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Atmospheric Chemistry and Physics
Discussions

The control of anthropogenic emissions contributed to 80% of the decrease in $PM_{2.5}$ concentrations in Beijing from 2013 to 2017

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Abstract

With the completion of the Beijing Five-year Clean Air Action Plan by the end of 2017, the annual mean $PM_{2.5}$ concentrations in Beijing dropped dramatically to 58.0 μg/m^3 in 2017 from 89.5 μg/m^3 in 2013. However, controversies exist to argue that favorable meteorological conditions in 2017 that helped pollution dispersion were the major factor for such rapid decrease in $PM_{2.5}$ concentrations. To comprehensively evaluate this five-year plan, we employed Kolmogorov-Zurbenko (KZ) filtering and a WRF-CMAQ model to quantify the relative contribution of meteorological conditions and the control of anthropogenic emissions to $PM_{2.5}$ reduction in Beijing from 2013 to 2017. For these five years, the relative contribution of emission-reduction measures to the decrease of $PM_{2.5}$ concentrations in Beijing calculated by KZ filtering and the WRF-CMAQ model was 80.6% and 78.6% respectively. The WRF-CMAQ model further revealed that local and regional emission-reduction measures contributed to 53.7% and 24.9% of the $PM_{2.5}$ reduction respectively. For local emission-reduction measures, the

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Atmospheric Chemistry and Physics
Discussions

regulation of coal boilers, increasing clean fuels for residential use, industrial restructuring, the regulation of raise dust and vehicle emissions contributed to 20.1 %, 17.4%, 10.8%, 3.0 % and 2.4% of $PM_{2.5}$ reduction respectively. Both models suggested that the control of anthropogenic emissions contributed to around 80% of the total decrease in $PM_{2.5}$ concentrations in Beijing, indicating that emission control was crucial for the notable improvement in air quality in Beijing from 2013 to 2017. Therefore, such long-term air quality clean plan should be continued for the future years to further reduce $PM_{2.5}$ concentrations in Beijing. Considering that different emission-reduction measures exert distinct effects on $PM_{2.5}$ reduction and existing emission-reduction measures work poorly to reduce ozone concentrations, future strategies for emission-reduction should be designed and implemented accordingly.

Keywords: PM2.5 reduction, anthropogenic emissions, meteorological conditions, Kolmogorov-Zurbenko (KZ) filtering, WRF-CMAQ

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1 Introduction

In December 2012, a heavy haze episode occurred in Beijing, during which the highest hourly $PM_{2.5}$ concentrations once reached 886 μg/m^3, a historical record. The extremely high $PM_{2.5}$ concentrations led to long-lasting black and thick fogs, which not only significantly influenced people's daily life (low-visibility induced traffic jam), but also exerted strong negative influences on public health (Brunekreef et al., 2002; Dominici et al., 2014; Nel et al., 2005; Zhang et al., 2012; Qiao et al., 2014). Since then, severe haze episodes have frequently occurred in Beijing and other regions in China (Chan et al., 2008; Huang, R., et al., 2014; Guo et al.,2014; Zheng et al.,2015), and $PM_{2.5}$ pollution has become one of the most concerned environmental issues in China. Since 2013, a national network of ground stations for monitoring hourly $PM_{2.5}$ concentrations has been established gradually, including 35 ground observation stations in Beijing, which provide important support for proper management and in-depth investigation of $PM_{2.5}$ concentrations. Meanwhile, for effectively reducing local $PM_{2.5}$ concentrations, the local government proposed the Beijing Five-year Clean Air Action Plan (2013-2017). This plan suggested the specific aim that the annual mean $PM_{2.5}$ concentrations in Beijing should be reduced from 89.5 μg/m^3 in 2013 to 60 μg/m^3 in 2017 and included a series of emission-reduction measures, including shutting down heavily polluting factories, restricting traffic emissions and replacing coal fuels with clean energies. Furthermore, for reducing high $PM_{2.5}$ concentrations during severe haze episodes, Beijing Municipal Government published the "Heavy Air Pollution Contingency Plan" in 2012, and further revised this plan in March 2015. According to this plan, a series of contingent emission reduction measures should be implemented according to the severeness of $PM_{2.5}$ pollution episodes. By the end of 2017, these long-term and contingent emission-reduction measures had worked together to reduce the annual mean $PM_{2.5}$ in Beijing to 58.0 μg/m^3, indicating a great success of $PM_{2.5}$ management during the past five years.

In addition to anthropogenic emissions, the strong meteorological influences on $PM_{2.5}$ concentrations in Beijing have been widely acknowledged (Cheng et al., 2017; Chen, Z. et al., 2016, 2017, 2018; UNEP, 2016; Wang et al., 2014; Zhao et al., 2013). For instance, Chen, Z et al. (2016) found that for 2014, more than 180 days in Beijing experienced a dramatic AQI (Air Quality Index) change (ΔAQI>50), compared with the previous day. Considering the total emission of airborne pollutants for a mega city hardly change significantly on a daily basis, the rapid variation of meteorological conditions in Beijing was one important driver for the dramatic change of daily air quality in Beijing. In this case，there arises the controversy that meteorology, instead of emission-reduction measures，made a major contribution to the remarkable reduction of $PM_{2.5}$ concentrations in Beijing from 2013 to 2017. With the completion of the five-year plan, it is highly necessary to quantify the relative contribution of meteorological conditions and emission-reduction measures to the remarkable decrease of $PM_{2.5}$ concentrations in Beijing.

To this end, we employ different approaches in this paper to comprehensively estimate adjusted $PM_{2.5}$ concentrations in Beijing while eliminating the influence from the variation in meteorological conditions and thus quantify the relative contribution of emission-reduction measures to the decrease of $PM_{2.5}$ concentrations. In this light, this research provides important insight for better designing and implementing successive clean air plans in the future to further mitigate $PM_{2.5}$ pollution in Beijing.

2 Data Sources

2.1 $PM_{2.5}$ and meteorological data

In this study, hourly $PM_{2.5}$ concentration data were acquired from the website PM25.in, which collects official data provided by China National Environmental Monitoring Center (CNEMC). Beijing has established an advanced air quality monitoring network with 35 ground stations across the city. Considering the major contribution of industry and traffic-induced emissions in urban areas, we selected all twelve urban stations to analyze the variation of $PM_{2.5}$ concentrations and quantify their influencing factors. In addition to these urban stations, we also selected two background stations, the DingLing Station located in the suburb and the MiYun Reservoir Station located in the outer suburb, one transportation station (the Qianmen station) located close to a main road, and one rural station (the Yufa Station) which is far away from central Beijing for the following analysis. The DingLing and MiYun Reservoir Stations were chosen as background stations by the Ministry of Environmental Protection of China. These two stations receive limited influence from anthropogenic emissions due to their location in suburban and outer suburban areas. Comparing the variation in $PM_{2.5}$ concentrations and its anthropogenic and meteorological driving factors in different type of stations provides a useful reference for comprehensively understanding the effects of emission-reduction measures on the reduction of $PM_{2.5}$ concentrations in Beijing in the past five years. The locations of these selected stations are shown in Fig 1. Meteorological data for this research were collected from the Guanxiangtai Station (GXT,54511, 116.46 ° E, 39.80 ° N), Beijing and were downloaded from the Department of

Atmospheric Science, College of Engineering, University of Wyoming (http://weather.uwyo.edu/upperair/sounding.html). Both the $PM_{2.5}$ and meteorological data were collected from January 1st, 2013 to December 31st, 2017.

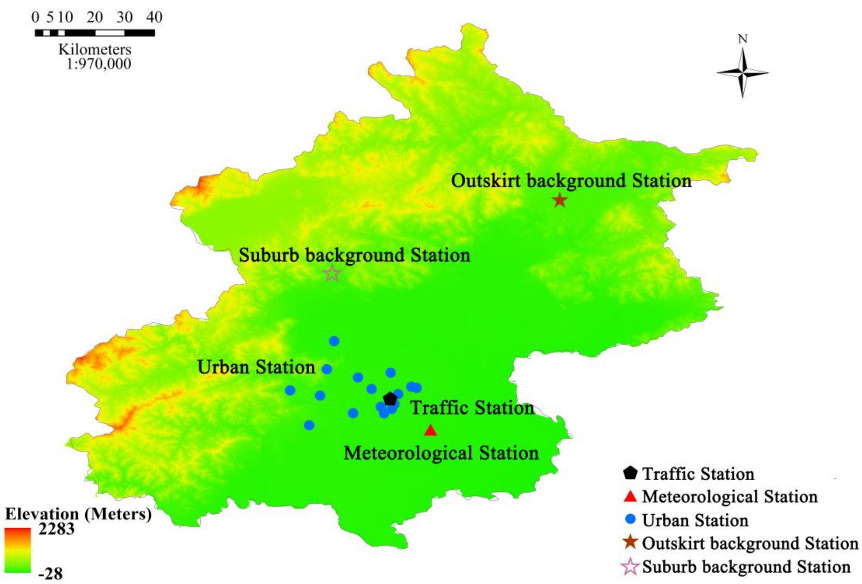


Fig 1. Locations of different ground monitoring stations.

2.2 Emission inventories

For this research, we employed both regional and local emission inventories for running model simulation. Multi-resolution Emission Inventory for China, MEIC, (http://meicmodel.org/) provided by Tsinghua University, were employed as the regional emission inventories. MEIC has been widely employed and verified as a reliable emission inventory by a diversity of studies (Hong et al., 2017; Saikawa et al., 2017; Zhou et al., 2017; etc.). Different from regional emission inventories, local emission inventories are usually produced independently by local institutes. The Beijing local-emission inventories employed for this research is produced and updated by Beijing Municipal Research Institute of Environmental protection fully according to the requirement of MEP on the production of local emission inventories within the Beijing-Tianjin-Hebei region. This local-emission inventory is produced by synthesizing local environmental statistical data and reported emission data, carrying out field investigations and conducting a series estimation according to Beijing Five-year Clean Air Action

Plan. This Beijing local-emission inventory has been formally employed for the implementation of recent "2017 Air Pollution Prevention and Management Plan for the Beijing-Tianjin-Hebei Region and its Surrounding Areas" (MEP, 2017).

3 Methods

A key step for quantifying the relative contribution of anthropogenic emissions to the decrease of $PM_{2.5}$ concentrations is to properly filter meteorological influences on $PM_{2.5}$ concentrations, which is highly challenging and rarely investigated by previous studies. Therefore, we employed both a statistical method and a chemical transport model in this study to comprehensively evaluate the role of anthropogenic emissions and meteorological conditions in the decrease of $PM_{2.5}$ concentrations in Beijing during the past five years.

3.1 Kolmogorov-Zurbenko (KZ) filtering

Since meteorological conditions exert a strong influence on $PM_{2.5}$ concentrations in Beijing, the removal of seasonal signals from time series of meteorological factors results in data sets suitable for understanding the trend of $PM_{2.5}$ concentrations mainly influenced by anthropic factors (Eskridge et al., 1997). To better analyze the trend of time series data without the disturbances from large variations of influencing variables, a statistical method called Kolmogorov-Zurbenko (KZ) filtering was proposed by Rao et al. (1994). The KZ filter is advantageous in removing high-frequency variations in the data set based on the iterative moving average. Eskridge et al. (1997) compared four major approaches for trend detection, including PEST, anomalies, wavelet transform, and the KZ filter, and suggested that the confidence in detecting long-term trend of the KZ filter was much higher than that of the other methods. Due to its reliable performance in trend detection in complicated ecosystems, the KZ filter has frequently been employed to remove seasonal signals of meteorological conditions and extract long-term trend of airborne pollutants (Zurbenko, et al., 1996; Eskridge, et al., 1997; Kang, et al., 2013). One potential limitation of the KZ filter is that iterative moving average (m) may impose an influence on detecting abrupt changes of variations. Therefore, Zurbenko et al.（1996）proposed an enhanced KZ filter that employed a dynamic variable m that decreases with the increase in changing rate, which is employed in this study to estimate the modified $PM_{2.5}$ concentrations in Beijing by removing large seasonal variations in meteorological conditions. The principle of the KZ filter is briefly introduced as follows.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

Atmospheric Chemistry and Physics
Discussions

The raw time-series data of airborne pollutants can be decomposed as:

$$X(t)=\mathrm{E}(t)+\mathrm{S(t)}+\mathrm{W(t)} \qquad (1)$$

$$X_b(\mathrm{t})=\mathrm{E(t)}+\mathrm{S(t)} \qquad (2)$$

$$\mathrm{E(t)}=\mathrm{KZ}_{365,3}(X) \qquad (3)$$

$$\mathrm{S(t)}=\mathrm{KZ}_{15,5}(X)-\mathrm{KZ}_{365,3}(X) \qquad (4)$$

$$\mathrm{W(t)}=\mathrm{X(t)}-\mathrm{KZ}_{15,5}(X) \qquad (5)$$

Where X (t) is the original time series of airborne pollutants, E(t) is the long-term trend component, S(t) is the seasonal variation, W(t) is the residue or synoptic-scale (short-term) variations. $KZ_{i,\ j}$(X) indicates a KZ filtering on the original dataset X with a moving wind size of i and j iterations.

X_b (t) stands for the base component, the sum of the long-term trend component and seasonal variation, presenting steady trend variation. E(t) is mainly effected by long-term anthropogenic emission and climate change. S(t) is mainly influenced by the seasonal variation of emission factors and meteorological conditions. The residue W(t) is caused by short-term and small-scale shifts of emissions and meteorological conditions.

The long-term trend component E(t) processed by KZ filtering still contains the influence of meteorological conditions, which can be removed by multiple regression models. Multiple linear relationships are established for the residue and baseline component respectively using strongly correlated meteorological factors.

We conducted correlation analysis between $PM_{2.5}$ concentrations and a series of meteorological factors, including temperature, wind speed, wind direction, precipitation, relative humidity, solar radiation, evaporation and air pressure. The correlation analysis revealed that wind speed, relative humidity, temperature, solar radiation and air pressure were strongly and significantly correlated with $PM_{2.5}$ concentrations in Beijing, which was consistent with the findings from previous studies (Sun et al., 2013; Chen, Z., et al., 2017, 2018; Wang et al., 2018). Therefore, we further established multiple linear regression equations between $PM_{2.5}$ concentrations and wind speed, relative humidity, temperature and solar radiation as follows.

$$W(\mathrm{t})=a_0+\sum a_i \mathrm{w}_i(t)+\varepsilon_w(t) \qquad (6)$$

$$X_b(\mathrm{t})=\mathrm{b}_0+\sum b_i x_i(t)+\varepsilon_b(t) \qquad (7)$$

$$\varepsilon(t)=\varepsilon_w(t)+\varepsilon_b(t) \qquad (8)$$

Where $w_i(\mathrm{t})$ and $x_i(\mathrm{t})$ stand for the different synoptic-scale variations and baseline component of the i[th] meteorological factor. ε_w and ε_b is the regression residue of the synoptic-scale variations and baseline component. $\varepsilon(\mathrm{t})$ indicates the total residue, including the short-term influence of local emission sources, meteorological influences neglected during the regression and noise.

Next, KZ filtering is conducted on the $\varepsilon(\mathrm{t})$ for its long-term component $\varepsilon_E(t)$. After the variation of meteorological influences was filtered, the reconstructed time series of airborne pollutants $X_{LT}(t)$ was calculated as the sum of $\varepsilon_E(t)$ and the average value of E(t) , $\overline{E(t)}$.

$$X_{LT}(t)=\overline{E(t)}+e_E(t) \tag{9}$$

After KZ filtering, the relative contribution of meteorological conditions to the variation in $PM_{2.5}$ concentrations can be calculated as follows:

$$P_{contrib}=\frac{K_{org}-K}{K_{org}}\times 100\% \tag{10}$$

Where $P_{contrib}$ is the relative contribution of meteorological conditions to the variation of $PM_{2.5}$ concentrations in Beijing, K_{org} is the variation slope of the original $PM_{2.5}$ time series; K is the variation slope of adjusted $PM_{2.5}$ time series after meteorological variations are removed.

3.2 WRF-CMAQ model

We employed the WRF-CMAQ model for simulating the effects of emission-reduction measures on the reduction of $PM_{2.5}$ concentrations. The WRF-CMAQ model includes three models: The middle-scale meteorology model (WRF), the source emission model (SMOKE) (http://www.cmascenter.org/smoke/) and the community multiscale air quality modeling system (CMAQ) (http://www.cmascenter.org/CMAQ). The center of the CMAQ was set at coordinate 35 °N, 110 °E and a bi-directional nested technology was employed, producing two layers of grids with a horizontal resolution of 36 km and 12 km respectively. The first layer of grids with 36km resolution and 200×160 cells covered most areas in East Asia (including China, Japan, North Korea, South Korea, and other countries). The second layer of grids with 12km resolution and 120×102 cells covered the North China Plain (including the Beijing-Tianjin-Hebei region, and Shandong and Henan Provinces). The vertical layer was divided into 20 unequal layers, eight of which were of a distance of less than 1km to the ground for better featuring the structure of atmospheric boundary. The height of the ground layer was 35m.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

We employed ARW-WRF3.2 to simulate the meteorological field. The setting of the center and the bi-directional nest for the WRF was similar to that of the CMAQ as mentioned above. There were 35 vertical layers for the WRF and the outer layer provided boundary conditions of the inner layer. The meteorological background field and boundary information with a FNL resolution of 1°×1° and temporal resolution of 6h were acquired from NCAR (National Center for Atmospheric Research, https://ncar.ucar.edu/) and NCEP (National Centers for Environmental Prediction) respectively. The terrain and underlying surface information was obtained from the USGS 30s global DEM (https://earthquake.usgs.gov/). The output from the WRF model was interpolated to the region and grid of the CMAQ model using the Meteorology-Chemistry Interface Processor (MCIP, https://www.cmascenter.org/mcip). The meteorological factors used for this model includes temperature, air pressure, humidity, geopotential height, zonal wind, meridional wind, precipitation, boundary layer heights and so forth. An estimation model for terrestrial ecosystem MEGAN (http://ab.inf.uni-tuebingen.de/software/megan/) was employed to process the natural emissions. Anthropogenic emission data were from the Multi-resolution Emission Inventory for China, MEIC 0.5°×0.5° emission inventory (http://www.meicmodel.org/) and Beijing emission inventory (http://www.cee.cn/). We input the processed natural and anthropogenic emission data into the SMOKE model and acquired comprehensive emission source files.

Scenario simulation is employed to estimate the contribution of emission-reduction to the variation in $PM_{2.5}$ concentrations.

$$P_{contrib} = \frac{C - C_{base}}{C} \times 100\% \qquad (11)$$

Where $P_{contrib}$, C and C_{base} are the contribution rate of emission reduction to $PM_{2.5}$ concentrations, the simulated $PM_{2.5}$ concentrations under the emission reduction scenario and simulated $PM_{2.5}$ concentrations in the baseline scenario respectively.

To evaluate the relative contribution of meteorological conditions and different emission-reduction measures to the decrease of $PM_{2.5}$ concentrations, we designed two baseline experiments and six sensitivity experiments. For the first baseline experiment, we employed the actual meteorological data in 2013. For the second baseline experiment, we employed the actual meteorological data in 2017 and emission inventory in 2017. Since no emission-reduction measures were conducted in 2013, the first baseline experiment was used for model verification and estimating the relative contribution of meteorological variations to the variation of $PM_{2.5}$ concentrations. By comparing the first and second baseline experiment, the relative contribution of all emission-reduction measures to the variation of $PM_{2.5}$ concentrations can be quantified. For the first sensitivity experiment, we employed the actual meteorological conditions in 2013 and emission inventory in 2017 and compared the simulation result with the baseline experiment, which demonstrated the relative contribution of meteorological variations to a $PM_{2.5}$ reduction in Beijing during the past five years. Since the WRF-CMAQ simulation simply considered the $PM_{2.5}$ concentrations and meteorological conditions in 2013 and 2017 without considering their variation process from 2013 to 2017, KZ filtering may perform better in quantifying the relative contribution of meteorological variations to a $PM_{2.5}$ reduction in Beijing. However, the output from this sensitivity experiment serves as a useful reference for understanding the reliability of the output from the KZ filtering. For the remaining five sensitivity-simulation experiments, we added the reduced emission amount induced by one specific emission-reduction measure to the actual emission amount in 2017 and kept other parameters unchanged, which quantified the relative contribution of one type of emission sources to the $PM_{2.5}$ reduction in Beijing during the past five years. Therefore, we acquired the influence of the relative contribution of each emission source on $PM_{2.5}$ reduction in Beijing (Table 1).

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

Table 1. The design and material for the baseline and five sensitivity experiments using WRF-CMAQ model

ID	Meteorological Data	Emission-reduction measures	Simulation Year	Major purposes
Baseline Experiment1	2013	No emission-reduction Measures	2013	**2013 baseline scenario**
Baseline Experiment2	2017	All emission-reduction Measures	2017	**2017 baseline scenario**
Sensitivity Experiment 1	2013	All emission-reduction Measures	2017	The relative contribution of meteorological variations to the decrease of $PM_{2.5}$ concentrations from 2013 to 2017
Sensitivity Experiment 2	2017	All emission-reduction measures except for industrial restructuring	2017	The relative contribution of industrial restructuring to the decrease of $PM_{2.5}$ concentrations from 2013 to 2017
Sensitivity Experiment 3	2017	All emission-reduction measures except for the regulation of coal boilers	2017	The relative contribution of the regulation of coal boilers to the decrease of $PM_{2.5}$ concentrations from 2013 to 2017
Sensitivity Experiment 4	2017	All emission-reduction measures except for increasing clean fuels for civil use	2017	The relative contribution of increasing clean fuels for civil use to the decrease of $PM_{2.5}$ concentrations from 2013 to 2017
Sensitivity Experiment 5	2017	All emission-reduction measures except for the regulation of vehicle emissions	2017	The relative contribution of the regulation of vehicle emissions to the decrease of $PM_{2.5}$ concentrations from 2013 to 2017

For emission data, all experiments employed Beijing local emissions inventory in 2017 for Beijing and regional emission inventory in 2017 for other regions.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

3.3 Model verification

3.3.1 Verification of the KZ filtering

For each station, the original time series of $PM_{2.5}$ data was processed by the KZ filter and the relative contribution of the long-term trend, seasonal variation and short-term variation to the total variance was shown as Table 2. The sum of the long-term trend, seasonal variation and short-term variation contributed to more than 93.6~95.3% of the total variance for different stations respectively. The larger the total variance, the three components are more independent to each other. According to Table 2, the large value of the total variation for each station indicated a satisfactory result from the KZ filtering. The relative contribution of short-term variation was much larger than that of the seasonal and long-term variation, suggesting that short-term variations of meteorological conditions and emission conditions exerted a strong influence on the rapid variation in $PM_{2.5}$ concentrations in Beijing. This result is consistent with findings from previous studies (Chen et al., 2016; Ma et al., 2016).

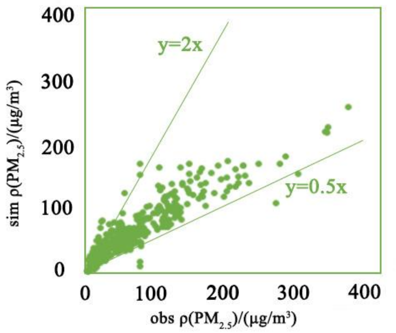
Table 2. The relative contribution of different components to the total variance of original time series of $PM_{2.5}$ concentrations from 2013-2017 at different stations

Stations	Long-term Trend(%)	Seasonal Variation(%)	Short-term Variation(%)	Total variance(%)
Yufa	2.1	23.8	66.8	94.0
Miyun Reservoir	1.4	9.0	83.8	95.2
Dingling	1.6	11.0	81.3	94.9
Qianmen	2.7	12.7	78.5	95.1
Olympic center	2.1	11.9	80.0	95.3
Xiangshan	1.2	10.3	83.4	94.9
Huayuan	2.2	15.9	75.6	93.7
Yungang	2.1	15.1	76.5	93.6
WanShouxigong	1.6	14.2	78.2	94.0
Dongsi	1.6	12.3	80.0	94.0
TianTan	2.1	13.2	78.6	93.8
NongZhanguan	1.8	13.7	78.6	94.1
Gucheng	1.8	13.5	78.5	93.7
Guanyuan	1.6	12.6	79.8	94.0
BeiBuxinqu	1.7	13.8	78.4	93.9
WanLiu	3.5	11.9	78.2	93.6

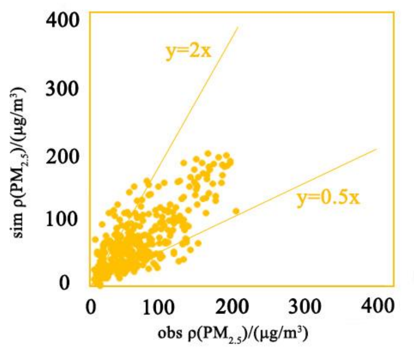
3.3.2 Verification of the WRF-CMAQ

We employed the emission inventory and meteorological data for 2013 to verify the accuracy of the WRF-CMAQ model. For three different stations (the DingLing background station, the Yufa rural station and the Olympic Center urban station), we compared the observed and estimated $PM_{2.5}$ concentrations (Fig 2). According to Fig 2, the general trend of the simulated $PM_{2.5}$ concentrations was similar to that of the observed value. A general agreement was found between the simulated and observed data with more than 85% of data points falling into the siege area of 1:2 and 2:1 lines. WRF-CMAQ slightly underestimated $PM_{2.5}$ concentrations due to the uncertainty in the emission inventory, meteorological field simulation errors and insufficient chemical reaction mechanisms. For three stations, the correlation coefficient R, normalized mean bias (NMB), normalized mean error (NME), mean fractional bias (MFB) and mean fractional error (MFE) between observed and simulated data was 0.69~0.74, 11%~17%, 20%~27%, -21%~-17%, and 15%~27% respectively, indicating a satisfactory simulation output (EPA，2005; Boylan et al., 2006)

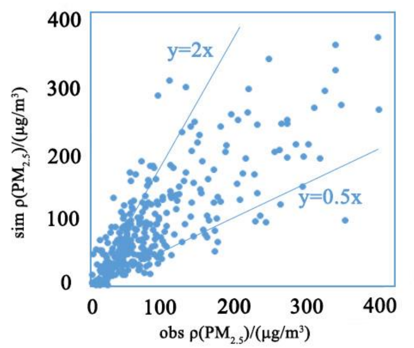
Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019



(a) Dingling background station



(b) Olympic center urban station



(c) Yufa rural station

Fig 2. The comparison between observed and WRF-CMAQ simulated $PM_{2.5}$ concentrations

4 Results

4.1 The relative contribution of emission-reduction measures and meteorological variations to the decrease of PM2.5 concentrations in Beijing from 2013 to 2017

4.1.1 Estimation based on KZ filtering

Through KZ filtering, the original time-series of $PM_{2.5}$ concentrations and adjusted time-series of $PM_{2.5}$ concentrations with filtered meteorological variations were acquired. Based on these, for each station, the actual variation of $PM_{2.5}$ concentrations and the adjusted variation in $PM_{2.5}$ concentrations without the influence of meteorological variations from 2013 to 2017 were calculated (as shown in Table 3), which indicate the relative contribution of anthropogenic emissions and meteorological conditions to

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
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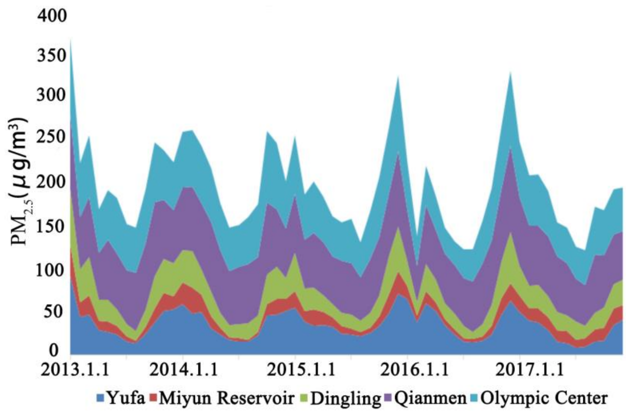
Discussion started: 31 January 2019

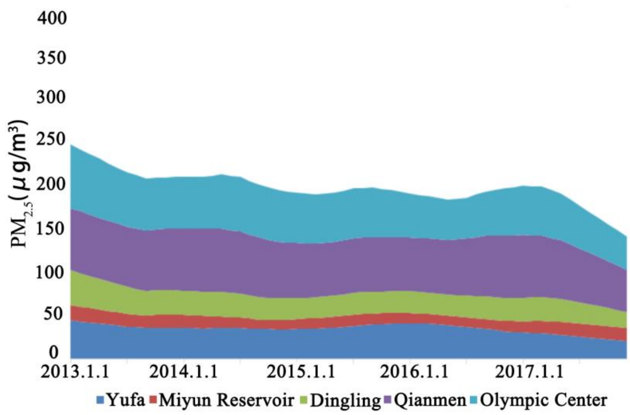
Atmospheric Chemistry and Physics
Discussions

the decrease in $PM_{2.5}$ concentrations in Beijing during the five-year period.

The original time series of $PM_{2.5}$ concentrations and adjusted time series of $PM_{2.5}$ concentrations processed using KZ filtering were illustrated using one urban station, one rural station, one transportation station, and two background stations (Fig 3). As shown in Fig 3, the most abrupt variations in $PM_{2.5}$ concentrations have been smoothed through KZ filtering.



a. Original time series of $PM_{2.5}$ concentrations from 2013 to 2017



b. Processed time series of PM2.5 concentrations from 2013 to 2017 using KZ filter

Fig 3. The comparison of original and KZ processed time series of $PM_{2.5}$ concentrations in Beijing from 2013 to 2017

According to Table 3, the annual mean $PM_{2.5}$ concentrations in Beijing in 2017 was 35.6% lower than that in 2013. By filtering the influence of meteorological variations, the adjusted annual mean $PM_{2.5}$ concentrations in Beijing in 2017 decreased by 31.7% when compared to that in 2013, indicating that

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

the variation in meteorological conditions exerted a moderate influence on the reduction of $PM_{2.5}$ concentrations during the past five years. Meteorological conditions in Beijing were generally favorable for $PM_{2.5}$ dispersion during the five years, especially the latter half of 2017, when there was a high frequency of strong Northerly winds and much lower wintertime $PM_{2.5}$ concentrations than previous years.

For the winter of 2017, frequent windy weather and successive clean sky had a strong influence on the reduction of $PM_{2.5}$ concentrations in Beijing. This led to a hot debate concerning whether the notable decrease in $PM_{2.5}$ concentrations was largely due to the favorable meteorological conditions or emission-reduction measures. Table 3 suggests that emission-reduction measures contributed to 75.2%~85.0% $PM_{2.5}$ decrease in the five-year period, indicating that emission-reduction measures worked effectively in all rural, urban and background stations. On average, the relative contribution of anthropogenic emissions and meteorological variations to $PM_{2.5}$ reduction in Beijing from 2013 to 2017 was 80.6% and 19.4% respectively. Therefore, in spite of more favorable meteorological conditions, properly designed and implemented emission-reduction measures were the dominant driver for the remarkable decrease of $PM_{2.5}$ concentrations in Beijing during the past five years.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

Table 3. Estimated relative contribution of emission-reduction and meteorological variations to $PM_{2.5}$ reduction in Beijing from 2013 to 2017 using KZ filter

Stations	$PM_{2.5}$ concentrations in 2013($\mu g \cdot m^{-3}$)	$PM_{2.5}$ concentrations in 2017 ($\mu g \cdot m^{-3}$)	Adjusted $PM_{2.5}$ concentrations in 2017($\mu g \cdot m^{-3}$)	$PM_{2.5}$ Decrease rate ($\mu g \cdot m^{-3} \cdot m^{-1}$)[1]	Adjusted $PM_{2.5}$ Decrease rate ($\mu g \cdot m^{-3} \cdot m^{-1}$)[2]	Contribution of emission reduction (%)[3]	Contribution of meteorological variations (%)[4]
Yufa	111.1	69.7	74.6	-0.78	-0.63	80.4	19.7
Miyun Reservoir	58.8	44.8	47.0	-0.40	-0.33	82.8	17.2
Dingling	69.6	47.1	50.6	-0.54	-0.44	80.8	19.2
Qianmen	103.9	64.0	68.9	-0.81	-0.69	85.0	15.0
Olympic center	90.4	57.2	61.7	-0.68	-0.55	80.8	19.2
Xiangshan	77.0	59.3	60.3	-0.46	-0.39	83.9	16.1
Huayuan	101.5	64.4	69.2	-0.77	-0.63	81.9	18.1
Yungang	91.8	60.2	64.0	-0.69	-0.55	79.6	20.4
WanShouxigong	93.7	62.0	66.8	-0.64	-0.50	78.2	21.8
Dongsi	94.9	62.4	67.5	-0.62	-0.49	78.9	21.1
TianTan	92.3	58.4	64.6	-0.68	-0.55	80.2	19.9
NongZhanguan	92.2	59.9	65.9	-0.66	-0.53	80.3	19.8
Gucheng	92.7	61.4	65.9	-0.65	-0.50	77.6	22.4
Guanyuan	89.6	59.5	64.6	-0.60	-0.48	79.6	20.4
BeiBuxinqu	86.6	59.5	63.3	-0.60	-0.45	75.2	24.8
WanLiu	98.1	56.2	60.4	-0.87	-0.73	84.2	15.8

[1] $PM_{2.5}$ decrease rate: the fitted variation slope of original monthly average $PM_{2.5}$ time series;
[2] Adjusted $PM_{2.5}$ decrease rate: the fitted variation slope of adjusted monthly average $PM_{2.5}$ time series;
[3]. Contribution of emission reduction = 1 - Contribution of meteorological variations;
[4]. Contribution of meteorological variations = ($PM_{2.5}$ decrease rate - Adjusted $PM_{2.5}$ decrease rate) / $PM_{2.5}$ decrease rate.

4.1.2 Estimation based on WRF-CMAQ model

In addition to the KZ filter, we also employed the WRF-CMAQ model to estimate the relative contribution of emission-reduction measures and meteorological conditions to the decrease of $PM_{2.5}$ concentrations in Beijing. The result is shown in Table 4.

Table 4. Estimated relative contribution of emission-reduction and meteorological variations to $PM_{2.5}$ reduction in Beijing from 2013 to 2017 using WRF-CMAQ model

Stations	Contribution of meteorological variations (%)	Contribution of emission-reduction(%)
Yufa	21.9	78.2
Miyun Reservoir	20.8	79.2
Dingling	21.7	78.3
Qianmen	21.2	78.8
Olympic center	21.2	78.8
Xiangshan	20.3	79.7
Huayuan	21.2	78.8
Yungang	21.2	78.8
WanShouxigong	21.2	78.8
Dongsi	21.2	78.8
TianTan	21.2	78.8
NongZhanguan	21.2	78.8
Gucheng	22.2	77.8
Guanyuan	21.2	78.8
BeiBuxinqu	22.2	77.8
WanLiu	22.2	77.8

As Table 4 shows, and based on the WRF-CMAQ model, the relative contribution of meteorological variations to the decrease in $PM_{2.5}$ concentrations in Beijing from 2013 to 2017 ranged from 20.3% to 22.2% in different stations, while emission-reduction measures contributed to about four-fifths of the decrease in $PM_{2.5}$ concentrations from 2013 to 2017. It is worth mentioning that the WRF-CMAQ model was a grid-based model and thus the calculated contribution of meteorological variations for some stations located in the same grid was the same. Instead, station-based KZ filtering led to more reliable analysis for each station and can better distinguish the differences between different stations.

Furthermore, the WRF-CMAQ model simply considered the differences between the meteorological conditions in 2013 and 2017 without considering their variations during the past five years while the KZ filtering analyzed the entire time series of $PM_{2.5}$ and meteorological data from 2013 to 2017. The averaged relative contribution of meteorological variations to $PM_{2.5}$ reduction in Beijing calculated using the WRF-CMAQ model was 21.4%, very similar to the 19.4% obtained by using KZ filtering. The slightly larger meteorological contribution calculated using the WRF-CMAQ model might be attributed to the favorable meteorological conditions in the winter of 2017.

Due to its fine spatial resolution and capability in providing a better understanding of the influence of meteorological conditions on $PM_{2.5}$ concentrations, KZ filtering provides a more reliable method for researchers and decision makers to understand the relative importance of emission-reduction measures and meteorological conditions in recent $PM_{2.5}$ reduction in Beijing. However, similar results from the WRF-CMAQ simulation provide complementary evidence for the fact that anthropogenic emissions exerted a much stronger influence on $PM_{2.5}$ concentrations than meteorological conditions. In the next subsection, and based on a detailed local emission inventory, we use the WRF-CMAQ model to further quantify the relative contribution of different emission-reduction measures to the decrease in $PM_{2.5}$ concentrations in Beijing.

4.2 The relative contribution of different emission-reduction measures to the decrease in PM2.5 concentrations in Beijing

Based on the WRF-CMAQ model, we simulated the scenario that no emission-reduction measures were implemented in Beijing from 2013 to 2017 and estimated that with emission-reduction measures, the total amount of reduction in SO_2, NO_x, VOCs, direct $PM_{2.5}$ and direct PM_{10} caused by these measures was 79000t, 93000t, 116000t, 44000t and 139000t respectively. The amount of reduced pollutants accounted for 83.2%、42.9%、42.4%、54.7% and 52.4% of the total emission of SO_2, NO_x, VOCs, direct $PM_{2.5}$ and direct PM_{10} respectively, indicating the remarkable effect of emission-reduction measures on $PM_{2.5}$ reduction during the past five years (UNEP,2018).

The observed annual average $PM_{2.5}$ concentrations in Beijing in 2017 was 58 mg/m^3, compared with 89.5 μg/m^3 in 2013. Based on the WRF-CMAQ simulation, meteorological conditions contributed a decrease of 6.7 μg/m^3 to the total decrease of 31.5 μg/m^3. Meanwhile, local and regional emission-reduction measures contributed 16.9 μg/m^3 and 7.8 μg/m^3 respectively. Amongst the

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

emission-reduction measures implemented in 2017, the regulation of coal boilers had the most significant effect on $PM_{2.5}$ reduction in Beijing and resulted in a decrease of 6.3 $\mu g/m^3$. Meanwhile, increasing clean fuels for residential use and industrial restructuring also exerted strong influences on $PM_{2.5}$ reduction and contributed to a decrease of 5.5 $\mu g/m^3$ and 3.4 $\mu g/m^3$ respectively. The relative contribution of the regulations on raise dust and vehicle emissions was relatively small, leading to a decrease of 1.7$\mu g/m^3$ in total.

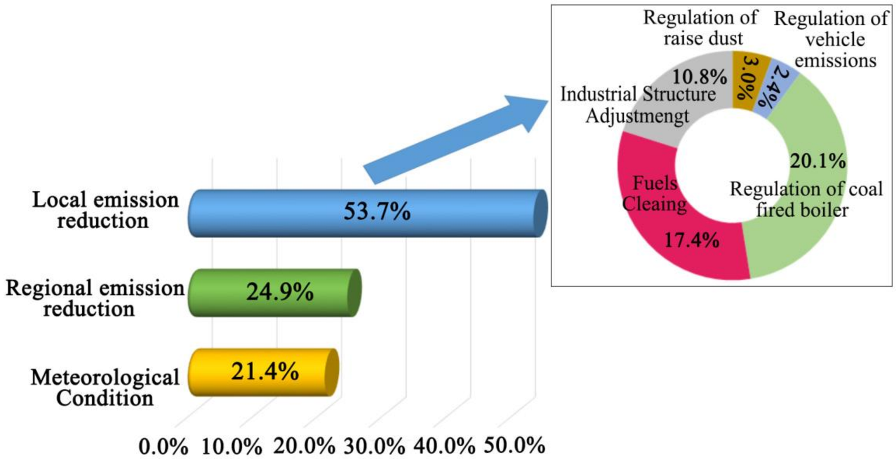


Fig 4. The relative contribution of different influencing factors to the decrease of PM2.5 concentrations in Beijing from 2013 to 2017

5 Discussion

By the end of 2017, the Beijing Five-year Clean Air Action Plan (2013-2017) was completed and achieved its primary goal of reducing the annual average $PM_{2.5}$ concentrations to less than 60 $\mu g/m^3$. Meanwhile, since November 2017, strong northerly winds in Beijing resulted in the cleanest winter for the past five years, raising arguments about whether the favorable meteorological conditions was primarily responsible for the $PM_{2.5}$ reduction or whether the significant improvement in air quality in Beijing was mainly due to the control of anthropogenic emissions. In this case, a quantitative comparison between the influence of meteorological conditions and emission-reduction measures on $PM_{2.5}$ reduction is necessary for comprehensively evaluating the effects of the Five-year Clean Air Action Plan. Based on two different approaches, results of this study revealed that the control of anthropogenic emissions contributed to around 80% of the decrease in $PM_{2.5}$ concentrations in Beijing from 2013 to 2017, indicating that the Five-Year Clean Air Plan exerted a much stronger influence on

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

Atmospheric Chemistry and Physics
Discussions

the improvement of air quality than meteorological conditions. The large contribution of some specific emission-reduction measures may be obscured in the presence of favorable meteorological conditions. For instance, many residents may attribute the clean winter of 2017 to the notable strong winds without noticing some of the major emission-reduction measures implemented during this period. A large-scale replacement of coal boilers with gas boilers was conducted in Beijing and its neighboring areas since 2013. As quantified by the WRF-CMAQ model，the regulation of coal boilers and increasing clean fuels for residential use in total contributed to an 11.8μg/m^3 decrease in $PM_{2.5}$ concentrations，much (almost twice) larger than the 6.7 μg/m^3 decrease brought about by favorable meteorological conditions. In general, although favorable meteorological conditions (e.g., strong winds) may lead to an instant improvement of air quality, regular emission-reduction measures exert a reliable and consistent influence on the long-term reduction of $PM_{2.5}$ concentrations in Beijing. Given the satisfactory performance of the Five-year Clean Air Action Plan in $PM_{2.5}$ reduction, such kind of long-term clean air plans should be further designed and implemented in the future.

Despite the major contribution of emission-reduction measures to $PM_{2.5}$ reduction in Beijing, meteorological influences, which contributed to 20% of $PM_{2.5}$ reduction, should also be considered as well. In addition to the control of anthropogenic emissions, the $PM_{2.5}$ reduction may be realized through meteorological means. For the winter of 2017, strong northwesterly winds led to instant improvement in air quality, suggesting wind was a dominant meteorological factor for the concentration or dispersion of $PM_{2.5}$ in Beijing. Meanwhile, previous studies (Chen et al., 2017) suggested that increasing wind speeds lead to increased evaporation, increased sunshine duration (SSD) and reduced humidity, which further reduced local $PM_{2.5}$ concentrations. In other words, strong winds help reduce $PM_{2.5}$ concentrations through direct and indirect measures. In this light, the forthcoming Beijing Wind-corridor Project (http://news.10jqka.com.cn/20170331/c597397500.shtml), which includes five 500m-width corridors and more than ten 80m-width corridors to bring in stronger wintertime northwesterly winds, can be a promising approach for promoting favorable long-term meteorological influences on $PM_{2.5}$ reduction in Beijing.

Despite the remarkable decrease in $PM_{2.5}$ concentrations, recent ground ozone pollution in Beijing has aroused growing concerns. In the past decade, ozone concentrations in Beijing demonstrated a notable increase and ozone even became the dominant pollutant in June 2017 (Cheng et al., 2018）. Current emission-reduction measures, even the wind-corridor project, have been designed and implemented to simply reduce $PM_{2.5}$ concentrations. Meanwhile, ozone concentrations even increased during specific

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

Atmospheric Chemistry and Physics Discussions

periods with strict emission-reduction measures, indicating that ordinary emission-reduction measures for $PM_{2.5}$ reduction were not suitable for reducing ozone concentrations. Due to complicated and unpredictable reactions between a diversity of ozone precursors, emission-reduction measures for reducing one specific precursor may conversely increase ozone concentrations (Cheng et al., 2018). Given the severe threat ground ozone exerts on public health, future emission-reduction measures should be comprehensively designed to reduce both ozone and $PM_{2.5}$ concentrations.

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Author contribution

Chen, Z., Gao, B. and Xu, B designed this research. Chen, Z wrote this manuscript. Chen, D., Zhuang, Y, Cheng, N. and Li, R. conducted data analysis. Chen, D and Zhuang, Y. produced the figures. Kwan, M., and Chen, B helped revise this manuscript.

References

1. Boylan J W, Russell A G.: PM and light extinction model performance metrics, goals and criteria for three-dimensional air quality models. Atmospheric Environment, 40 (26): 4946-4959,2006.
2. Brunekreef B, Holgate S.: Air pollution and health Lancet, 360:1233-1242,2002.
3. Chan, C.K., Yao, X.: Air pollution in mega cities in China. Atmos. Environ., 42: 1-42,2008.
4. Chen, Z. Y., Cai, J., Gao, B. B., Xu, B., Dai, S., He, B., Xie, X. M.: Detecting the causality influence of individual meteorological factors on local PM2:5 concentrations in the Jing-Jin-Ji region, Scientific Reports, 7 , 40735,2017.
5. Chen, Z.Y., Xie, X., Cai, J., Chen, D., Gao, B., He, B., Cheng, N., Xu, B.: Understanding meteorological influences on PM2.5 concentrations across China: a temporal and spatial perspective, Atmos. Chem. Phys., 18, 5343-5358,2018.
6. Chen，Z.Y., Xu, B., Cai，J., Gao, B.B.: Understanding temporal patterns and characteristics of air quality in Beijing: A local and regional perspective. Atmospheric Environment. 127, 303-315,2016.
7. Cheng, N., Chen, Z., Sun, F., Sun, R., Dong, X., & Xie, X.: Ground ozone concentrations over beijing from 2004 to 2015: variation patterns, indicative precursors and effects of emission-reduction. Environmental Pollution, 237, 262-274,2018.
8. Cheng,N.L.,Zhang,D.W.,Li,Y.T.: Spatio-temporal variations of PM2.5 concentrations and the evaluation of emission reduction measures during two red air pollution alerts in Beijing. Scientific Reports7, 8220 (2017),doi:10.1038/s41598-017-08895-x,2017.
9. Dominici F,Greenstone M, Sunstein C.: Particulate matter matters Science, 344:257-259,2014.
10. Eskridge R E, Ku J Y, Rao S T.: Separating Different Scales of Motion in Time Series of Meteorological Variables, B. Am. Meteorol. Soc., 78, 1473–1483,1997.
11. Guo S, Hu M, Zamora M L.: Elucidating severe urban haze formation in China. Proceedings of the National Academy of Sciences, 111(49): 17373-17378,2014.
12. Hong C, Zhang Q, He K.: Variations of China's emission estimates: response to uncertainties in energy statistics. Atmospheric Chemistry and Physics, 17(2): 1227-1239,2017.
13. Huang,R.,Zhang,Y.,Bozzetti,C.,Ho,K.F.,Cao,J.J.,Han,Y.: High secondary aerosol contribution to particulate pollution during haze events in China. Nature,514: 218–222,2014.
14. Kang, D., Hogrefe, C., Foley, K. L., Napelenok, S. L., Mathur, R., Rao, S. T.: Application of the kolmogorov–zurbenko filter and the decoupled direct 3d method for the dynamic evaluation of a regional air quality model. Atmospheric Environment, 80(23), 58-69,2013.

15. Ma, Z. Q.,Xu, J.,Quan, W. J.: Significant increase of surface ozone at a rural site, north of eastern China.Atmospheric Chemistry and Physics,16(6):3969-3977,2016.
16. MEP.: 2017 air pollution prevention and management plan for the Beijing-Tianjin-Hebei region and its surrounding areas,2017.
17. Nel A.: Air pollution-related illness effects of particles Science, 308:804-806,2015.
18. Qiao, L.P., Cai, J., Wang, H.L., Wang, W.L., Zhou, M., Lou, S.R., Chen, R.J., Dai, H.X., Chen, C.H., Kan, H.D.: PM2.5 Constituents and Hospital Emergency-Room Visits in Shanghai, China. Environmental Science and Technology. 48 (17), 10406–10414,2014.
19. Rao S T and Zurbenko I G.: Detecting and Tracking Changes in Ozone Air Quality. Air Waste, 44, 1089–1092, doi:10.1080/10473289.1994.10467303,1994.
20. Saikawa E, Kim H, Zhong M.: Comparison of emissions inventories of anthropogenic air pollutants and greenhouse gases in China. Atmospheric Chemistry and Physics, 17(10): 6393-6421,2017.
21. Sun Y, Song T, Tang G.: The vertical distribution of PM 2.5, and boundary-layer structure during summer haze in Beijing. Atmospheric Environment, 74(2):413-421,2013.
22. UNEP.: A Review of Air Pollution Control in Beijing: 1998-2013.United Nations Environment Programme (UNEP), Nairobi, Kenya,2016.
23. UNEP.: A Review of Air Pollution Control in Beijing: 1998-2017.United Nations Environment Programme (UNEP), Nairobi, Kenya,2018.
24. US EPA.: Guidance on the use of models and other analyses in attainment demonstrations for the 8-hour ozone NAAQS, North Carolina, US Environmental Protection Agency ,2005.
25. Wang X, Wei W, Cheng S.: Characteristics and classification of PM2.5 pollution episodes in Beijing from 2013 to 2015. Science of the Total Environment, 612:170-179,2018.
26. Wang, S.X., Xing, J., Zhao, B., Jang, C., Hao, J.M.: Effectiveness of national air pollution control policies on the air quality in metropolitan areas of China. J. Environ. Sci. 26 (1):13–22. http://dx.doi.org/10.1016/s1001-0742(13)60381-2,2014.
27. Zhang, Q., He, K., Huo,H.: Cleaning China's air.Nature,484:161-162,2012.
28. Zhao, B., Wang, S.X., Wang, J.D., Fu, J., Liu, T.H., Xu, J.Y., Fu, X., Hao, J.M.: Impact of national NOX and SO2 control policies on particulate matter pollution in China. Atmos. Environ. 77, 453–463,2013.
29. Zheng, G. J., Duan, F. K., Su, H., Ma, Y. L., Cheng, Y., Zheng, B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions,

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1112
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 31 January 2019

Atmos. Chem. Phys., 15, 2969-2983, doi:10.5194/acp-15-2969-2015,2015.

30. Zhou Y, Zhao Y, Mao P.: Development of a high-resolution emission inventory and its evaluation and application through air quality modeling for Jiangsu Province, China[J]. Atmospheric Chemistry and Physics, 17(1): 211-233,2017.

31. Zurbenko I, Chen J, Rao S T.: Detecting discontinuities in time series of upper air data: Demonstration of an adaptive filter technique, J. Climate, 9,3548–3560,1996.