Article (refereed) - postprint

Song, Wei; Chang, Yunhua; Liu, Xuejun; Li, Kaihui; Gong, Yanming; He, Guixiang; Wang, Xiaoli; Christie, Peter; Zheng, Mei; Dore, Anthony J.; Tian, Changyan. 2015. **A multiyear assessment of air quality benefits from China's emerging shale gas revolution: Urumqi as a case study.** *Environmental Science & Technology,* 49 (4). 2066-2072. 10.1021/es5050024

A Multi-year Assessment of Air Quality Benefits from China's Emerging Shale Gas Revolution: Urumqi as a Case Study

Wei Song[1, 3 #], Yunhua Chang[2 #], Xuejun Liu[1,4 *], Kaihui Li[1], Yanming Gong[1], Guixiang He[1,3], Xiaoli Wang[1,3], Peter Christie[4], Mei Zheng[5], Anthony J. Dore[6], Changyan Tian[1]

1. State Key Laboratory of Desert and Oasis Ecology, Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences, Urumqi 830011, China

2. Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China

3. Graduate University of the Chinese Academy of Sciences, Beijing, 100039, China

4. College of Resources and Environmental Sciences, China Agricultural University, Beijing 100193, China

5. College of Environmental Sciences and Engineering, Peking University, Beijing 100871, China

6. Centre for Ecology and Hydrology Edinburgh, Bush Estate, Penicuik, Midlothian EH26 0QB, UK

[#] Equal contribution; * Corresponding author: liu310@cau.edu.cn.

Telephone: +86 10 62733459; Fax: +86 10 62731016

The manuscript contained 6206 words and 4 figures and 1 table.

Abstract

China is seeking to unlock its shale gas in order to curb its notorious urban air pollution, but robust assessment of the impact on $PM_{2.5}$ pollution of replacing coal with natural gas for winter heating is lacking. Here, using a whole-city heating energy shift opportunity offered by substantial reductions in coal combustion during the heating periods in Urumqi, northwest China, we conducted a four-year study to reveal the impact of replacing coal with natural gas on the mass concentrations and chemical components of $PM_{2.5}$. We found a significant decline in $PM_{2.5}$, major soluble ions and metal elements in $PM_{2.5}$ in January of 2013 and 2014 compared with the same periods in 2012 and 2011, reflecting the positive effects on air quality of using natural gas as a whole-city heating fuel. This occurred following complete replacement with natural gas for heating energy in October 2012. The weather conditions during winter did not show any significant variation over the four years of the study. Our results reveal that China and other developing nations will benefit greatly from a change in energy source, i.e., increasing the contribution of either natural gas or shale gas to total energy consumption with a concomitant reduction in coal consumption.

Introduction

Concerns about the effects of atmospheric particulate matter (hereafter referred to as PM) range from their influence on biogeochemical processes [1, 2] and climate change [3, 4] to public health.[5, 6] Recent interest in China has focused on $PM_{2.5}$ (fine particles; PM with aerodynamic diameter $\leq$ 2.5 μm) because numerous studies have provided convincing evidence of an association between ambient $PM_{2.5}$ and regional haze[7-9] as well as human mortality/morbidity.[10-12]

Existing field observations in China reveal that $PM_{2.5}$ levels in urban sites show a distinct seasonal pattern, with much higher mass concentrations in winter than in

summer.[13-15] This is particularly true in northern China where coal-based heating systems in winter have become the dominant sources of SO_2, NO_x and primary PM. According to Chen et al.,[16] extended-use heating systems that substantially intensify total suspended particulate (TSP) pollution have resulted in an average loss of more than five years of life expectancy for the 500 million residents of the northern China. Moreover, Meng et al.[11] have explored the association between size-fractionated particle number concentrations and daily mortality in Shenyang (capital city of Liaoning province, northeast (NE) China) and conclude that adverse health effects may increase with decreasing particle size. In Xi'an (capital city of Shaanxi province, northwest (NW) China) Huang et al.[10] observed a greater risk from $PM_{2.5}$ and selected species on all causes of mortality during periods of heating from 2004 to 2008. Although coal can be expected to continue playing a vital role as an abundant and economic energy source in the foreseeable future, these studies clearly indicate that a strategy of switching away from coal to other energy sources for heating in China is urgently needed.

The environmental benefits of replacing coal with other energy sources (e.g., biofuels, wind, hydro, solar, and nuclear power) are well established in various scenarios.[17-19] Natural gas is increasingly considered to be a promising fossil fuel in China in the transition to renewable sources. It is regarded as a fuel that can reduce concentrations of greenhouse gases (GHG) and PM and its precursors (mainly SO_2 and NO_x).[20] Official data show that Chinese domestic proven recoverable reserves of (conventional) natural gas are 1.5% of the world total, and as such have been viewed as a luxury. However, the past decade has witnessed the rapid development of new technologies allowing the widespread recovery of natural gas from shale formations in the US and this has brought about economic revival during the 2008-2009 global

financial crisis and has also reshaped the energy landscape of the US with profound geopolitical implications.[21] It is also worth noting that there is continuing concern about the adverse environmental risks related to air, water, and geology as well as public health from shale energy development which highlights the importance of effective governance.[22, 23]

A study shows that China has a total of 25.08 trillion cu m of proven reserves of shale gas, equivalent to nearly 200 times its annual gas consumption.[24] Mounting public pressure regarding air pollution is pushing the government to embrace natural gas to reduce atmospheric (e.g., SO_2, NO_x, primary PM) emissions. However, China is still in the nascent stage of shale gas development and the shale gas revolution is still a dream. One of the unanswered questions behind this proposed development is to what extent natural gas can achieve improvements in air quality. Although we have witnessed several successful examples of improvements in air quality (e.g., during the 2008 Beijing Olympic Games and the 2010 Shanghai World Expo) after introducing a series of aggressive emissions control plans (including phasing out coal-fired power plants, powering more cars and buses with natural gas, and raising standards for vehicle emissions),[25-29] a city-wide systematic assessment of the benefits to air quality from switching to natural gas is clearly lacking.

Xinjiang province in NW China is known for its vast oil and gas reserves and has become the biggest energy base in China. Since 2011 Urumqi, the capital city of Xinjiang, has implemented a large-scale policy of shifting from coal to natural gas to improve temporarily its air quality during the winter heating period (usually from 15 October to 15 March the following year). By taking advantage of this unique opportunity, we conducted a study over four consecutive years (2011-2014) to examine the relationships between substantial changes in energy use and changes in

levels of polluting gases and PM concentrations in an attempt to provide solid evidence for the potential role that shale gas might play in the national air quality control strategy in the future.

Materials and Methods

Site description

Sampling was conducted at Shengdisuo (SDS) (43°51′N, 87°33′E, 775 m above mean sea level) in Urumqi city, Xinjiang Uygur Autonomous Region, northwest China (Fig. S1, Supporting Information). Urumqi has a population of 2.2 million with a total area of approximately 14000 km^2 and is surrounded by the Tianshan Mountains. The SDS site is an urban monitoring site surrounded by a business district, residential areas and major roads. The sampling site is about 100 m west of a heating supply station which supplies heating from mid-October every year. Heating in winter with natural gas gradually replaced coal in the main city of Urumqi during the 2013 to 2014 heating period (Fig. S2, Supporting Information).

Sampling procedure and sample analysis

Airborne $PM_{2.5}$ were sampled using a particulate sampler (TH-16A, Tianhong Inc., Wuhan, China) with a flow rate of 16.7 L min^{-1}, and 25-28 daily samples of $PM_{2.5}$ were collected at SDS site during the month of January in 2011, 2012, 2013 and 2014. The sampler was placed about 2 m above the ground and ran for 24 h to obtain a particulate matter sample on 47 mm quartz filters (Whatman, Maidstone, UK). Before and after sampling, each filter was conditioned for at least 24 h inside an artificial climate chamber at a relative humidity of 50% and a temperature of 25 °C, and then weighed (Sartorius, Göttingen, Germany; precision 10 μg). $PM_{2.5}$ mass concentrations were determined from the mass difference and the sampled air volume. Each sampling filter was extracted with 10 ml deionized water (18.2 MΩ) by ultrasonication for 30

min, and the extract solution was filtered through a syringe filter (0.22 μm, Tengda Inc., Tianjin, China) then stored in a refrigerator. Ammonium and nitrate in $PM_{2.5}$ were measured with a continuous flow analyzer (AutoAnalyzer 3, Germany), sulfate, chloride and the major cations (Ca^{2+}, Mg^{2+}, K^{+}, and Na^{+}) by an ion chromatography system (ICS5000, Dionex, USA), and metal (loid) elements Cr, As, Ni, Mn, Cu, Cd, Pb and Al by ICP Mass Spectrometry (ELAN DRC Ⅱ, PerkinElmer, USA). However, only ammonium and nitrate were measured in $PM_{2.5}$ samples in January 2011. Monthly mean air temperature, wind speed, relative humidity and precipitation in the month of January in 2011, 2012, 2013 and 2014 at the SDS site are shown in Fig. S3 and synoptic weather maps at the surface (1000 mb) around Urumqi during the same periods are given in Fig. S4 (Supporting Information), data downloaded from the Internet.[30]

Quality Assurance/Quality Control

We analyzed three field blanks at every batch of samplers to monitor for contamination or interferences. Sample concentrations were determined from external calibration curves prepared at concentrations ranging from 1 to 1000 μg L^{-1} for As, Cr, Mn, Ni, Cu, Cd, Pb and Al, and 0.5 to 50 mg L^{-1} for SO_4^{2-}, Cl^{-}, Na^{+}, K^{+}, Mg^{2+} and Ca^{2+}, and 0.4 to 2.0 mg L^{-1} for NH_4^{+} and NO_3^{-}. The limits of quantification were restricted to water soluble ions or metals and the limits of detection were ± 0.01 mg L^{-1} for major anion and cation ions, and ±10 ng L^{-1} for metal elements. The lab belongs to Chinese State Key Laboratory for Oasis and Arid Ecosystems and has a complete quality control system. We have monitored the blank (treated syringe filters without sampling) and standard (designed specific concentrations of various ions and metal elements) samples at each measurement event. Normally, the results from the blank samples were around or below the detection limits while the differences

between the measured and 'theoretical' results from the standard samples were controlled to be lesser than ±5%.

Statistical analysis

Values of $PM_{2.5}$, NH_4^+, NO_3^-, SO_4^{2-}, Cl^-, Na^+, K^+, Mg^{2+}, Ca^{2+}, Cr, As, Ni, Mn, Cu, Cd, Pb and Al concentrations in each month at SDS site are shown as mean ± standard error (s.e.). All statistical analysis (Pearson correlation analysis and one-way analysis of variance) was performed using the SPSS 16.0 statistical package (SPSS Inc., Chicago, IL).

Results

Mass concentrations of $PM_{2.5}$

As shown in Fig. 1, daily $PM_{2.5}$ concentrations ranged from 101 to 568, 69.8 to 688, 35.6 to 265 and 19.2 to 207 μg m^{-3} in January 2011, 2012, 2013 and 2014, respectively. The monthly average $PM_{2.5}$ concentrations at SDS site were 322 (±26.1), 323 (±37.6), 120 (±10.1) and 78.9 (±8.1) μg m^{-3} in January 2011, 2012, 2013 and 2014, respectively. Compared with the average of January 2011 and 2012, monthly mean $PM_{2.5}$ concentrations in January 2013 and 2014 decreased by 62.8 and 75.5%, showing a highly significant decline (P<0.01). We also found similar decrease in $PM_{2.5}$ in Winter (from October in the first year to April in the next year) in 2012/2013 and 2013/2014 compared with in 2010/2011 and 2011/2012 (Fig. S5, Supporting Information). We summarized weather condition (wind speed, relative humidity and air temperature) for several groups and compared $PM_{2.5}$ concentrations in January from 2011 to 2014 (across four years) under the same or similar weather conditions (Fig. 2). The results reveal significant decrease in $PM_{2.5}$ levels in 2013 and 2014 compared with 2011 and 2012 at the same wind speed, relative humidity and/or air temperature in most cases.

Concentrations of particulate water-soluble inorganic ions, arsenic and metal elements

Concentrations of particulate water-soluble inorganic ions and metal elements in $PM_{2.5}$ at the SDS site in January 2012, 2013 and 2014 are shown in Figs. 3 and 4. Most of the water-soluble inorganic ion (e.g., NH_4^+, NO_3^-, SO_4^{2-}, Cl^-, Na^+, K^+, Mg^{2+}, Ca^{2+}) concentrations decreased significantly (P<0.01) in January 2013 and 2014 compared with those in January 2012, with the sole exception of K^+. Similarly, metal (Cr, Mn, Ni, Cu, Cd, Pb and Al) and arsenic (As) concentrations decreased significantly in January 2013 and 2014 compared with January 2012 (P<0.01).

Discussion

Daily $PM_{2.5}$ concentrations ranged from 19.2 to 688 μg m^{-3} in January from 2011 to 2014 at the SDS site in Urumqi. The Chinese second class standard of daily $PM_{2.5}$ concentrations is 75 μg m^{-3}. The concentrations of $PM_{2.5}$ in all the 27 days during which samples were collected in January 2011 exceeded the Chinese second class standard of Ambient Air Quality Standard. Also, twenty-four day $PM_{2.5}$ concentrations during the 25 days of monitoring in January 2012 exceeded the second class standard of Ambient Air Quality for the $PM_{2.5}$ Standard. By comparison, the situation of $PM_{2.5}$ pollution changed in January 2013 and 2014. Only 6 days of $PM_{2.5}$ concentration measurement in 28 days of monitoring in January 2013 and 12 days during 26 days in January 2014 exceeded the second class standard of Ambient Air Quality Standard. Both monthly $PM_{2.5}$ concentrations and daily $PM_{2.5}$ concentrations in January 2011 and 2012 were significantly higher than those in January 2013 and 2014. We analyzed the meteorological factors in January of 2011, 2012, 2013 and 2014 (Fig. S3, Supporting Information) and found significantly low temperature in January 2011, high relative humidity in 2012, high wind speeds and relatively low precipitation in

2013 compared with the same month (January) in the other three years. However, we observed that weather data for at least one year in each group (2011-2012 or 2013-2014) were not significantly different from the other group when we compared any single weather parameter in January 2011 and 2012 with January 2013 and 2014. In addition, we found the same significant decreasing trend for $PM_{2.5}$ levels in January 2013 and 2014 compared with the same period of 2011 and 2012 when we grouped and compared all monitoring days in different years based on similar weather conditions (e.g., wind speed, relative humidity and air temperature) (Fig. 2). For example, $PM_{2.5}$ concentrations decreased when the wind speed increased from > 1 m s^{-1} to > 2 m s^{-1} during the January from 2011 to 2014, showing that winds were conductive of the spread of $PM_{2.5}$. However, under the same or similar wind speed, $PM_{2.5}$ concentrations were significantly lower in January 2013 and 2014 than in January 2011 and 2012 in most cases. Therefore it can be inferred that meteorological conditions are not the main factors responsible for the significant changes in $PM_{2.5}$ concentrations. Synoptic weather maps at the surface (1000 mb) of northwest China (Fig. S4) show that uniform high pressure and low wind speed prevail over Urumqi through January of each year, further indicating that local pollutant emissions dominate the pollution evolution in our study periods.

The large-scale project “shifting from coal to natural gas” was implemented after the 2012-2013 heating season. The natural gas heating area occupied only a small part of the total heating area during the 2010-2011 and 2011-2012 heating seasons, and in the 2012-2013 heating season the natural gas heating area occupied 76% of the total heating area, and heating with natural gas gradually replaced coal in the main city of Urumqi during the heating period from 2013 to 2014 (Fig. S2, Supporting Information). Since the implementation of the project “shifting from coal to natural

gas", the consumption of natural gas has increased rapidly during the heating season (Figs. S6 and S7, Supporting Information). Meanwhile, coal consumption has decreased by about 5,000,000 tons, sulfur dioxide emissions (SO_2) by about 35,000 tons and soot by about 17,000 tons in the 2012-2013 heating season compared with the 2011-2012 heating season. A further coal consumption saving up to 7 million tons will be achieved in the 2013-2014 heating season.[31] We can conclude that heating with natural gas as a replacement for coal can significantly reduce the winter concentrations of $PM_{2.5}$ in Urumqi.

Coal combustion has been the major source of $PM_{2.5}$.[32] In our study the concentrations of $PM_{2.5}$ were reduced by 62.8% and 75.6% after the introduction of heating with natural gas in January 2013 and 2014 instead of coal in January 2012. Although the project "shifting from coal to natural gas" has significantly reduced the concentration of $PM_{2.5}$, pollution levels are still very high in Urumqi. $PM_{2.5}$ concentrations in Urumqi are much higher than those in Bishkek, where monthly $PM_{2.5}$ concentrations average only 11.7 μg m^{-3}.[33] Both Urumqi and Bishkek are located in the northern Tianshan Mountains, central Asia. The monthly $PM_{2.5}$ concentrations in Urumqi are moderate compared to other cities in China. The $PM_{2.5}$ concentration (120 μg m^{-3}) in January 2013 (after the shift from coal to natural gas for winter heating) in Urumqi was lower than that in Beijing (158 μg m^{-3}) or Xi'an (345 μg m^{-3}) but was still higher than in Shanghai (90.7 μg m^{-3}) or Guangzhou (69.1 μg m^{-3}) during the same period.[34]

Together with the reduction in $PM_{2.5}$ concentrations the water-soluble chemical components in $PM_{2.5}$ (except for K^+ ions) were significantly reduced (Fig. 3). K^+ is derived mainly from biomass burning.[20] It follows that heating with natural gas or coal will not significantly influence the concentration of K^+ in $PM_{2.5}$.

Sulfate ions (SO_4^{2-}) and nitrate ions (NO_3^-) are formed by the oxidation of sulfur dioxide (SO_2) and nitrogen dioxide (NO_2).[25] Most of the SO_2 and NO_x emissions are from coal combustion.[33, 35] Burning coal is known to produce much higher NO_x and SO_2 emissions than burning natural gas.[36] Compared with the 2011-2012 heating period (heating with coal), coal consumption fell by 5,000,000 tons and sulfur dioxide emissions by 200,000 tons during the 2012-2013 heating period (heating with natural gas).[37] The decreased SO_4^{2-} and NO_3^- concentrations in January 2013 and 2014 are attributable to heating with natural gas instead of coal.

In our study, we found that NH_3 concentrations in January in 2013 and 2014 were significant higher than in January in 2011 and 2012 (Fig. S8, Supporting Information). Ammonium ions (NH_4^+) were formed by the reaction of ammonia (NH_3) with acid gases (SO_2, NO_x and HCl). The implementation of the project "shifting from coal to natural gas" reduced emissions of acid gases (SO_2, NO_x and HCl). There were not enough acid gases in the air to react with NH_3 to form ammonium ions. NH_3 in the air was mainly in gaseous form. We infer that supply of heating with natural gas instead of coal can significantly decrease the concentration of NH_4^+ in $PM_{2.5}$ indirectly. In our study, we found a good relationship between SO_4^{2-} and NH_4^+. The correlation coefficients were 0.997 and 0.899 in January 2012 and 2013.

We found a good association between Na^+ with Cl^-, with correlation coefficients of 0.941 and 0.785 in January 2012 and 2013. We can infer that the two ions may have the same sources. Studies by Dou et al.[38] suggested that burning coal can produce HCl and SO_2. In order to reduce SO_2 and HCl emissions, CaO and $NaHCO_3$ are used in power stations to react with H_2SO_4 and HCl.[39] As a result, some PM in the forms of $CaSO_4$, Na_2SO_4, $CaCl_2$ and NaCl are produced. Natural gas combustion produces less SO_2 than does coal burning.[36] There is no evidence to indicate that burning natural gas

can produce HCl. Supply of heating with natural gas instead of coal can therefore decrease the concentrations of Cl^-, Na^+ and Ca^{2+} in $PM_{2.5}$ significantly. Kong et al.[40] noted that coal contains a certain amount of magnesium (Mg). This can be emitted into the atmosphere together with the smoke dust during the combustion of coal. However, burning natural gas cannot lead to this problem. Supply of heating with natural gas instead of coal can therefore significantly decrease the concentration of Mg^{2+} in $PM_{2.5}$.

Lead (Pb) in the urban atmosphere was derived mainly from vehicle exhausts and coal combustion.[41, 42] The number of motor vehicles in Urumqi was about 359 000, 466 000, and 591 000 in March of 2011 and 2012 and June 2013.[43, 44] Theoretically, the concentration of Pb in $PM_{2.5}$ in the atmosphere would be stable or slightly increase with the increasing number of motor vehicles. This is because China has adopted ULP (Un-Leaded Petrol, Pb ≤ 13 mg L^{-1}) since 1 January 2000 and all petrol engines with ULP must be with the EURO III emission standard since 31 December 2009. Therefore per vehicle Pb emissions will be stable but total vehicle Pb emissions may increase slightly during our study period (2011-2014). In fact, the concentration of Pb in the atmosphere in January 2013 was significantly reduced compared with January 2012. Clearly, automobile exhausts were not a major source of Pb in the atmosphere in Urumqi. The decrease in Pb in $PM_{2.5}$ in Urumqi city should be mainly from the decreased Pb emissions from coal combustion due to the replacement of coal by natural gas in winter heating period. We conclude that coal combustion emission of Pb was the major source of the metal in the atmosphere in Urumqi. In contrast, burning natural gas does not generate emissions of Pb to the atmosphere. The concentration of Pb in $PM_{2.5}$ in the atmosphere decreased significantly due to supply of heating with natural gas instead of coal in January 2013.

Compared with burning natural gas, burning coal produced heavy metal emissions (e.g., Cu, Cd, Cr, Mn, Ni, Pb), arsenic (As) and aluminum (Al).[42, 45-49] Supplying heating with natural gas also significantly reduces the concentration of As, Cd, Cu, Cr, Mn, Ni and Al in $PM_{2.5}$ (Fig. 3).

Natural gas comprised 4.73% and coal 68.5% of total energy consumption in 2012 in China. This compares with a global average of 23.9% for gas and 29.9% for coal (Fig. S9, Supporting Information). If we increase the contribution of natural gas to the total energy consumption in China to the world average, we can reduce the consumption of coal by 751 million tons and increase the consumption of natural gas by 583.2 billion cubic meters. Thus, we can reduce emissions of 4.57 million tons of SO_2, 2.87 million tons of NO_x and 0.734 million tons of dust. Compared with the national emissions of SO_2, NO_x and dust, increasing the proportion of natural gas consumption can reduce SO_2 emissions by 21.6%, NO_x emissions by 12.3% and dust emissions by 5.94% (Table 1) and this will help to improve atmospheric quality. If we increase the proportion of natural gas consumption from 4.73% to 23.9%, this will result in an additional consumption of 583.2 billion cubic meters of natural gas every year. The proven recoverable reserves of natural gas are only 3.1 trillion cubic meters. Fortunately, China has rich shale gas resources for which the proven recoverable reserves are 25.08 trillion cubic meters[24]. Shale gas is therefore likely to occupy an important place in the energy supply in the future and may represent an ideal transitional energy source which can reduce air pollution. Of course, the adverse effects of shale energy development (e.g., fugitive emissions of methane, groundwater contamination) should be considered carefully when large scale exploration of shale gas in the future.[50] Meanwhile the proportion of other clean energy sources in particular renewable energy sources (e.g., solar, wind and biomass energy) should also

be increased for better air quality in the future.

Therefore, government decisions or policies will play a positive role in improving air quality. It would be worthwhile for China, especially in rapidly developing regions (e.g. the Yangtze River Delta, the Pearl River Delta and the North China Plain) to take more stringent measures (e.g. stricter emission standards and closure of heavily polluting enterprises) to control the emission of pollutants. Our case study reveals that haze in China will be greatly alleviated in the near future if we take action now.

Acknowledgements: This work was financially supported by the Chinese National Basic Research Program (2014CB954202), the distinguished Talent Program of National Natural Science Foundation of China (40425007), the West Light Foundation of the Chinese Academy of Sciences (XBBS201206) and the research foundation for outstanding young talents of Xinjiang, China (2013721048).

Supporting Information Available

The manuscript entitled 'A Multi-year Assessment of Air Quality Benefits from China's Emerging Shale Gas Revolution: Taking Urumqi as an Example' by Wei Song, Yunhua Chang, Xuejun Liu, Kaihui Li, Yanming Gong, Guixiang He, Xiaoli Wang and Changyan Tian. Our supporting information included 7 pages and 10 figures. This information is available free of charge via the Internet at http://pubs.acs.org/.

References:

(1) Andreae, M. O.; Crutzen, P. J. Atmospheric aerosols: Biogeochemical sources and role in atmospheric chemistry. *Science.* **1997**, *276*, 1052-1058.

(2) Mahowald, N.; Ward, D. S.; Kloster, S.; Flanner, M. G.; Heald, C. L.; Heavens, N. G.; Hess, P. G.; Lamarque, J.-F.; Chuang, P. Y. Aerosol impacts on climate and biogeochemistry. *Annu. Rev. Env. Resour.* **2011**, *36*, 45-74.

(3) Lambert, F.; Kug, J. S.; Park, R. J.; Mahowald, N.; Winckler, G.; Abe-Ouchi, A.; O'ishi, R.; Takemura, T.; Lee, J. H. The role of mineral-dust aerosols in polar temperature amplification. *Nat Clim Change.* **2013**, *5*, 487-491.

(4) Paasonen, P.; Asmi, A.; Petaja, T.; Kajos, M. K.; Aijala, M.; Junninen, H.; Holst, T.; Abbatt, J. P. D.; Arneth, A.; Birmili, W.; van der Gon, H. D.; Hamed, A.; Hoffer, A.; Laakso, L.; Laaksonen, A.; Leaitch, W. R.; Plass-Dulmer, C.; Pryor, S. C.; Raisanen, P.; Swietlicki, E.; Wiedensohler, A.; Worsnop, D. R.; Kerminen, V. M.; Kulmala, M. Warming-induced increase in aerosol number concentration likely to moderate climate change. *Nat Geosci*. **2013**, *6*, 438-442.

(5) Dockery, D. W.; Pope, C. A.; Xu, X.; Spengler, J. D.; Ware, J. H.; Fay, M. E.; Ferris, B. G.; Speizer, F. E. An association between air pollution and mortality in six U.S. cities. *N Engl J Med*. **1993**, *329*, 1753-1759.

(6) Shah, A. S. V.; Langrish, J. P.; Nair, H.; McAllister, D. A.; Hunter, A. L.; Donaldson, K.; Newby, D. E.; Mills, N. L. Global association of air pollution and heart failure: A systematic review and meta-analysis. *Lancet*. **2013**, *382*, 1039-1048.

(7) Cao, J. J.; Wang, Q. Y.; Chow, J. C.; Watson, J. G.; Tie, X. X.; Shen, Z. X.; Wang, P.; An, Z. S. Impacts of aerosol compositions on visibility impairment in Xi'an,

China. *Atmos. Environ.* **2012**, *59*, 559-566.

(8) Wang, Y.; Li, L.; Chen, C.; Huang, C.; Huang, H.; Feng, J.; Wang, S.; Wang, H.; Zhang, G.; Zhou, M.; Cheng, P.; Wu, M.; Sheng, G.; Fu, J.; Hu, Y.; Russell, A. G.; Wumaer, A. Source apportionment of fine particulate matter during autumn haze episodes in Shanghai, China. *J Geophys Res-Atmos*. **2014**, *119*, 1903-1914.

(9) Zhao, X.; Zhao, P.; Xu, J.; Meng, W.; Pu, W.; Dong, F.; He, D.; Shi, Q. Analysis of a winter regional haze event and its formation mechanism in the North China Plain. *Atmos Chem Phys*. **2013**, *13*, 5685-5696.

(10) Huang, W.; Cao, J.; Tao, Y.; Dai, L.; Lu, S. E.; Hou, B.; Wang, Z.; Zhu, T. Seasonal variation of chemical species associated with short-term mortality effects of $PM_{2.5}$ in Xi'an, a central city in China. *Am J Epidemiol*. **2012**, *175*, 556-566.

(11) Meng, X.; Ma, Y.; Chen, R.; Zhou, Z.; Chen, B.; Kan, H. Size-fractionated particle number concentrations and daily mortality in a Chinese city. *Environ Health Persp*. **2013**, *121*, 1174-1178.

(12) Gong, J.C.; Zhu, T.; Kipen, H.; Wang, G.F.; Hu, M.; Guo, Q.F.; Ohman-Strickland, P.; Lu, S.E.; Wang, Y.D.; Zhu, P.; Rich, Q.R.; Huang, W.; Zhang, J.F. Comparisons of ultrafine and fine particles in their associations with biomarkers reflecting physiological pathways. *Environ Sci Technol*. **2014**, *48*, 5264-5273.

(13) He, K.; Zhao, Q.; Ma, Y.; Duan, F.; Yang, F. Spatial and seasonal variability of $PM_{2.5}$ acidity at two Chinese megacities: Insights into the formation of secondary inorganic aerosols. *Atmos Chem Phys*. **2012**, *12*, 1377-1395.

(14) Zhang, R.; Jing, J.; Tao, J.; Hsu, S. C.; Wang, G.; Cao, J.; Lee, C. S. L.; Zhu, L.;

390 Chen, Z.; Zhao, Y.; Shen, Z. Chemical characterization and source
391 apportionment of $PM_{2.5}$ in Beijing: Seasonal perspective. *Atmos Chem Phys*.
392 **2013**, *13*, 7053-7074.
393 (15) Tao, J.; Gao, J.; Zhang, L.; Zhang, R.; Che, H.; Zhang, Z.; Lin, Z.; Jing, J.; Cao,
394 J.; Hsu, S. C., $PM_{2.5}$ pollution in a megacity of southwest China: Source
395 apportionment and implication. *Atmos Chem Phys Discus*. **2014**, *14*,
396 5147-5196.
397 (16) Chen, Y. Y.; Ebenstein, A.; Greenstone, M.; Li, H. B. Evidence on the impact of
398 sustained exposure to air pollution on life expectancy from China's Huai River
399 policy. *Proc. Natl. Acad. Sci*. **2013**, *10*, 12936-12941.
400 (17) McElroy, M. B.; Lu, X.; Nielsen, C. P.; Wang, Y. X. Potential for
401 wind-generated electricity in China. *Science*. **2009**, *325*, 1378-1380.
402 (18) Ou, X. M.; Zhang, X. L.; Chang, S.Y. Scenario analysis on alternative fuel /
403 vehicle for China's future road transport: Life-cycle energy demand and GHG
404 emissions. *Energy Policy*. **2010**, *38*, 3943–3956.
405 (19) Hu, Y.; Cheng, H. Development and bottlenecks of renewable electricity
406 generation in China: A critical review. *Environ Sci Technol*. **2013**, *47*,
407 3044-3056.
408 (20) Cheng, Y.; Engling, G.; He, K. B.; Duan, F. K.; Ma, Y. L.; Du, Z. Y.; Liu, J. M.;
409 Zheng, M.; Weber, R. J. Biomass burning contribution to Beijing aerosol.
410 *Atmos Chem Phys*. **2013**, *13*, 7765-7781.
411 (21) Wang, Q.; Chen, X.; Jha, A. N.; Rogers, H. Natural gas from shale formation –
412 The evolution, evidences and challenges of shale gas revolution in United
413 States. *Renew Sust Energ Rev*. **2014**, *30*, 1–28.
414 (22) Vidic, R. D.; Brantley, S. L.; Vandenbossche, J. M.; Yoxtheimer, D.; Abad, J. D.

Impact of shale gas development on regional water quality. *Science*. **2013**, *340*, 1-9.

(23) Small, M.J.; Stern, P. C.; Bomberg, E.; Christopherson, S. M.; Goldstein, B. D.; Israel, A. L.; Jackson, R. B.; Krupnick, A.; Mauter, M. S.; Nash, J.; North, D. W.; Olmstead, S. M.; Prakash, A.; Rabe, B.; Richardson, N.; Tierney, S.; Webler, T.; Wong-Parodi, G.; Zielinska, B., Risks and risk governance in unconventional shale gas development. *Environ Sci Technol*. **2014**, *48*, 8289-8297.

(24) Chang, Y. H.; Liu, X. J.; Christie, P. Emerging shale gas revolution in China. *Environ Sci Technol*. **2012**, *46*, 12281-12282.

(25) Shen, J. L.; Tang, A. H.; Liu, X. J.; Kopsch, J.; Fangmeier, A.; Goulding, K.; Zhang, F. S. Impacts of pollution controls on air quality in Beijing during the 2008 Olympic Games. *J Environ Qual*. **2011**, *40*, 37-45.

(26) Guo, S.; Hu, M.; Guo, Q.; Zhang, X.; Schauer, J. J.; Zhang, R. Quantitative evaluation of emission control of primary and secondary organic aerosol sources during Beijing 2008 Olympics. *Atmos Chem Phys*. **2013**, *13*, 8303-8314.

(27) Li, X. H.; He, K. B.; Li, C. C.; Yang, F. M.; Zhao, Q.; Ma, Y. L.; Cheng, Y.; Ouyang, W. J.; Chen, G. C. $PM_{2.5}$ mass, chemical composition and light extinction before and during the 2008 Beijing Olympics. *J Geophys Res-Atmos*. **2013**, *118*, 12158-12167.

(28) Zhang, M.; Chen, J. M.; Chen, X. Y.; Cheng, T. T.; Zhang, Y. L.; Zhang, H. F.; Ding, A. J.; Wang, M.; Mellouki, A. Urban aerosol characteristics during the World Expo 2010 in Shanghai. *Aerosol Air Qual Res*. **2013**, *13*, 36-48.

(29) Huang, K.; Zhuang, G.; Lin, Y.; Wang, Q.; Fu, J. S.; Fu, Q.; Liu, T.; Deng, C.

How to improve the air quality over megacities in China: Pollution characterization and source analysis in Shanghai before, during, and after the 2010 World Expo. *Atmos Chem Phys*. **2013**, *13*, 5927-5942.

(30) http://www.wunderground.com

(31) http://news.iyaxin.com/content/2013-09/29/content_4234760.htm

(32) Reddy, M. S.; Venkataraman, C. Inventory of aerosol and sulphur dioxide emissions from India: I - Fossil fuel combustion. *Atmos Environ*. **2002**, *36*, 677-697.

(33) Chen, B. B.; Sverdlik, L. G.; Imashev, S. A.; Solomon, P. A.; Lantz, J.; Schauer, J. J.; Shafer, M. M.; Artamonova, M. S.; Carmichael, G. Empirical relationship between particulate matter and aerosol optical depth over Northern Tien-Shan, Central Asia. *Air Qual Atmos Hlth*. **2013**, *6*, 385-396.

(34) Huang, R. L.; Zhang, Y. L.; Bozzetti, C.; Ho, K. F.; Cao, J. J.; Han, Y. M.; Daellenbach, K. R.; Slowik, J. G; Platt, S. M.; Canonaco, F.; Zotter, P.; Wolf R.; Pieber, S. M.; Bruns, E. A.; Crippa, M.; Ciarelli, G.; Piazzalunga, A.; Schwikowski, M.; Abbaszade, G.; Schnelle-Kreis, J.; Zimmermann, R.; An, Z.; Szidat, S.; Baltensperger, U.; Haddad, I. E.; Prévôt, A. S. H. High secondary aerosol contribution to particulate pollution during haze events in China. *Nature*. **2014**, *514*, 218-222.

(35) Zhao, Y.; Wang, S. X.; Duan, L.; Lei, Y.; Cao, P. F.; Hao, J. M. Primary air pollutant emissions of coal-fired power plants in China: Current status and future prediction. *Atmos Environ*. **2008**, *42*, 8442-8452.

(36) Huo, H.; Zhang, Q.; Liu, F.; He, K. B. Climate and environmental effects of electric vehicles versus compressed natural gas vehicles in China: A life-cycle analysis at provincial level. *Environ Sci Technol*. **2013**, *47*, 1711-1718.

(37) http://www.iyaxin.com

(38) Dou, B. L.; Wang, C.; Chen, H. S.; Song, Y. C.; Xie, B. Z.; Xu, Y. J.; Tan, C. Q. Research progress of hot gas filtration, desulphurization and HCl removal in coal-derived fuel gas: A review. *Chem Eng Res Des*. **2012**, *90*, 1901-1917.

(39) Shemwell, B. E.; Ergut, A.; Levendis, Y. A. Economics of an integrated approach to control SO_2, NO_x, HCl, and particulate emissions from power plants. *J Air Waste Manage*. **2002**, *52*, 521-534.

(40) Kong, S. F., Ji, Y. Q.; Lu, B.; Chen, L.; Han, B.; Li, Z. Y.; Bai, Z. P. Characterization of PM_{10} source profiles for fugitive dust in Fushun-a city famous for coal. *Atmos Environ*. **2011**, *45*, 5351-5365.

(41) Monaci, F.; Moni, F.; Lanciotti, E.; Grechi, D.; Bargagli, R. Biomonitoring of airborne metals in urban environments: New tracers of vehicle emission, in place of lead. *Environ Pollut*. **2000**, *107*, 321-327.

(42) McConnell, J. R.; Edwards, R. Coal burning leaves toxic heavy metal legacy in the Arctic. *Proc. Natl. Acad. Sci*. **2008**, *105*, 12140-12144.

(43) http://www.iyaxin.com

(44) http://www.urumqi.gov.cn

(45) Xie, R. K.; Seip, H. M.; Wibetoe, G.; Nori, S.; McLeod, C. W. Heavy coal combustion as the dominant source of particulate pollution in Taiyuan, China, corroborated by high concentrations of arsenic and selenium in PM_{10}. *Sci Total Environ*. **2006**, *370*, 409-415.

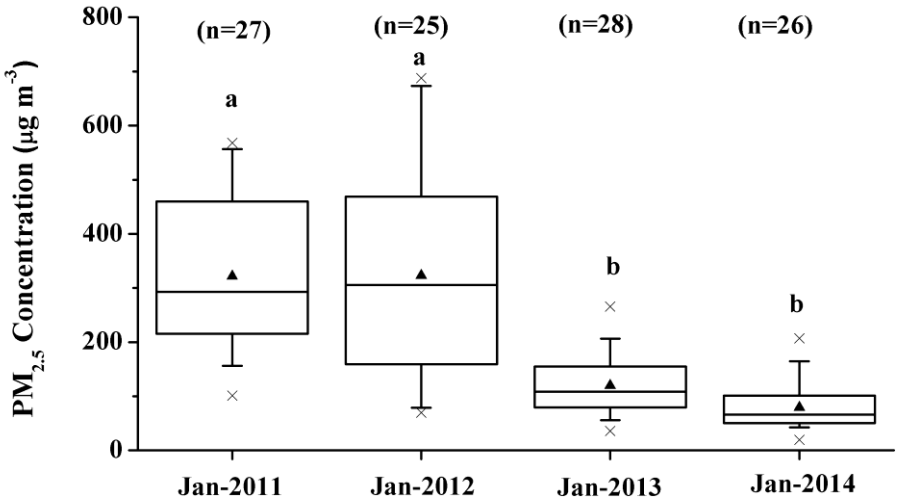
(46) An, D.; Li, D. S.; Liang, Y.; Jing, Z. J. Unventilated indoor coal-fired stoves in Guizhou province, China: Reduction of arsenic exposure through behavior changes resulting from mitigation and health education in populations with arsenicosis. *Environ Health Persp*. **2007**, *15*, 659-662.

(47) Huggins, F. E.; Senior, C. L.; Chu, P.; Ladwig, K.; Huffman, G. P. Selenium and arsenic speciation in fly ash from full-scale coal-burning utility plants. *Environ Sci Technol*. **2007**, *41*, 3284-3289.

(48) Qiao, X. C.; Si, P.; Yu, J. G. A systematic investigation into the extraction of aluminum from coal spoil through Kaolinite. *Environ Sci Technol*. **2008**, *42*, 8541-8546.

(49) Galiulin, R. V.; Galiulina, R. A. Heavy metal pollution in the territory of Chelyabinsk upon coal combustion. *Solid Fuel Chem*. **2013**, *47*, 129-131.

(50) Stern, P. C.; Webler, T.; Small, M. T.; Special issue: Understanding the risks of unconventional shale gas development. *Environ Sci Technol*. **2014**, *48*, 8287-8288.



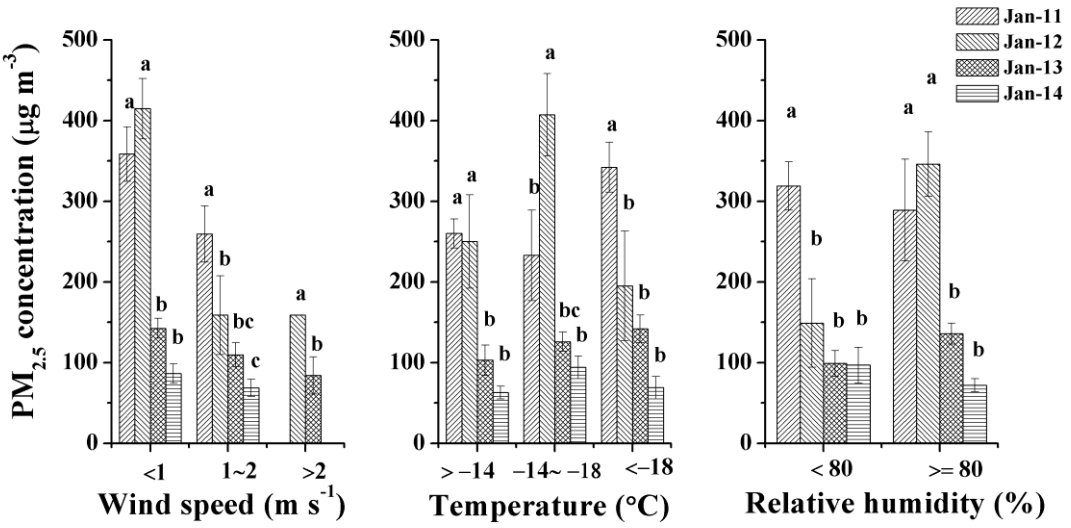
503

504 **Fig. 1.** Comparison of the monthly mean $PM_{2.5}$ concentrations in January 2011, 2012,
505 2013 and 2014. The black line and triangle, lower and upper edges, bars and forks in
506 or outside the boxes represent median and mean values, 25th and 75th, 5th and 95th,
507 and 5th and 95th percentiles of all data, respectively. (Values in row without same
508 letters are significantly different at $p<0.01$).

509 n: sample size

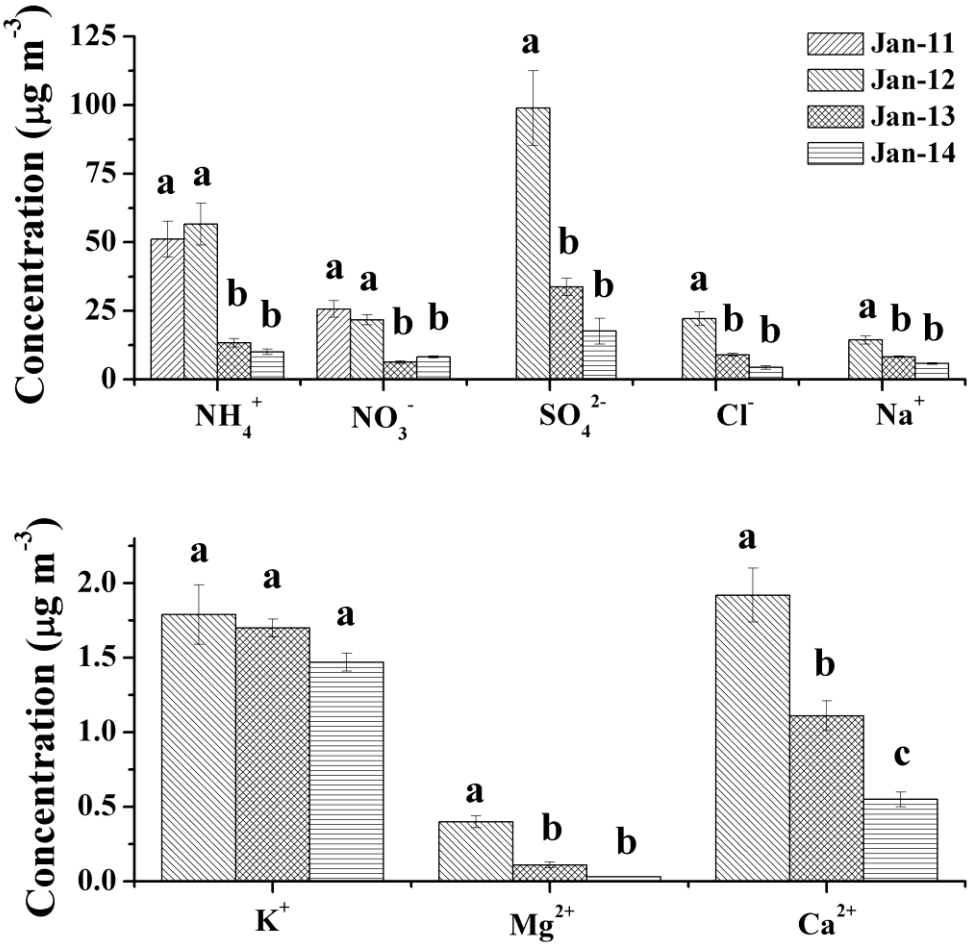
510

511



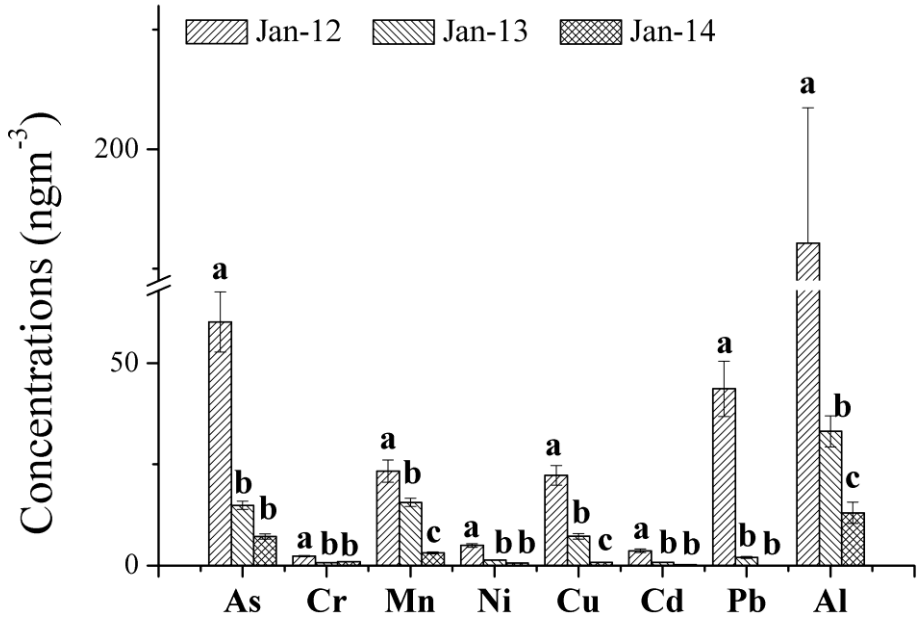
512

513 **Fig. 2.** Comparison of $PM_{2.5}$ concentrations under the conditions of the same wind
514 speed, a similar range of temperature and humidity changes. (Values in row without
515 same letters are significantly different at $p<0.01$).
516



517

518 **Fig. 3.** Concentrations of water-soluble inorganic ions in $PM_{2.5}$ at SDS site during
519 January 2011, January 2012, January 2013 and January 2014 in Urumqi (Values in
520 row without same letters are significantly different at $p<0.01$).



521

522 **Fig. 4.** Concentrations of metal elements in $PM_{2.5}$ at SDS site during January 2012,

523 January 2013 and January 2014 in Urumqi (Values in row without same letters are

524 significantly different at $p<0.01$).

	Coal			Natural gas					
	751 million tons (coal saving potential if its use reduced to 49.3% of national energy supply)			583.2 billion cubic meters (additional gas required if its use raised to 23.9% of national energy supply)					
	Emission factor (T / tce) [1,2]	Removal efficiency (%) [3-5]	Emissions (million tons)	Emission factor (T / one million cubic meters) [2]	Removal efficiency (%)	Emission (million tons)	Reduction (million tons)	National emissions in 2012 (million tons) [6]	Reduction rate (%)
SO_2	0.016	60.8	4.71	0.63	60.8	0.144	4.57	21.18	21.6
NO_x	0.009	40.0	4.06	3.40	40.0	1.19	2.87	23.38	12.3
Dust	0.01	90.0	0.75	0.29	90.0	0.0167	0.733	12.36	5.94

Table 1. SO_2, NO_x and dust emissions from reduced coal and increased natural gas, SO_2, NO_x and dust reduction and reduction rate

1. Chemical Energy Saving Technical Manual. Chemical Industry Press, 2006
2. http://www.fchbw.com/Item/949.aspx
3. Wang, F.Q., Du, Y.G., Liu, Y., Wang, X.M., Development Status and Recommendations on Flue Gas Denitration in Coal-fired Power Plant in China. *Research Progress*, **2007**, *1*, 18-22 (in Chinese)
4. Liu, Z.Q., Chen, T., Liu, J., Status of Coal Utilization in Shenyang and Analysis on Countermeasures in Pollution Control of Coal Burning. *Environmental Science*, **2009**,*35*, 9-12 (in Chinese)
5. Li, M.S., Zhang, J.H., Luo, H.J., Lin, L.Y., Li, Q., Zhang, Y.J., Sulfur Dioxide Reduction and Potential in China. *Scientia Geographica Sinica* **2011**,*31*, 1065-1071 (in Chinese)
6. China Statistical Yearbook 2013. China Statistics Press, Beijing