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TECHNICAL PAPER

Winter and Summer $PM_{2.5}$ Chemical Compositions in Fourteen Chinese Cities

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$PM_{2.5}$ in 14 of China's large cities achieves high concentrations in both winter and summer with averages >100 μg m^{-3} being common occurrences. A grand average of 115 μg m^{-3} was found for all cities, with a minimum of 27 μg m^{-3} measured at Qingdao during summer and a maximum of 356 μg m^{-3} at Xi'an during winter. Both primary and secondary $PM_{2.5}$ are important contributors at all of the cities and during both winter and summer. While ammonium sulfate is a large contributor during both seasons, ammonium nitrate contributions are much larger during winter. Lead levels are still high in several cities, reaching an average of 1.68 μg m^{-3} in Xi'an. High correlations of lead with arsenic and sulfate concentrations indicate that much of it derives from coal combustion, rather than leaded fuels, which were phased out by calendar year 2000. Although limited fugitive dust markers were available, scaling of iron by its ratios in source profiles shows ∼20% of $PM_{2.5}$ deriving from fugitive dust in most of the cities. Multipollutant control strategies will be needed that address incomplete combustion of coal and biomass, engine exhaust, and fugitive dust, as well as sulfur dioxide, oxides of nitrogen, and ammonia gaseous precursors for ammonium sulfate and ammonium nitrate.

Implications: $PM_{2.5}$ mass and chemical composition show large contributions from carbon, sulfate, nitrate, ammonium, and fugitive dust during winter and summer and across fourteen large cities. Multipollutant control strategies will be needed that address both primary $PM_{2.5}$ emissions and gaseous precursors to attain China's recently adopted $PM_{2.5}$ national air quality standards.

Introduction

Suspended particulate matter (PM) is the major pollutant in many Chinese cities (Chan and Yao, 2008; Tie and Cao, 2009). Coal combustion to generate electricity and for domestic cooking and heating constitutes ∼70% of the national energy budget (NAE et al., 2008). Total biomass burning in China, which includes domestic cooking and residential heating, field burning of crop residue, forest fires, and grassland fires, is estimated at 511.3 Tg yr^{-1} (Yan et al., 2006). Improved engines and tighter emission standards are being offset by rapid growth in the motor vehicle fleet (Han and Hayashi, 2008). Paved and unpaved roads, construction, agricultural operations, and wind-blown soil eject geological material into the atmosphere (Du et al., 2008; Xuan et al., 2004). These and other emitters are contributing to high PM levels in Chinese cities, both through direct PM emissions and through conversion of sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3), and volatile organic compound (VOC) gases to secondary sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), and organic carbon (OC).

The Chinese government issued a national $PM_{2.5}$ standard on February 29, 2012, that requires cities to have concentrations below 35 μg m^{-3} annual average and <75 μg m^{-3} for 24 hr, beginning in 2016 (http://cleanairinitiative.org/portal/node/8163). These standards were adopted owing to recognized adverse effects of $PM_{2.5}$ chemical components on human health, visibility, and materials (Hu et al., 2009; Mauderly and Chow, 2008; Pope and Dockery, 2006; Watson, 2002). Elements, ions, and carbon fractions are often measured in $PM_{2.5}$ to better evaluate the adverse effects and to indicate contributing sources. Several studies have reported these measurements in China (Cao et al., 2011; Chow et al., 2006; Deng et al., 2011; Duan et al., 2006; Gu et al., 2011; Guinot et al., 2007; He et al., 2001; Ho et al., 2006; Hu et al., 2010; Louie et al., 2005; Louie et al., 2005; Shen et al., 2007; So et al., 2007; Song et al., 2007; Sun et al., 2004; Wang et al., 2006; Wang et al., 2007; Wu et al., 2003; Xu

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et al., 2004; Yang et al., 2011; Zhang et al., 2010; Zhang and Friedlander, 2000; Zhao et al., 2010), but the areas studied, sampling site zones of representation, sampling periods, variables measured, and analysis methods are of insufficient consistency to evaluate similarities and differences. Reported here are consistently characterized simultaneous winter and summer $PM_{2.5}$ mass and chemical concentrations obtained during 2003 at receptors with neighborhood and urban scale (Chow et al., 2002) in 14 of China's major cities. These measurements are used to compare and contrast the situation across a broad range of emissions and meteorology, examine seasonal changes, and assess contributions from coal combustion using elemental concentration ratios. These measurements from nearly a decade ago provide a baseline against which to evaluate future speciated $PM_{2.5}$ measurements that will be needed to create and evaluate the multipollutant (Chow and Watson, 2011) control strategies required to attain the national standards.

Materials and Methods

As shown in Figure 1, measurement sites were located in 14 economically developed and developing cities across China. The neighborhood- and urban-scale sites were located on the campuses of schools and research institutes, as previously described (Cao et al., 2007; Cao et al., 2011; Han et al., 2009; Ho et al., 2007; Wang et al., 2006). Filter samplers were located on rooftops at 6 to 20 m above ground level for around 2 weeks of sampling during winter (January 6–20) and summer (June 3–July 30) of 2003.

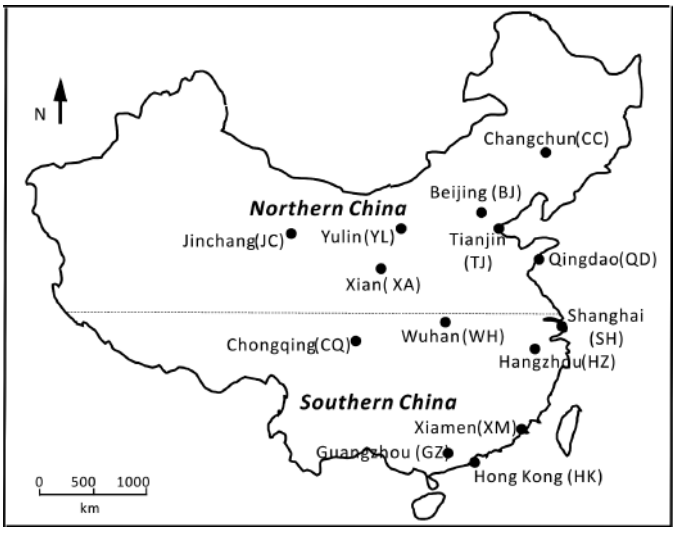

Figure 1. $PM_{2.5}$ samples were taken in seven southern China cities: Chongqing (CQ), Guangzhou (GZ), Hong Kong (HK), Hangzhou (HZ), Shanghai (SH), Wuhan (WH), and Xiamen (XM); and seven northern China cities: Beijing (BJ), Changchun (CC), Jinchang (JC), Qingdao (QD), Tianjin (TJ), Xi'an (XA), and Yulin (YL). Filter samples were obtained from 0900 to 0900 LST the next morning over 2-week periods during winter (January 6–20) and summer (June 3 – July 30) of 2003. Cities are classified as representing northern and southern China since: (1) precipitation events are more frequent and intense in southern China, and (2) northern China cities have lower wintertime temperatures, resulting in a greater amount of domestic heating, often using coal, along with shallower and more prolonged surface inversions at night and early morning.

$PM_{2.5}$ samples were obtained on prefired (900°C, 3 h) 47-mm Whatman QM-A quartz-fiber filters by mini-volume air samplers (Airmetrics, Eugene, OR) at 5 L min^{-1} flow rates. The exposed filters were stored at ~4°C after sampling, including shipping to the Xi'an laboratory, to minimize evaporation of volatile components. Filters were weighed before and after sampling with a ±1-μg sensitivity Sartorius MC5 electronic microbalance (Sartorius, Göttingen, Germany) after 24-hr equilibration at 20 to 23°C and 35 to 45% relative humidity (RH). Each filter was weighed at least three times before and after sampling. The maximum differences among the three repeated weights were less than 10 μg for blank filters and less than 20 μg for exposed filters. The collected PM was the difference between the average of exposed weights and the average of unexposed weights. Field blanks were also collected at each sampling site every seventh day by exposing filters in the sampler without drawing air through them; these were used to account for passive deposition or artefacts introduced between sample changing.

Elemental concentrations of Fe, Ti, Mn, Zn, As, Br, and Pb in filter deposits were determined by energy-dispersive x-ray fluorescence (ED-XRF) spectrometry (PANalytical Epsilon 5, Almelo, The Netherlands) (Chow and Watson, 2012; Watson et al., 2012). Other elements, such as Si, Ca, Al, and Mg, were not quantified owing to high and variable blank values on quartz-fiber filters and potential biases caused by absorption of low-energy x-rays from particles penetrating into the filter. XRF measurements on nine collocated Teflon-membrane and quartz-fiber filters from Xi'an were comparable for these elements, with correlations (r) ranging from 0.982 for Fe and Zn (with slopes of 1.054 and 1.062, respectively) to 0.915 for As (with slope of 1.204). Measurement precision was determined as the standard deviation of several analyses of the same samples, yielding ±7.6% for Fe, ±8.6% for Ti, ±12.5% for Mn, ±7.6% for Zn, ±23.5% for As, ±33.3% for Br, and ±7.9% for Pb at typical concentration levels. Instrumental detection limits are 24.0 ng m^{-3} for Fe, 14.0 ng m^{-3} for Ti, 25.0 ng m^{-3} for Mn, 24.0 ng m^{-3} for Zn, 26.0 ng m^{-3} for As, 9.0 ng m^{-3} for Br, and 21.0 ng m^{-3} for Pb based on the uncertainties of blank filter counts. Replicate measurements were taken for every eight samples, and no differences were found that exceeded the precision intervals.

Following XRF analysis, the filter was sectioned with a precision cutter and one-fourth was extracted in 10 mL of distilled deionized water; the extract was submitted to ion chromatographic (IC) analysis (Shen et al., 2008; Shen et al., 2009) for cations Na^+, NH_4^+, and K^+ and anions SO_4^{2-}, NO_3^-, and Cl^-. Detection limits were 4.6 μg L^{-1} for Na^+, 4.0 μg L^{-1} for NH_4^+, 10.0 μg L^{-1} for K^+, 0.5 μg L^{-1} for Cl^-, 15 μg L^{-1} for NO_3^-, and 20 μg L^{-1} for SO_4^{2-}. Reference materials (National Research Center for Certified Reference Materials, China) agreed with analyses values within ± 4%. One in 10 extracts was reanalyzed and none of the differences between these replicates exceeded precision intervals. Blank values were also subtracted from sample concentrations.

Organic carbon (OC) and elemental carbon (EC) were determined on a 0.5-cm^2 punch from each filter by a DRI model 2001 carbon analyzer (Atmoslytic, Inc., Calabasas, CA) following the IMPROVE thermal/optical reflectance (TOR) protocol (Cao

et al., 2003; Chow et al., 1993; Chow et al., 2007; Chow et al., 2011). This produced four OC fractions (OC1, OC2, OC3, and OC4 at 120, 250, 450, and 550°C, respectively, in a helium [He] atmosphere); OP (a pyrolyzed carbon fraction determined when reflected 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 550, 700, and 800°C, respectively, in a 2% O_2/98% He atmosphere). OC is defined as OC1 + OC2 + OC3 + OC4 + OP, and EC is defined as EC1 + EC2 + EC3 − OP.

Results and Discussion

$PM_{2.5}$ mass concentrations

Figure 2 shows the wide distribution of concentrations observed across seasons and cities. The grand average of 115 $\mu g\ m^{-3}$ is more than 3 times the annual standard, and the highest 24-hr value of 543.9 $\mu g\ m^{-3}$, found in Xi'an during winter, is more than 7 times the 24-hr standard. OC, SO_4^{2-} NO_3^-, NH_4^+, and EC are the most abundant species, all with averages exceeding 5 $\mu g\ m^{-3}$. Elemental averages are less than the averages for carbon and ions, with Fe having the highest average of 2.4 $\mu g\ m^{-3}$ at Chongqing during winter. Concentrations ranged over several orders of magnitude, with the range increasing as the average concentration decreased. This variability indicates large spatial and temporal differences across the network.

Table 1 summarizes winter and summer $PM_{2.5}$ averages for each city. Standard deviations are typically 25% to 50% of the averages, indicating that these averages are not highly influenced by extreme events. Standard errors (standard deviation divided by the square root of the number of samples) of the averages are in the range of 6% to 13%.

In every city except Beijing and Xiamen (no summer data), wintertime $PM_{2.5}$ exceeded those of summertime, in many cases by a factor of 2 or more. Seasonal averages for $PM_{2.5}$ mass were similar in Beijing, with a winter/summer ratio of 0.88, in contrast to the highest ratio of 4.9 at Qingdao, a coastal city in northern China. The lack of difference in the mass ratio for Beijing is partially due to the lack of change in the OC concentrations, which were 23.9 ± 12.4 $\mu g\ m^{-3}$ in winter, only 20% higher than summer. The winter/summer mass ratios for other cities are reflected in the major chemical component averages, which are 2 to 3 for OC, EC, and SO_4^{2-} in most cities, with NO_3^- and NH_4^+ showing even higher winter/summer differences.

Average wintertime $PM_{2.5}$ was lowest in Xiamen (74.2 $\mu g\ m^{-3}$) and highest in Xi'an (356.3 $\mu g\ m^{-3}$). $PM_{2.5}$ was higher at inland cities (e.g., Xi'an, Wuhan, and Chongqing), and lower at the coastal (e.g., Xiamen and Hong Kong) and desert (i.e., Jinchang) cities. For the summer samples, average $PM_{2.5}$ was lowest in Qingdao (27.3 $\mu g\ m^{-3}$), and highest in Beijing (131.6 $\mu g\ m^{-3}$).

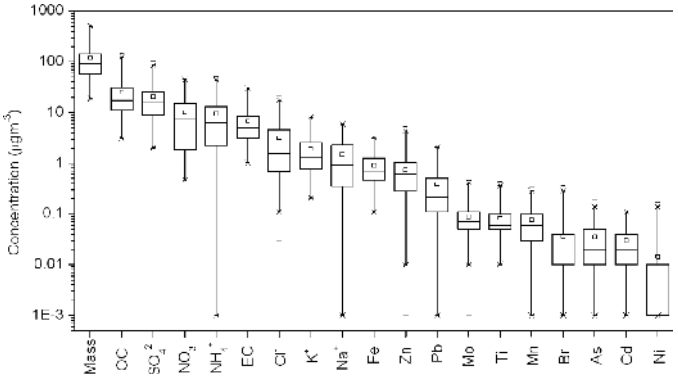

Figure 2. Average (square), median (central horizontal bar), 25th and 75th percentiles (lower and upper bars), 1st and 99th percentiles (lower and upper x), and minimum and maximum (—) concentrations for each chemical component across all cities and seasons. Average chemical components are ordered by abundance, with OC (24.5 $\mu g\ m^{-3}$), SO_4^{2-} (19.9 $\mu g\ m^{-3}$), NO_3^- (9.9 $\mu g\ m^{-3}$), NH_4^+ (9.2 $\mu g\ m^{-3}$), EC (6.5 $\mu g\ m^{-3}$), Cl^- (3.1 $\mu g\ m^{-3}$), K^+ (1. 9 $\mu g\ m^{-3}$), and Na^+ (1.5 $\mu g\ m^{-3}$) all being at important levels.

$PM_{2.5}$ composition

OC and EC exhibited winter maxima and summer minima. OC was the most abundant wintertime $PM_{2.5}$ constituent in all cities except Hangzhou and Hong Kong, ranging from 13.3 (Hong Kong) to 95.8 $\mu g\ m^{-3}$ (Xi'an). Wintertime EC levels vary with OC concentrations, which ranged from 4.6 (Jinchang) to 21.5 $\mu g\ m^{-3}$ (Xi'an). This co-occurrence is expected, as OC and EC typically result from incomplete combustion of solid and liquid fuels (Lighty et al., 2000). OC and EC concentrations were highest in the inland cities, such as Changchun, Xi'an, Wuhan, and Chongqing, and lower in the coastal cities, such as Qingdao, Xiamen, and Hong Kong.

Wintertime SO_4^{2-} was the second most abundant component of $PM_{2.5}$ for all the cities except Hong Kong, varying from 11.5 $\mu g\ m^{-3}$ in Jinchang to 60.9 $\mu g\ m^{-3}$ in Chongqing. This was followed by NO_3^-, ranging from 2.1 $\mu g\ m^{-3}$ (Jinchang) to 29 $\mu g\ m^{-3}$ (Xi'an), and NH_4^+ ranged from 6.6 $\mu g\ m^{-3}$ (Jinchang) to 29.8 $\mu g\ m^{-3}$ (Xi'an). These high secondary ammonium sulfate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3) levels imply the need for precursor gas, as well as primary PM, emission reductions to reduce $PM_{2.5}$ mass. The higher NH_4NO_3 values in winter than summer are consistent with a shift in equilibrium from the gas to particle phase with lower temperatures and higher RH (Stelson et al., 1979).

K^+ is considered a marker for biomass burning (Andreae, 1983; Duan et al., 2004), although it is also a component of certain soils and sea spray (Pytkowicz and Kester, 1971). Wintertime K^+ levels exceeded 3 $\mu g\ m^{-3}$ at Xi'an, Wuhan, Chongqing, and Hangzhou. The inland cities experience cold temperatures during winter and have abundant biomass available for residential heating.

Fe is a marker for fugitive dust, although it also originates from heavy industry. The wintertime Fe concentration was highest at 2.4 $\mu g\ m^{-3}$ in Chongqing, followed by Xi'an (1.8 $\mu g\ m^{-3}$), with the lowest wintertime average of 0.6 $\mu g\ m^{-3}$ at Xiamen. The two arid-region cities had low Fe concentrations, 1.2 $\mu g\ m^{-3}$ at Jinchang and 0.7 $\mu g\ m^{-3}$ at Yulin. The wintertime Fe averages did not correlate well with other soil components such as Ti and Mn across the sites, which may indicate additional Fe sources or variability in the fugitive dust compositions.

Table 1. Arithmetic averages ± standard deviations (μg m^{-3}) for $PM_{2.5}$ mass and chemical components by city and season. See Figure 1 for city codes. Each average contains ~14 values

City	Mass	OC	EC	Na^+	NH_4^+	K^+	Cl^-	NO_3^-	SO_4^{2-}	Ti	Mn	Fe	Zn	As	Br	Pb
	Winter	(June 6–20, 2003)														
Beijing (BJ)	115.6 ± 46.6	23.9 ± 12.4	6.2 ± 2.7	0.4 ± 0.4	9.4 ± 4.1	0.6 ± 0.4	2.3 ± 1.8	13.1 ± 4.5	20.0 ± 4.2	0.09 ± 0.03	0.07 ± 0.03	0.99 ± 0.32	0.39 ± 0.33	0.04 ± 0.04	0.01 ± 0.01	0.28 ± 0.23
Changchun (CC)	148.1 ± 21.3	40.4 ± 7.04	13.4 ± 3.2	1.4 ± 0.8	9.4 ± 2.1	1.1 ± 0.3	4.5 ± 2.5	11.7 ± 4.1	17.0 ± 5.1	0.17 ± 0.05	0.08 ± 0.03	1.25 ± 0.32	1.33 ± 0.46	0.09 ± 0.02	0.01 ± 0.01	0.46 ± 0.09
Jinchang (JC)	86.8 ± 14.2	17.6 ± 5.4	4.6 ± 1.1	1.4 ± 0.4	6.6 ± 1.4	0.8 ± 0.3	4.3 ± 1.3	2.1 ± 1.4	11.6 ± 2.6	0.15 ± 0.09	0.03 ± 0.02	1.19 ± 0.62	0.11 ± 0.02	0.05 ± 0.01	0.01 ± 0.01	0.16 ± 0.03
Qingdao (QD)	134.8 ± 43.0	26.3 ± 10.3	6.2 ± 2.5	3.2 ± 1.6	15.3 ± 5.2	2.8 ± 1.3	6.5 ± 2.2	19.3 ± 9.2	21.1 ± 7.7	0.09 ± 0.02	0.09 ± 0.05	0.95 ± 0.28	0.43 ± 0.23	0.02 ± 0.01	0.17 ± 0.12	0.27 ± 0.15
Tianjin (TJ)	203.1 ± 76.2	43.0 ± 16.1	8.9 ± 2.5	2.9 ± 2.0	22.2 ± 9.8	2.5 ± 1.0	11.4 ± 5.6	25.5 ± 10.3	32.5 ± 15.1	0.11 ± 0.03	0.17 ± 0.06	1.55 ± 0.74	1.34 ± 0.50	0.06 ± 0.02	0.08 ± 0.07	0.63 ± 0.28
Xi'an (XA)	356.3 ± 118.4	95.8 ± 27.7	21.5 ± 5.9	1.9 ± 0.7	29.8 ± 11.5	4.9 ± 1.3	9.5 ± 4.4	29.0 ± 10.0	53.8 ± 25.6	0.2 ± 0.09	0.19 ± 0.07	1.75 ± 0.52	2.41 ± 1.50	0.11 ± 0.04	0.07 ± 0.03	1.68 ± 0.60
Yulin (YL)	141.3 ± 56.0	31.3 ± 8.24	8.3 ± 3.0	3.8 ± 0.8	10.6 ± 3.5	1.4 ± 0.5	1.3 ± 1.5	10.9 ± 4.4	20.2 ± 7.5	0.04 ± 0.01	0.03 ± 0.01	0.66 ± 0.27	0.05 ± 0.03	0.01 ± 0.00	0 ± 0.01	0.07 ± 0.02
Chongqing (CQ)	316.6 ± 101.2	75.2 ± 21.9	17.2 ± 5.0	1.5 ± 1.0	28.8 ± 8.9	6.2 ± 2.4	10.8 ± 4.7	18.1 ± 6.4	60.9 ± 19.6	0.15 ± 0.02	0.18 ± 0.07	2.41 ± 0.75	0.92 ± 0.29	0.08 ± 0.03	0.14 ± 0.07	0.72 ± 0.26
Guangzhou (GZ)	110.2 ± 79.6	24.2 ± 22.1	8.4 ± 6.6	1.5 ± 1.6	8.5 ± 7.6	1.8 ± 1.2	1.9 ± 2.0	11.5 ± 7.8	20.6 ± 10.8	0.06 ± 0.03	0.06 ± 0.04	0.78 ± 0.51	0.59 ± 0.32	0.04 ± 0.02	0.06 ± 0.04	0.42 ± 0.26
Hong Kong (HK)	88.37 ± 23.14	13.3 ± 5.1	6.9 ± 2.8	0.6 ± 0.5	8.0 ± 3.5	0.8 ± 0.4	1.2 ± 1.5	9.5 ± 2.7	21.4 ± 5.6	0.05 ± 0.03	0.04 ± 0.03	0.62 ± 0.15	0.35 ± 0.19	0.01 ± 0.01	0.01 ± 0.01	0.19 ± 0.09
Hangzhou (HZ)	177.3 ± 59.5	30.5 ± 8.1	9.1 ± 2.1	1.2 ± 0.4	19.1 ± 10.7	4.4 ± 1.3	3.7 ± 1.8	25.7 ± 14.8	33.4 ± 16.7	0.09 ± 0.02	0.13 ± 0.04	1.35 ± 0.31	1.35 ± 0.43	0.04 ± 0.00	0.08 ± 0.05	0.75 ± 0.17
Shanghai (SH)	139.4 ± 50.6	26.7 ± 7.7	8.6 ± 2.0	1.9 ± 1.3	14.5 ± 5.9	2.1 ± 1.1	6.6 ± 3.1	17.5 ± 8.7	21.6 ± 12.3	0.09 ± 0.02	0.18 ± 0.07	1.18 ± 0.38	0.93 ± 0.39	0.03 ± 0.01	0.05 ± 0.01	0.48 ± 0.17
Wuhan (WH)	172.3 ± 67.0	33.8 ± 15.8	8.0 ± 3.4	1.7 ± 0.7	18.4 ± 10.2	3.4 ± 1.3	4.1 ± 1.2	22.2 ± 10.7	31.4 ± 15.6	0.08 ± 0.03	0.11 ± 0.05	1.56 ± 0.58	1.11 ± 0.48	0.08 ± 0.03	0.03 ± 0.02	1.1 ± 0.48
Xiamen (XM)	74.2 ± 27.2	15.5 ± 5.7	4.9 ± 1.5	0.8 ± 0.6	7.0 ± 3.9	0.9 ± 0.5	1.4 ± 0.5	7.6 ± 4.1	13.5 ± 8.4	0.06 ± 0.02	0.05 ± 0.01	0.6 ± 0.10	0.71 ± 0.31	0.02 ± 0.01	0.02 ± 0.01	0.22 ± 0.11

(*Continued*)

Table 1. (Cont.)

City	Mass	OC	EC	Na^+	NH_4^+	K^+	Cl^-	NO_3^-	SO_4^{2-}	Ti	Mn	Fe	Zn	As	Br	Pb
	Summer	(June 3–July 30, 2003)														
Beijing (BJ)	131.6 ± 28.0	19.7±4.7	5.7 ± 4.1	0.5 ± 0.3	9.8 ± 4.2	2.5 ± 0.6	1.3 ± 0.6	13.7 ± 6.4	22.6 ± 9.2	0.06 ± 0.02	0.06 ± 0.02	0.8 ± 0.24	0.44 ± 0.15	0.02 ± 0.01	0.02 ± 0.02	0.18 ± 0.09
Changchun (CC)	51.0 ± 19.8	10.6 ± 2.4	2.6 ± 0.8	0.2 ± 0.1	2.1 ± 2.5	0.9 ± 0.3	0.6 ± 0.2	2.7 ± 1.0	8.4 ± 6.6	0.05 ± 0.02	0.04 ± 0.02	0.55 ± 0.24	0.5 ± 0.20	0.01 ± 0.00	0.01 ± 0.00	0.07 ± 0.06
Jinchang (JC)	49.7 ± 2.3	7.1 ± 0.9	1.6 ± 0.2	3.6 ± 0.1	0.4 ± 0.5	0.7 ± 0.2	1.0 ± 0.2	0.9 ± 0.2	7.8 ± 2.5	0.25 ± 0.27	0.02 ± 0.02	0.58 ± 0.02	0.15 ± 0.11	0.05 ± 0.03	0.01 ± 0.00	0.8 ± 0.88
Qingdao (QD)	27.3 ± 11.0	5.1 ± 3.1	1.5 ± 0.7	0.2 ± 0.4	1.6 ± 1.4	0.4 ± 0.2	0.6 ± 0.7	1.8 ± 1.0	6.7 ± 3.1	0.03 ± 0.03	0.01 ± 0.01	0.26 ± 0.13	0.03 ± 0.01	0 ± 0.00	0.01 ± 0.00	0.03 ± 0.02
Tianjin (TJ)	101.7 ± 26.2	16.4 ± 3.9	3.8 ± 1.5	3.5 ± 1.7	7.7 ± 3.8	1.3 ± 0.5	3.8 ± 2.3	8.7 ± 3.4	22.1 ± 9.1	0.05 ± 0.02	0.05 ± 0.03	0.76 ± 0.31	0.6 ± 0.24	0.01 ± 0.01	0.04 ± 0.03	0.24 ± 0.13
Xi'an (XA)	102.8 ± 32.9	24.5 ± 6.7	6.5 ± 1.4	2.5 ± 1.9	4.3 ± 3.9	1.7 ± 0.4	1.1 ± 0.5	5.1 ± 2.8	16.8 ± 10.0	0.1 ± 0.03	0.07 ± 0.03	1.08 ± 0.48	1.22 ± 0.62	0.05 ± 0.04	0.01 ± 0.00	0.75 ± 0.49
Yulin (YL)	45.6 ± 8.5	10.6 ± 1.7	3.4 ± 1.3	4.1 ± 0.5	0.7 ± 1.0	0.5 ± 0.1	0.8 ± 0.2	1.5 ± 0.9	9.0 ± 3.9	0.06 ± 0.04	0.02 ± 0.01	0.41 ± 0.12	0.07 ± 0.02	0.01 ± 0.01	0.01 ± 0.00	0.07 ± 0.03
Chongqing (CQ)	106.9 ± 40.4	23.7 ± 8.2	7.4 ± 1.6	0.8 ± 0.3	7.0 ± 3.6	2.3 ± 0.7	2.2 ± 1.4	4.2 ± 3.0	20.4 ± 9.4	0.07 ± 0.03	0.1 ± 0.06	0.79 ± 0.40	0.67 ± 0.25	0.03 ± 0.01	0.04 ± 0.02	0.2 ± 0.07
Guangzhou (GZ)	39.7 ± 6.6	9.9 ± 1.3	3.3 ± 0.5	0.6 ± 0.8	1.2 ± 0.8	1.0 ± 0.5	0.8 ± 0.7	1.2 ± 0.4	6.4 ± 1.5	0.06 ± 0.01	0.02 ± 0.01	0.26 ± 0.07	0.66 ± 0.16	0.02 ± 0.01	0.01 ± 0.01	0.12 ± 0.05
Hong Kong (HK)	30.4 ± 7.3	6.6 ± 1.9	3.3 ± 1.1	0.72 ± 0.3	0.3 ± 0.2	0.6 ± 0.1	1.2 ± 0.4	1.2 ± 0.5	4.3 ± 1.2	0.04 ± 0.02	0.04 ± 0.04	0.27 ± 0.11	0.19 ± 0.15	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.02
Hangzhou (HZ)	80.0 ± 40.3	15.2 ± 3.5	3.2 ± 1.2	0.3 ± 0.4	5.3 ± 5.4	2.9 ± 1.1	0.6 ± 0.3	5.5 ± 7.8	16.5 ± 9.4	0.06 ± 0.01	0.05 ± 0.02	0.51 ± 0.14	1.12 ± 0.26	0.02 ± 0.01	0.01 ± 0.01	0.33 ± 0.17
Shanghai (SH)	54.0 ± 23.1	13.4 ± 5.4	3.3 ± 1.9	0.6 ± 0.1	3.5 ± 2.5	0.9 ± 0.4	0.6 ± 0.2	2.6 ± 2.1	12.0 ± 5.7	0.06 ± 0.02	0.06 ± 0.02	0.46 ± 0.28	0.78 ± 0.23	0.02 ± 0.01	0.02 ± 0.01	0.16 ± 0.11
Wuhan (WH)	70.7 ± 19.9	14.1 ± 3.7	2.7 ± 0.6	0.2 ± 0.2	2.9 ± 2.0	2.3 ± 1.2	0.5 ± 0.1	2.7 ± 1.0	12.4 ± 7.1	0.05 ± 0.01	0.08 ± 0.07	0.93 ± 0.28	0.69 ± 0.32	0.05 ± 0.04	0.01 ± 0.00	0.36 ± 0.25
Xiamen (XM)	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

Note: NA, data not available.

The highest wintertime As (0.11 $\mu g\ m^{-3}$) and Pb (1.7 $\mu g\ m^{-3}$) concentrations were found at Xi'an. As and Pb are found in Chinese coal (Tian et al., 2011; Want et al., 2006), while Pb gasoline additives were discontinued in 2000 (Xu et al., 2012). The highest Br average was found at a coastal city, Qingdao (0.17 $\mu g\ m^{-3}$), consistent with a potential marine aerosol contribution.

Summertime averages were lower than those for winter for nearly all chemical components. In most cases, this can be attributed to warmer weather that improved dispersion and shifted the NH_4NO_3 from the particle to gas phase. Lower OC and EC averages are probably less related to domestic biomass and coal combustion, which is consistent with lower K^+ and As averages. Engine exhaust and agricultural burning emissions are expected to contribute larger portions of OC and EC during summer.

NO_3^- and NH_4^+ show the biggest contrast between winter and summer, consistent with the change in equilibrium. SO_4^{2-} levels were also much lower during summer than winter. This would be consistent with more nearby SO_2 to SO_4^{2-} conversion during winter, possibly in conjunction with reactive fogs and clouds (Pandis et al., 1992) and with local accumulation under stagnant conditions. The summer values could be more influenced by standard photochemical mechanisms occurring during long-range transport (Qian et al., 2001).

The Fe and Ti fugitive dust markers do not show a clear winter/summer pattern, being higher in some cities during summer and lower in others. The sampling periods did not include the April/May Asian dust storms (Gong and Zhang, 2008; Li et al., 2008) that are causes of high $PM_{2.5}$ during these periods. The other elements do not show major or consistent differences between winter and summer, except that the summertime averages are generally lower. The summer Pb average in Xi'an decreased by more than a factor of two (0.75 $\mu g\ m^{-3}$).

Chemical ratios as source indicators

Several potential sources of different chemical components were mentioned earlier. These can be better understood by examining some of the elemental ratios available from the data set that might correspond to similar ratios in the source profiles. OC/EC ratios across the 14 cities are compared in Cao et al. (2007). Given the large role of domestic and industrial coal use, the SO_4^{2-}/OC, SO_4^{2-}/EC, NO_3^-/SO_4^{2-}, As/Fe, and Pb/Fe ratios are compared with ratios from other cities in Table 2. The 2003 SO_4^{2-}/OC ratio found in this study (0.90 ± 0.43) is much higher than that for the other cities, as is the SO_4^{2-}/EC ratio (3.42 ± 2.06). Only Toronto had a higher SO_4^{2-}/EC ratio (i.e., 4.93), mostly due to low EC levels. As noted earlier, there are spatial and seasonal variations in these ratios that reflect local and regional contributions.

NO_3^-/SO_4^{2-} ratios have been used to evaluate relative contributions from coal-burning emissions, which abound in NO_x and SO_2, and engine exhaust, which is a major NO_x emitter but contains little SO_2 (Hu et al., 2002; Wang et al., 2005; Yao et al., 2002). Average NO_3^-/SO_4^{2-} ratios were 0.61 in winter and 0.30 in summer. The NO_3^-/SO_4^{2-} ratio for Toronto (0.81) was >75% higher than the value found in this study (i.e., 0.46 ± 0.27), while the ratios in Seattle, WA (0.43), and Mexico City (0.45) were comparable. As/Fe and Pb/Fe ratios were 0.04 ± 0.03 and 0.39 ± 0.32, respectively, much higher than those for the other cities and indicative of the ash in uncontrolled coal combustion. Figure 3 shows a reasonably good association of Pb and SO_4^{2-} concentrations with the As marker for coal ash. The scatter (e.g., Figure 3d) is typical of different ash composition and SO_2 to SO_4^{2-} transformation rates. The Pb/As correlation indicates that the Pb more probably derives from the coal ash than from the remnants from leaded gasoline, as also indicated by differences in abundances for Pb isotopic ratios (Xu et al., 2012; Widory et al., 2010; Zheng et al., 2004).

Material balance

Material balances estimating organic matter and soil from their marker species are shown in Figures 4 and 5 for the winter and summer seasons. Consistent with the previous discussion, organic material (OM), SO_4^{2-}, NO_3^-, and NH_4^+ are large components. OC takes on an even larger role when its unmeasured hydrogen and oxygen components are taken into account as OM. The role of geological material is also enhanced when the Fe marker is leveraged by reasonable assumptions about its abundance in Chinese soils. Approximately 0 to 15% of the measured mass is not quantified by the chemical analysis, which is potentially due to unmeasured species, underestimations for weighting factors for OM and geological material, and uncertainties in filter equilibration and gravimetric analysis (Malm et al., 2011; Kajino et al., 2006).

For winter samples, contributions in order of importance were OM > geological material > sulfate > nitrate> ammonium > elemental carbon at major cities such as Beijing, Tianjin, Wuhan, Chongqing, Hangzhou, and Xiamen. Compositions

Table 2. Comparison of $PM_{2.5}$ chemical component ratios for the 14 Chinese cities with ratios from selected cities in Europe, Canada, Mexico, and the United States

Cities	SO_4^{2-}/OC	SO_4^{2-}/EC	NO_3^-/SO_4^{2-}	As/Fe	Pb/Fe	Reference
14 Chinese cites (winter and summer)	0.90 ± 0.43	3.42 ± 2.06	0.46 ± 0.27	0.04 ± 0.03	0.39 ± 0.32	This study
Over Europe	0.50	1.51	0.92	NA	NA	Schaap et al. (2004)
Toronto, ON, Canada	0.62	4.93	0.81	0.007	0.062	Lee et al. (2003)
Mexico City, Mexico	0.32	0.71	0.45	0.006	0.068	Vega et al. (2004)
Seattle, WA	0.45	1.13	0.43	0.015	0.099	Maykut et al. (2003)

Note: NA, data not available.

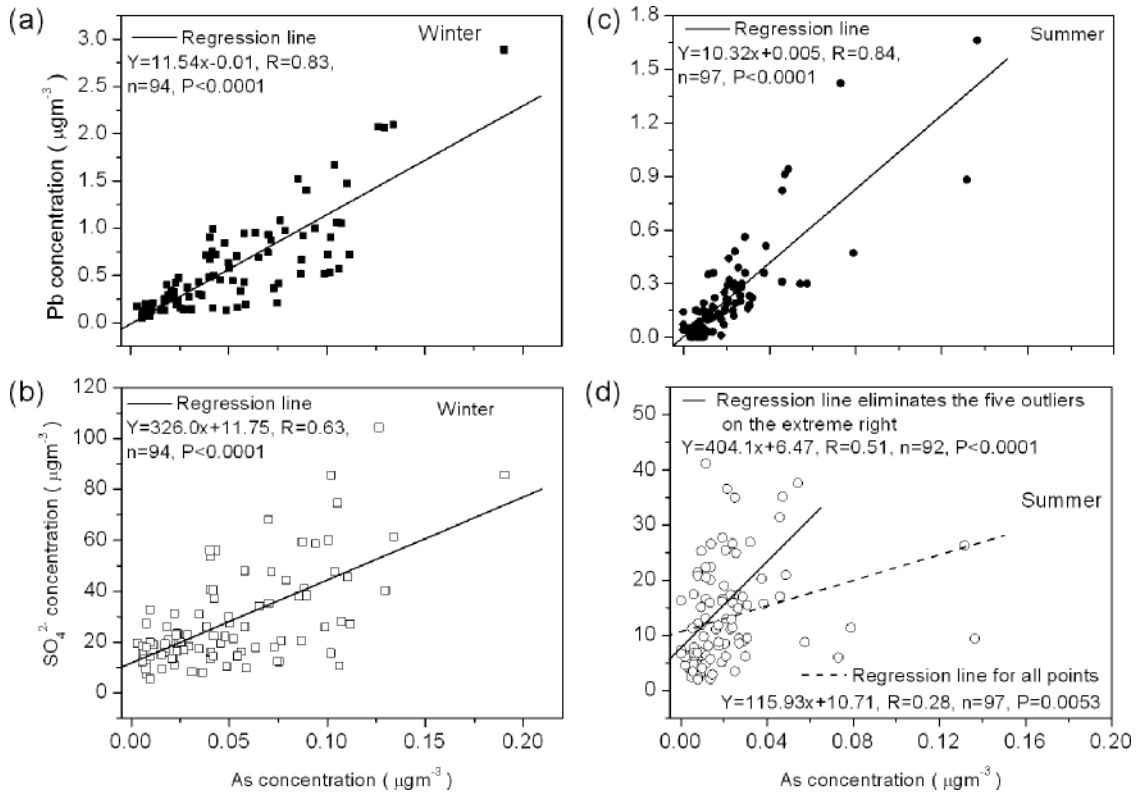

Figure 3. Relationships between $PM_{2.5}$ As, Pb, and SO_4^{2-} concentrations from the 14 cities during winter and summer, 2003.

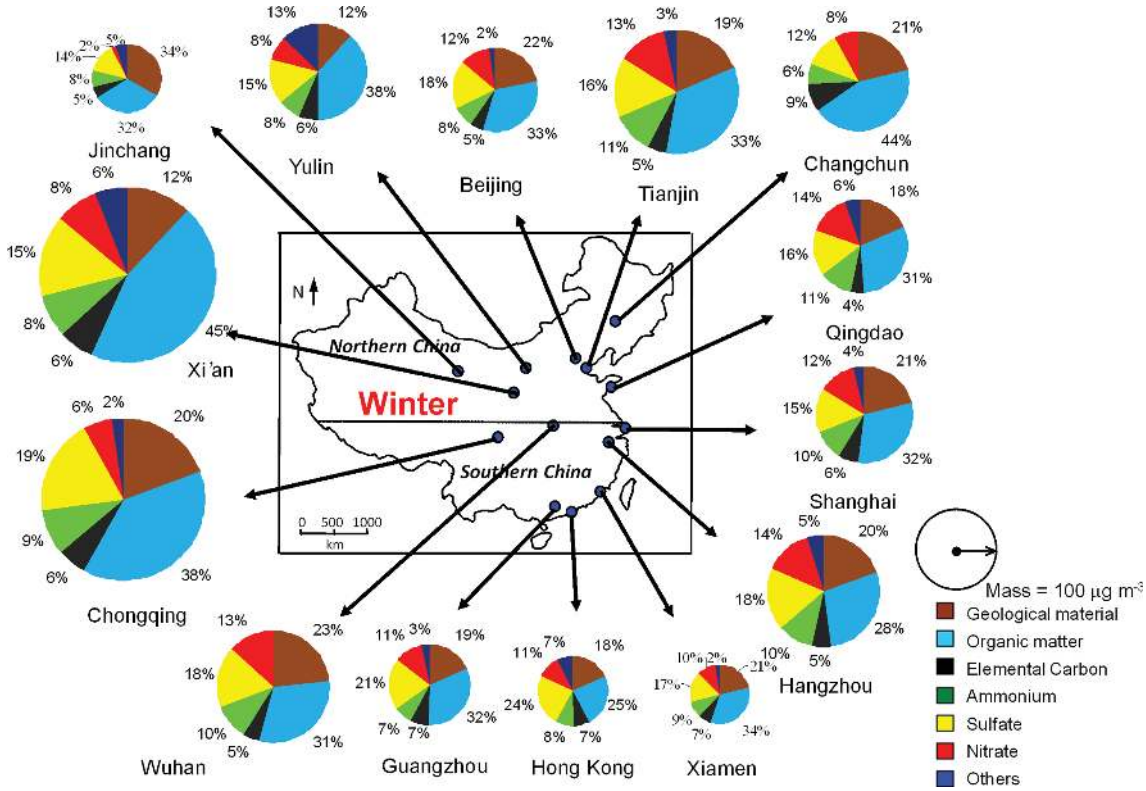

Figure 4. Wintertime material balance of $PM_{2.5}$ for the 14 Chinese cities. Organic matter (OM) is estimated as 1.6 × OC (Chen and Yu, 2007; El-Zanan et al., 2005; El-Zanan et al., 2009) to account for unmeasured hydrogen and oxygen. Geological material is estimated as 25 × Fe (Cao et al., 2008; Wu et al., 2011) to account for unmeasured oxygen and non-iron minerals. "Others" is the remaining unaccounted-for mass after subtracting the sum of measured components from the $PM_{2.5}$ mass. Unaccounted-for mass can be potentially composed of unmeasured geological material (e.g., calcium carbonate), a higher fraction of oxygen in OM, and liquid water associated with NH_4^+, NO_3^-, and SO_4^{2-} at the 35% to 45% relative humidity filter weighing conditions.

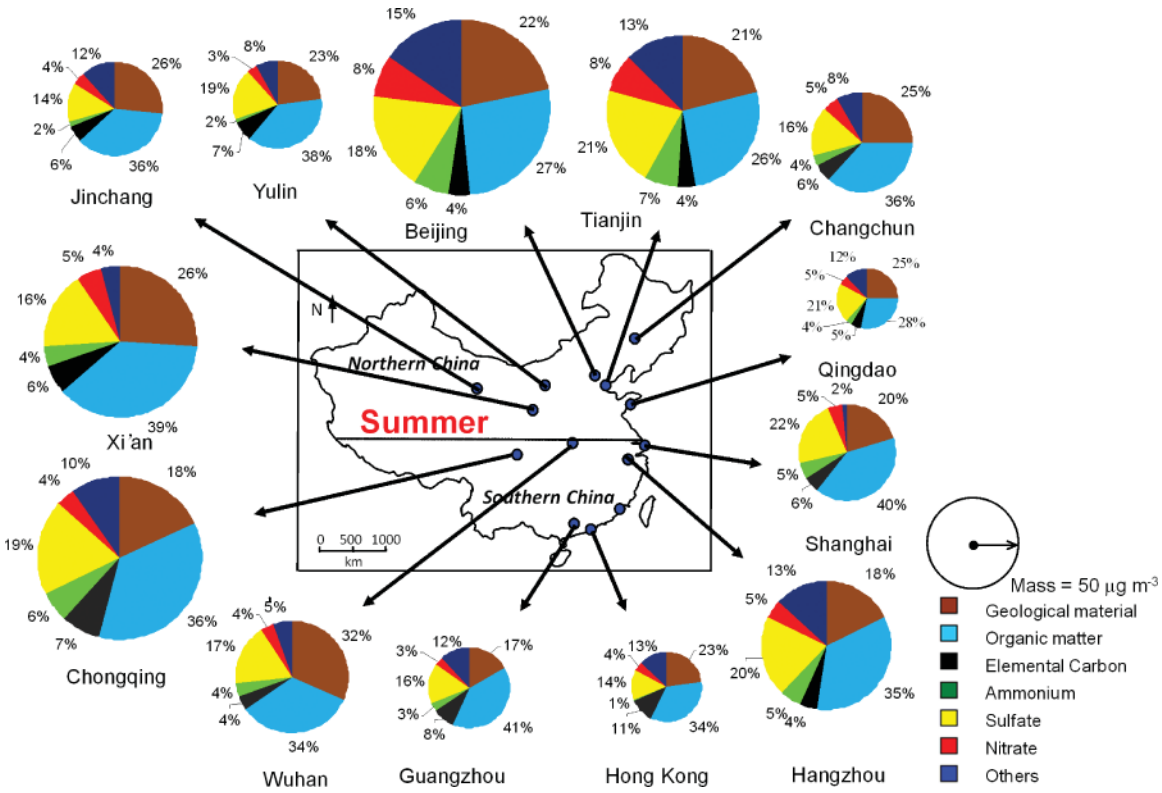

Figure 5. Summertime material balance of $PM_{2.5}$ for the 14 Chinese cities. Organic matter, geological material, and others are explained in the Figure 4 caption.

differed for Yulin, Xi'an, and Hong Kong, where the wintertime SO_4^{2-} contribution exceeded that from geological material. At arid Jinchang, the geological material contribution exceeded the SO_4^{2-} and OM contributions.

During summer, most cities follow the general trend of OM > geological material > sulfate > nitrate, with elemental carbon contributions higher than ammonium contributions at all cities but Beijing and Tianjin. The contribution from SO_4^{2-} in Hong Kong differed between winter (25%) and summer (14%). At Shanghai and Hangzhou, SO_4^{2-} contributions exceeded those of geological material.

Comparison with other $PM_{2.5}$ speciation studies in Chinese cities

Table 3 compares city-specific results from this study with chemical concentrations from other major cities (i.e., Beijing, Xi'an, Shanghai, and Guangzhou). Although there are differences in magnitude owing to the differences in measurement periods, zones of representation, and measurement methods, the major components are similar in magnitude and order of importance for nearly all of the studies. There is no evidence of major upward or downward trends in mass and chemical composition from 1999 to 2006, but this is expected, given the short durations of the measurement programs and the large variability in emissions and meteorology expected over this time period. Trends in the United States have only been associated with emission reductions over long periods of a decade or more using chemically speciated measurements that are specific to those emissions.

$PM_{2.5}$ in Chinese cities versus non-Chinese cities

Table 4 compares the major components from Chinese cities with $PM_{2.5}$ compositions in other countries. The geological material contribution is on the order of 10% in $PM_{2.5}$ from the non-Chinese cities, about half of that estimated from this study (19.5%). Roadside sites in St. Louis, MO, and Barcelona, Spain, showed more comparable geological material contributions (15.4 and 15.2%). The OM fractions in the Chinese cities are similar to most of the other cities, although the absolute OM concentrations are much higher in China. SO_4^{2-}, NO_3^-, and NH_4^+ are important in $PM_{2.5}$ in all of the cities, but their fractions are more variable and their absolute values are generally lower than those found in the 14 Chinese cities.

Average $PM_{2.5}$ concentrations for this study ranged from 3 to 9 times higher than the values in Seoul, Yokohama, St. Louis, Indianapolis, Toronto, Mexico City, Barcelona, and Milan, with corresponding 2 to 10 times higher levels of OM. The average Chinese secondary aerosol concentrations for SO_4^{2-}, NO_3^-, and NH_4^+ were 2–5, 1–10, and 2–7 times higher, respectively, than those in other cities in the world. As and Pb were 10 times and average geological material was 5–43 times those found in other cities.

Conclusions

$PM_{2.5}$ in 14 of China's large cities achieved high concentrations in both winter and summer of 2003 with averages >100 μg m^{-3} being common occurrences. A grand average of 115 μg m^{-3} was

Table 3. Comparison of $PM_{2.5}$ and major chemical concentrations ($\mu g\ m^{-3}$) from this study with measurements from other $PM_{2.5}$ studies in Beijing (BJ), Xi'an (XA), Shanghai (SH), and Guangzhou (GZ)

Location	Site description	Period (MM/YY)	$PM_{2.5}$	OC	EC	SO_4^{2-}	NO_3^-	NH_4^+	K^+	Fe	As	Pb	Reference
Beijing (BJ)	Semiresidential (Tsinghua)	09/99–09/00	127.0	29.1	10.1	14.1	9.9	6.5	2.2	1.1	NA	0.34	He et al. (2001)
	Urban (Chegongzhuang)	09/99–09/00	115.0	21.5	8.7	14.5	10.3	6.2	2.2	1.1	NA	0.30	He et al. (2001)
	Semiresidential (Tsinghua)	08/01–09/02	106.9	28.8	9.6	10.6	7.8	5.5	1.4	1.2	0.06	0.21	Duan et al. (2006)
	Urban (Chegongzhuang)	08/01–09/02	96.6	22.9	10.3	9.9	6.9	5.8	1.7	1.1	0.06	0.17	Duan et al. (2006)
	Traffic (Beijing Normal University)	2002–2003	106.5	22.4	8.1	23.2	14.6	11.7	1.6	0.8	0.03	0.21	Sun et al. (2004)
	Industrial (Capital Steel Company)	2002–2003	111.5	22.8	8.2	21.2	13.4	12.2	1.7	1.5	0.05	0.24	Sun et al. (2004)
	Residential (Yihai Garden)	2002–2003	128.8	24.4	13.9	24.8	16.3	15.0	2.7	1.1	0.04	0.21	Sun et al. (2004)
	Urban	01/03, 06/03–07/03	123.6	21.8	5.9	21.3	13.4	9.6	1.5	0.9	0.03	0.23	This study
	Suburban	06/05–08/05	68.0	8.2	4.9	22.5	9.7	5.4	NA	NA	NA	NA	Pathak et al. (2011)
	Semiresidential (TSinghua) and rural sites (Miyun)	03/05–02/06	118.5	24.5	8.2	15.8	10.1	7.3	NA	1.1	0.02	0.24	Yang et al. (2011)
Xi'an (XA)	Urban	01/03, 06/03–07/03	229.6	60.2	14.0	35.3	17.0	17.0	3.3	1.4	0.08	1.22	This study
	Urban	09/03–02/04	199.4	48.0	13.3	NA	NA	NA	NA	NA	NA	NA	Cao et al. (2005)
	Urban	10/05–10/06	NA	NA	NA	19.5	8.3	8.7	1.8	NA	NA	NA	Shen et al. (2008)
	Urban	03/06–03/07	194.1	NA	NA	35.6	16.4	11.4	NA	NA	NA	NA	Zhang et al. (2011)
Shanghai (SH)	Urban	03/99–02/00	67.6	16.8	6.5	13.0	5.8	5.7	NA	0.9	NA	0.29	Ye et al. (2003)
	Urban	01/03, 06/03–07/03	96.7	20.0	6.1	16.8	10.1	9.0	1.5	0.8	0.03	0.32	This study
	Urban (Fudan and Taopu)	09/03–01/05	94.6	NA	NA	10.4	6.2	3.8	0.6	NA	NA	NA	Wang et al. (2006)
	Urban (Baoshan, Putuo, Huangpu, Jiading)	04/04–05/04	NA	NA	NA	NA	NA	NA	NA	0.9	0.03	0.11	Chen et al. (2009)
	Urban	06/05–08/05	67.0	16.9	10.0	15.6	7.2	4.2	NA	NA	NA	NA	Pathak et al. (2011)
	Urban (Zhabei district)	10/05–07/06	90.3	14.7	2.8	NA	NA	NA	NA	NA	NA	NA	Feng et al. (2009)
	Subsurb (Jiading district)	10/05–07/06	95.5	17.5	3.0	NA	NA	NA	NA	NA	NA	NA	Feng et al. (2009)
Guangzhou (GZ)	Urban, industrial, and background mixed	06/02–07/02	78.1	15.8	5.9	NA	NA	NA	NA	NA	NA	NA	Cao et al. (2004)
	Urban, industrial, and background mixed	01/02–02/02	105.9	22.6	8.3	NA	NA	NA	NA	NA	NA	NA	Cao et al. (2004)
	Urban	10/02–06/03	70.6	17.6	4.4	14.7	4.0	4.5	NA	NA	NA	NA	Hagler et al. (2006)
	Urban	01/03, 06/03–07/03	75.0	17.1	5.8	13.5	6.3	4.8	1.4	0.5	0.03	0.27	This study
	Urban	10/04–11/04	102.9	22.4	7.1	27.8	4.3	12.1	2.6	NA	NA	NA	Andreae et al. (2008)
	Suburban	06/05–08/05	59.0	14.9	10.5	12.7	5.8	5.0	NA	NA	NA	NA	Pathak et al. (2011)
	Residential and commercial mixed (Wushan)	12/08–02/09	81.7	17.5	4.1	5.6	12.0	4.7	NA	1.9	0.04	0.45	Yang et al. (2011)

Note: NA, data not available.

Table 4. Comparison of $PM_{2.5}$ mass and major chemical components from this study with measurements from other major cities worldwide

City	Site type	Period (MM/YY)	Concentration ($\mu g\ m^{-3}$)									Reference
			Mass	SO_4^{2-}	NO_3^-	NH_4^+	OM	EC	Geological matter	As	Pb	
14 Cities, China	Urban	01/03, 06/03–07/03	115.2	19.89 (17.3%)	9.98 (8.7%)	9.20 (8.0%)	29.48 (25.6%)	6.58 (5.7%)	22.4 (19.5%)	0.04	0.39	This study
Seoul, Korea	Urban	03/03–12/06	37.6	5.77 (15.3%)	5.17 (13.8%)	3.70 (9.8%)	11.36 (30.2%)	2.91 (7.7%)	2.78[a] (7.4%)	NA	0.03	Heo et al. (2009)
Yokohama, Japan	Urban	09/07–08/08	20.6	3.80 (18.4%)	0.96 (4.7%)	2.27 (11.0%)	4.50 (21.8%)	1.94 (9.4%)	NA	NA	NA	Khan et al. (2010)
St.Louis, MO												
Blair	Traffic	02/00–12/03	16.4[b]	4.23 (25.8%)	2.48 (15.1%)	1.94 (11.8%)	3.25 (19.8%)	0.84 (5.1%)	2.53[a] (15.4%)	0.003	0.013	Lee and Hopke (2006)[87]
Arnold	Traffic	02/00–12/03	15.5	4.16 (26.8%)	1.9 (12.4%)	1.61 (10.4%)	3.3 (21.3%)	0.58 (3.7%)	0.52[a] (3.4%)	0.005	0.019	
Indianapolis, IN	Urban	12/00–11/03	17.8[a]	7.12 (40.0%)	2.85 (16.0%)	2.28 (12.8%)	2.86 (16.0%)	0.66 (3.7%)	0.94[a] (5.3%)	0.002	0.006	Zhao and Hopke (2006)
Toronto, Canada	Urban	02/00–02/01	12.7	2.27 (17.9%)	1.84 (14.5%)	1.19 (9.4%)	4.39 (34.6%)	0.46 (3.6%)	1.38 (10.8)	0.0004	0.003	Lee et al. (2003)
Mexico City, Mexico	Urban	2000–2002	35.05	5.90 (13.4%)	2.67 (5.9%)	2.95 (6.7%)	21.88 (48.9%)	8.35 (17.0%)	2.62 (3.5%)	0.01	0.12	Vega et al. (2004)
Northwestern Europe	Urban	1996–2007	NA	NA (21%)	NA (16%)	NA	NA (25%)	NA (7%)	NA (5%)	NA	NA	Putaud et al. (2010)
Southern Europe	Urban	1996–2007	—	— (15%)	— (7%)	—	— (23%)	— (8%)	(11%)			Putaud et al. (2010)
Central Europe	Urban	1996–2007	NA	NA (19%)	NA (13%)	NA	NA (22%)	NA (14%)	NA (5%)	NA	NA	Putaud et al. (2010)
Barcelona, Spain	Traffic	1999–2001	27.6	4.20 (15.2%)	2.30 (8.3%)	2.00 (7.2%)	12.24 (44.3%)	NA	4.20 (15.2%)	NA	NA	Querol et al. (2004)
Milan, Italy	Urban	08/02–12/03	40.0	4.70 (11.8%)	8.70 (21.8%)	3.00 (7.5%)	15.48 (38.7%)	1.40 (3.5%)	1.50 (3.8%)	—	—	Lonati et al. (2008)

Notes: Figures in the parentheses are the mass percentages of each chemical component. NA, data not available. [a]Estimated by mass balance model. [b]Median values.

found for all cities, with a minimum of 27.3 $\mu g\ m^{-3}$ measured at coastal Qingdao during summer and a maximum of 356.3 $\mu g\ m^{-3}$ at inland Xi'an during winter. Both primary and secondary $PM_{2.5}$ are important contributors at all of the cities during both winter and summer. While ammonium sulfate is a large contributor during both seasons, ammonium nitrate contributions are much larger during winter. Lead levels are still high in several cities, reaching an average of 1.68 $\mu g\ m^{-3}$ in Xi'an during winter. High correlations of lead with arsenic and sulfate concentrations indicate that much of it derives from coal combustion rather than leaded fuels that were phased out by calendar year 2000. Although limited fugitive dust markers were available, scaling of iron by its ratios in source profiles shows ~20% of $PM_{2.5}$ deriving from fugitive dust in most of the cities. Multipollutant control strategies will be needed that address incomplete combustion of coal and biomass, engine exhaust, and fugitive dust, as well as sulfur dioxide, oxide of nitrogen, and ammonia gaseous precursors for ammonium sulfate and ammonium nitrate.

Acknowledgments

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