fig. 3** Time series of visibility, scattering coefficients, temperature, relative humidity (RH), wind speed and solar radiation at WQS during the sampling period.

Tan et al., 2009) and that these chemical species have much lower mass scattering efficiencies, in the present study we only considered the contribution of AS, AN, POM and

LAC. Calculated contributions of these species to  $b_{\text{ext}}$  derived from measured visibility range are presented in Fig. 5. In average, AS, AN, POM and LAC altogether

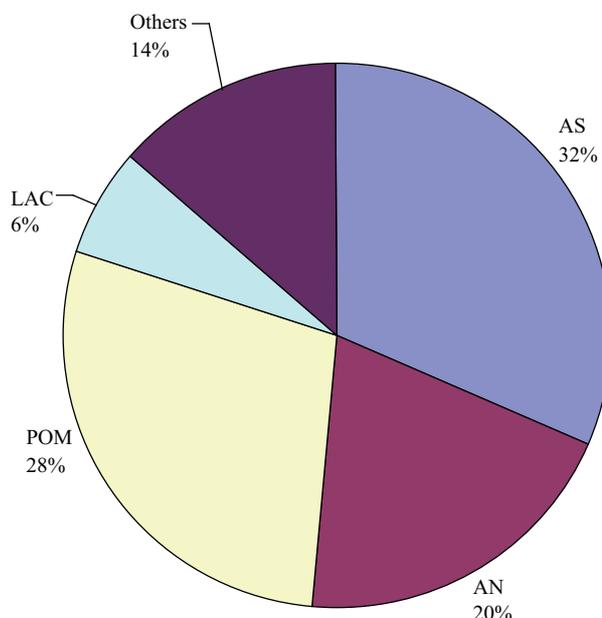


**Fig. 4** Linear regression of scattering coefficients and  $PM_{2.5}$  mass concentrations.

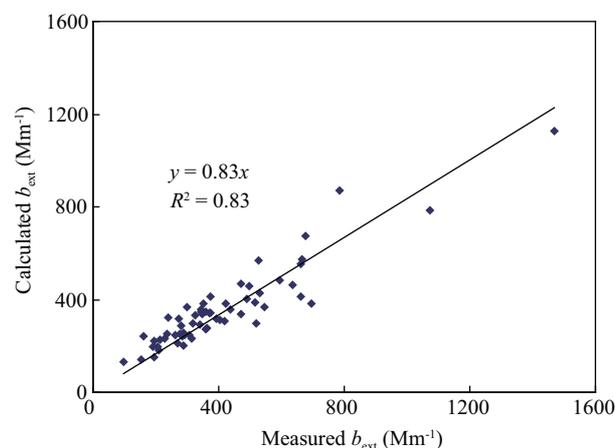
accounted for  $352 \text{ Mm}^{-1}$  or 86% of the overall  $b_{\text{ext}}$  (408) observed during the sampling period. As shown in Fig. 6, calculated  $b_{\text{ext}}$  only considering AS, AN, POM and LAC had a very good linear correlation with those from measured visibility, and they altogether could explain about 83% of  $b_{\text{ext}}$  from the slope of linear correlation in Fig. 6. Light scattering fine mode particles (AS, POM, AN) contributed over 80% to the light extinction efficiencies while light absorbing particles like LAC and other gases had minor contributions (Fig. 5). As the biggest contributors, sulfate, organic matter and nitrate, are by large secondarily formed in the atmosphere, effective control of their precursors like  $SO_2$ ,  $NO_x$  and VOCs is prerequisite to reducing haze episodes and improving visibility in the PRD region. It worth noting that the IMPROVE approach has an assumption that particles are externally mixed, this was in fact not the case in PRD region (Bi et al., 2011). Moreover, the specific mass scattering efficiencies for aerosol chemical species might be different from those from IMPROVE study. Therefore, the present study is only kind of rough estimation although calculated and measured  $b_{\text{ext}}$  seemed to match very well, and further study are needed to infer the local specific mass scattering efficiencies to assess the role of aerosols in radiative forcing and visibility degradation.

### 3 Conclusions

In the PRD region the haze episodes occur more frequently during November to January. In the present study we measured aerosol chemical and optical properties at a regional monitoring station in the central PRD from November 2 to December 25, 2010. Filter-based chemical analysis revealed that sulfate, nitrate, ammonium and carbonaceous aerosols contributed about 85% of  $PM_{2.5}$  mass and organic matter was the dominant component with an average share of 42%. A relatively higher mass scattering efficiency of  $5.3 \text{ m}^2/\text{g}$  indicated that visibility degradation was very sensitive to fine particle mass concentrations in the region. Based on the IMPROVE approach, sum of calculated light extinction coefficients by ammonium sulfate, OM,



**Fig. 5** Chemical budget of light extinction coefficients at WQS during the sampling period. AS: ammonium sulfate; AN: ammonium nitrate, POM: particulate organic material; LAC: light absorbing carbon.



**Fig. 6** Comparison of measured and calculated light extinction coefficients.

ammonium nitrate and EC showed highly significant correlation with measured overall light extinction coefficients, and they could explain over 80% of the chemical extinction budget. In average ammonium sulfate, OM, ammonium nitrate and EC contributed about 32%, 28%, 20% and 6% to the light extinction coefficients, and light scattering particles (ammonium sulfate, OM, ammonium nitrate) were the most important constituents controlling the visibility degradation in the region.

### Acknowledgments

This work was supported by the National Natural Science Foundation of China (No. 40821003, 41025012), NSFC-Guangdong Joint Funds (No. U0833003), the Natural Science Foundation of Guangdong (No. 7118013) and the Bureau of Science, Technology and Information of Guangzhou (No. 2010U1-E00601-2).

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