Supporting Information

Metallic elements and Pb isotopes in $PM_{2.5}$ in three Chinese typical megacities: spatial distribution and source apportionment

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Figure S1 Percentages of coal and natural gas in total energy consumption in Beijing from 2010 to 2014.

Sampling sites description

Beijing, the capital of China, is one of the world's most populous cities with a population of over 20 million in an area of 16,410 km^2. It is located in the northern part of the country with a typical continental monsoon climate. Annual rainfall averages nearly 700 mm, most of it comes in July and August. The average temperature throughout the whole year is 11.7 ℃. Winter is dry and cold. Beijing is also located in a basin with multiple emission sources, promoting accumulation of air pollutants and limiting their dispersion. Coal burning accounts for about 40% of $PM_{2.5}$ in Beijing before 2012. Since 2012, the city has been converting coal-fired power stations to burn natural gas. The percentages of coal in total energy consumption was decreasing from 29.6% to 20.4% in Beijing from 2010 to 2014, and natural gas was increasing from 14.6% to 21.1%, as shown in Figure S1. At the end of 2013, the number of motor vehicles in Beijing was 5.437 million.[1] $PM_{2.5}$ was sampled on the roof of an experimental building in the Tower Division of the Institute of Atmospheric Physics, Chinese Academy of Sciences (N 39.59°, E 116.22°).

Changchun, the capital city of Jilin province, is an industrial city, with a population of ~8 million in 2013 in an area of 20,565 km^2. Changchun belongs to the temperate continental semi-humid monsoon climate type, on a flat plain in northeast China. It is characterized by four distinct seasons. Winter is long (lasting from November to March), cold, and windy with a January mean temperature of -15.1 °C. And the average of air temperature in five months of the year is below 0 °C. Changchun is an old industrial base in China with a solid industrial foundation and a cradle for the industrial development of automobiles, rail cars, optoelectronic information, and applied chemistry. In 2013, the total output value of industrial enterprises above designated size was 921.34 billion CNY. Among them, the total output value of the automobile manufacturing industry was 549.26 billion CNY, accounting for 59.6%. In 2014, the coal consumption in Changchun was 25.8 million tons of standard coal, accounting for ~70% of total energy consumption. At the end of 2013, the number of motor vehicles was 1.445 million in Changchun.[2] $PM_{2.5}$ was

sampled on the roof of the building in Changchun Meteorological Bureau (N 43.54°, E 125.13°).

Chengdu, the capital city of Sichuan province, is located at the western edge of the Sichuan Basin, with a population of over 14 million in an area of 14,312 km^2. It has a mild, humid, subtropical climate with the annual average temperature of 16 ℃ and rainfall of ~1000 mm. Chengdu has one of the lowest annual sunshine totals nationally with monthly percent possible sunshine ranging from 16% in December to 38% in August. The city receives 1,073 hours of bright sunshine annually. Chengdu achieved a gross domestic product (GDP) of 910.89 billion CNY in 2013, an increase of 10.2% over the previous year. In 2012, coal consumption in Chengdu was 8.591 million tons of standard coal, accounting for ~25% of total energy consumption. At the end of 2013, the number of motor vehicles in Chengdu was 3.386 million, increasing at 366% and 30% compared to 2000 and 2010, respectively.[3] Chengdu is the fastest growing city in China.[4] $PM_{2.5}$ was sampled on the roof of the building in the Institute of Plateau Meteorology, China Meteorological Administration (N 30.39°, E 104.00°).

Moreover, the meteorological parameters and air quality indexes of each sampling site are shown in Tables S1 and S2.

Reference

1. Beijing Statistical Yearbook. Statistics Communique on the National Economy and Social Development of the City of Beijing. Beijing Municipal Bureau of Statistics and NBS Survey Office in Beijing. China Statistics Press, 2014.
2. Changchun Statistical Yearbook. Statistics Communique on the National Economy and Social Development of the City of Changchun. Changchun Municipal Bureau of Statistics and NBS Survey Office in Changchun. China Statistics Press, 2014.
3. Chengdu Statistical Yearbook. Statistics Communique on the National Economy and Social Development of the City of Chengdu. Chengdu Municipal Bureau of Statistics and NBS Survey Office in Chengdu. China Statistics Press, 2014.
4. Blue book of world cities. Annual report on world cities (2018) No.7. Social Sciences Academic Press (China), 2018.

Table S1 Meteorological parameters (mean±1σ) of the studied three cities in China during $PM_{2.5}$ sampling period*.

	Temperature (°C)	Sunlight time (h)	RH (%)	Pressure (hPa)	Boundary layer height (m)	Visibility (km)	Wind speed (km h^{-1})
Beijing	-0.8±2.5	192.4	36.8±16.2	1025.3±6.7	1080±207	10.4±7.6	10.1±5.4
Changchun	-14.2±4.5	198.3	57.9±8.1	1022.8±8.1	892±185	8.0±3.8	12.0±4.6
Chengdu	8.0±2.1	98.1	69.1±9.3	1023.2±5.2	707±163	4.2±2.0	3.9±1.8

*The meteorological parameters including temperature, sunlight time, relative humidity (RH), pressure, visibility, and wind speed during the sampling periods were obtained from the meteorological bureau of each city. Boundary layer height data were obtained from dataset of "ERA5 monthly averaged data on single levels from 1979 to present" in ECMWF (https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels-monthly-means?tab=form).

Table S2 AQI and the concentrations of gas pollutants (CO, SO_2, NO_2, and O_3) in the studied three cities during $PM_{2.5}$ sampling period*.

	AQI	CO (mg m^{-3})	SO_2 (μg m^{-3})	NO_2 (μg m^{-3})	O_3 (μg m^{-3})
Beijing	122	2	51	58	20
Changchun	118	1	68	50	34
Chengdu	212	2	35	86	22

*AQI (Air Quality Index), CO, SO_2, NO_2, and O_3 in Beijing, Changchun and Chengdu were retrieved from the Real-time Monitoring Platform of Chinese Air Quality (https://www.aqistudy.cn/).

Table S3 Minimum detection limits (MDL) and levels of blank samples of the 40 elements detected in this study.

Element	MDL (ng m^{-3})	Acid blank (ng m^{-3})	Field blanks (ng m^{-3})		
			Beijing	Changchun	Chengdu
Na	6.06	1.90	74.4	117	76.1
Mg	0.0702	1.47	6.29	16.7	7.19
Al	0.0998	1.10	4.54	21.5	5.31
Ca	0.187	17.1	81.3	89.8	47.5
Ti	0.0222	0.0747	0.115	0.543	0.105
V	0.000425	0.00941	0.00802	0.0788	0.00954
Cr	0.000189	0.636	0.361	1.20	0.925
Mn	0.00713	0.108	0.0241	0.937	0.0589
Fe	0.0666	3.51	1.21	31.2	3.11
Co	0.00134	0.0179	0.00238	0.0207	0.00768
Ni	0.162	1.42	0.417	0.707	0.672
Cu	0.00475	0.114	0.121	2.91	0.139
Zn	0.0152	0.199	1.06	8.72	0.948
Rb	0.00209	0.010	0.0149	0.110	0.00677
Sr	0.00909	0.0557	0.129	0.344	0.0846
Zr	0.00688	0.000557	0.0251	0.0910	0.0203
Nb	0.000424	0.00230	0.00631	0.0335	0.0205
Mo	0.0218	0.0851	0.238	0.326	0.257
Cd	0.000669	0.000886	0.00128	0.0447	0.00247
Sn	0.00584	0.155	0.271	0.176	0.338
Sb	0.00388	0.0239	0.0220	0.185	0.0276
Ba	0.00110	0.0365	0.267	1.610	0.639
La	0.000149	0.000808	0.00292	0.0126	0.00190
Ce	0.000223	0.000859	0.00385	0.0251	0.00245
Pr	0.000108	0.000108	0.000547	0.00303	0.000400
Nd	0.000161	0.000528	0.00170	0.0117	0.00143
Sm	0.000208	0.000197	0.000569	0.00220	0.000429
Eu	0.000146	0.000236	0.000277	0.000855	0.000196
Gd	0.0000943	0.000224	0.000620	0.00223	0.000260
Tb	0.000143	0.0000278	0.000140	0.000320	0.000104
Dy	0.0000944	0.000132	0.000612	0.00204	0.000577
Ho	0.0000676	0.0000343	0.000136	0.000405	0.0000636
Er	0.0000920	0.0000576	0.000341	0.000908	0.000332
Tm	0.0000504	0.0000451	0.0000409	0.000133	0.0000391
Yb	0.0000429	0.0000677	0.000271	0.000963	0.000197
Lu	0.0000539	0.0000275	0.0000649	0.000122	0.0000651
Tl	0.000482	0.00164	0.00400	0.0281	0.00140
Pb	0.00192	0.0130	0.0520	2.05	0.0415

Th	0.00189	0.000159	0.000896	0.00234	0.000319
U	0.0000414	0.0000947	0.00263	0.00497	0.00174

Table S4 Mean (±1σ) mass concentrations of $PM_{2.5}$ (μg m^{-3}) and bounded elements (ng m^{-3}) in the studied three cities.

	Beijing		Changchun		Chengdu	
	Mean	**Stdev***	**Mean**	**Stdev***	**Mean**	**Stdev***
$PM_{2.5}$	171	59.7	162	65.8	171	63.1
ΣElement**	1145	710	1272	991	857	291
Na	181	97.5	142	91.8	123	35.5
Mg	107	86.5	82.9	109	38.5	17.6
Al	161	146	280	306	134	82.2
Ca	293	285	325	297	247	113
Ti	5.25	4.44	8.30	6.48	4.05	1.94
V	0.784	0.292	0.876	0.493	0.531	0.215
Cr	2.16	1.09	2.18	1.79	2.29	2.46
Mn	12.5	4.94	11.3	7.88	12.0	4.15
Fe	237	150	218	190	167	64.2
Co	0.213	0.087	0.194	0.115	0.221	0.192
Ni	1.27	0.424	1.12	0.886	4.64	11.8
Cu	17.4	9.22	7.92	9.76	10.8	4.42
Zn	69.9	40.1	142	99.2	72.1	29.6
Rb	1.37	0.565	0.891	0.490	1.05	0.358
Sr	7.43	15.0	5.03	11.9	1.74	0.907
Zr	0.211	0.133	0.313	0.208	0.135	0.058
Nb	0.066	0.042	0.109	0.078	0.040	0.030
Mo	0.592	0.361	1.80	1.85	0.605	0.287
Cd	0.689	0.421	0.414	0.219	0.917	0.482
Sn	1.85	0.794	1.94	0.863	7.27	8.20
Sb	2.63	1.57	2.01	1.02	2.39	1.11
Ba	26.6	51.7	21.6	55.7	7.34	4.64
La	0.208	0.152	0.234	0.203	0.080	0.040
Ce	0.340	0.318	0.463	0.426	0.151	0.079
Pr	0.035	0.038	0.049	0.049	0.016	0.009
Nd	0.130	0.141	0.190	0.185	0.061	0.034
Sm	0.023	0.023	0.035	0.033	0.012	0.007
Eu	0.006	0.006	0.008	0.009	0.003	0.002
Gd	0.021	0.022	0.034	0.031	0.012	0.006
Tb	0.003	0.003	0.005	0.004	0.002	0.001
Dy	0.017	0.016	0.028	0.024	0.009	0.005
Ho	0.003	0.003	0.005	0.004	0.002	0.001
Er	0.008	0.007	0.014	0.011	0.004	0.002
Tm	0.001	0.001	0.002	0.001	0.001	0.000
Yb	0.007	0.006	0.012	0.009	0.004	0.002
Lu	0.001	0.001	0.002	0.001	0.001	0.000

Tl	0.378	0.167	0.226	0.066	0.208	0.075
Pb	31.3	17.4	14.7	11.5	18.8	6.50
Th	0.025	0.032	0.039	0.042	0.015	0.008
U	0.025	0.012	0.048	0.021	0.020	0.006

105 * Standard deviation.

106 ** Sum concentration of all tested elements (40) in the table.

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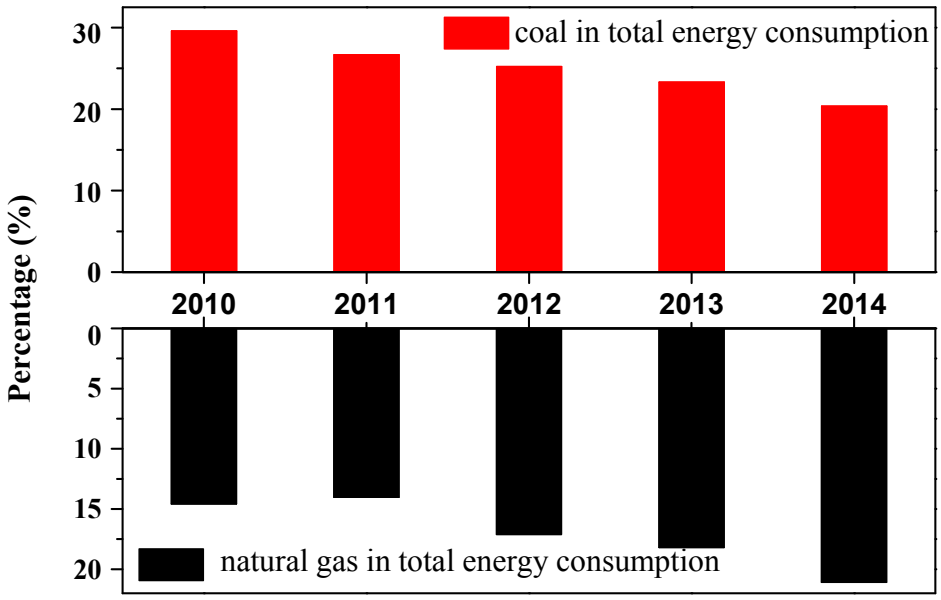

Figure S1 Percentages of coal and natural gas in total energy consumption in Beijing from 2010 to 2014.