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Control of $PM_{2.5}$ in Guangzhou during the 16th Asian Games period: Implication for hazy weather prevention

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HIGHLIGHTS

- Air quality, aerosol chemical properties and visibility were characterized.
- Relationships between $PM_{2.5}$ and visibility were investigated under different RH.
- New $PM_{2.5}$ standard for haze prevention was recommended.

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ABSTRACT

To evaluate the effectiveness of the integrated control measures for reducing $PM_{2.5}$ (aerosol particles with an aerodynamic diameter of less than 2.5 μm) and hazy weather, day- and night-time $PM_{2.5}$ samples were collected at an urban site in Guangzhou during the 16th Asian Games period in November 2010. $PM_{2.5}$ samples were subject to chemical analysis for major water-soluble ions, organic carbon (OC), element carbon (EC), and biomass burning tracers–anhydrosugar levoglucosan (LG). In addition, aerosol scattering coefficient (b_{sp}) at dry condition and aerosol absorption coefficient (b_{ap}) and visibility at ambient condition were measured. The seven major control measures were effective for reducing $PM_{2.5}$ mass concentration and improving visibility during the Asian Games period. All monitored air pollutants except $PM_{2.5}$ satisfied the National Ambient Air Quality Standards (NAAQS). However, daily $PM_{2.5}$ concentrations still exceeded the NAAQS on 47% of the days and hazy weather also occurred on 80% of the days during this period.

One factor causing the high frequency of hazy weather occurrence was the increased relative humidity during the Asian Games period. To avoid hazy weather occurrence, new $PM_{2.5}$ standard was recommended based on visibility calculations using three available aerosol hygroscopic curves previously obtained for this city. The recommended $PM_{2.5}$ standard was 63 μg m^{-3} under dry condition and lower than 42 μg m^{-3} under humid condition (RH ≥ 70%). These recommended values were much stricter than the NAAQS value of 75 μg m^{-3}. To reach the new standard, more rigorous control measures for coal industries should be established in the Pearl River Delta (PRD) region.

1. Introduction

Hazy weather is generally characterized by visibility degradation (<10 km) when RH <90% and is a result of light extinction — the sum of scattering and absorption, from gaseous and particulate pollutants (Sun et al., 2006). Light extinction by particles accounted for more than 90% of visibility degradation (Malm et al., 1994; Jung et al., 2009; Cao et al., 2012). The scattering ability of hygroscopic particulate species (e.g. sulfate, nitrate) can be evidently enhanced under high relative humidity conditions (Choi and Chan, 2002; Malm et al., 2003; Tang, 1996). Particle concentration and relative humidity need to be considered together for hazy weather prevention.

According to the Mie theory, fine particulate matter with an aerodynamic diameter of less than 2.5 μm ($PM_{2.5}$) is a key group of air pollutants causing hazy weather (Watson, 2002). The IMPROVE formula,

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developed by the Interagency Monitoring of Protected Visual Environments (IMPROVE) network, can predict light extinction from chemically-resolved $PM_{2.5}$ concentrations (Pitchford et al., 2007). Hazy weather has also been proven to be closely related to high levels of $PM_{2.5}$ in China (Liu et al., 2013a; Pui et al., 2014; Wang et al., 2014; Zhang et al., 2015). The national and provincial governments in China have developed controlling policies (http://www.gov.cn/) for $PM_{2.5}$, but not for haze weather prevention. It is unknown if the existing control measures for $PM_{2.5}$ is also effective to alleviate hazy weather.

The project of air quality assurance in Guangzhou during the 16th Asian Games period provided an opportunity to evaluate this issue. To ensure air quality that satisfies the NAAQS during the Asian Games period, seven major measures were exerted in Guangzhou and surrounding cities in Guangdong province (e.g., Shenzhen, Zhuhai, Foshan, Huizhou, Dongguan, Zhongshan, Jiangmen, Zhaoqing and Qingyuan, see Fig. 1) from 1 November to 20 December, 2010 (http://www.gdep.gov.cn). Accordingly, the concentrations of air pollutants, especially $PM_{2.5}$, evidently decreased during this period (Liu et al., 2013b). These control measures seemed to have also reduced the mortalities during the same period (Lin et al., 2014a). However, the frequency of hazy weather occurrence actually increased during this period. Thus, a better understanding of the relationship between $PM_{2.5}$ and hazy weather is needed for enacting collaborative control measures for both of the concerns. The present study aims to accomplish the following goals: to systematically characterize air quality and haze weather during the Asian Games period, to identify the relationship between $PM_{2.5}$ and hazy weather, to provide recommendations for new $PM_{2.5}$ standard for haze prevention under different RH conditions, and to recommend control measures based on analysis results.

2. Methodology

2.1. Site description

The instruments used in this study were installed on the roof (50 m above ground) of an office building of the South China Institute of Environmental Science (SCIES) which is located in an urban district of Guangzhou, the largest megacity of South China (23°07′N, 113°21′E; see Fig. 1). Detailed description of this station can be found elsewhere (Tao et al., 2012a).

2.2. Continuous measurements of $PM_{2.5}$ and gaseous pollutants

Tapered element oscillating microbalance (Rupprecht & Patashnick Company, Inc.; Model 1400a) was used to continuously measure $PM_{2.5}$ mass concentrations at 5 min resolution. Trace gasses including sulfur dioxide (SO_2), nitrogen oxides (NO_x), ozone (O_3) and carbon monoxide (CO) were also measured every 5 min by gas analyzers (Thermo Fisher Scientific Inc., Franklin, MA; Model 43i, Model 42i; Model 49i and 48i, respectively). All gas analyzers were calibrated weekly (Tao et al., 2012b). In addition to the above measurements, NO_2 and PM_{10} mass concentrations were also measured at the Guangdong Shangxueyuan national environmental monitoring site, located 2.0 km south of the SCIES site (Tao et al., 2014b).

2.3. Continuous measurements of aerosol scattering and absorption coefficients, and meteorological parameters

b_{sp} was measured using a single wavelength integrating nephelometer (Ecotech Pty Ltd, Australia, Model Aurora1000G) at the wavelength of 520 nm. The scattering intensity over angles from 7° to 170° is measured and integrated to yield b_{sp}. To obtain dry b_{sp}, RH of inflow air was controlled by heated inlet to be less than 40% to minimize the influence of water vapor. The nephelometer was equipped with a conventional total suspended particulate (TSP) cyclone. The flow rate was set at 5 L min^{-1}. Zero calibration was performed once every day with particle-free air, and span check was done on weekly basis using particle-free HFC-R134a gas (Tao et al., 2014a). Total b_{sp} was corrected for angular nonidealities following the method described in Anderson and Ogren (1998).

b_{ap} of 532 nm was converted from black carbon concentration measured at 880 nm wavelength by an aethalometer (Magee Scientic Company, Berkeley, CA, U.S.A., Model AE-31). The converted coefficient was 8.28 m^2 g^{-1} (Wu et al., 2009; Yan et al., 2008). The aethalometer was equipped with a conventional total suspended particulate (TSP) cyclone and a set of silica-gel diffusion driers (RH <20%) was placed at the upstream of the aethalometer. The flow rate was set at 5 L min^{-1}. The aethalometer was calibrated to zero by replacing the filter in the canister inlet with a clean filter every week (Tao et al., 2014a).

Meteorological parameters, including visibility (VIS), wind direction (WD), wind speed (WS), relative humidity (RH), temperature (TEMP), solar radiation (SR) and precipitation (PR) were measured every

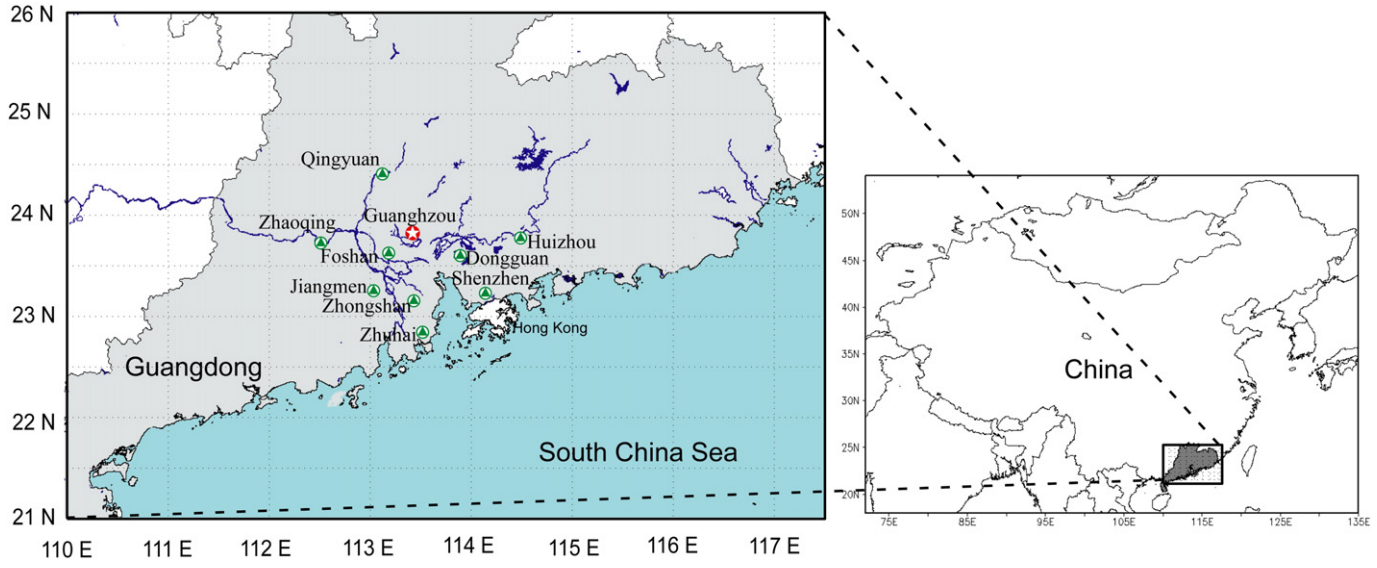

Fig. 1. The sampling location (23°07′N, 113°21′E) in Guangzhou and the locations of participating cities in Guangdong province taking the emission control measures during the Asian Games period.

30 min. Two meteorological instruments were mounted 3 m above the roof of the station (53 m above ground) (Tao et al., 2012b).

2.4. Sampling

The study period was divided into three intervals — before-, during- and post-game periods. The pre-game period covered from day-time of 4 November to night-time of 11 November, the during-game period covered from day-time of 12 November to night-time of 27 November, and the post-game period covered from day-time of 28 November to night-time of 30 November. $PM_{2.5}$ samples were collected with a Gravisol Sequential Ambient Particulate Monitor (Casella Monitor Inc., UK), which uses a cyclone to separate $PM_{2.5}$ and was operated at a flow rate of 16.7 L min^{-1} with mass flow control (MFC) system. The air stream was connected to a 47 mm Whatman quartz-fiber filter (QM/A, Whatman Inc., UK). Before sampling, the quartz filters were baked at 800 °C for at least 4 h to remove adsorbed organic vapors. In total, 27 day-time (from 8:00 a.m. to 7:00 p.m. local time) and 26 night-time (from 8:00 p.m. to 7:00 a.m. the next day) samples were collected. In this study, day- and night-$PM_{2.5}$ samples were collected separately to increase the number of samples so that the uncertainties of regression results can be limited to a reasonable range. Three field blanks were collected and used to account for any artifacts caused by gas absorption. The aerosol-loaded filter samples were stored in a freezer at −20 °C before analysis to prevent the volatilization of particles.

2.5. Gravimetric weighing

Quartz filters were measured gravimetrically for particle mass concentration using a Sartorius MC5 electronic microbalance with a sensitivity of ±1 μg (Sartorius, Göttingen, Germany) after 24 h equilibration at 23 ± 1 °C with relative humidity at 40 ± 5%. Each filter was weighed at least three times before and after sampling. Differences among replicate weights were mostly less than 20 μg for each sample. Net mass was obtained by subtracting pre-weight from post-weight (Xu et al., 2013).

2.6. Chemical analysis

An area of 0.526 cm^2 punched from each quartz filter was analyzed for eight carbon fractions following the IMPROVE_A thermal/optical reflectance (TOR) protocol on a DRI model 2001 carbon analyzer (Atmoslytic, Inc., Calabasas, CA, USA). This analysis produced four OC fractions (OC1, OC2, OC3, and OC4 at 140 °C, 280 °C, 480 °C, and 580 °C, respectively, in a helium [He] atmosphere); OP (a pyrolyzed carbon fraction determined when transmitted laser light attained its original intensity after oxygen [O_2] was added to the analysis atmosphere); and three EC fractions (EC1, EC2, and EC3 at 580 °C, 740 °C, and 840 °C, respectively, in a 2% O_2/98% He atmosphere). IMPROVE_TOR OC is operationally defined as OC1 + OC2 + OC3 + OC4 + OP and EC is defined as EC1 + EC2 + EC3-OP (Chow et al., 2007). Inter-laboratory comparison of samples between IMPROVE_TOR protocol and the TMO (thermal manganese dioxide oxidation) approach has shown a difference of <5% for TC and <10% for OC or EC (Cao et al., 2003). Average field blanks that had OC and EC were subtracted from each sample filter.

An area of 2.0 cm^2 punched from each quartz filter was extracted in 2 mL of ultrapure water under ultrasonic agitation for 1 h. Extracts were filtered through a syringe filter (pore size 0.25 μm, PTFE, Whatman, USA) to remove insoluble materials. The anhydrosugar levoglucosan was measured by a Dionex ICS-3000 system consisting of SP (quaternary pump), DC (column compartment), and ED (electrochemical detector with gold working electrode) (Dionex Corp., Sunnyvale, CA, USA). Instrumental controls, data acquisition, and chromatographic integration were performed using Dionex Chromeleon software. A calibration was performed for each analytical sequence (Engling et al., 2006; Iinuma et al., 2009; Tao et al., 2014a).

One-fourth of each quartz filter sample was used to determine the water-soluble ion concentrations. Four anions (SO_4^{2-}, NO_3^-, Cl^-, and F^-) and five cations (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) were determined in aqueous extracts of the filters by ion chromatography (IC, Dionex DX-600, Dionex Corp., Sunnyvale, CA, USA). The extraction of water-soluble species from the quartz filters was achieved by placing the cut portion (1/4) of each filter into a separate 20 mL vial, followed by 10 mL distilled-deionized water (with a resistivity of >18 MΩ), and then subjected to ultrasonic agitation for 1 h, as well as additional shaking (using a mechanical shaker) for 1 h, for complete extraction of the ionic compounds. The extract solutions were filtered (0.25 μm, PTFE, Whatman, USA) and stored at 4 °C in pre-cleaned tubes until analysis. Cation (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) concentrations were determined by using a CS12A column (Dionex Corp.) with 20 mM MSA eluent. Anions (SO_4^{2-}, NO_3^-, Cl^-, and F^-) were separated on an AS11-HC column (Dionex Corp.), using 20 mM KOH as the eluent. A calibration was performed for each analytical sequence. Standard Reference Materials (SRMs) obtained from the National Research Center for Certified Reference Materials in China were analyzed for quality assurance purposes. Procedural blank values were subtracted from sample concentrations (Tao et al., 2014b).

2.7. Data analysis methods

2.7.1. $PM_{2.5}$ mass reconstruction

To further evaluate whether the determined chemical components can represent the measured $PM_{2.5}$, the measured $PM_{2.5}$ mass was reconstructed based on $(NH_4)_2SO_4$ (1.375 SO_4^{2-}), NH_4NO_3 (1.29 NO_3^-), OM, sea salt (SS, 1.8 Cl^-), fine soil (FS), EC and water (Pitchford et al., 2007). Water concentrations were estimated by the online version of the E-AIM model (Model III) (http://www.aim.env.uea.ac.uk/aim/model3/model3a.php) at a fixed relative humidity (40%) and temperature (25 °C). The converting factor between OC and OM should vary with the dominant sources when reconstructing $PM_{2.5}$ mass. The good correlation ($R^2 = 0.73$) between OC and EC was found and the slope was 1.2 (Fig. 2a), which suggested that OC and EC should have common sources and the vehicle emission should be the dominant source (Watson et al., 2001). Moreover, the large intercept (5.8), high O_3 (53.2 μg m^3) and the moderate biomass burning tracer LG (145 ng m^{-3}) indicated that the secondary organic carbon (SOC) should have contributed greatly to OC (Cabada et al., 2004; Hennigan et al., 2011; Zhang et al., 2008). Thus, the converting factor was chosen to be 1.6 in this study. Due to the lack of soil element measurements, we assumed that Ca^{2+} is 5% of FS mass based on many previous soil source profiles (Aldabe et al., 2011; Amato and Hopke, 2012). Thus, [FS] = 20 [Ca^{2+}].

The reconstructed $PM_{2.5}$ mass can be calculated through the following formula:

$$[PM_{2.5}] = [(NH_4)_2SO_4] + [NH_4NO_3] + [OM] + [EC] + [SS] + [FS] + [Water]. \quad (1)$$

A good correlation between measured and reconstructed $PM_{2.5}$ mass concentrations was found ($R^2 = 0.90$), and the reconstructed mass accounted for 92% of the measured $PM_{2.5}$ mass (Fig. 3), indicating that $(NH_4)_2SO_4$, NH_4NO_3, OM, EC, SS, FS, and water were dominant species and the total mass of these species could closely represent the measured $PM_{2.5}$.

2.7.2. b_{sp} and b_{ap} reconstruction

According to Section 2.7.1, $(NH_4)_2SO_4$, NH_4NO_3, OM, EC, SS, and FS were the dominant chemical species in $PM_{2.5}$, which were also the major contributors to b_{sp} (Pitchford et al., 2007). Moreover, coarse matter (CM) was also a non-negligible contributor to b_{sp}, although its contribution should be lower than 5% in Guangzhou (Jung et al.,

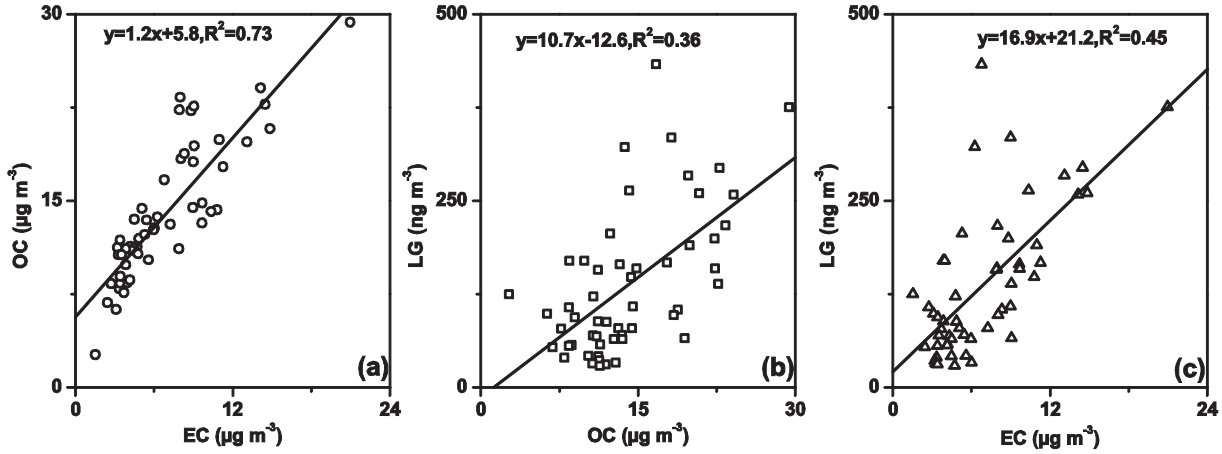

Fig. 2. Scatter plots of OC versus EC (a), LG versus OC (b) and LG versus EC (c).

2009). EC is the dominant absorption chemical component of aerosols and mainly distributes in $PM_{2.5}$ (Yu et al., 2010).

Due to the lack of information on the size distributions of these chemical species, multiple linear regressions of measured b_{sp} against the dominant species ($(NH_4)_2SO_4$, NH_4NO_3, OM, SS and FS) mass concentrations in $PM_{2.5}$ and CM were conducted to obtain their mass scattering efficiencies (MSEs) (Tao et al., 2014a,b). Considering that EC in $PM_{2.5}$ was the dominant species absorbing light, mass absorption efficiency (MAE) of EC can be obtained by regression of b_{ap} against EC. An external mixing of individual species is assumed in the above analysis.

The amount of scattering and absorption associated with individual species can be estimated statistically using (Tao et al., 2014b):

$$b_{sp} = a_1 f_L(RH)[(NH_4)_2SO_4] + a_2 f_L(RH)[NH_4NO_3] + a_3[OM] + a_4 f_{SS}(RH)\,[SS] + a_5[FS] + a_6[CM] \quad (2)$$

$$b_{ap} = b_1[EC] \quad (3)$$

b_{sp} and mass concentrations of chemical species are given in units of Mm^{-1} and μg m^{-3}, respectively. RH growth curves of $f_L(RH)$ and f_{SS} (RH) of sulfate, nitrate, and SS can be referred to IMPROVE net results (Pitchford et al., 2007). Considering that most of the sulfate and nitrate mass distributed in droplet mode (Yu et al., 2010), we used $f_L(RH)$ rather than $f_S(RH)$ in this study.

3. Results and discussion

3.1. Characterization of air quality and visibility

Daily or 8 hour average concentrations of gas pollutants (e.g. SO_2, NO_2, O_3 and CO) basically satisfied the NAAQS during the Asian Games period due to the strict control measures in Guangzhou and surrounding cities. However, $PM_{2.5}$ still exceeded the NAAQS on 47% of the days, although this was slightly improved compared to the pre-game period (63%). It is noticed that SO_2 and $PM_{2.5}$ concentrations gradually decreased with time since the control measures took effect in 1 November and then sharply dropped in 12 November when the opening ceremony began (Fig. 4). This trend was not found for NO_2, O_3 and CO. Overall, the air quality was significantly improved during the Asian Games period.

The average $PM_{2.5}$ concentration during the Asian Games period was 77 ± 24 μg m^{-3}, which was much lower than those observed in the same season but without control measures in Guangzhou (Jahn et al., 2013; Tao et al., 2014b). The concentrations of the dominant components in $PM_{2.5}$ (e.g. NH_4^+, SO_4^{2-}, NO_3^-, OC, and EC) also decreased during the Asian Games period compared with the pre-game period (Table 1 and Fig. 5). The concentrations of $PM_{2.5}$, NH_4^+, SO_4^{2-}, OC, and EC were cut down by 28%, 43%, 37%, 33%, and 31%, respectively. The 71% reduction in SO_2 concentration resulted in the large reduction of SO_4^{2-}. In contrast, NO_3^- was only cut down by 10%, which was likely due to the little change in NO_2 concentration between the before- and during-

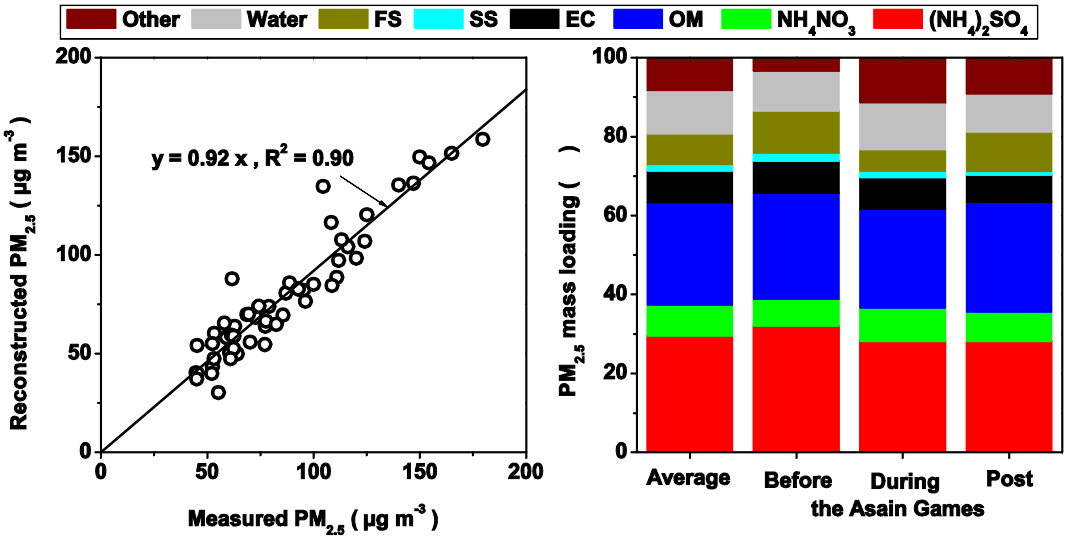

Fig. 3. Correlation between reconstructed and measured $PM_{2.5}$ mass concentrations (a) and mass fraction of major chemical components in $PM_{2.5}$ (b).

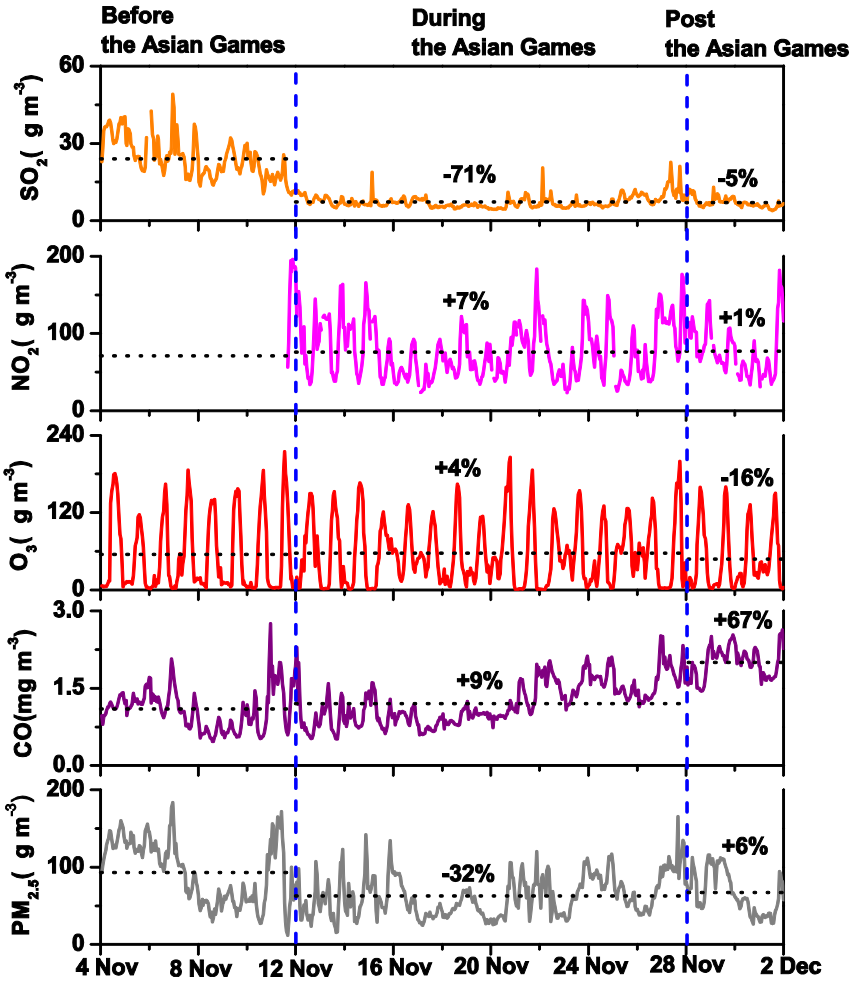

Fig. 4. Temporal variations of hourly concentrations of several air pollutants in November 2010 in Guangzhou.

game periods. The control of NO_x seemed to be not as effective as for SO_2, resulting high concentrations of its photochemical products (e.g. NO_2, O_3 and NO_3^-). The decrease in concentrations of other inorganic ions (e.g. Na^+, K^+, Ca^{2+} and Cl^-) was somewhat different than the dominant components discussed above due to their different sources.

The average visibility was 5.3 km during the Asian Games period which was improved by 31% compared with the before-game period, despite that RH was higher during the Games period (51% compared to 40%, Table 1). Dry b_{sp} and b_{ap} were 362 Mm^{-1} and 75 Mm^{-1} respectively, during the Games period, which were decreased by 31% and 23%, respectively, compared with those during the before-game period. These numbers were also lower than those measured in October 2004 in the same city, which were 418 Mm^{-1} and 91 Mm^{-1}, respectively (Andreae et al., 2008). These results should be mainly attributed to the decrease of $PM_{2.5}$ concentrations. The concentrations of the dominant species (e.g. $(NH_4)_2SO_4$, OM and EC) in $PM_{2.5}$, which were also the main contributors for visibility degradation, decreased at different extents. These phenomena demonstrate that the seven measures taken by Guangzhou municipal and the Guangdong provincial governments were effective to improve air quality and visibility.

Although visibility was significantly improved during the Asian Games period, the frequency of hazy weather occurrence (visibility less than 10 km and RH less than 90%) actually increased. Haze occurred on 80% of the days during the Games period due to increased RH and occurred on 75% of the days during before-game period. High RH generally enhances aerosol scattering ability (Malm et al., 2003). Thus, the improvement of visibility did not warrant the decrease of haze occurrence if RH increases significantly.

3.2. Contribution of air pollutants to visibility degradation

Visibility degradation is mainly caused by light scattering and absorption by particles (b_{sp} and b_{ap}) and gasses (b_{sg} and b_{ag}). To investigate the contribution of b_{sp}, b_{sg}, b_{ap} and b_{ag} to visibility degradation, visibility can be converted into a b_{ext} (550 nm) using a transfer function ($b_{ext} = 3000$/visibility) (Tao et al., 2012a; Lin et al., 2014b). The average visibility was 5.5 km during the Asian Games period and the conversed b_{ext} was 545 Mm^{-1}. Nitrogen dioxide is the only gas pollutant in the atmosphere that absorbs light. The average b_{ag} was 12 Mm^{-1} as calculated based on NO_2 concentration (Tao et al., 2012a). b_{sg} is Rayleigh scattering and was about 12 Mm^{-1} at the SCIES site (Lin et al., 2014b). The average b_{ap} was 75 Mm^{-1}converted from black carbon concentration. b_{sp} at ambient condition was 446 Mm^{-1}, estimated from b_{ext} subtracting b_{sg}, b_{ap} and b_{ag}. This number was much higher than that at dry condition (RH ≤40%) (362 Mm^{-1}) and should be ascribed to the hygroscopic properties of aerosols. In summary, b_{sp}, b_{sg}, b_{ap} and b_{ag} contributed 82%, 2%, 14% and 2%, respectively, to b_{ext} during the Asian Games period. Evidently, light extinction by particles was the key factor for visibility degradation and hazy weather.

Based on Mie theory, $PM_{2.5}$ is the major contributor to b_{sp}. To identify source contributions to visibility degradation, relative contributions of chemical components of $PM_{2.5}$ to particle light extinction (b_{sp} and b_{ap}) need to be first known. The MSEs and MAE of all the dominant chemical species are the key parameters used to connect their mass concentrations to their contributions to b_{sp} and b_{ap}. The MSEs of $(NH_4)_2SO_4$, NH_4NO_3, OM, SS, FS and CM were 5.8 ± 0.6 $m^2\ g^{-1}$, 5.9 ± 1.1 $m^2\ g^{-1}$, 6.4 ± 1.2 $m^2\ g^{-1}$, 1.9 ± 0.7 $m^2\ g^{-1}$, 0.8 ± 0.5 $m^2\ g^{-1}$ and

Table 1
Summary of air pollutant concentrations, aerosol optical properties and meteorological parameters in Guangzhou during the Asian Games period.

Components	Before the Asian Games	During the Asian Games	Post the Asian Games
$PM_{2.5}$/μg m^{-3}[a]	94 ± 41	63 ± 26	67 ± 28
SO_2/μg m^{-3}	24.0 ± 7.9	7.4 ± 2.5	7.0 ± 1.9
NO_2/μg m^{-3}	64.6 ± 21.5	76.0 ± 35.0	77.4 ± 32.9
O_3/μg m^{-3}	55.2 ± 59.8	56.7 ± 49.1	47.8 ± 48.3
CO/mg m^{-3}	1.1 ± 0.4	1.2 ± 0.4	2.0 ± 0.3
$PM_{2.5}$/μg m^{-3}[b]	107 ± 43	77 ± 24	82 ± 29
LG/ng m^{-3}	138 ± 102	130 ± 96	168 ± 103
OC/μg m^{-3}	17.9 ± 5.7	12.0 ± 4.2	14.2 ± 5.4
EC/μg m^{-3}	8.8 ± 4.8	6.1 ± 3.3	5.7 ± 2.3
Na^+/μg m^{-3}	0.6 ± 0.2	0.5 ± 0.2	0.6 ± 0.2
NH_4^+/μg m^{-3}	6.7 ± 3.7	3.8 ± 1.6	4.3 ± 1.7
K^+/μg m^{-3}	1.9 ± 0.8	1.0 ± 0.4	1.4 ± 0.8
Mg^{2+}/μg m^{-3}	0.0 ± 0.0	0.1 ± 0.1	0.0 ± 0.0
Ca^{2+}/μg m^{-3}	0.6 ± 0.2	0.2 ± 0.2	0.4 ± 0.2
Cl^-/μg m^{-3}	1.2 ± 1.1	0.7 ± 1.5	0.4 ± 0.4
SO_4^{2-}/μg m^{-3}	24.9 ± 11.9	15.6 ± 5.4	16.8 ± 5.5
NO_3^-/μg m^{-3}	5.6 ± 5.4	5.1 ± 4.8	4.8 ± 2.4
Water/μg m^{-3}	10.9 ± 4.8	9.1 ± 3.9	8.0 ± 2.4
b_{sp}/M m^{-1}	517 ± 255	362 ± 145	412 ± 155
b_{ap}/M m^{-1}	96 ± 64	75 ± 47	78 ± 35
VIS/km	4.2 ± 6.2	5.5 ± 10.6	4.7 ± 12.6
RH/%	40 ± 15	51 ± 13	56 ± 9
Temp/°C	22 ± 3	22 ± 3	21 ± 2
WS/m s^{-1}	0.4 ± 0.5	0.4 ± 0.4	0.3 ± 0.3
PR/mm	2.4	0	0
SR/W m^{-2}	170 ± 260	164 ± 247	138 ± 216

[a] TEOM.
[b] Filters.

0.5 ± 0.3 m^2 g^{-1}, respectively, which were estimated using Eq. (2). The MAE of EC was 10.6 ± 0.5 m^2 g^{-1} as estimated from Eq. (3). Good correlations ($R^2 > 0.90$) were found between the reconstructed and measured dry b_{sp} or b_{ap} (Fig. 6a and b). Moreover, the slopes of regression were close to 1.0. To further investigate the application of MSEs and MAE of chemical species in ambient conditions, b_{ext} was reconstructed from b_{sp}, b_{ap}, b_{sg} and b_{ag} (only b_{sp} needed to be reconstructed using Eq. (2)). We excluded the visibility data in 5 and 21 November because of a short rain and fog event on those days. A good correlation (R^2 = 0.87) was also found between the reconstructed and measured b_{ext} in ambient condition, although the slope (0.94) was slightly lower than 1.00 (Fig. 6c). These results suggested that the estimated MSEs and MAE should be reasonable.

According to the MSEs and MAE of the chemical species and the regression equations shown in Fig. 6, the contributions from the major air pollutants to b_{ext} are summarized in Table 2. $(NH_4)_2SO_4$, NH_4NO_3, OM, SS, FS, CM, EC and NO_2 accounted for 39 ± 8%, 12 ± 5%, 25 ± 5%, 1 ± 1%, 1 ± 0%, 4 ± 1%, 14 ± 3% and 2 ± 1%, respectively, of the estimated b_{ext} during the Asian Games period. Although the variations of the percentage contributions for all the air pollutants were small during the period, the contributions of $(NH_4)_2SO_4$, OM, FS and EC decreased by 27%, 33%, 56% and 30%, respectively, compared with those during before-game period. The percentage reductions in b_{ext} were similar to those in their respective concentrations for OM, FS and EC. However, this was not the case for $(NH_4)_2SO_4$ and NH_4NO_3 for which the decrease in their b_{ext} due to the decrease in their concentrations was offset by the increased RH (from 42% to 51%) due to their hygroscopic growth potentials. The estimated b_{sp} at ambient condition (RH = 51%) increased by 9 ± 19% compared to the measured b_{sp} at dry condition during the Asian Games period due to the hygroscopic growth of aerosols.

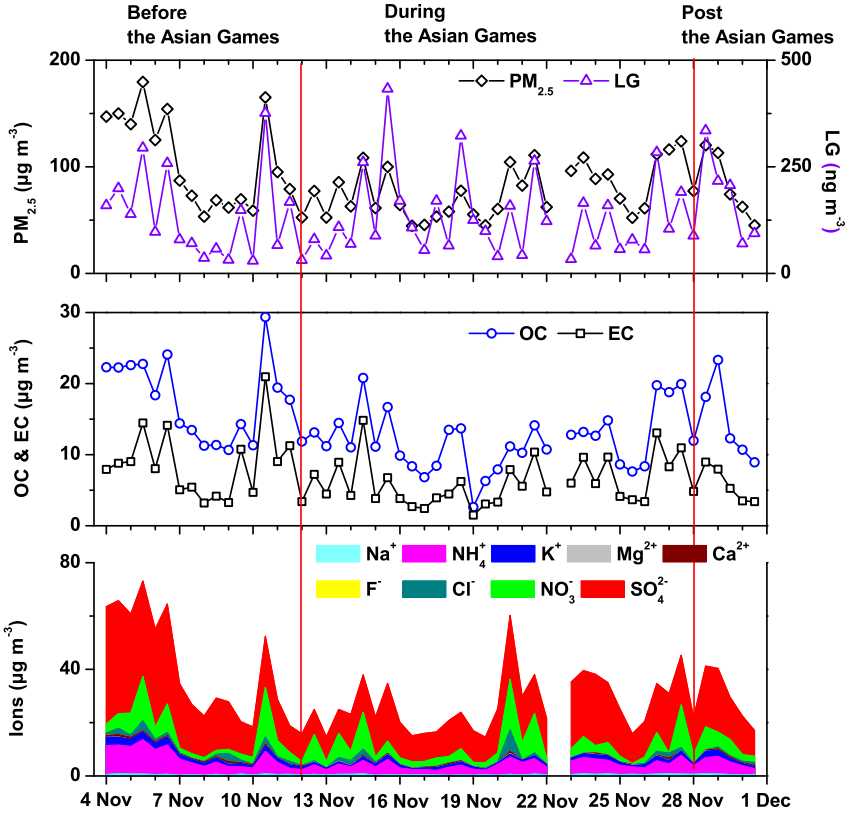

Fig. 5. Temporal variations of several chemical components of $PM_{2.5}$ in November 2010 in Guangzhou.

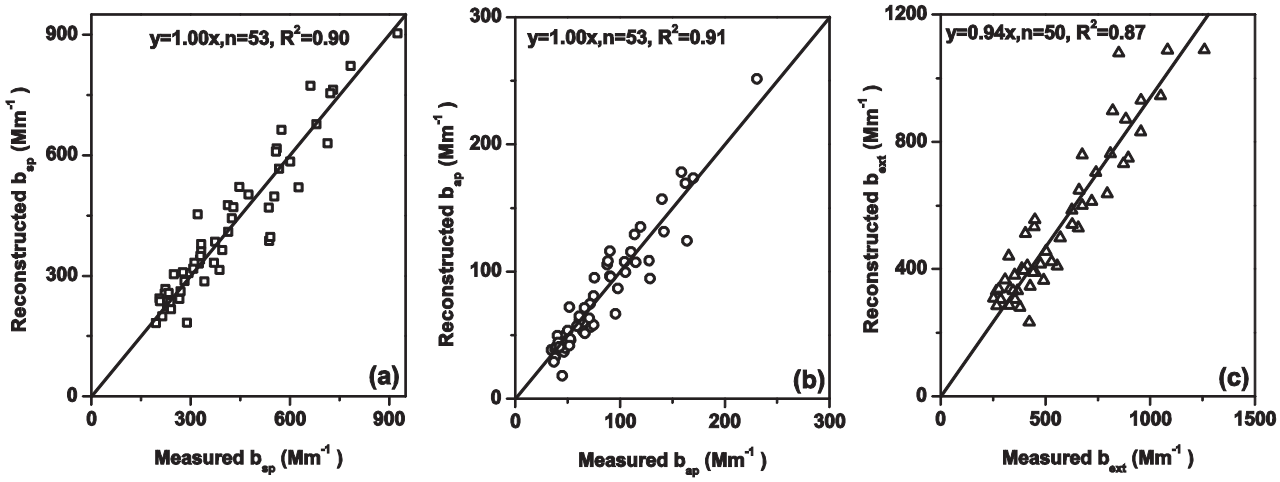

Fig. 6. Reconstructed versus measured b_{sp}, b_{ap} and b_{ext} under dry and ambient conditions.

3.3. Recommended $PM_{2.5}$ standard for haze prevention at different RH conditions

As discussed above, hazy weather should be mainly ascribed to light extinction by $PM_{2.5}$. Even if daily $PM_{2.5}$ concentration met the NAAQS, haze weather could still occur at higher RH conditions. Thus, stricter standard than the NAAQS is needed for $PM_{2.5}$ to improve visibility and reduce haze occurrence. Because of the dependence of b_{sp} by $PM_{2.5}$ on ambient RH, the $PM_{2.5}$ level under which no haze occurs (here after referred to critical $PM_{2.5}$ level) will also depend on RH. The critical $PM_{2.5}$ level under different RH conditions can be estimated according to Liu et al. (2013a):

$$b_{ext}(RH) = Q_{sp}[PM_{2.5}]f(RH) + Q_{ap}[PM_{2.5}] + 0.5[CM] + 24 \quad (4)$$

where Q_{sp} and Q_{ap} are the MSE and MAE of $PM_{2.5}$, respectively. The estimated or measured f(RH) in Guangzhou can be referred to previous studies (Cheng et al., 2008; Liu et al., 2008; Lin et al., 2014b). Q_{sp}, Q_{ap} and $PM_{2.5}$ mass concentrations are in units of $m^2\ g^{-1}$, $m^2\ g^{-1}$ and $\mu g\ m^{-3}$, respectively. The average b_{ag} and b_{sg} were both 12 Mm^{-1} during the observation period; thus, the light extinction by gas was 24 Mm^{-1}. In fact, the concentrations of NO_2 in autumn were higher than those in other seasons (Tao et al., 2012b); thus b_{ag} was likely an overestimation to some extent. As a result, b_{ext} should be less than 391 Mm^{-1} corresponding to 10 km.

The Q_{sp} or Q_{ap} of $PM_{2.5}$ can be calculated by linear regression between b_{sp} or b_{ap} and $PM_{2.5}$ mass concentrations (Fig. 7a and b). Considering CM also contributed a small portion to the total b_{sp}, b_{sp} by $PM_{2.5}$ could be estimated by measured b_{sp} minus calculated b_{sp} by CM. EC mainly distributed in fine particles and its MAE in fine particles was much higher than that in coarse particles (Yu et al., 2010). Therefore, Q_{ap} of $PM_{2.5}$ should approximately be equal to Q_{ap} of TSP. The estimated average Q_{sp} and Q_{ap} of $PM_{2.5}$ were 4.6 $m^2\ g^{-1}$ and 1.0 $m^2\ g^{-1}$, respectively. The estimated Q_{sp} was in the range of literature values generated for this and other cities in previous years (Andreae et al., 2008; Jung et al., 2009; Liu et al., 2013a; Jing et al., in press; Tao et al., 2014a,b). However, the estimated Q_{ap} was in the upper range of literature values (Liu et al., 2013a; Jing et al., in press) due to the higher EC concentrations during the observation period (Fig. 2c).

Three different, but comparable f(RH) curves were available in literature for suburban or urban areas of Guangzhou (Cheng et al., 2008; Liu et al., 2008; Lin et al., 2014b). They were all used here to estimate critical $PM_{2.5}$ using Eq. (4), results from which are summarized in Table 3. Apparently, the recommended $PM_{2.5}$ level decrease with increasing f(RH). The recommended $PM_{2.5}$ levels were similar when using f(RH) of Liu et al. (2008) and Lin et al. (2014b), both of which were originally generated for urban sites, but were slightly lower than those from using the f(RH) of Cheng et al. (2008), which was originally generated for a suburban site. According to the results in Table 3, daily $PM_{2.5}$ concentration should be controlled below 53 $\mu g\ m^{-3}$ to avoid haze occurrence at RH of 51%, as was the case during the Asian Games period. On annual scale, daily $PM_{2.5}$ concentrations should be less than 42 $\mu g\ m^{-3}$ at ambient conditions (RH = 70%) in Guangzhou and be less than 55 $\mu g\ m^{-3}$ at ambient conditions (RH = 51%) in Beijing (Liu et al., 2013a; Lin et al., 2012). And at dry condition (RH <30%), the recommended $PM_{2.5}$ concentration is 63 $\mu g\ m^{-3}$ in Guangzhou and 60 $\mu g\ m^{-3}$ in Beijing.

Table 2
Contributions to b_{ext} from major air pollutants under ambient conditions in Guangzhou.

Air pollutants	Before the Asian Games		During the Asian Games		Post the Asian Games	
	Contribution/Mm^{-1}	Percentage/%	Contribution/Mm^{-1}	Percentage/%	Contribution/Mm^{-1}	Percentage/%
$(NH_4)_2SO_4$	263 ± 184	37 ± 13	192 ± 65	39 ± 8	214 ± 55	41 ± 9
NH_4NO_3	58 ± 54	7 ± 5	63 ± 43	12 ± 5	58 ± 24	10 ± 2
OM	183 ± 47	29 ± 6	123 ± 38	25 ± 5	146 ± 42	28 ± 1
SS	7 ± 7	1 ± 1	6 ± 10	1 ± 1	4 ± 3	1 ± 1
FS	9 ± 3	2 ± 1	4 ± 2	1 ± 0	7 ± 4	1 ± 1
CM	15 ± 10	3 ± 2	19 ± 8	4 ± 1	9 ± 12	2 ± 2
EC	106 ± 39	17 ± 5	74 ± 28	14 ± 3	68 ± 23	13 ± 1
NO_2	12 ± 5	2 ± 1	13 ± 3	2 ± 1	11 ± 3	2 ± 0
Rayleigh	12 ± 0	2 ± 1	12 ± 0	2 ± 1	12 ± 0	2 ± 1
Estimated b_{ext}	663 ± 292	–	504 ± 152	–	529 ± 147	–

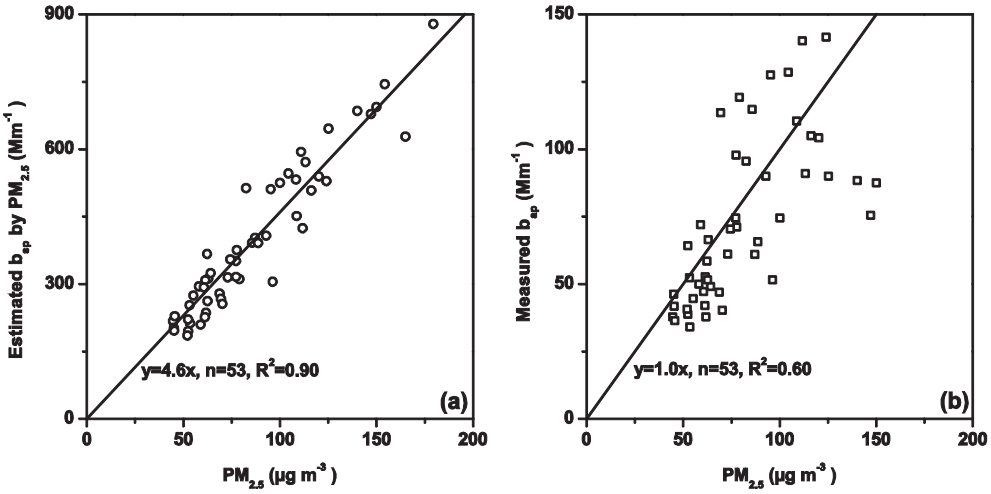

Fig. 7. Correlations between b_{sp} and $PM_{2.5}$ (a) and between b_{ap} and $PM_{2.5}$ (b).

Apparently, the NAAQS of $PM_{2.5}$ (75 μg m^{-3}) cannot prevent hazy weather occurrence at ambient conditions or dry conditions in either Guangzhou or Beijing.

3.4. Implication for haze prevention from $PM_{2.5}$ control measures

As discussed in Section 3.1, air quality and visibility were apparently improved due to a series of control measures during the Asian Games period. The dominant species in $PM_{2.5}$, which were the main contributors for b_{ext}, were also reduced. MSEs or MAE of $(NH_4)_2SO_4$, NH_4NO_3, OM and EC were higher than those of the other $PM_{2.5}$ components and CM. A reduction of these species should be more effective than reducing other species for hazy weather prevention. Reduction of the non-hygroscopic dominant species (e.g. OM and EC) can linearly decrease their contribution to b_{ext} because RH will not have an effect on their b_{ext}. Undoubtedly, air quality or statistical models should be useful tools to evaluate the control effectiveness of air pollution or hazy weather during the Asian Games period (Liu et al., 2013b; Liu et al., 2012; Valipour et al., 2012, 2013). For example, the exertion of seven major emission control measures has been proved to be effective based on a study using an air quality model (Liu et al., 2013b). The present study had a different focus from existing studies in that we investigated the causes of the increased haze weather occurrence despite the decreased $PM_{2.5}$ level and provided recommendations for future control measures.

According to the results of Section 2.7.1 and in a previous study (Tao et al., 2014b), vehicle emission should be the dominant source of carbonaceous aerosols in Guangzhou. The slope between OC and EC was 1.2 during the Asian Games period (Fig. 2), slightly lower than those found in previous years and for the same season, e.g., 1.4 in 2004 (Andreae et al., 2008) and 1.5 in 2009 (Tao et al., 2014b). This implies that the control for industrial emission related to coal combustion has taken effect and reduced carbonaceous aerosols. However, OC from non-combustion processes accounted for 38% of the total OC during the Asian Games period based on the regression equation between OC and EC (Fig. 2). This implies that the control measures for SOC formation were limited or not very effective. The high O_3 concentrations also suggested high SOC levels to some extent (Grivas et al., 2012).

In fact, vehicle emission controls were executed in Guangzhou during the Asian Games, including improvements in gasoline quality and temporary traffic controls (e.g., the so-called Odd–Even rule which allowed vehicles on road on every second day) (Zhang et al., 2013). Moreover, the enhanced I/M program and implementation of European IV emission standards for new light-duty vehicles have also been taken in the past several years. Thus, further stricter control for vehicle emission, such as the Odd–Even rule, might be a tough choice for Guangzhou government. Other controlling methods (e.g. biomass burning) should be considered to further reduce OC and EC emissions. Although the correlations between LG and OC or EC were moderate (Fig. 2b and c), several biomass burning episodes occurred during the Asian Games period (Fig. 2). The release of OC and EC by biomass burning can be estimated by typical biomass burning chemical profiles for cereal straw and wheat straw smoke (Zhang et al., 2007). The average LG/OC and LG/EC for cereal straw and wheat straw smoke were 0.07 and 0.79, respectively, in China. Then OC and EC concentrations from biomass burning accounted for 13 ± 9% and 3 ± 2%, respectively, of the total of OC and EC. If biomass burning is under control, the sum of

Table 3
Recommended $PM_{2.5}$ mass concentrations under different RH conditions in Guangzhou.

RH/%	*f*(RH) Cheng et al. (2008)	Recommended $PM_{2.5}$/μg m^{-3}	*f*(RH) Liu et al. (2008)	Recommended $PM_{2.5}$/μg m^{-3}	*f*(RH) Lin et al. (2014b)	Recommended $PM_{2.5}$/μg m^{-3}	Final recommended $PM_{2.5}$/μg m^{-3}
≤30	1.000	63	1.000	63	1.000	63	63
31–35	1.001	63	1.011	63	1.021	62	62
36–40	1.007	63	1.036	61	1.057	60	60
41–45	1.019	62	1.070	60	1.095	57	57
46–50	1.039	61	1.117	58	1.135	55	55
51–55	1.068	60	1.178	55	1.179	53	53
56–60	1.108	58	1.257	52	1.230	51	51
61–65	1.162	56	1.355	49	1.292	49	49
66–70	1.237	53	1.475	46	1.363	46	46
71–75	1.340	50	1.621	42	1.451	43	42
76–80	1.488	45	1.795	38	1.588	45	38
81–85	1.711	40	2.001	35	1.807	38	35
86–90	2.085	34	2.241	31	2.217	32	31

OC and EC could be cut by up to 16%. This is still not enough for hazy weather prevention.

Further reduction of $(NH_4)_2SO_4$ and NH_4NO_3 seemed to be the only choice to prevent hazy weather. $(NH_4)_2SO_4$ and NH_4NO_3 should be cut by 60% to avoid hazy weather occurrence if biomass burning is under control. These measures are still conservative because MSEs of these chemical species should decrease with their mass concentrations (Malm et al., 2003). In conclusion, OM, EC, $(NH_4)_2SO_4$ and NH_4NO_3 concentrations should have been reduced by an additional 15%, 3%, 60% and 60%, respectively, to prevent hazy weather during the Asian Games period. Accordingly, $PM_{2.5}$ concentration should be decreased to about 53 μg m^{-3}, which is equal to the recommended value of 53 μg m^{-3} at RH = 51%. Obviously, the coal consumption should be further reduced in the future for further reducing SO_2 and NO_x emissions from power plants and industrial boilers in Guangdong province, especially in the PRD region.

Moreover, although FS concentrations decreased by 62% during the Asian Games period compared with that during the before-game period, its MSE was lower than those of the other species in $PM_{2.5}$ and thereby had an insignificant impact on b_{ext}. Thus, the control measures for FS were more effective for $PM_{2.5}$ than for hazy weather.

4. Conclusions

Air pollutants including SO_2, NO_2, O_3, CO and $PM_{2.5}$, chemical compositions of $PM_{2.5}$, b_{sp}, b_{ap} and visibility were synchronously measured in November 2010 in an urban environment in Guangzhou. $PM_{2.5}$ and visibility were improved during this period when the Asian Games were held due to the exertion of seven major emission control measures. The occurrence frequency of hazy weather actually increased slightly during this period due to the increase in humidity. Light extinction by $PM_{2.5}$ and aerosol hygroscopic growth were the major factors needed to be considered in order to decrease haze weather. New $PM_{2.5}$ standard was recommended based on the measured dominant $PM_{2.5}$ components and existing aerosol hygroscopic growth curves and was much stricter than the current NAAQS. To satisfy the new standard, the total of OC and EC needs to be further reduced by up to 18% and $(NH_4)_2SO_4$ and NH_4NO_3 each needs to be reduced by 60%. This requires a large reduction of SO_2 and NO_x emissions from coal consumption and other emission sectors in the PRD region. Exact percentage reduction of SO_2 and NO_x needs to be further investigated, e.g., from using air equality model simulations. Results presented here provided scientific evidence for making future emission control policies and also provided new research questions.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.11.074.

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