Evaluation of the Changes of the Concentration, Composition and Possible Sources of Size-Resolved Particulate Matter between 2010 and 2011 in a Western Chinese Mega City

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ABSTRACT

In this work, PM_{2.5} and PM₁₀ samples were collected from four functional regions in Chengdu city from Jan. to Feb. in 2010 and 2011. The chemical concentrations of nineteen elements, two water-soluble ions and TC were measured. The results showed that the levels of particulates and their chemical components were relatively higher in 2010 than those in 2011: for PM₁₀, PM (39%), Si (27%), TC (76%), NO₃⁻ (16%), SO₄²⁻ (19%); for PM_{2.5}, PM (33%), Si (11%), TC (67%), NO₃⁻ (32%), SO₄²⁻ (17%). Ternary diagram analysis, enrichment and other statistical methods were employed to study the spatial and temporal variations of the concentrations of PM and their compositions. Moreover, relative changes (%) of concentrations combined with the relative reported data from statistical yearbook and environmental quality reports are discussed to evaluate the effects of pollution control strategies for different functional regions. Finally, back-trajectory and factor-cluster analysis are proposed as an aid for the discussion of the meteorological influences and the possible source categories. The reduction of crustal dust may be attributed from the influence of long-range air transport. All the methods used above resulted in an overall agreement that showed the pollution control strategies were effective in reducing particulate air pollution: the total carbon (TC) reduction, accounting for 40–49% (PM₁₀) and 35–48% (PM_{2.5}), was mostly attributed to the strategy of banning high-emission yellow-tag vehicles, especially in commercial regions; the reduction of SO₂ and NO_x emissions had an impact on the sulphate (17.03% for PM₁₀ and 15.37% for PM_{2.5}) and nitrate (12.48% for PM₁₀ and 21.58% for PM_{2.5}) decrease. The findings of this study can provide useful information for developing effective control strategies.

Keywords: PM₁₀; PM_{2.5}; Control strategies; Enrichment; Factor-cluster analysis.

INTRODUCTION

Atmospheric particulate matter (PM), which has been studied by numerous researchers, is an important air pollutant in many large cities (Zheng *et al.*, 2005; Moorhouse, 2007; Thurston *et al.*, 2011; Niyobuhungiro and Blottnitz, 2013; Tian *et al.*, 2013). A large amount of PM not only affects visibility and causes acidification but also has adverse effects on human health, such as increased respiratory and

cardiovascular symptoms and diseases (Harrison and Yin, 2000; Tie *et al.*, 2009; Cheng *et al.*, 2012; Vernile *et al.*, 2013). Moreover, some studies have showed that PM can change the radiation balance, and have important effect on climate change (Bellouin *et al.*, 2005; Tiwari *et al.*, 2012; Tahir *et al.*, 2013).

Due to these adverse effects caused by PM, many countries have taken various measures to control particle pollution to drastically reduce PM levels (Zhao *et al.*, 2006; Heinberg and Fridley, 2010; Shao *et al.*, 2011; Wang and Hao, 2012). Therefore, to establish highly efficient measures for PM control and prevention, it is imperative to evaluate the effectiveness of these control strategies.

Some studies have focused on the evaluation of policy implementation (O’Ryan, 1996; Gianini *et al.*, 2012; Li *et al.*, 2012). Such studies have shown that pollution control strategies can be effective in decreasing particulate pollution in urban air. Although most of these studies have already

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analysed the changes of PM concentrations under the influence of control measures, few studies have examined their chemical characteristics, especially those of size-resolved PMs. What's more, the studies about different regions are quite limited. Because urban areas represent an assembly of different land use types and different source categories, investigation of the effectiveness of control strategies for various functional regions is necessary.

In this paper, in order to evaluate the effectiveness of control strategies, we have chosen the same time in two years and the same monitoring regions in Chengdu. The changes of the concentration, composition and possible sources of size-resolved particulate matter between 2010 and 2011 in four functional regions of Chengdu have been studied. To discuss the chemical characteristic of PMs, methods of enrichment (RE), relative change (RC), ternary plots and back-trajectory were employed. Additionally, a recently proposed factor-cluster analysis (FCA) method was applied to identify the possible sources in monitoring regions (Masiol *et al.*, 2010, 2012). To the best of our knowledge, this is the first time FCA has been used to study both temporary and spatial variation simultaneously. Moreover, this study represents a first effort to investigate the effectiveness of control strategies for different land use types. The findings in this work can be used to provide effective measures that can greatly enhance PM environmental management for different functional regions.

MATERIALS AND METHODS

Sampling Site Description

Chengdu (102°54'–104°53'E, 30°05'–31°26'N), the capital of Sichuan Province in southwest China, is one of the most important economic, transportation and communication centres in Western China. The population is more than 10 million, and its area is approximately 12121 km². Chengdu was recently named one of the 13 emerging megacities or megalopolises in China in a July 2012 report by the Economist Intelligence Unit. Chengdu is located at the western edge of the Sichuan Basin and sits on the Chengdu Plain. The area has a monsoon-influenced humid subtropical climate, with distinct seasons accompanied by abundant rainfall and protection from both sweltering summers and freezing winters. The cities' favourable climate contributes to the development of agriculture and animal husbandry, making Chengdu a comfortable place for sustained human habitat as well as a suitable home for giant pandas.

Ambient Sampling Method

PM₁₀ and PM_{2.5} samples were collected from four functional regions: Background, Commercial, Industrial and Residential, as shown in Fig. S1 (in the supplementary material). The four sampling sites were named BGY (Background), HBDL (Commercial), KJY (Industrial) and SWY (Residential). The samplers were located on the top of the buildings, and the heights of the building are 10–15 meters. Two sampling campaigns were conducted from Jan. to Feb. in 2010 and 2011. In this work, a total of 126 samples (57 in 2010, 69 in 2011) of PM₁₀ and 128 (59 in 2010, 69 in 2011)

of PM_{2.5} were obtained. The PM₁₀ and PM_{2.5} samples were collected by filtration using a medium-volume air sampler (Wuhan Tianhong Intelligence Instrumentation Facility, TH-150 Medium Volume Sampler), with the pump set at 100 L/min and running continuously for 24 h, from 8:00 am to 8:00 am on the next day. For both PM₁₀ and PM_{2.5} at each site, two parallel samplers were used, one for polypropylene membrane filters and the other for quartz-fibre filters.

Chemical Analysis

Concentrations of nineteen elements, two water-soluble ions and TC (total carbon) were measured. Briefly, the elements (including Na, Mg, Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg and Pb) were analysed by an Inductively Coupled Plasma (IRIS Intrepid II, Thermo Electron) spectrometer (Baldwin *et al.*, 1994; Shi *et al.*, 2012; Tian *et al.*, 2013). Water-soluble ions, NO₃[−] and SO₄^{2−}, were detected by ion chromatography (DX-120, Dionex Ltd., USA) after extraction by an ultrasonic extraction system (AS3120, AutoScience). A clip in the shape of a circle (15 mm in diameter) from each quartz-fibre filter was used to measure TC following the IMPROVE thermal/optical reflectance (TOR) protocol implemented in DRI/OGC carbon analysers (Chow *et al.*, 1993; Watson *et al.*, 1994). A detailed description of sampling and chemical analyses was provided in our previous studies (Bi *et al.*, 2006; Wu *et al.*, 2009).

RESULTS AND DISCUSSION

PM₁₀ and PM_{2.5} Concentrations

The concentrations of size-resolved PM and chemical species are listed in Table 1. In 2010, the average concentrations of PM₁₀ and PM_{2.5} were found to be in the range of 193.64–331.17 µg/m³ and 140.01–198.50 µg/m³, respectively. In 2011, the average concentrations at four different functional regions varied from 134.29 to 231.85 µg/m³ for PM₁₀ and from 107.56 to 165.63 µg/m³ for PM_{2.5}. The concentrations of PM₁₀ and PM_{2.5} in 2011 were significantly lower than in 2010, with a p value < 0.05 (T-test). Additionally, the concentrations of fine and coarse particulates in Chengdu were compared with those in other cities. As shown in Table S1 (in the supplementary material), it is obvious that the concentrations of the PMs in Chengdu were higher than in most of the other cities in the world (Park and Kim, 2005; Ho *et al.*, 2006; Querol *et al.*, 2008; Lim *et al.*, 2010; Thurston *et al.*, 2011; Murillo *et al.*, 2012; Xu *et al.*, 2012), though slightly lower than in some Chinese cities, such as Beijing (He *et al.*, 2001; Wang *et al.*, 2003; Zeng *et al.*, 2010).

Concentrations Variability in Different Regions

The PM spatial distribution between 2010 and 2011 differed: in 2010, the PM₁₀ concentration was highest in commercial regions and lowest in residential regions; in 2011, the PM₁₀ concentration was highest in residential regions and lowest in commercial regions. Unlike PM₁₀, the PM_{2.5} concentration exhibited the highest levels in industrial regions for both years.

Table 1(a). Concentration of PM₁₀ and PM_{2.5} as well as their chemical species in 2010 (μg/m³).

	2010							
	PM ₁₀				PM _{2.5}			
	Background	Commercial	Residential	Industrial	Background	Commercial	Residential	Industrial
PM	193.64 ± 46.05	331.17 ± 128.96	283.70 ± 75.18	309.05 ± 99.70	140.01 ± 38.01	196.44 ± 88.50	180.03 ± 92.09	198.50 ± 75.89
Na	2.22 ± 0.78	4.00 ± 1.57	3.77 ± 0.85	4.06 ± 1.35	0.97 ± 0.41	1.19 ± 0.61	1.09 ± 0.55	1.38 ± 0.61
Mg	0.31 ± 0.32	2.51 ± 1.31	1.59 ± 1.62	1.88 ± 1.33	0.15 ± 0.08	0.37 ± 0.35	0.58 ± 0.88	0.71 ± 1.03
Al	14.99 ± 4.81	16.45 ± 6.44	13.56 ± 3.11	15.84 ± 7.29	6.03 ± 2.70	7.84 ± 4.04	5.67 ± 3.42	6.29 ± 2.89
Si	27.83 ± 8.58	34.86 ± 12.82	27.52 ± 6.50	33.18 ± 14.14	12.96 ± 4.32	12.20 ± 6.06	13.53 ± 8.17	13.22 ± 5.80
K	3.45 ± 1.19	5.42 ± 2.55	3.73 ± 1.58	4.30 ± 2.13	1.76 ± 0.85	2.42 ± 1.58	1.91 ± 1.04	2.23 ± 1.14
Ca	4.24 ± 1.45	16.72 ± 7.28	11.00 ± 5.50	14.14 ± 5.99	1.01 ± 0.56	3.17 ± 1.50	4.07 ± 4.12	3.60 ± 7.29
Ti	1.09 ± 0.53	0.47 ± 0.36	0.42 ± 0.22	0.53 ± 0.28	0.48 ± 0.28	0.21 ± 0.13	0.19 ± 0.20	0.22 ± 0.16
V	0.00 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
Cr	0.02 ± 0.00	0.06 ± 0.04	0.03 ± 0.02	0.03 ± 0.02	0.01 ± 0.00	0.02 ± 0.03	0.02 ± 0.02	0.10 ± 0.26
Mn	0.08 ± 0.03	0.27 ± 0.12	0.14 ± 0.07	0.22 ± 0.11	0.05 ± 0.02	0.15 ± 0.06	0.17 ± 0.19	0.14 ± 0.14
Fe	4.03 ± 1.19	7.76 ± 3.39	5.96 ± 2.17	7.45 ± 2.77	2.00 ± 0.77	2.45 ± 1.23	2.08 ± 1.17	2.37 ± 0.94
Co	0.03 ± 0.02	0.02 ± 0.02	0.01 ± 0.01	0.02 ± 0.02	0.02 ± 0.01	0.01 ± 0.01	0.00 ± 0.00	0.01 ± 0.01
Ni	0.04 ± 0.38	0.02 ± 0.01	0.02 ± 0.02	0.02 ± 0.01	0.04 ± 0.24	0.01 ± 0.01	0.02 ± 0.02	0.01 ± 0.01
Cu	0.08 ± 0.05	0.12 ± 0.05	0.48 ± 0.35	0.48 ± 1.08	0.05 ± 0.07	0.09 ± 0.05	0.20 ± 0.11	0.19 ± 0.42
Zn	0.43 ± 0.21	1.21 ± 0.90	1.16 ± 0.90	0.98 ± 0.73	0.35 ± 0.18	1.15 ± 0.73	0.77 ± 0.74	0.93 ± 1.28
As	0.01 ± 0.01	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
Cd	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
Hg	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
Pb	0.33 ± 0.18	0.08 ± 0.05	0.06 ± 0.07	0.11 ± 0.32	0.28 ± 0.14	0.06 ± 0.04	0.06 ± 0.06	0.08 ± 0.10
TC	32.43 ± 8.66	92.37 ± 40.02	59.48 ± 19.15	71.08 ± 26.76	29.03 ± 6.91	74.21 ± 28.61	54.09 ± 24.18	61.92 ± 21.02
NO ₃ ⁻	9.42 ± 13.33	20.44 ± 10.50	20.68 ± 9.95	22.61 ± 15.75	8.54 ± 5.25	15.21 ± 18.57	18.28 ± 15.10	17.15 ± 16.40
SO ₄ ²⁻	26.89 ± 9.70	48.28 ± 36.51	48.99 ± 18.71	76.88 ± 43.20	23.52 ± 7.58	38.33 ± 30.22	32.69 ± 26.13	47.18 ± 23.58
Crustal Dust	105.56 ± 29.03	147.51 ± 51.18	113.60 ± 21.08	137.42 ± 55.88	46.31 ± 15.90	52.14 ± 22.76	50.93 ± 30.21	51.63 ± 24.97

The concentrations and fractions of the chemical species collected at the four different functional regions in 2010 and 2011 are shown in Table 1 and Fig. S2. Similarly to other studies (Ho *et al.*, 2006; Querol *et al.*, 2008), carbonaceous species and secondary inorganic ions (sulphate, nitrate) (15–38% and 19–36% of PMs) are the main contributors to the PM_{2.5} and PM₁₀ concentrations. Other components, such as Si, Al and Ca, showed fractions of 6–15%, 3–8% and 1–6% in the PMs, respectively.

The PM_{2.5}/PM₁₀ ratios for the concentrations of individual chemical species in the different functional regions are shown in Table S2, with a range from 0.58 to 0.80. The ratio for TC was more than 80%, due to most of the carbonaceous compositions mainly being enriched in fine PM (Wang *et al.*, 2003). The PM_{2.5}/PM₁₀ ratios for nitrate and sulphate were relatively lower than TC, at more than 60%, due to SO₄²⁻ and NO₃⁻ being more abundant in PM_{2.5} (Oh *et al.*, 2011; Thurston *et al.*, 2011; Wang *et al.*, 2012). In contrast, crustal elements such as Ca, Mg, Al and Na appeared to have very low ratios of PM_{2.5}/PM₁₀. Studies have shown that the majority of crustal elements are likely to be enriched in coarse particulates (Querol *et al.*, 2008). Overall, in each functional region, the size distribution of PM and chemical species showed similar characteristics (Gianini *et al.*, 2012).

Levels of Major Constituents in PM

According to Table 1, chemical species such as Al, Si, Ca, TC, SO₄²⁻ and NO₃⁻ presented relatively high concentrations in all regions for both fine and coarse particles. Among these major constituents, Al, Si and Ca are the markers of crustal dust (Shi *et al.*, 2009; Begum *et al.*, 2010; Shi *et al.*, 2011), indicating crustal dust may contribute to the PM in

these regions. Previous studies have shown that crustal dust can be estimated by the “sum of oxides” method (Andrews *et al.*, 2000), assuming that the crustal dust is composed of SiO₂, Al₂O₃, Fe₂O₃, TiO₂, CaO and K₂O (Hedberg *et al.*, 2005). The concentration of crustal dust was calculated according to Hedberg *et al.* (2005):

$$[\text{Crustal dust}] = 2.14 [\text{Si}] + 1.89 [\text{Al}] + 1.43 [\text{Fe}] + 1.67 [\text{Ti}] + 1.40 [\text{Ca}] + 1.20 [\text{K}] \quad (1)$$

where the square brackets are the concentrations of chemical species. The concentrations of crustal dust for all regions are also listed in Table 1.

Fig. 1 describes the fractions of major constituents for both PMs from all regions. Crustal dust exhibited the highest levels among the major constituents, accounting for 40–55% (in 2010) and 43–58% (in 2011) in PM₁₀ and 26–33% (in 2010) and 28–36% (in 2011) in PM_{2.5}. These levels suggest that crustal dust may be an important source category in the study regions. TC, a marker of vehicle exhaust (Pant and Harrison, 2012), showed the second most abundant levels, indicating that vehicle exhaust may be a potential source category. The fractions of SO₄²⁻ and NO₃⁻ were approximately 20% and 10%, respectively, indicating that the secondary sulphate and nitrate may occur. Additionally, SO₄²⁻ has also been reported to be a good marker for coal combustion (Pant and Harrison, 2012) due to its precursor gas (SO₂) being emitted from fossil fuel combustion (Shon *et al.*, 2012). Thus, industrial coal combustion cannot be ignored. The annual total industrial coal consumption (million tons) and SO₂ emission in Chengdu during 2010 and 2011 can be found in Table S3 in the supplementary material.

Fig. 1. Fractions (%) of major constituents in size resolved PM from different regions, in 2010 and 2011.

The fractions of major constituents in ambient PM are influenced by the profile of the source category. To analyse the fractions of major constituents, the ternary diagrams (in Fig. S3) for the fractions (%) of crustal dust, TC and sulphate + nitrate are plotted. For PM₁₀ in 2010, it can be found that the points of the commercial regions exhibit relatively higher fractions (%) of TC, indicating a more frequent impact of traffic in commercial regions than in other regions, which is reasonable due to the high traffic density present in the commercial regions. The points of the industrial regions were characterised by sulphate + nitrate, which suggested that secondary sources and industrial coal combustion might be the important sources categories in this region. For the background points, crustal dust was the richest component, as indicated by lack of the anthropogenic sources. The possible sources for the residential regions were complex because of the lack of outstanding constituents. In 2011, PM₁₀ exhibited a similar plot. However, the fractions of TC were clearly decreased for all the points, suggesting the influence of vehicle exhaust may have been relatively weaker in 2011 than in 2010.

The ternary diagrams of PM_{2.5} are consistent with those of PM₁₀ in some aspects. Nevertheless, in comparison with the PM₁₀ points, the fractions of TC and sulphate+nitrate in PM_{2.5} were relatively higher, while those of crustal dust in PM_{2.5} were lower. This result is due to crustal dust usually having a more pronounced coarse particle size fraction than coal combustion, vehicle emissions and other particles (Ehrlich *et al.*, 2007; Lin *et al.*, 2010), while NO₃⁻ and SO₄²⁻ are rich in the fine particles (Lin *et al.*, 2010; Zhang *et al.*, 2011).

Enrichment of Major Constituents for Different Regions

As discussed above, the different regions are characterised by discriminative constituents. In this section, therefore, the enrichment of major constituents is calculated to further analyse the temporal and spatial variations of the chemical characteristics of PM. For the commercial region, the enrichment (RE) was calculated as follows (Gianini *et al.*, 2012):

$$RE_i = ([Commercial]_i - [Background]_i) / [Commercial]_i \times 100 \quad (2)$$

where [Commercial]_i and [Background]_i are the average concentrations (μg/m³) of the *i*th measured constituents in the commercial and background regions. For calculations of residential and industrial REs, [Commercial]_i in Eq. (2) should be replaced by [Residential]_i and [Industrial]_i, respectively.

The temporal and spatial REs of size resolved PM and their major constituents are described in Fig. 2. It can be clearly found that the TC-REs presented the highest level for the commercial regions; sulphate-REs were important for the industrial regions; nitrate-REs were prominent for the residential regions; and the crustal dust-REs were quite low for all regions. The spatial variations of the REs may be attributable to the different important source categories in the different regions.

To further discuss the spatial differences of the REs, RE diagrams were made, which are scatter plots of the major constituent RE values for one region against those of another (Wongphatarakul *et al.*, 1998; Zhang and Friedlander, 2000; Gianini *et al.*, 2012). Fig. 3 gives a comprehensive comparison of the REs for all regions. According to all the scatter plots in Fig. 3, for both PM₁₀ and PM_{2.5}, it can be concluded that TC was higher in the commercial region than the other regions; sulphate was outstanding in the industrial regions; and for the residential regions, the prominent constituents were inconsistent for all the plots. The results of Fig. 3 are in reasonable agreement with those of the ternary diagram (in Fig. S3).

The Changes of Chemical Concentrations

Several studies have shown that pollution control strategies can positively affect atmospheric pollution reduction (Wang *et al.*, 2009; Gianini *et al.*, 2012). In this work, the changes of chemical concentrations between 2010 and 2011, combined with the relevant reported data in the Chengdu Statistical Yearbook and Chengdu Environmental Quality Reports (see Table S3), are discussed to evaluate the effects of pollution control strategies (Table S4). The relative changes (RCs) of the individual major constituents of PM are calculated according to:

$$RC(\%) = ([2011]_i - [2010]_i) / [2010]_i \times 100 \quad (3)$$

where [2011]_i and [2010]_i are the average concentrations (μg/m³) of the *i*th measured constituents in different sampling years. Fig. 4 describes the relative changes of the concentrations of PM₁₀ and PM_{2.5} as well as their major constituents in 2010 and 2011 for the background, commercial, industrial and residential regions.

The mass concentrations of PM₁₀ and PM_{2.5} were obviously reduced for all the regions, accounting for 18–37% (PM₁₀) and 17–38% (PM_{2.5}). Additionally, the major constituents exhibited various changes. First, the largest decrease was found for TC, accounting for 40–49% (PM₁₀) and 35–48% (PM_{2.5}). According to the Chengdu Statistical Yearbook (Table S3), the vehicle population increased from 2.36 (2010) to 2.59 (2011) million, which contradicts the results indicating the high reduction of TC. Hence, the positive change of TC is certainly attributable to the pollution control strategies (Strategy 2 in Table S4). As shown in Table S4, high-emission yellow-tag vehicles have been banned from roads (from 7:30 to 19:30) in urban areas since 1 Oct. 2010. According to Fig. 4, the relative changes of TC presented the highest level in the commercial regions, showing the most outstanding effect of strategy 2 in these regions compared with the other regions.

Second, the concentrations of sulphate and nitrate decreased in most regions. The implementation of the continuing reduction of SO₂ (the precursor of sulphate) emissions might be an important factor for the reduction of sulphate (Strategy 1 in Table S4). During the Chinese Eleventh Five-year period (2006–2010), the continuing reduction of SO₂ emissions was an important pollution control strategy in Chinese cities, and it played a key role in decreasing these

Fig. 2. Temporal and spatial REs (%) of size resolved PM and their major constituents.

emissions. According to Table S3, the annual total industrial SO₂ emissions decreased from 127 to 96.8 thousand tons (the relative change was –24%), and the industrial coal consumption ranged from 8690 to 7957 thousand tons (the relative change was –8%). Additionally, SO₄^{2–} can also be product from the long-term transport. So the variety of transport in different years might be another factor for sulfate variation in the city. The NO_x (the precursor of nitrate (Shon *et al.*, 2012)) reduction may have been a chief influencing factor in the nitrate reduction. In Table S3, the annual total industrial NO_x emissions decreased from 57.8 to 52.4 thousand tons. The yellow-tag vehicle ban also resulted in reduced NO_x emissions.

Third, for most regions the concentrations of crustal dust decreased obviously. As found in some studies, long-range air transport usually contributes to crustal dust in addition to local pollution (Wang and Hao, 2012). To better understand the air flow and long-range transport patterns, the back trajectory model was applied. 3-D 72-hour back trajectories were calculated using the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT-4.9) model from 1 Jan. to 28 Feb. in 2010 and 2011, respectively. For each day, the 3D 72-h backward trajectories were calculated every 6 h at 08, 14, 20, and 02 UTC (coordinated universal time) at a height of 500 m above the ground level (Hwang and Hopke, 2007). A total of 236 trajectories were calculated

and inputted for cluster analysis to identify the common atmospheric transport patterns. As shown in Fig. S4(a) and S4(b), six clusters were obtained according to their overall wind speed and direction for each year. Note that in the cluster map for 2010, Cluster 1 (13%) came from the direction of Xinjiang province. It is well known that the Taklimakan Desert (the largest desert in China) is in the southwest portion of Xinjiang province. In 2011, the trajectories (Cluster 1) originating from Xinjiang province were only 3% of the total trajectories, much lower than those in 2010; and the cluster from the Inner Mongolia province only accounted for 5%. Thus, the changes of crustal dust may be attributable from the influence of long-range air transport.

Source Identification by FCA

As discussed above, the spatial and temporal variations of the major constituents were attributed to the important source categories at the different regions in the two years. In this work, factor-cluster analysis (FCA) was performed to identify the potential source categories and then select groups of samples on the basis of their similar chemical compositions and origins (Masiol *et al.*, 2010). The FCA included two steps. First, the ambient PM data were analysed by FA (factor analysis). The possible source categories were identified by the varimax rotated factor loading matrix. Factor scores (n samples and p extracted factors) associated

with each sample were also obtained by FA. Second, the factor score matrix was applied as input for cluster analysis (CA) to group samples by similarity in source contributions

(Masiol *et al.*, 2010). In recent studies, FCA had been proposed and proved as a useful tool for source identification (Masiol *et al.*, 2010, 2012).

Fig. 3. Enrichment diagrams: region against region, for PM_{10} and $PM_{2.5}$, in 2010 and 2011.

Fig. 4. Relative change (%) for mean concentration of major constituents between the 2010 and 2011.

In this work, for the each size-resolved PM, the samples from all regions in the two years were combined to be used as input data for the FA. Amato and Hopke (2012) suggested that it is possible to use the spatial variability of the source contributions to enhance the source apportionment. In our previous and other works, using combined data for the factor model also produced reasonable results (Escrig *et al.*, 2009; Shi *et al.*, 2009). Thus, for PM₁₀, a 126 × 12 input dataset was analysed by FA, where 126 is the number of PM₁₀ samples and 12 is the number of chemical species (some trace elements were removed to avoid disturbance). SPSS 12.0 software was applied for the FA.

Table 2 lists the rotated factor loadings for the PM₁₀ samples. Four factors were extracted by FA. The first factor (49% of the total variance) obtained high loadings for Si, Al, Fe and TC. Si and Al are the source markers for crustal dust, and TC is associated with the vehicle exhaust emission. Thus, this factor may indicate mixed sources, including crustal dust and steel manufacture and vehicle exhaust emissions. The result of these sources being combined into a single factor most likely reflects the contribution of some regions with the influence of these sources, and therefore, FA cannot separate them (Minguillon *et al.*, 2012). Factor 2 (16% of the total variance) strongly correlated with Ca and can be identified as cement dust (Morishita *et al.*, 2011). According to the field survey, cement manufactories located in the study areas, and this is in agreement with the identified source category. Additionally, Mg also got high loading, so the Magnesium alloy industrial manufacture source might be included in this factor. Factor 3 (10% of the total variance) and Factor 4 (9% of the total variance) were weighted by SO₄²⁻ and NO₃⁻, so the two factors can be identified as secondary sulphate and nitrate, respectively.

After FA, the factor scores were used as input for CA using Ward's method and Euclidean distance (Masiol *et al.*, 2010). As shown in Fig. 5, five groups were clustered by FCA. The concentrations (mean ± standard deviation) of

PM₁₀ and the individual chemical species for each group are listed in Table 2. Group 1 (n = 19), with high concentrations of Al, Si, Fe and TC, can be associated with Factor 1. Group 2 (n = 24) showed high fraction (%) of Ca compared with other factors, which can be interpreted as cement dust (Factor 2). Group 3 (n = 15) and Group 4 (n = 33) had high concentrations of SO₄²⁻ and NO₃⁻, which can be related to secondary sulphate (Factor 3) and secondary nitrate (Factor 4). Group 5 had no prominent high-level chemical species, might be the unknown sources. The FA explains 84% of the total variance of the PM₁₀ dataset, so the residual 16% may be represented by Group 5, which in turn may be interpreted as background pollution.

Additionally, the results of FCA can show the source changes between 2010 and 2011 for the different regions. The samples clustered in Group 1 were mostly from the commercial and residential regions in 2010, while those in Group 2 were from the commercial and residential regions in 2011, showing the changes of the important source categories between the two years. These changes may be due to the yellow-tag vehicles being banned from the road and to building construction at the commercial and residential regions in 2011. For Group 2, most of the samples were from industrial regions in 2010 and 2011, indicating that the important source categories may not vary in industrial regions. The samples belonging to Group 4 were relatively complex, including the commercial, residential and industrial regions in 2010 and 2011. The emitted precursor gas NO_x may influence the samples in Group 4. For Group 5, samples from the background and residential regions were included.

The results of the FCA for PM_{2.5} are shown in Table 3. Three factors were identified by FA. The first factor (39% of the total variance), with a high loading of Al, Si, Fe, TC and NO₃⁻, can be explained as mixed sources, including crustal dust, vehicle exhaust and secondary nitrate. The second factor (20% of the total variance) can be associated with secondary sulphate due to the high level of SO₄²⁻, and

Table 2. The rotated factor loadings and concentrations ($\mu\text{g}/\text{m}^3$) of PM_{10} and individual chemical species for each group

Factor1	Factor2	Factor3	Factor4	Group1	Group2	Group3	Group4	Group5
				Mixed Source (Factor 1) n = 19 $\mu\text{g}/\text{m}^3$	Cement Dust (Factor 2) n = 24 $\mu\text{g}/\text{m}^3$	Secondary Sulfate (Factor 3) n = 15 $\mu\text{g}/\text{m}^3$	Secondary Nitrate (Factor 4) n = 33 $\mu\text{g}/\text{m}^3$	Unknown source n = 35 $\mu\text{g}/\text{m}^3$
PM_{10}				352.57 $\pm$ 110.70	186.05 $\pm$ 61.64	288.27 $\pm$ 47.60	257.06 $\pm$ 77.94	196.26 $\pm$ 7.97
Na	0.80			4.7 $\pm$ 1.28	2.87 $\pm$ 1.05	3.76 $\pm$ 0.63	3.06 $\pm$ 1.24	2.37 $\pm$ 0.92
Mg	-0.11		0.20	2.5 $\pm$ 1.42	6 $\pm$ 2.69	2.63 $\pm$ 2.78	1.7 $\pm$ 1.48	0.62 $\pm$ 0.48
Al	0.87		0.05	18.38 $\pm$ 7.15	9.9 $\pm$ 2.57	13.6 $\pm$ 2.29	12.77 $\pm$ 3.97	11.27 $\pm$ 3.83
Si	0.82		0.15	37.13 $\pm$ 12.34	22.09 $\pm$ 7.1	30.83 $\pm$ 6.42	28.5 $\pm$ 10.52	23.61 $\pm$ 7.85
K	0.90		0.22	5.63 $\pm$ 2.36	1.16 $\pm$ 0.58	2.73 $\pm$ 1.71	2.46 $\pm$ 1.75	2.02 $\pm$ 1.39
Ca	0.34		0.00	16.86 $\pm$ 6.59	18.18 $\pm$ 6.91	11.61 $\pm$ 4.22	9.55 $\pm$ 5.95	5.5 $\pm$ 3.12
Mn	0.83		0.05	0.29 $\pm$ 0.1	0.15 $\pm$ 0.06	0.15 $\pm$ 0.05	0.14 $\pm$ 0.08	0.1 $\pm$ 0.04
Fe	0.90		0.03	8.53 $\pm$ 2.98	4.16 $\pm$ 1.66	6.6 $\pm$ 1.18	5.66 $\pm$ 2.1	4.25 $\pm$ 1.45
Zn	0.75		0.18	1.52 $\pm$ 0.93	0.67 $\pm$ 0.7	0.77 $\pm$ 0.38	0.77 $\pm$ 0.58	0.52 $\pm$ 0.25
TC	0.84		0.10	83.31 $\pm$ 36.2	33.05 $\pm$ 13.01	62.98 $\pm$ 19.95	52.87 $\pm$ 19.99	36.67 $\pm$ 16.29
NO_3^-	0.21		0.08	12.14 $\pm$ 7.92	14.99 $\pm$ 8.49	17.9 $\pm$ 6.46	28.42 $\pm$ 11.06	10.64 $\pm$ 4.4
SO_4^{2-}	0.36		0.96	56.13 $\pm$ 33.13	37.34 $\pm$ 24.77	102.87 $\pm$ 30.34	52.41 $\pm$ 21.46	29.86 $\pm$ 11.54
Variation (%)	49	16	9

Fig. 5. Hierarchical clustering of factor scores, for PM_{10} samples.

Table 3. The rotated factor loadings and concentrations ($\mu\text{g}/\text{m}^3$) of $\text{PM}_{2.5}$ and individual chemical species for each group.

	Factor 1	Factor 2	Factor 3	Group 1	Group 2	Group 3	Group 4
	Mixed source	Secondary sulfate	Cement dust	Unknown source 1 $n = 52$ $\mu\text{g}/\text{m}^3$	Mixed Source (Factor 1 and 2) $n = 44$ $\mu\text{g}/\text{m}^3$	Cement Dust (Factor 3) $n = 27$ $\mu\text{g}/\text{m}^3$	Unknown source 2 $n = 5$ $\mu\text{g}/\text{m}^3$
PM_{10}				119.03 ± 31.64	214.79 ± 64.76	150.80 ± 67.46	131.05 ± 42.56
Na	0.78	0.31	0.38	0.77 ± 0.24	1.38 ± 0.46	1.32 ± 0.54	0.77 ± 0.24
Mg	0.05	-0.10	0.93	0.44 ± 0.41	0.40 ± 0.37	2.79 ± 1.07	0.11 ± 0.06
Al	0.84	0.39	0.03	4.03 ± 1.08	7.74 ± 2.73	5.57 ± 2.38	5.18 ± 1.75
Si	0.84	0.30	0.12	9.31 ± 3.73	16.43 ± 5.43	12.48 ± 5.33	10.36 ± 3.37
K	0.57	0.65	-0.02	0.92 ± 0.50	2.00 ± 1.22	1.31 ± 1.15	1.46 ± 0.65
Ca	0.00	0.28	0.78	2.33 ± 1.56	2.02 ± 1.56	6.89 ± 3.66	0.68 ± 0.41
Mn	0.13	0.18	0.20	0.09 ± 0.12	0.11 ± 0.07	0.16 ± 0.18	0.05 ± 0.02
Fe	0.84	0.45	0.07	1.50 ± 0.44	2.65 ± 0.84	1.96 ± 0.97	1.82 ± 0.90
Zn	0.24	0.72	0.15	0.50 ± 0.34	0.77 ± 0.64	0.72 ± 1.16	0.37 ± 0.20
TC	0.71	0.47	-0.15	32.52 ± 10.75	64.01 ± 22.71	36.90 ± 19.77	27.85 ± 8.53
NO_3^-	0.87	-0.05	-0.12	10.00 ± 4.43	21.49 ± 15.78	11.34 ± 5.35	6.61 ± 6.16
SO_4^{2-}	0.50	0.72	0.09	23.34 ± 9.69	49.65 ± 22.75	36.31 ± 24.00	21.75 ± 7.97
Eiengvalue	4.64	2.36	1.75				
Variation(%)	39	20	15				

the third factor (15% of the total variance) may be the cement dust, as indicated by the high level of Ca. Additionally, similar to the results of PM₁₀, Magnesium alloy industrial manufacture source might be included in this factor. Four groups were discriminated by FCA, as shown in Fig. 6. The concentration of PM_{2.5} and the individual chemical species for each group were calculated accordingly and are listed in Table 3. According to Table 3, Groups 1 and 4 may be explained as background pollution due to the lack of outstanding species; So Group 1 and Group 4 might be unknown sources. Group 2, with high fractions (%) of Al, Si, Fe, TC, NO₃⁻ and SO₄²⁻, may be a mixture of factors 1 and 2; and Group 3 may be related to factor 3 due to high concentration of Ca. Concerning the origins of the samples included in each group, the samples in Group 1 were from the commercial and residential regions in 2010 and 2011 as well as the background in 2011; the samples in Group 2 were mainly from the commercial and residential regions in 2010 as well as the industrial regions in 2010 and 2011; the samples in Group 3 were from the commercial and residential regions in 2011; and the samples in Group 4 were from the background in 2010. Overall, the result of PM_{2.5} was similar to that of PM₁₀ for FCA, and the conclusions drawn from the FCA are in agreement with the discussion above.

CONCLUSIONS

In this study, PM₁₀ and PM_{2.5} samples were collected from four functional regions in Chengdu, China, during 2010 and 2011. The changes of the concentrations of PM and their major constituents were analysed to evaluate the effects of pollution control strategies. The following can be concluded. The strategy of banning high-emission yellow-tag vehicles was the most important reason for the TC reduction. The reduction of SO₂ and NO_x emissions played a key role in the decrease of sulphate and nitrate. The reduction of crustal dust may be attributed from the meteorological factor (long-range transport). Finally, FCA was employed to analyze the changes of possible source categories for the different years. The results showed that for commercial and residential regions, vehicle emissions may have been the important source in 2010, while their influence may have been relatively weaker in 2011; and for industrial and background regions, the dominant source categories may not have changed.

ACKNOWLEDGMENTS

This study is supported by Natural Science Foundation of China (21207070, 41205089, 41375132), Special Funds for Research on Public Welfares of the Ministry of Environmental Protection of China (201409003), and the Combined Laboratory of the Tianjin Meteorological Bureau and Nankai University.

SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

Fig. 6. Hierarchical clustering of factor scores, for PM_{2.5} samples.

REFERENCES

- Amato, F. and Hopke, P.K. (2012). Source Apportionment of the Ambient PM_{2.5} across St. Louis Using Constrained Positive Matrix Factorization. *Atmos. Environ.* 46: 329–337.
- Andrews, E., Saxena, P., Musarra, S., Hildemann, L.M., Koutrakis, P., McMurry, P.H., Olmez, I. and White, W.H. (2000). Concentration and Composition of Atmospheric Aerosols from the 1995 Seavs Experiment and a Review of the Closure between Chemical and Gravimetric Measurements. *J. Air Waste Manage. Assoc.* 50: 648–664.
- Baldwin, D.P., Zamzow, D.S. and D'Silva, A.P. (1994). Aerosol Mass Measurement and Solution Standard Additions for Quantization in Laser Ablation–Inductively Coupled Plasma Atomic Emission Spectrometry. *Anal. Chem.* 66: 1911–1917.
- Begum, B.A., Biswas, S.K., Markwitz, A. and Hopke, P.K. (2010). Identification of Sources of Fine and Coarse Particulate Matter in Dhaka, Bangladesh. *Air Aerosol Qual. Res.* 10: 345–353.
- Bellouin, N., Boucher, O., Haywood, J. and Reddy, M.S. (2005). Global Estimate of Aerosol Direct Radiative Forcing from Satellite Measurements. *Nature* 438: 1138–1141.
- Bi, X.H., Feng, Y.C., Wu, J.H. and Wang, Y.Q. (2007). Source Apportionment of PM₁₀ in Six Cities of Northern China. *Atmos. Environ.* 41: 903–912.
- Cheng, Y.H., Chang, H.P. and Yan, J.W. (2012). Temporal Variations in Airborne Particulate Matter Levels at an Indoor Bus Terminal and Exposure Implications for Terminal Workers. *Air Aerosol Qual. Res.* 12: 30–38.
- Chow, J.C., Watson, J.G., Pritchett, L.C., Pierson, W.R., Frazier, C.A. and Purcell, R.G. (1993). The DRI Thermal/Optical Reflectance carbon analysis system: Description, evaluation and applications in U.S. air quality studies. *Atmos. Environ.* 27A: 1185–1201.
- Ehrlich, C., Noll, G., Kalkoff, W.D., Baumbach, G. and Dreiseidler, A. (2007). PM₁₀, PM_{2.5}, and PM_{10-2.5}–Emissions, from Industrial Plants–Results from Measurement Programmes in Germany. *Atmos. Environ.* 44: 6236–6254.
- Escrig, A., Monfort, E., Celades, I., Querol, X., Amato, F., Minguillón, M.C. and Hopke, P.K. (2009). Application of Optimally Scaled Target Factor Analysis for Assessing Source Contribution of Ambient PM₁₀. *J. Air Waste Manage. Assoc.* 59: 1296–1307.
- Gianini, M.F.D., Gehrig, R., Fischer, A., Ulrich, A., Wichser, A. and Hueglin, C. (2012). Chemical Composition of PM₁₀ in Switzerland: An Analysis for 2008/2009 and Changes Since 1998/1999. *Atmos. Environ.* 54: 97–106.
- Harrison, R.M. and Yin, J. (2000). Particulate Matter in the Atmosphere: Which Particle Properties are Important for Its Effects on Health? *Sci. Total Environ.* 249: 85–101.
- He, K., Yang, F., Ma, Y., Zhang, Q., Yao, X., Chan, C.K., Cadle, S., Chan, T. and Mulawa, P. (2001). The Characteristics of PM_{2.5} in Beijing, China. *Atmos. Environ.* 35: 4959–4970.
- Hedberg, E., Gidhagen, L. and Johanson, C. (2005). Source Contributions to PM₁₀ And Arsenic Concentrations in Central Chile Using Positive Matrix Factorization. *Atmos. Environ.* 39: 549–561.
- Heinberg, R. and Fridley D. (2010). The End of Cheap Coal. *Nature* 468: 367–369.
- Ho, K.F., Lee, S.C., Cao, J.J., Chow, J.C., Watson, J.G. and Chan, C.K. (2006). Seasonal Variations and Mass Closure Analysis of Particulate Matter in Hong Kong. *Sci. Total Environ.* 355: 276–287.
- Hwang, I. and Hopke, P.K. (2007). Estimation of Source Apportionment and Potential Source Locations of PM_{2.5} at a West Coastal IMPROVE Site. *Atmos. Environ.* 41: 506–518.
- Li, X.R., Wang, L.L., Wang, Y.S., Wen, T.X., Yang, Y.J., Zhao, Y.A. and Wang, Y.F. (2012). Chemical Composition and Size Distribution of Airborne Particulate Matters in Beijing during the 2008 Olympics. *Atmos. Environ.* 50: 278–286.
- Lim, J.M., Lee, J.H., Moon, J.H. and Chung, Y.S. (2010). Source Apportionment of PM₁₀ at a Small Industrial Area Using Positive Matrix Factorization. *Atmos. Res.* 95: 88–100.
- Lin, P., Huang, X.F., He, L.Y. and Yu, J.Z. (2010). Abundance and Size Distribution of HULIS in Ambient Aerosols at a Rural Site in South China. *J. Aerosol Sci.* 41: 74–87.
- Masiol, M., Rampazzo, G., Ceccato, D., Squizzato, S. and Pavoni, B. (2010). Characterization of PM₁₀ Sources in a Coastal Area near Venice (Italy): An Application of Factor-cluster Analysis. *Chemosphere* 80: 771–778.
- Masiol, M., Squizzato, S., Ceccato, D., Rampazzo, G. and Pavoni, B. (2012). A Chemometric Approach to Determine Local and Regional Sources of PM₁₀ and Its Geochemical Composition in a Coastal Area. *Atmos. Environ.* 54: 127–133.
- Minguillón, M.C., Shembari, A., Triguero-Mas, M., de Nazelle, A., Dadvand, P., Figueras, F., Salvado, J.A., Grimalt, J.O., Nieuwenhuijsen, M. and Querol, X. (2012). Source Apportionment of Indoor, Outdoor and Personal PM_{2.5} Exposure of Pregnant Women in Barcelona, Spain. *Atmos. Environ.* 59: 426–436.
- Moorhouse, S. (2007). US Air Pollution is Harmful and Fine Particles Can Kill. *Nature* 445: 709–709.
- Morishita, M., Keeler, G.J., Kamal, A.S., Wagner, J.G., Harkema, J.R., and Rohr, A.C. (2011). Identification of Ambient PM_{2.5} Sources and Analysis of Pollution Episodes in Detroit, Michigan Using Highly Time-resolved Measurements. *Atmos. Environ.* 45: 1627–1637.
- Murillo, J.H., Ramos, A.C., García, F.Á., Jiménez, S.B., Cárdenas, B. and Mizohata, A. (2012). Chemical Composition of PM_{2.5} Particles in Salamanca, Guanajuato Mexico: Source Apportionment with Receptor Models. *Atmos. Res.* 107: 31–41.
- Niyobuhungiro, R.V. and Blottnitz, H.V. (2013). Investigation of Arsenic Airborne in Particulate Matter around Caterers' Wood Fires in the Cape Town Region. *Air Aerosol Qual. Res.* 13: 219–224.
- Oh, M.S., Lee, T.J. and Kim, D.S. (2011). Quantitative Source Apportionment of Size-segregated Particulate

- Matter at Urbanized Local Site in Korea. *Air Aerosol Qual. Res.* 11: 247–264.
- O’Ryan, R.E. (1996). Cost-Effective Policies to Improve Urban Air Quality in Santiago, Chile. *J. Environ. Manage.* 31: 302–313.
- Pant, P. and Harrison, R.M. (2012). Critical Review of Receptor Modelling for Particulate Matter: A Case Study of India. *Atmos. Environ.* 49: 1–12.
- Park, S.S. and Kim, Y.J. (2005). Source Contributions to Fine Particulate Matter in an Urban Atmosphere. *Chemosphere* 59: 217–226.
- Querol, X., Alastuey, A., Moreno, T., Viana, M.M., Castillo, S., Pey, J., Rodríguez, S., Artiñano, B., Salvador, P., Sánchez, M., Garcia Dos Santos, S., Herce Garraleta, M.D., Fernandez-Patier, R., Moreno-Grau, S., Negral, L., Minguillón, M.C., Monfort, E., Sanz, M.J., Palomo-Marín, R., Pinilla-Gil, E., Cuevas, E., de la Rosa, J. and Sánchez de la Campa, A. (2008). Spatial and Temporal Variations in Airborne Particulate Matter (PM₁₀ and PM_{2.5}) across Spain 1999–2005. *Atmos. Environ.* 42: 3964–3979.
- Shao, M., Wang, B., Lu, S., Yuan, B. and Wang, M. (2011). Effect of Beijing Olympics Control Measures on Reducing Reactive Hydrocarbon Species. *Environ. Sci. Technol.* 45: 514–519.
- Shi, G.L., Li, X., Feng, Y.C., Wang, Y.Q., Wu, J.H., Li, J. and Zhu, T. (2009). Combined Source Apportionment, Using Positive Matrix Factorization-Chemical Mass Balance and Principal Component Analysis/Multiplelinear Regression-Chemical Mass Balance Models. *Atmos. Environ.* 43: 2929–2937.
- Shi, G.L., Zeng, F., Li, X., Feng, Y.C., Wang, Y.Q., Liu, G.X. and Zhu, T. (2011). Estimated Contributions and Uncertainties of PCA/MLR–CMB Results: Source Apportionment for Synthetic and Ambient Datasets. *Atmos. Environ.* 45: 2811–2819.
- Shi, G.L., Tian, Y.Z., Han, S.Q., Zhang, Y.F., Li, X., Feng, Y.C., Wu, J.H. and Zhu, T. (2012). Vertical Characteristics of Carbonaceous Species and Their Source Contributions in a Chinese Mega City. *Atmos. Environ.* 60: 358–365.
- Shon, Z.H., Kim, K.H., Song, S.K., Jung, K., Kim, N.J. and Lee, J.B. (2012). Relationship between Water-soluble Ions in PM_{2.5} and Their Precursor Gases in Seoul Megacity. *Atmos. Environ.* 59: 540–550.
- Tahir, N.M., Suratman S., Fong, F.T., Hamzah, M.S. and Latif, M.T. (2013). Temporal Distribution and Chemical Characterization of Atmospheric Particulate Matter in the Eastern Coast of Peninsular Malaysia. *Air Aerosol Qual. Res.* 13: 584–595.
- Thurston, G.D., Ito, K. and Lall, R. (2011). A Source Apportionment of U.S. Fine Particulate Matter Air Pollution. *Atmos. Environ.* 45: 3924–3936.
- Tian, Y.Z., Xiao, Z.M., Han, B., Shi, G.L., Wang, W., Hao, H.Z., Li, X., Feng, Y.C. and Zhu, T. (2013). Seasonal Study of Primary and Secondary Sources of Carbonaceous Species in PM₁₀ from Five Northern Chinese Cities. *Air Aerosol Qual. Res.* 13: 148–161.
- Tian, Y.Z., Wu, J.H., Shi, G.L., Wu, J.Y., Zhang, Y.F., Zhou, L.D., Zhang, P. and Feng, Y.C. (2013). Long-term Variation of the Levels, Compositions and Sources of Size-resolved Particulate Matter in a Megacity in China. *Sci. Total Environ.* 463–464: 462–468.
- Tie, X., Wu, D. and Brasseur, G. (2009). Lung Cancer Mortality and Exposure to Atmospheric Aerosol Particles in Guangzhou, China. *Atmos. Environ.* 43: 2375–2377.
- Tiwari, S., Chate, D.M., Pragma, P., Ali, K. and Bisht, D.S. (2012). Variations in Mass of the PM₁₀, PM_{2.5} and PM₁ during the Monsoon and the Winter at New Delhi. *Air Aerosol Qual. Res.* 12: 20–29.
- Vernile, P., Tutino, M., Bari, G., Amodio, M., Spagnuolo, M., Gennaro, G.D. and Lillo, E.D. (2013). Particulate Matter Toxicity Evaluation Using Bioindicators and Comet Assay. *Air Aerosol Qual. Res.* 13: 172–178.
- Wang, G.H., Wang, H., Yu, Y.J., Gao, S.X., Feng, J.F., Gao, S.T. and Wang, L.S. (2003). Chemical Characterization of Water-soluble Components of PM₁₀ and PM_{2.5} Atmospheric Aerosols in Five Locations of Nanjing, China. *Atmos. Environ.* 37: 2893–2902.
- Wang, S.X. and Hao, J.M. (2012). Air Quality Management in China: Issues, Challenges, and Options. *J. Environ. Sci.* 24: 2–13.
- Wang, W., Primbs, T., Tao, S. and Simonich, S.L.M. (2009a). Atmospheric Particulate Matter Pollution during the 2008 Beijing Olympics. *Environ. Sci. Technol.* 43: 5314–5320.
- Wang, Z.S., Wu, T., Shi, G.L., Fu, X., Tian, Y.Z., Feng, Y.C., Wu, X.F., Wu, G., Bai, Z.P. and Zhang, W.J. (2012). Potential Source Analysis for PM₁₀ and PM_{2.5} in Autumn in a Northern City in China. *Air Aerosol Qual. Res.* 12: 39–48.
- Watson, J.G., Chow, J.C., Lowenthal, D.H., Pritchett, L.C., Frazier, C.A., Neuroth, G.R. and Robbins, R. (1994). Differences in the Carbon Composition of Source Profiles for Diesel- and Gasoline-powered Vehicles. *Atmos. Environ.* 28: 2493–2505.
- Wongphatarakul, V., Friedlander, S.K. and Pinto, J.P. (1998). A Comparative Study of PM_{2.5} Ambient Aerosol Chemical 614 Databases. *Environ. Sci. Technol.* 32: 3926–3934.
- Wu, L., Feng, Y.C., Wu, J.H., Zhu, T., Bi, X.H., Han, B., Yang, W.H. and Yang, Z.Q. (2009). Secondary Organic Carbon Quantification and Source Apportionment of PM₁₀ in Kaifeng, China. *J. Environ. Sci.-China* 21: 1353–1362.
- Xu, L.L., Chen, X.Q., Chen, J.S., Zhang, F.W., He, C., Du, K. and Wang, Y. (2012). Characterization of PM₁₀ Atmospheric Aerosol at Urban and Urban Background Sites in Fuzhou city, China. *Environ. Sci. Pollut. Res. Int.* 19: 1443–1453.
- Zeng, F., Shi, G.L., Li, X., Feng, Y.C., Bi, X.H., Wu, J.H. and Xue, Y.H. (2010). Application of a Combined Model to Study the Source Apportionment of PM₁₀ in Taiyuan, China. *Air Aerosol Qual. Res.* 10: 177–184.
- Zhang, Z. and Friedlander, S.K. (2000). A Comparative Study of Chemical Databases for Fine Particle Chinese Aerosols. *Environ. Sci. Technol.* 34: 4687–4697.
- Zhao, C., Tie, X.X. and Lin, Y.P. (2006). Reduction of Precipitation and Increase in Aerosols; a Positive

Feedback in Eastern-Central China. *Geophys. Res. Lett.* 33: L11814.

Environ. 39: 3967–3976.

Zheng, M., Salmon, L.G., Schauer, J.J., Zeng, L., Kiang, C.S., Zhang, Y. and Cass, G.R. (2005). Seasonal Trends in PM_{2.5} Source Contributions in Beijing, China. *Atmos.*

Received for review, April 2, 2013

Accepted, September 23, 2013

Supplementary Materials

Evaluation of the Changes of the Concentration, Composition and Possible Sources of Size-Resolved Particulate Matter between 2010 and 2011 in a Western Chinese Mega City

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Table S1. PM and chemical species in other cities.

City	PM	$\mu\text{g}/\text{m}^3$	Period	Reference
Chengdu	PM ₁₀	279.39 ± 99.95	2010	This study
Chengdu	PM ₁₀	200.62 ± 69.18	2011	This study
Taiyuan	PM ₁₀	305.46 ± 192.17	2001–2002	Zeng <i>et al.</i> (2010)
Nanjing	PM ₁₀	316.20 ± 162.12	2001	Wang <i>et al.</i> (2003)
Fuzhou ^a	PM ₁₀	69.92 ± 30.10	2008 Winter	Xu <i>et al.</i> (2012)
Hong Kong ^b	PM ₁₀	55.75 ± 23.84	2000–2001 Winter	Ho <i>et al.</i> (2006)
Korea	PM ₁₀	81.0 ± 41.0	2002	Lim <i>et al.</i> (2010)
Spain ^c	PM ₁₀	50	1999–2005	Querol <i>et al.</i> (2008)
Switzerland ^d	PM ₁₀	29.4	2008–2009	Giannini <i>et al.</i> (2012)
Chengdu	PM _{2.5}	178.75 ± 80.52	2010	This Study
Chengdu	PM _{2.5}	134.62 ± 46.09	2011	This Study
Nan Jing	PM _{2.5}	222.20 ± 103.56	2001	Wang <i>et al.</i> (2003)
Beijing ^e	PM _{2.5}	175.9	1999–2000	He <i>et al.</i> (2001)
Seoul	PM _{2.5}	41.8 ± 3.30	1998–1999	Park <i>et al.</i> (2005)
Hong Kong	PM _{2.5}	42.59 ± 21.03	2000–2001 Winter	Ho <i>et al.</i> (2006)
Spain ^c	PM _{2.5}	35	1999–2005	Querol <i>et al.</i> (2008)
US	PM _{2.5}	14.2 ± 0.2	2000–2005	Thurston <i>et al.</i> (2011)

^a Urban - Ziyang site.^b KT site.^c Traffic - L'Hospital site.^d Bern site.^e Chegongzhuang site.

Table S2. Average concentration ratio between PM_{2.5} and PM₁₀.

	2010 PM _{2.5} /PM ₁₀				2011 PM _{2.5} /PM ₁₀			
	Background	Commercial	Residential	Industrial	Background	Commercial	Residential	Industrial
Mass conc.	0.72	0.59	0.63	0.64	0.80	0.58	0.62	0.73
Na	0.44	0.30	0.29	0.34	0.52	0.34	0.34	0.38
Mg	0.50	0.15	0.36	0.38	0.53	0.27	0.40	0.45
Al	0.40	0.48	0.42	0.40	0.45	0.48	0.47	0.46
Si	0.47	0.35	0.49	0.40	0.55	0.34	0.49	0.56
K	0.51	0.45	0.51	0.52	0.53	0.63	0.58	0.89
Ca	0.24	0.19	0.37	0.25	0.32	0.20	0.50	0.30
Ti	0.44	0.45	0.45	0.41	0.31	0.43	0.53	0.34
V	0.70	0.69	0.67	0.64	0.89	0.57	0.62	0.78
Cr	0.73	0.35	0.57	3.72	0.95	0.64	0.63	0.50
Mn	0.65	0.55	1.24	0.62	0.76	0.89	0.59	0.73
Fe	0.50	0.32	0.35	0.32	0.51	0.31	0.35	0.47
Co	0.48	0.46	0.43	0.40	0.69	0.74	0.38	0.31
Ni	0.80	0.68	0.82	0.80	0.59	0.67	0.60	0.44
Cu	0.64	0.74	0.42	0.39	0.46	0.63	0.48	0.52
Zn	0.82	0.95	0.66	0.95	0.79	0.80	0.71	0.91
As	0.74	0.47	0.72	0.51	0.48	0.69	0.66	0.82
Cd	0.66	0.61	0.81	0.59	0.81	0.73	0.67	0.56
Hg	0.44	0.31	0.55	0.66	0.41	0.43	0.74	0.42
Pb	0.85	0.70	0.91	0.74	0.84	0.67	0.74	0.44
TC	0.90	0.80	0.91	0.87	0.92	0.82	0.97	0.94
NO ₃ ⁻	0.91	0.74	0.88	0.76	0.94	0.63	0.72	0.67
SO ₄ ²⁻	0.87	0.79	0.67	0.61	0.91	0.66	0.72	0.70

Table S3. The related reported data for Chengdu, in 2009 and 2010.

	2009	2010	Relative change (%)
Population (million)	11.4	11.5	0.88
Annual total industrial coal consumption (thousand tons)	8690	7957	−8.43
Annual total industrial SO ₂ emission (thousand tons)	127	96.8	−23.78
Annual total industrial NO _x emission (thousand tons)	57.8	52.4	−9.34
Vehicle population (million)	2.36	2.59	9.75

The reported data was referred to the Chengdu Statistical Yearbook 2009, 2010.

The relative change (%) was calculated according to:

$$[Relative\ change] = ([2010] - [2009]) / [2009] \times 100$$

where, *[2010]* is the data in 2010 and *[2009]* is the data in 2009

References

Bureau of Statistics of Chengdu. Chengdu Statistical Yearbook, 2009, 2010 (in Chinese).

Environmental Monitoring Center of Chengdu). Chengdu Environmental Quality Reports, 2006-2010 (in Chinese).

Table S4. The pollution control strategies.

Strategies		time
Strategy 1	Continuing reduction of SO ₂ emission	Target: 10% every year
Strategy 2	Yellow-tag vehicles banned from the road	High-emission yellow-tag vehicles have been banned from roads (from 7:30 to 19:30) in urban areas since 1 Oct. 2010
Strategy 3	Other pollution control strategies	Forbidding the lawless industrial emission; Updating the outdated environmental protection technology in factories

Fig. S1. Map of monitoring areas.

Annotate:

BaiGongYan (BGY: Background): 30°34'N, 104°17'E;

HuanBaoDaLou (HBDL: Commercial): 30°39'N, 104°02'E;

KeJiYuan (KJY: Industrial): 30°37'N, 104°03'E;

SanWaYao (SWY: Residential): 30°36'N, 104°04'E.

Fig. S2. Fractions (%) of chemical species in size resolved PM from different regions, in 2010 and 2011.

Fig. S3(a). Ternary plots for the fractions of crustal dust, TC and sulfate + nitrate in PM_{10} from different regions.

Fig. S3(b). Ternary plots for the fractions of crustal dust, TC and sulfate + nitrate in $PM_{2.5}$ from different regions

Fig. S4(a). 3D 72-h back trajectories cluster maps from 1 Jan. to 28 Feb. 2010 for Chengdu.

Fig. S4(b). 3D 72-h back trajectories cluster maps from 1 Jan. to 28 Feb. 2011 for Chengdu