

# Dominant role of emission reduction in PM<sub>2.5</sub> air quality improvement in Beijing during 2013-2017: a model-based decomposition analysis

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## Abstract.

In 2013, China's government published the Air Pollution Prevention and Control Action Plan (APPCAP) with a specific target for Beijing, which aims to reduce annual mean PM<sub>2.5</sub> concentrations in Beijing to 60  $\mu\text{g m}^{-3}$  in 2017. During 2013-2017, the  
15 air quality in Beijing was significantly improved following the implementation of various emission control measures locally and regionally, with the annual mean PM<sub>2.5</sub> concentration decreasing from 89.5  $\mu\text{g m}^{-3}$  in 2013 to 58  $\mu\text{g m}^{-3}$  in 2017. As meteorological conditions were more favourable to the reduction of air pollution in 2017 than in 2013 and 2016, the "real" effectiveness of emission control measures on the improvement of air quality in Beijing has frequently been questioned.

In this work, by combining a detailed bottom-up emission inventory over Beijing, the MEIC regional emission inventory, and  
20 the WRF-CMAQ model, we attribute the improvement in Beijing's PM<sub>2.5</sub> air quality in 2017 (compared to 2013 and 2016) to the following factors: changes in meteorological conditions, reduction of emissions from surrounding regions, and seven specific categories of local emission control measures in Beijing. We collect and summarize data related to 32 detailed control measures implemented during 2013-2017, quantify the emission reductions associated with each measure using the bottom-up local emission inventory in 2013, aggregate the measures into seven categories, and conduct a series of CMAQ simulations  
25 to quantify the contribution of different factors to the PM<sub>2.5</sub> changes.

We found that, although changes in meteorological conditions partly explain the improved PM<sub>2.5</sub> air quality in Beijing in 2017 compared to 2013 (3.8  $\mu\text{g m}^{-3}$ , 12.1% of total), the rapid decrease in PM<sub>2.5</sub> concentrations in Beijing during 2013-2017 was dominated by local (20.6  $\mu\text{g m}^{-3}$ , 65.4%) and regional (7.1  $\mu\text{g m}^{-3}$ , 22.5%) emission reductions. The seven categories of emission control measures, i.e., *Coal-fired boiler control*, *Clean fuels in the residential sector*, *Optimize industrial structure*,  
30 *Fugitive dust control*, *Vehicle emission control*, *Improved end-of-pipe control*, and *Integrated treatment of VOCs*, reduced the PM<sub>2.5</sub> concentrations in Beijing by 5.9, 5.3, 3.2, 2.3, 1.9, 1.8, and 0.2  $\mu\text{g m}^{-3}$ , respectively, during 2013-2017. We also found that, changes in meteorological conditions could explain roughly 30% of total reduction in PM<sub>2.5</sub> concentration during 2016-2017 with more prominent contribution in winter months (November and December). If the meteorological conditions in 2017

had remained the same as those in 2016, the annual mean  $PM_{2.5}$  concentrations would have increased from  $58 \mu\text{g m}^{-3}$  to  $63 \mu\text{g m}^{-3}$ , exceeding the target established in the APPCAP. Despite the remarkable impacts from meteorological condition changes, local and regional emission reductions still played major roles in the  $PM_{2.5}$  decrease in Beijing during 2016-2017, and *Clean fuels in the residential sector*, *Coal-fired boiler control*, and *Optimize industrial structure* were the three most effective local measures (contributing reductions of 2.1, 1.9 and  $1.5 \mu\text{g m}^{-3}$ , respectively). Our study confirms the effectiveness of clean air actions in Beijing and its surrounding regions and reveals that a new generation of control measures and strengthened regional joint emission control measures should be implemented for continued air quality improvement in Beijing because the major emitting sources have changed since the implementation of the clean air actions.

## 1 Introduction

Most countries inevitably undergo and tackle severe air pollution in the development process. In recent years, severe  $PM_{2.5}$  pollution in China has gradually become an urgent challenge to the government (Wang et al., 2012; Li et al., 2017). It not only posed a threat to human health but has also badly influenced the social economy and ecological environment (Menon et al., 2002; Chan et al., 2006; Ming et al., 2009; Zheng et al., 2015; Zhu et al., 2015). Beijing, as the capital of China, has suffered especially severe air quality problems. In 2013, the annual average  $PM_{2.5}$  concentrations in Beijing reached  $90 \mu\text{g m}^{-3}$ , which was nearly three times higher than China's National Ambient Air Quality Standard (NAAQS) of  $35 \mu\text{g m}^{-3}$  (MEP, 2012). In addition to the high annual average  $PM_{2.5}$  concentrations, several frequent and severe heavy haze episodes in January 2013 made the situation even worse and caused great public concern (Zhang et al., 2014; Zheng et al., 2016).

To address the increasingly serious  $PM_{2.5}$  pollution, the Chinese government released the Air Pollution Prevention and Control Action Plan (APPCAP) in September 2013, which aimed to mitigate severe PM pollution across China, especially in some typical regions. In particular, the average  $PM_{2.5}$  concentrations of Beijing should be reduced to less than  $60 \mu\text{g m}^{-3}$  by 2017. Based on the ambition and guidance of the APPCAP, Beijing has made further efforts and formulated the *Beijing 2013-2017 Clean Air Action Plan* (referred to as the *Beijing Action Plan*) to mitigate air pollutions. The *Beijing Action Plan* represents the most important and systematic set of local air pollution control and management policies in the past five years. After implementing a series of air pollution control policies and measures, the annual mean  $PM_{2.5}$  concentrations in Beijing decreased to  $58 \mu\text{g m}^{-3}$  in 2017 (BMEP, 2018), 35.2% lower than that in 2013 and surpassing the air quality goals of the APPCAP. Meanwhile, the surrounding regions of Beijing, such as Tianjin, Hebei, Shandong, Shanxi and Henan province, also implemented the APPCAP, and the air quality of the whole region has attained marked improvements, which have also been confirmed by satellite-based and ground-based observations (Liu et al., 2016; Cai et al., 2017; Zhao et al., 2017; Wang et al., 2017; Zheng et al., 2017).

The  $PM_{2.5}$  concentrations in the atmosphere are affected by several factors, while pollutant emissions, regional transport and meteorological conditions play dominant roles (He et al., 2001; Chen et al., 2018). In general, local pollutant emissions contribute most to the air pollution for a given city, and the control of emissions is always one of the most effective ways to

mitigate air pollution. Regarding the influence of regional transport, considering that Beijing is embraced on three sides by mountains except for the south and southeast direction, the transport of air pollutants from the south and southeast can easily affect the PM<sub>2.5</sub> concentrations in Beijing (Sun et al., 2015; Wang et al., 2015; Chen et al., 2017). However, regional cities in these two directions, such as Baoding, Changzhou, Hengshui, Shijiazhuang, Tangshan, and Tianjin, suffer even worse air pollution (Li et al., 2017). A combination of PM<sub>2.5</sub> formation and transport resulted in the regionally complex air pollution characteristics in Beijing (Lang et al., 2013; Chen et al., 2016; Zhang et al., 2018; Zhong et al., 2018). Besides the impact of pollutant emissions and transport, PM<sub>2.5</sub> concentrations are also highly influenced by some other factors, including atmospheric advection, atmospheric diffusion and secondary aerosol formulation. (Sun et al., 2015; Yin et al., 2016; Zhang et al., 2016). Several studies have also reported that frequent stable meteorological conditions play an important role in severe pollution episodes (Elser et al., 2016; Ma et al., 2017; Zhang et al., 2018). Based on emission inventories and air quality models, existing studies have established a mature sensitivity decomposition framework to assess the contributions of emission control to air quality improvements (Zhao et al., 2013; Cai et al., 2017).

The mitigation of PM<sub>2.5</sub> pollution in Beijing was significant during 2013-2017, especially during 2016-2017. This impact resulted from the integrated effects of various factors, including the local emission control through the *Beijing Action Plan*, the surrounding emission reductions through the APPCAP, and the impacts of meteorological condition changes. Several studies have researched the roles of meteorology, regional and local emissions in Beijing's PM<sub>2.5</sub> pollution; however, most of these studies analysed a single factor or focused on heavy pollution episodes (Wang et al., 2013; Zeng et al., 2014; Zhang et al., 2015; Zheng et al., 2016; Liu et al., 2017; Ma et al., 2017; Wang et al., 2017). There was no systematic and decomposed attribution analysis of Beijing's air quality improvements at an annual scale, especially during the periods of 2013-2017 and 2016-2017, on which the PM<sub>2.5</sub> concentrations in Beijing decreased significantly. To better understand the great progress in air cleaning of Beijing in recent years, a more comprehensive analysis covering the periods of 2013-2017 and 2016-2017 is urgently needed. In this study, based on several sensitivity simulations, we established a decomposition analysis framework to evaluate the impacts of local control policies, surrounding emission reductions and the meteorological changes on PM<sub>2.5</sub> abatements in Beijing during 2013-2017 and 2016-2017. First, the emission reductions of Beijing and its surroundings were estimated based on the quantification of air pollution control measures; meanwhile, a new multiple-pollutant emission inventory of Beijing and its surroundings, covering the periods of 2013-2017, was updated and developed. Second, based on a "zero-out" method, we designed a set of sensitivity experiments under different local and regional emission control measures and different meteorological conditions. Third, we used the Weather Research and Forecasting Model (WRF) and Community Multiscale Air Quality Model (CMAQ) to reproduce and simulate the air quality under different meteorological conditions and emission scenarios. Finally, based on zero-out approach, an integrated and decomposed attribution analysis of PM<sub>2.5</sub> abatements in Beijing was developed to quantify the impacts of local pollution control, surrounding emission reductions, and meteorological changes. The study also identified the key point of next steps for air pollution control, which would be beneficial for future policy-making.

## 2 Methodology and Data

A model-based decomposition attribution analysis of PM<sub>2.5</sub> abatements in Beijing during 2013-2017 and 2016-2017 was developed under the framework shown in Figure 1. First, we used the observation data from 12 national observation stations in Beijing to review the air quality during 2013-2017, especially the monthly PM<sub>2.5</sub> concentrations in this period. The contributions of total PM<sub>2.5</sub> abatements in Beijing in 2017 were decomposed into three basic parts, including meteorology change, surrounding emission control and local emission control. Then, we use the WRF-CMAQ modelling system and observed PM<sub>2.5</sub> concentrations to quantify the contributions of these three factors. To further evaluate the effect of local control policies, we divided the *Beijing Action Plan* into seven specific policy types, estimated the corresponding emission reductions and updated the emission inventory during 2013-2017 based on Beijing's local emission inventory (BJ-EI, described in Sect.2.2) and the framework of the MEIC model (Zhang et al., 2007; <http://www.meicmodel.org/>). The contributions from local emission control to the PM<sub>2.5</sub> air quality improvements in Beijing were also decomposed into specific measures with the WRF-CMAQ model and measure-related sensitivity experiments.

### 2.1 Ground-based and satellite-based observational data

The study first reviewed the air quality in Beijing from 2013 to 2017 with both ground observation data and satellite observation data. Since 2012, Beijing has maintained an automated air quality monitoring network with 35 stations spatially distributed in the 16 administrative districts and counties (the specific locations of the air quality monitors are shown in the supporting information (SI), Figure S1). Since 2013, hourly concentrations of SO<sub>2</sub>, NO<sub>2</sub>, CO, PM<sub>2.5</sub>, PM<sub>10</sub>, and O<sub>3</sub> have been continuously measured and recorded by Beijing's Environment Protection Bureau (EPB). In this study, we reviewed the changes in SO<sub>2</sub>, NO<sub>2</sub>, CO, O<sub>3</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> in Beijing annually during 2013-2017 and analysed the monthly PM<sub>2.5</sub> concentrations during this period. The hourly observation data we used were from the 12 national observation stations in Beijing, which are included among the above 35 stations (SI, Figure S1).

Satellite observation data can provide the objective evidence of the emission reductions and the air quality improvements. Based on satellite observations, we analysed the changes of tropospheric vertical column NO<sub>2</sub>, boundary layer SO<sub>2</sub> and Aerosol Optical Depth (AOD) in Beijing during 2013-2017 to back up the estimated emission trends and reductions (Sect. 3.5). Based on the Ozone Monitoring Instrument (OMI) sensor, the NO<sub>2</sub> tropospheric vertical column density product, DOMINO, is produced by KNMI; and the SO<sub>2</sub> boundary layer vertical column density product, OMI SO<sub>2</sub>, is produced by NASA. The daily AOD observation data comes from the Terra and Aqua, based on the MODIS sensor. To reduce uncertainties in the satellite-based evaluation, DOMINO NO<sub>2</sub> and OMI SO<sub>2</sub> columns with cloud fraction higher than 0.3, surface reflectance higher than 0.3 were discarded. Based on our previous research and methods (Zheng et al., 2016), we estimated the ground-level PM<sub>2.5</sub> concentrations in Beijing using the satellite-derived AOD data.

To evaluate the accuracy of PM<sub>2.5</sub> compositions simulation, we collected the PM<sub>2.5</sub> compositions data from the Surface PARTICulate mAtter Network (SPARTAN, [www.spartan-network.org](http://www.spartan-network.org)). SPARTAN is a global, long-term project that

observed and analysed the particulate matter mass, water-soluble ions, black carbon and metals since 2013 (Snider et al., 2015). Major SPARTN measurements include the Air Photon 3-wavelength integrating nephelometer and the Air Photon SS4i automated air sampler. SPARTAN monitors are located in nearly 20 highly populated regions in the world, such as Beijing, Hanoi, Singapore, Dhaka, Pretoria, Toronto, Bondville. Detailed measurements and sites information can be found in Snider's research (Snider et al., 2015; Snider et al., 2016). In this research, we used the reconstructed PM<sub>2.5</sub> speciation data in Beijing Site (located in the Department of Earth System Science, Tsinghua University) (<https://www.spartan-network.org/beijing-china>) to validate the model simulation results of PM<sub>2.5</sub> compositions, and to verify the variation of PM<sub>2.5</sub> compositions in Beijing during 2013-2017. Additionally, we collected the observational PM<sub>2.5</sub> compositions from the reported research (Shao et al., 2018), to verify the simulated variations of PM<sub>2.5</sub> compositions in CMAQ model. In Shao's research, they observed and analysed the multi-size PM samples at an urban site on Beijing Normal University, and compared the chemical compositions changes from January 2013 to 2016-2017 winter in Beijing.

## 2.2 Estimates of local emission reduction from specific control measures

In this study, the anthropogenic emission inventory of Beijing was provided by the Beijing Municipal Environmental Monitoring Center (BMEMC). Based on the bottom-up method, the BMEMC developed a high-resolution emission inventory for Beijing (BJ-EI) of 2013 and 2017. BJ-EI basically had the same source classification as the MEIC model (described in Sect.2.3), however, the investigation and calculation process of BJ-EI were conducted at the county level, which of MEIC were conducted at the provincial level. The power, heating, industry (such as cement, iron, steel, chemical industry, manufacturing industry) and most solvent use (such as vehicle paint, ink, paint and coating) sectors were treated as point sources, with a higher accuracy of emission facility locations. In addition, fugitive dust emissions, including bare soil dust, road dust and construction dust, were added in BJ-EI, which were missing in MEIC model because of the lack of activity rates data. More detailed comparisons of BJ-EI and MEIC model can be found in SI, Table S1. Therefore, the spatial distribution and emission source allocation of BJ-EI were more accurate than those of the MEIC model, which can significantly improve the air quality modelling, especially when modelling with finer resolutions (Zheng et al., 2017). Meanwhile, more detailed and objective activity rate, technology distribution and removal efficacy data at the county-level were collected from BJ-EI, which can largely reduce the uncertainty in estimating the emission reductions of each local control policy.

Beijing started the air quality protection process in 1998 and has focused most on the control of SO<sub>2</sub> and NO<sub>x</sub> (Wang et al., 2012; Zhang et al., 2016). The Chinese government released the APPCAP nationwide in 2013 and committed to reducing PM<sub>2.5</sub> pollution for the first time ever (Zheng et al., 2018). The APPCAP aimed to reduce the annual PM<sub>2.5</sub> concentrations of the Beijing-Tianjin-Hebei region by 25% compared with 2013, and particularly, the PM<sub>2.5</sub> concentrations of Beijing should be controlled to less than 60 µg m<sup>-3</sup>. To fulfil the air quality targets, Beijing released its own *Beijing Action Plan* under the framework and guidance of the *Nation Action Plan*, which contained much more ambitious and stricter control measures than ever before. We summarized and classified all measures in the *Beijing Action Plan* into seven types, including *Coal-fired*

*boiler control, Clean fuels in the residential sector, Optimized economic structure, Improved end-of-pipe control, Vehicle emission control, Integrated treatment of VOCs and Fugitive dust control.* All the quantifiable control measures were listed in Table 1.

Following several previous studies (Jiang et al., 2015), air pollution control policies and measures could be quantified by adjusting the emission calculation parameters, then the emission reduction associated with a single policy can then be estimated from the emission difference between before and after the implementation of this specific policy.

## **2.3 Regional emission inventory data**

The air pollutant emission inventory of Beijing's surrounding regions (including Tianjin, Hebei, Henan, Shandong, Shanxi, Inner Mongolia) for the period of 2013-2017 was obtained from the MEIC model and Zheng's updating work (Zheng et al., 2018). MEIC is a bottom-up emission inventory model developed for China by Tsinghua University, covering 31 provinces in China and the year range of 1990 up to now. More than 700 emission sources were developed in MEIC model. The methodologies and data on which the MEIC is based, as well as the continues updating process of pollutants emission factors, have been introduced in our previous studies (Zhang et al 2007; Liu et al., 2010; Lei et al., 2011; Li et al., 2014; Shen et al., 2014; Hong et al., 2017; Qi et al., 2017; Zheng et al., 2017; Li et al., 2017a; Li et al., 2017b). For the updated emission inventory and emission reductions of the surrounding regions, we referred to our previous work (Zheng et al., 2018). In their work, Zheng et al collected the latest Chinese Energy Statistics data from National Bureau of Statistics, the industrial production and technology penetration data, and the unpublished data from the Ministry of Ecology and Environment. Then they estimated and updated China's anthropogenic emissions from 2010 to 2017 under the framework of MEIC model. Particularly, they calibrated the emission calculation parameters (such as technology penetrations and removal efficiencies) in major sectors, such as power, cement, steel, iron, of each province based on emission control policies. Based on their updated China anthropogenic emission inventory, we collected and analysed the detailed sectoral emission reductions and trends of Beijing's surrounding regions during 2013-2017.

## **2.4 WRF-CMAQ model**

### **2.4.1 Model configuration**

In this work, WRFv3.8 and CMAQ 5.1 were used to build up the air quality modelling system. WRF model provided the meteorological conditions, while CMAQ model simulated the air quality and main pollutant concentrations.

For the simulation area, three nested domains were designed in this study (SI, Figure S2), with horizontal resolution of 36 km  $\times$  36 km, 12km  $\times$  12km and 4 km  $\times$  4 km respectively. The first domain covered the entire China area and some parts of south and east Asia; the second one covered the majority of eastern and northern China, while the third one focused on Beijing and its surrounding regions, including the municipality of Tianjin and the provinces of Hebei, Henan, Shandong, Shanxi and Inner Mongolia. To reduce uncertainty of meteorological boundary condition, the simulation size of WRF model was three grid cells

larger than that of CMAQ model in each domain. The vertical resolution was designed as 23 sigma levels from surface to tropopause (about 100mb) for WRF simulation (with 10 layers below 3-km), while collapsed into 14 CTM layers by Meteorology-Chemistry Interface Processor (MCIP). The 14 sigma levels for CMAQ model vertical resolution were 1.000, 0.995, 0.988, 0.980, 0.970, 0.956, 0.938, 0.893, 0.839, 0.777, 0.702, 0.582, 0.400, 0.200 and 0.000.

5 For the WRF model configuration, we chose the New Goddard scheme (Chou et al., 1998) and RRTM (Mlawer et al., 1997) for shortwave and longwave radiation options, the Kain-Fritsch cloud parameterization (version 2, Kain, 2004), the ACM2 PBL scheme (Pleim, 2007), the Pleim-Xiu land-surface scheme (Xiu and Pleim, 2000) and WSM6 cloud microphysics (Hong and Lim, 2006). Analysis nudging, observational nudging and soil nudging were adopted, and FDDA data was from the U.S. National Centers for Environmental Prediction (NCEP, <http://rda.ucar.edu/datasets/>), Automated Data Processing surface  
10 (ds461.0) and upper (ds351.0) air data. The meteorological initial and boundary conditions were derived from the final analysis data (FNL). We made a continuous meteorology simulation during 2013-2017, with a ten-days spin-up before this period.

For the CMAQ model configuration, we applied the CB05 as gas-phase chemical mechanism and AERO6 as particulate matter chemical mechanisms. The on-line computation of photolytic rates was adopted using the simulated aerosol and ozone concentrations. The chemical initial and boundary conditions of the first domain were interpolated from the output of GEOS-  
15 Chem model (Bay. I, et al., 2001; Geng et al., 2015). We firstly simulated a complete time series of all pollutant's concentrations during 2013-2017 (as base cases), and then modelled the 18 sensitivity experiments (described in Sect.2.5). A ten-days spin-up period was adopted for each sensitivity simulation to mitigate the initial condition impacts. To make the analysis and evaluation more comprehensive, we simulated all sensitivity scenarios for the whole year in cases where the severe pollution period was missing. Similar configurations for WRF and CMAQ model were applied in our previous studies,  
20 and exhibited good agreement with observations (Zheng et al., 2015; Zheng et al., 2017).

For the emission inputs of the model system, the anthropogenic emission inventory for Beijing was taken from BMEMC, and the inventories for other regions in China were provided by the MEIC model, which has been updated to 2017 level based on Zheng's work (Zheng et al., 2018). Emissions for the other Asian countries were derived from the MIX emission inventory (Model Inter-comparison Study Asia Phase III (MICS-ASIA III); Li et al., 2017a). The biogenic emissions were taken from  
25 the Model of Emission of Gases and Aerosols from Nature (MEGAN v2.1). For the dust emission, bare lands dust was calculated by the in-line windblown dust in CMAQ model. As Sect.2.2 described, other dust source, such as road dust and construction dust were added in BJ-EI, while missed in MEIC model. It brought uncertainty of air quality simulation, especially for the PM<sub>10</sub> simulation in other regions (discussed in 2.4.2).

#### **2.4.2 Model validation**

30 To evaluate the meteorology results simulated by the WRF model, we collected the hourly observed meteorology data from the Computational and Information Systems Laboratory at the National Center for Atmospheric Research in Boulder (NCAR, <https://rda.ucar.edu/>) and calculated the mean bias (MB), mean error (ME), correlation coefficient (Corr), root mean square

error (RMSE), normalized mean bias (NMB), and normalized mean error (NME). The evaluation results showed that the simulation basically reproduced the meteorological conditions in 2013 and 2017, and the temperature simulation especially featured a high accuracy. The monthly evaluation results of the simulated temperature, relative humidity, wind speed and wind direction for Beijing in 2013 to 2017 were shown in the SI (Table S2 (a)-(f)).

5 To evaluate the pollutant concentrations simulated by the CMAQ model, we collected the hourly observed major pollutant concentration data from the Beijing Municipal Environmental Protection Bureau. The decrement of PM<sub>2.5</sub> concentration is the most important target in APPCAP, as well as the major pollutants this research focused on. We emphatically analysed the accuracy of PM<sub>2.5</sub> simulations, and listed the monthly descriptive statistics (MB, ME, Corr, RMSE, NMB, and NME) of the hourly observational PM<sub>2.5</sub> and CMAQ model simulation for 12 national stations in Beijing during 2013-2017 (SI, Table S3(a)).

10 The time series of PM<sub>2.5</sub> from observations and CMAQ simulations in three base cases ( $E_{L13S13}M_{13}$ ,  $E_{L16S16}M_{16}$ , and  $E_{L17S17}M_{17}$ , described in Sect.2.5) for Beijing were showed in Figure 2. Furthermore, we calculated the annual descriptive statistic characters of the observational and simulated PM<sub>2.5</sub> with other five major pollutants (SO<sub>2</sub>, NO<sub>2</sub>, PM<sub>2.5</sub>, PM<sub>10</sub>, CO, and O<sub>3</sub>) in Beijing during 2013-2017, which can be found in SI, Table S3 (b). The time series and evaluation results indicated that the CMAQ model and simulation results in this work can relatively well reproduce the temporal and spatial distribution of air

15 pollutants in Beijing and its surroundings. As for the simulated PM<sub>2.5</sub> of 2017, the monthly Corr of PM<sub>2.5</sub> concentrations varied from 0.53 (in May) to 0.89 (in October), and the annual Corr of PM<sub>2.5</sub> concentrations varied from 0.65 (in 2016) to 0.81 (in 2014). The NMB and NME of monthly PM<sub>2.5</sub> simulations were within  $\pm 45\%$  and  $\pm 55\%$ , respectively. According to the observation data, the annual average PM<sub>2.5</sub> concentrations in Beijing decreased by 31.5  $\mu\text{g m}^{-3}$  from 2013 to 2017, while the simulated PM<sub>2.5</sub> decreased by 32.8  $\mu\text{g m}^{-3}$  (Table 2). Compared with 2016, the observed and simulated PM<sub>2.5</sub> decreased by

20 14.9 and 16.6  $\mu\text{g m}^{-3}$ , respectively (Table 2). The evaluation results suggested that the modelling system in this work can be used to quantify and analyse the attribution of PM<sub>2.5</sub> mitigations in Beijing. As for the simulation results of other pollutants in Beijing, the Corr varied from 0.61-0.74 for SO<sub>2</sub>, 0.59-0.68 for NO<sub>2</sub>, 0.62-0.78 for CO, 0.64-0.74 for O<sub>3</sub>, 0.62-0.74 for PM<sub>10</sub> (Table S3 (b)), which was acceptable for the research. The SO<sub>2</sub> simulation was overestimated in the five years, especially during 2013-2015, which indicated the SO<sub>2</sub> emission in BJ-EI might be higher than the reality. The added fugitive dust

25 emission (road and construction dust) in BJ-EI has improved the PM<sub>10</sub> simulation of Beijing obviously, with an overestimation range of 4% - 12% (Table S3 (b)). However, the PM<sub>10</sub> simulation of other cities in the third domain were underestimated (-8%) - (34%), especially in some heavy industry cities as Tangshan (-34%), Baoding (-25%), and Handan (-29%). This might be attributed to lack of construction and road dust emissions in these regions, as well as the uncertainty of the dust model (Todd et al., 2008; Foroutan et al., 2018). It might introduce the uncertainty of simulation, but given that our research was

30 focused on the attribution analyses of anthropogenic emission changes in Beijing, this uncertainty was relatively small. The O<sub>3</sub> was underestimated in this WRF-CMAQ model system, with the range of (-8.3%) - (-22.6%). The rough vertical layers, the underestimation of nature source emissions, the defect of upper boundary simulation in regional model and the uncertainties of VOCs emission inventories might all lead this underestimation.



Besides PM<sub>2.5</sub> concentrations, we also evaluated the simulated PM<sub>2.5</sub> compositions. We compared the PM<sub>2.5</sub> compositions of the observations from Beijing Site, SPARTAN and the simulations from the same grid, which can be found in SI (Table S4). Generally speaking, secondary inorganic aerosol (SIA) was overestimated in most periods (0.1%-56.6%), especially for the nitrate (NO<sub>3</sub><sup>-</sup>) simulation, which overestimated 6.56%-89.9%. Organic matter (OM) was under estimated in most periods, with the underestimation range of (-3.0%) - (-41.8%). This might be caused by the missing mechanism and insufficient simulation of secondary organic aerosols (SOA) formulations in CMAQ model. The NMB of simulated and observed black carbon (BC) varied from (-52.9%) – (38.2%). Similar evaluation results can be found in Weagle’s research, that he compared the GEOS-Chem simulation with SPARTAN observation data, with an average 45.7% overestimation of SIA, -58.9% underestimation of BC, and -19.2% underestimation of OM. However, the variation trends of simulated PM<sub>2.5</sub> compositions were basically consistent with the SPARTAN data, both have the remarkable decrement in OM and increment in NO<sub>3</sub><sup>-</sup>. In Shao’s observational results, that SO<sub>4</sub><sup>2-</sup> proportion in 2016-2017 winter in Beijing reduced by 11% compared with January,2013; while NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> proportions increased by 77.9% and 47.3% (Shao et al., 2018). And in our research, the relative change ratio of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> proportions in Beijing from January, 2013 to 2016-2017 winter were -29.1%, 89.2% and 11.7% respectively. In general. The simulated compositions basically captured the variations in observation results, which can support the reasonable analyse of the chemical composition changes.

## 2.5 Scenario design and decomposition analysis

To decompose the attribution of PM<sub>2.5</sub> abatements in Beijing from 2013-2017 and 2016-2017, we set up 18 sensitivity simulations based on zero-out approach and quantified the contributions of meteorology changes, emission reductions in surrounding areas and seven types of local emission control policies. Given that the nonlinearity between the response of PM<sub>2.5</sub> concentrations and the meteorology or emission changes, we discussed uncertainties and limitations in Sect.3.5. The description and details of all scenarios were listed in Table 2, and the direct simulation results were listed in Table 2, column *Simulated PM<sub>2.5</sub> (μg m<sup>-3</sup>)*.

All scenario cases were labelled as  $E_{LiSj}M_k$ .  $M_k(k)$  represents the meteorological period the case adopted and  $E_{LiSj}(i, j)$  represents the emission period. Total emission inventories of China consisted of two parts, that the BJ-EI from BMEC and the regional (all parts of China except for Beijing) emission inventories from MEIC model. The adopted emission period of these two parts were labelled as  $Li(i)$  and  $Sj(j)$  respectively.

$E_{L13S13}M_{13}$ ,  $E_{L16S16}M_{16}$ , and  $E_{L17S17}M_{17}$  were three base cases, driven by the actual emission inventories and meteorology of 2013, 2016 and 2017, respectively, to reproduce the air quality of the corresponding year.  $E_{L17S17}M_{13}$  and  $E_{L17S17}M_{16}$  were designed to investigate the impact of meteorology. These two cases were driven by varying meteorological conditions (meteorology of 2013 and 2016, respectively) and the same emission inventory (for the year 2017).  $E_{L17S13}M_{17}$  and  $E_{L17S16}M_{17}$  were designed to quantify the impact of surrounding emission reduction during 2013-2017 and 2016-2017. In these two cases,

the emission inventory of Beijing was set to the 2017 level, while the regional emission inventory was set to the 2013 and 2016 levels, respectively.

Another fourteen simulations were designed to quantify the air quality improvements contributed by seven types of local control policies during two periods. Cases for 2013-2017 and 2016-2017 were labelled as  $E_{L_{pi}S17}M_{17}$  and  $E_{L_{qi}S17}M_{17}$  respectively, where  $i$  represents the number of each policy (described and listed in Table 1). The meteorological conditions and regional emission inventories of these fourteen cases were set to 2017. For each simulation, emission reductions introduced by the corresponding policy type were added to the 2017 baseline, equivalent of “turning off” this type of policy during this period. And then the derived emission inventory was applied to drive the corresponding air quality modelling.

A linear additive relationship was assumed among all contributors to perform a decomposition analysis, and the simulated contributions of all sensitivity cases were then normalized by the difference in observed  $PM_{2.5}$  concentrations from 2013-2017 and 2016-2017. The normalization process of 2013-2017 period were calculated by the following equations, while the simulated results for period of 2016-2017 can be normalized with the similar process.

$$SCon(M) = SPM_{2.5}(E_{L17S17}M_{13}) - SPM_{2.5}(E_{L17S17}M_{17}) \quad (1)$$

$$SCon(S) = SPM_{2.5}(E_{L17S13}M_{17}) - SPM_{2.5}(E_{L17S17}M_{17}) \quad (2)$$

$$SCon(pi) = SPM_{2.5}(E_{L_{pi}S17}M_{17}) - SPM_{2.5}(E_{L17S17}M_{17}) \quad (3)$$

$$NCon(M) = (PM_{2.5OBS2013} - PM_{2.5OBS2017}) \times \frac{SCon(M)}{SCon(M) + SCon(S) + \sum_{i=1}^7 SCon(pi)} \quad (4)$$

$$NCon(S) = (PM_{2.5OBS2013} - PM_{2.5OBS2017}) \times \frac{SCon(S)}{SCon(M) + SCon(S) + \sum_{i=1}^7 SCon(pi)} \quad (5)$$

$$NCon(pi) = (PM_{2.5OBS2013} - PM_{2.5OBS2017}) \times \frac{SCon(pi)}{SCon(M) + SCon(S) + \sum_{i=1}^7 SCon(pi)} \quad (6)$$

where  $SCon(M)$  represents the simulated contribution of meteorology change during 2013-2017, which equals the balance of simulated  $PM_{2.5}$  ( $\mu g m^{-3}$ ) from case  $E_{L17S17}M_{13}$  and case  $E_{L17S17}M_{17}$ . Similarly,  $SCon(M)$  and  $SCon(pi)$  represent the simulated contributions of regional emission reductions and each local control policy type.  $NCon(M)$  represents the normalized contribution of meteorology change during 2013-2017, which equals the product of the observational  $PM_{2.5}$  balance (from 2013-2017) and the proportion of simulated meteorology contribution (in the simulated contributions of all factors). Similarly,  $NCon(M)$  and  $NCon(pi)$  represent the normalized contribution of regional emission reductions and each local control policy type.

### 3 Results and discussion

#### 3.1 Observed changes in surface air quality in Beijing during 2013-2017

In 2013, air pollution was the major environmental problem in Beijing and its surrounding regions (Zhang et al., 2016). In addition to the severe and persistent haze events, the annual mean  $PM_{2.5}$  concentration was  $89.5 \mu\text{g m}^{-3}$  in Beijing. Furthermore, the concentrations of other major air pollutants were also at fairly high levels, with  $56.0 \mu\text{g m}^{-3}$  for  $NO_2$ ,  $26.5 \mu\text{g m}^{-3}$  for  $SO_2$ ,  $183.4 \mu\text{g m}^{-3}$  for  $O_3$  (the statistic refers to the annual average of daily maximum eight-hour sliding, 90th percentile concentration) and  $3.4 \text{mg m}^{-3}$  for CO (the statistic refers to the annual average of daily 24-hour, 95th percentile concentration) (BMEP, 2014).

During 2013-2017, the annual average concentrations of  $SO_2$ ,  $NO_2$ ,  $PM_{2.5}$  and  $PM_{10}$  decreased steadily in Beijing (SI, Figure S3(a)).  $SO_2$  had the most significant decrease of -69.8% (SI, Figure S3(b)), indicating the great effectiveness of the clean air actions on  $SO_2$  emission control.  $PM_{2.5}$  had the second greatest decrement of 35.2%, and the annual concentration of  $PM_{2.5}$  in 2017 was  $58 \mu\text{g m}^{-3}$ , overfulfilling the air quality targets in the *APPCAP*.

Although the annual average  $PM_{2.5}$  concentrations decreased remarkably during 2013-2017, the monthly concentration varied substantially in different years, as shown in Figure 3. Compared with the 2013 level, the average  $PM_{2.5}$  concentrations of each month in 2017 all had a notable decline and presented a similar trend from Jul to Dec. However, compared with the 2016 level, the  $PM_{2.5}$  pollution in Jan and Feb were more severe than those in 2017, while improved obviously after Oct and decreased by nearly 66.3% in Dec (from  $130.7 \mu\text{g m}^{-3}$  in 2016 to  $44.0 \mu\text{g m}^{-3}$  in 2017). The monthly  $PM_{2.5}$  concentrations in Nov and Dec in 2016 were also much higher than those in 2013. A heavy  $PM_{2.5}$  pollution episode occurred in the autumn of 2016 and the winter of 2016-2017. However, the  $PM_{2.5}$  concentrations decreased notably after Sep in 2017 compared with both 2013 and 2016. The observed  $PM_{2.5}$  trends indicate that the emission trend and intensity are major factors in the variation in  $PM_{2.5}$  concentrations, while the meteorology changes also play an important role. The quantification of the contributions of emission control and meteorology changes will benefit numerous future applications.

#### 3.2 Attribution of the 2013-2017 emission reduction in Beijing to specific measures

Based on the MEIC model and the detailed local bottom-up emission inventory, Beijing's atmospheric emissions were updated by year and source sector, as shown in Figure 4. Furthermore, the attribution of emission reductions in Beijing to specific control measures during 2013-2017 and 2016-2017 was displayed in Figure 5.

The major air pollutant emissions in Beijing in 2013 are estimated as follows: 95 kt of  $SO_2$ , 218 kt of  $NO_x$ , 273 kt of volatile organic compounds (VOCs) and 81 kt of  $PM_{2.5}$ . The power and heating sector and the residential sector were the major sources of  $SO_2$  emissions, accounting for 45.1% and 40.6%, respectively.  $NO_x$  emissions mainly came from mobile sources, which contributed 67.2%. Solvent use, mobile sources and industry made notable contributions to VOCs emissions, accounting for 32.0%, 23.8% and 23.7%, respectively. Fugitive dust and the residential sector were the major emitters of  $PM_{2.5}$ , with proportions of 48.7% and 26.2%. However, the implementation of the *Beijing Action Plan* had a significant impact in terms

of local emission reductions. Compared with 2013, Beijing's anthropogenic emissions in 2017 were estimated to have decreased by 83.6% for SO<sub>2</sub>, 42.9% for NO<sub>x</sub>, 42.4% for VOCs, and 54.7% for PM<sub>2.5</sub>. Furthermore, the structure of the emission proportions also changed. For NO<sub>x</sub> emissions, transportation still remained the largest emitter of NO<sub>x</sub> in 2017 but represented a much higher proportion in 2017 than in 2013. The contributions of other sectors, especially power and heating, decreased.

5 With notable contributions of VOCs emission reductions in the residential and industrial sectors, the proportions of these two sectors decreased obviously in 2017, and solvent use as well as transportation became the major emitters. For PM<sub>2.5</sub>, through the effective measures implemented in the residential, industrial, and power and heating sectors, these sectors emitted less PM<sub>2.5</sub> in 2017 than in 2013, and the majority of PM<sub>2.5</sub> emissions came from fugitive dust.

In general, the power and heating, industrial, and residential sectors exhibited the most notable emission reductions during

10 2013-2017. The variations in emissions by sector and year are mainly attributable to air pollution control policies and measures. As previously mentioned, seven types of air pollution control measures were simultaneously contributing to the emission reduction process. According to our research, during 2013-2017, *Coal-fired boiler control* and *Clean fuels in the residential sector* had the most notable effects on SO<sub>2</sub> emission reductions and reduced SO<sub>2</sub> emissions by 35 and 28 kt, respectively, accounting for 44.0% and 35.2% of the total (Figure 6 (a); SI, Table S5). Coal combustion was regarded as the major source

15 of SO<sub>2</sub> emissions in Beijing, where coal is primarily used for residential heating and cooking, coal-fired boilers and power plants. The great emission reduction in SO<sub>2</sub> indicated accurate source identification and effective emission control in the past five years. However, we should also notice that end-of-pipe controls on coal combustion in Beijing have been developed and almost finished recently, leaving little room for further emission reduction. Therefore, the adjustment and optimization of the energy structure would be the most effective and dominant pathway for mitigating coal combustion pollution in the future.

20 According to several studies on the source apportionment of PM<sub>2.5</sub> in Beijing, the transportation sector accounted for a major part of PM<sub>2.5</sub> pollution in 2013, and its contribution has become much higher since then (Li et al., 2015; Li et al., 2017; Hua et al., 2018; Zhang et al., 2018). *Vehicle emission control*, including both on-road and off-road vehicles, was the biggest contributor to NO<sub>x</sub> emission reductions with an estimated total reduction of 44 kt NO<sub>x</sub>, accounting for 47% of the total reductions (Figure 6 (a); SI, Table S5). *Improve end-of-pipe control* reduced the NO<sub>x</sub> emissions by 10 kt in total and accounted

25 for 10.3% of the total NO<sub>x</sub> reductions. In view of the widespread conversion of combustion equipment from coal-based to oil/gas-based equipment, several measures were taken to improve the end-of-pipe control in response to the potential increase in NO<sub>x</sub> emissions, including the application of low-nitrogen-burning (LNB) technologies. A large number of gas-fired or oil-fired boilers, equivalent to 34,000 MV, have been renovated, decreasing NO<sub>x</sub> emissions by nearly 7.5 kt (SI, Table S5). Benefitting from the advanced planning of VOCs pollution control and scientific source apportionment, VOCs control

30 measures were also as effective as other pollutant control measures in Beijing during 2013-2017. *Integrated treatment of VOCs* had the most prominent achievement in reducing VOCs emissions, with a reduction of 57 kt and a proportion of 49.3% (Figure 6 (a); SI, Table S5). *Vehicle emission control* and *Optimize industrial structure* also effectively reduced VOCs emissions, accounting for 16.1% and 11.4%, respectively (Figure 6 (a); SI, Table S5). For PM<sub>2.5</sub> emission control, *Clean fuels in the residential sector*, *Fugitive dust control*, *Coal-fired boiler control* and *Optimize industrial structure* all made obvious

contributions, which reduced the PM<sub>2.5</sub> emissions by 13, 11, 10 and 6 kt, respectively, and accounted for 90.3% of the total (Figure 6 (a); SI, Table S5). In recent years, fugitive dust has gradually become the most dominant source of PM<sub>2.5</sub> emissions, but the relevant control measures are considered less effective than measures focused on coal combustion and the industry sector. Moreover, as the PM<sub>2.5</sub> emissions from the industry sector and coal combustion have gradually decreased and become better managed, fugitive dust, including road dust, construction dust and stock dump dust, has become the most challenging target for future PM<sub>2.5</sub> emission control. In general, *Coal-fired boiler control*, *Clean fuels in the residential sector*, *Optimize industrial structure* and *Vehicle emission control* made significant contributions to pollutant emission reductions in Beijing during 2013-2017 overall, while *Integrated treatment of VOCs* and *Fugitive dust control* achieved prominent reductions in VOCs and PM<sub>2.5</sub> emissions.

To ensure that the national air quality targets of the APPCAP could be achieved as scheduled, Beijing implemented a series of stronger and more targeted pollution control policies and measures since 2016. For energy structure adjustment, measures associated with *Clean fuels in the residential sector* were enhanced. A total of 92,000 households in urban areas and 369,000 households in rural areas converted the scattered coal-used fuels into clean fuels, close to the total amount of 2013-2016. For industrial structure adjustment, Beijing strengthened the elimination and management of small, cluttered and heavily polluting factories. More than 6,500 factories were eliminated during 2016-2017, approaching one and a half times compared with the total amount during 2013-2016. During 2016-2017, SO<sub>2</sub>, NO<sub>x</sub>, VOCs and PM<sub>2.5</sub> were estimated to have decreased by 19.6 kt, 29.0 kt, 42.9 kt and 15.7 kt, respectively (SI, Table S5). *Clean fuels in the residential sector*, *Coal-fired boiler control*, and *Optimize industrial structure* were the top three most effective local measures during this period. In addition, with the enhanced management of non-point air pollution sources, including fugitive dust, heavily polluting vehicles and domestic solvent use, the relevant control measures generated more remarkable emission reductions in this period than the previous periods. For instance, *Fugitive dust control* was estimated to have decreased the PM<sub>2.5</sub> emissions by 4.7 kt during 2016-2017, representing 42.5% of the total PM<sub>2.5</sub> emission reductions by *Fugitive dust control* during 2013-2017.

### 3.3 Emission reduction in surrounding regions during 2013-2017

According to our previous research (Zheng et al., 2018), the implementation of national clean air actions has brought conspicuous emission reductions in Beijing's surrounding regions (including Tianjin, Hebei, Henan, Shandong, Shanxi and Inner Mongolia) from 2013-2017 (China State Council, 2018). Figure 6 showed the updated emission inventory of Beijing's surrounding regions by year and sector during 2013-2017.

According to Figure 6, SO<sub>2</sub> and PM<sub>2.5</sub> emissions presented rapid decreasing trends from 2013-2017, while the trend of VOCs emissions remained steady with a slight increment. Compared with 2013, SO<sub>2</sub>, NO<sub>x</sub> and PM<sub>2.5</sub> emissions were estimated to decrease by 59.5%, 22.9% and 36.6%, respectively, while VOCs emissions increased slightly by 0.2% (Zheng et al., 2018). During 2013-2017, the industrial and the power and heating sectors made the most prominent contributions to SO<sub>2</sub> emission reductions, which decreased SO<sub>2</sub> emissions by 3,021 and 2,000 kt, respectively, indicating that the SO<sub>2</sub> emissions control measures were quite effective (Cai et al., 2017; Shao et al., 2018). Reductions in the NO<sub>x</sub> emissions mainly came from the

power and heating sector, with a reduction of 1,541 kt within five years. For PM<sub>2.5</sub> emission reductions, industrial sectors were the greatest contributors, with reductions of 621 and 528 kt. The VOCs emissions in the surrounding regions continued to increase, especially in the solvent use sectors. During the process of implementing national clean air actions, the surrounding regions also carried out several valid measures to control VOCs emissions, such as the supervision and repair of gasoline stations, oil tankers and oil transfer processes and the integrated treatment and management of petrochemical and refinery industries (Hui et al., 2019). The VOCs emissions from the residential and transportation sectors in 2017 decreased by 20.7%. However, due to the lack of thorough regulation of chemical industries and the ineffective end-of-pipe control of solvent use sources, the total VOCs emissions increased slightly in 2017 (Zheng et al., 2018).

### 3.4 Decomposition of PM<sub>2.5</sub> concentration changes in Beijing during 2013-2017

#### 10 3.4.1 Modelled PM<sub>2.5</sub> air quality changes in Beijing during 2013-2017

According to the base simulation results, the annual average PM<sub>2.5</sub> concentrations of Beijing decreased by 32.8 µg m<sup>-3</sup> from 2013-2017 and by 16.6 µg m<sup>-3</sup> from 2016-2017, which agrees well with the observed decreases (31.5 µg m<sup>-3</sup> from 2013-2017 and 14.9 µg m<sup>-3</sup> from 2016-2017). Although there was a steady decline in PM<sub>2.5</sub> concentrations of Beijing during 2013-2017, the trends of PM<sub>2.5</sub> compositions varied differently. The simulation results of base cases (which adopted the real meteorology and emissions of each year) showed that the sulfate (SO<sub>4</sub><sup>2-</sup>) and organic matter (OM) were dominate species for the decline in PM<sub>2.5</sub> concentrations during 2013-2017, with the decrement of 7.5 µg m<sup>-3</sup> (56.6%) and 9.6 µg m<sup>-3</sup> (40.5%) respectively. The contribution of SO<sub>4</sub><sup>2-</sup> to the total PM<sub>2.5</sub> also decreased obviously, from 15.3% in 2013 to 10.7% in 2017; and OM proportion decreased from 27.5% in 2013 to 26.5% in 2017. The rapid decrement of SO<sub>4</sub><sup>2-</sup> was consistent with the remarkable SO<sub>2</sub> emission reductions in Beijing during 2013-2017. Along with the effective SO<sub>2</sub> emission control measures, the SO<sub>4</sub><sup>2-</sup> was basically no longer the key contributor leading to heavy pollution in Beijing while the nitrate-driven haze pollution has become more dominate in Beijing in recent years, especially in the summertime (Li et al., 2018). The decrement of OM was mainly caused by the prominent emission reductions of primary organic carbon (mainly come from the residential burning and other coal combustion sources). VOCs emission reductions also contributed to the OM decreasing, however, due to the insufficient simulation of secondary organic aerosols (SOA) formulations in CMAQ model, the contributions of VOCs emission control might be underestimated. In contrast, nitrate (NO<sub>3</sub><sup>-</sup>) increased in 2014-2016, and kept basically the same concentration level in 2017 (10.4 µg m<sup>-3</sup>) as 2013 (10.9 µg m<sup>-3</sup>). However, the contribution of NO<sub>3</sub><sup>-</sup> to the total PM<sub>2.5</sub> increased a lot, from 12.7% in 2013 to 19.4% in 2017. The specific concentration and proportion trends of PM<sub>2.5</sub> concentrations can be found in SI, Table S6.

Figure 7 shows the spatial distribution of PM<sub>2.5</sub> concentrations in Beijing and the surrounding areas in 2013, 2016 and 2017 (panels (a)-(c)), along with the total PM<sub>2.5</sub> changes and the changes due to major contributing factors from 2013-2017 (panels (d)-(f)) and 2016-2017 (panels (g)-(i)). In 2013, some typical regions, such as the southern Beijing and most of the cities of

Tianjin, Tangshan, Baoding, Shijiazhuang, Handan, and Anyang, suffered intense PM<sub>2.5</sub> pollution. After implementing the APPCAP and local air pollution control policies, severe pollution was mitigated in most regions, although several heavily polluted spots still existed. However, Beijing had successfully removed itself from the list of heavily polluted areas. According to the base simulation results (Figure 7, panels (a)-(c)), Beijing, especially the southern area, had the most notable PM<sub>2.5</sub> decrease among all parts of the third nested simulation domain. The municipality of Tianjin and southwestern Hebei Province also achieved prominent abatements in PM<sub>2.5</sub> pollution. Based on the spatial distributions of total PM<sub>2.5</sub> changes and changes due to major contributing factors in Beijing (Figure 7, panels (d)-(i)), the control of emissions dominated the PM<sub>2.5</sub> changes in both 2013-2017 and 2016-2017; however, the favourable effects of meteorological changes during 2016-2017 were much more remarkable. We further estimated and quantified the contributions of each factor as follows.

### 10 3.4.2 Contribution from changes in meteorological conditions

According to the simulation results of the base cases and emission-fixed sensitivity experiments (Table 2; Table S6), the meteorological conditions in 2017 were found to be more favourable than those in the previous periods, especially 2016. During 2013-2017, changes in meteorological conditions contributed 3.8  $\mu\text{g m}^{-3}$  to the PM<sub>2.5</sub> air quality improvements, accounting for 12.1% of the total abatements. Under the meteorological conditions of 2013 and the emission level of 2017, the annual average PM<sub>2.5</sub> concentration of Beijing would have decreased from 90  $\mu\text{g m}^{-3}$  to 62-62.5  $\mu\text{g m}^{-3}$  and would not have achieved the air quality targets established in the APPCAP. During 2016-2017, the favourable effects of meteorology changes became much more striking and contributed 4.4  $\mu\text{g m}^{-3}$ , accounting for 29.5% of the total PM<sub>2.5</sub> abatements from 2016-2017. Similarly, under the meteorological conditions of 2016, the PM<sub>2.5</sub> level in Beijing in 2017 would have decreased to 62.5-63.0  $\mu\text{g m}^{-3}$ , still in excess of the APPCAP target.

20 From the perspective of annual average analysis, changes in meteorology generally had a beneficial effect on air pollution mitigation in 2017; however, the impact varied greatly in the monthly analysis. Figure 8 showed the monthly average simulated PM<sub>2.5</sub> concentrations in the two fixed-emission sensitivity experiments. Compared with the meteorological conditions of 2013 (Figure 8 (a)), the meteorological conditions of 2017 became better in winter, especially in Jan and Feb. Under the anthropogenic emissions of 2017, the meteorological conditions of Jan and Feb 2013 would have increased the PM<sub>2.5</sub> concentration by 22.5% and 37.7%, respectively. However, the meteorological conditions of 2017 were worse than those of 2013 in spring and summer, especially in Apr, May and Jul. The air quality was good in the first few months of 2016, and the simulation results also indicated that the meteorological conditions during this period in 2017 were much worse than those in 2016, increasing the PM<sub>2.5</sub> concentrations by 51.6  $\mu\text{g m}^{-3}$  in Jan and 28.6  $\mu\text{g m}^{-3}$  in Feb (Figure 8 (b)). However, the conditions improved in the following months, especially during Oct to Dec. If the meteorological conditions remained the same as those in 2016, the monthly average PM<sub>2.5</sub> concentrations of Oct, Nov, and Dec would have increased by 25.4%, 58.0% and 92.4%, respectively. It is worth noting that severe PM<sub>2.5</sub> pollution and haze events always occur in winter in North China; therefore,

remarkable improvements in the meteorological conditions in Jan, Feb, Nov and Dec would contribute greatly to the mitigation of annual  $\text{PM}_{2.5}$  concentrations.

### 3.4.3 Contribution from local and regional emission reduction measures

Although the changes in meteorological conditions were favourable for  $\text{PM}_{2.5}$ -related air quality improvements in Beijing, the control of emissions was still the dominant factor in  $\text{PM}_{2.5}$  abatement in recent years and contributed the reductions of  $27.7 \mu\text{g m}^{-3}$  (accounting for 87.9%) and  $10.5 \mu\text{g m}^{-3}$  (accounting for 70.5%) in 2013-2017 and 2016-2017, respectively.

According to the simulation results of the regional emission-fixed sensitivity experiments (Table 2; Table S6), the contributions of regional emission reductions to the  $\text{PM}_{2.5}$  abatements in Beijing were  $7.1 \mu\text{g m}^{-3}$  during 2013-2017 and  $2.5 \mu\text{g m}^{-3}$  during 2016-2017, accounting for 22.5% and 16.8%, respectively. The results indicated that by implementing the APPCAP, regional provinces and cities around Beijing achieved notable emission control effects. In particular, emission reductions in the industrial and power sectors have made striking contributions to  $\text{PM}_{2.5}$ -related air quality improvements in regional areas.

In addition to the impacts of meteorology changes and regional emission reductions, the contributions of local emission control to the  $\text{PM}_{2.5}$  abatements in Beijing were estimated to be  $20.6 \mu\text{g m}^{-3}$  (2013-2017) and  $8.0 \mu\text{g m}^{-3}$  (2016-2017), accounting for 65.4% and 53.7%, respectively. According to the results of the measure-related sensitivity experiments (Table S6), we further decomposed the contributions due to local emission control into each specific measure. As Figure 9 shows, during 2013-2017, *Coal-fired boiler control* made the largest contribution of  $5.9 \mu\text{g m}^{-3}$ , accounting for 18.7% of the total decrement. *Clean fuels in the residential sector* was the second greatest contributor after *Coal-fired boiler control*, decreasing  $\text{PM}_{2.5}$  concentrations by  $5.3 \mu\text{g m}^{-3}$ . Measures associated with *Optimize industrial structure* also effectively reduced  $\text{PM}_{2.5}$  concentrations, with a decrease of  $3.2 \mu\text{g m}^{-3}$  and a proportion of 10.2%. Measures associated with *Fugitive dust control*, *Vehicle emission control*, *Improved end-of-pipe control*, and *Integrated treatment of VOCs* had relatively minor contributions and reduced the  $\text{PM}_{2.5}$  concentrations in Beijing by 7.3%, 6.0%, 5.7% and 0.6%, respectively, from 2013-2017 (Figure 9 (a)). During 2016-2017, *Clean fuels in the residential sector*, *Coal-fired boiler control* and *Optimize industrial structure* were the top three contributors to the  $\text{PM}_{2.5}$  abatements among all local policies, accounting for 14.1%, 12.8% and 10.1% of the total (Figure 9 (b)). These results highlight the great enhancement in the control of bulk coal use and the elimination of small, clustered, heavily polluting factories during this period.

In summary, the improvement in the  $\text{PM}_{2.5}$ -related air quality in Beijing was decomposed, and the results were shown in Figure 10. During 2013-2017, meteorology changes, surrounding emission reductions and local emission control contributed  $3.8$ ,  $7.1$  and  $20.6 \mu\text{g m}^{-3}$ , respectively, accounting for 12.1%, 22.5%, and 65.4%. *Coal-fired boiler control*, *Clean fuels in the residential sector* and *Optimize industrial structure* were the top three contributors among all local emission control policies. Emission reduction was the most dominant factor in the air quality improvements in Beijing during this period. For 2016-2017, the contributions of meteorology changes, surrounding emission reductions and local emission control were  $4.4$ ,  $2.5$  and  $8.0 \mu\text{g m}^{-3}$ , respectively. The favourable meteorological conditions during this period had a remarkable effect, accounting for 29.5% of



the total pollution reduction. The top three local control measures of this period were same as those of 2013-2017 but had a different order. *Clean fuels in the residential sector* and *Optimize industrial structure* made larger contributions.

### 3.5 Uncertainties and limitations

We built a model-based decomposition framework and attributed the PM<sub>2.5</sub> abatements in Beijing during 2013-2017 and 2016-2017, however certain uncertainties and limitations remain. The major uncertainties and limitations of this work are discussed below.

#### 3.5.1 Uncertainties of zero-out approach

Although various methods have been developed to quantify the source of PM<sub>2.5</sub> and evaluate their contributions, such as receptor-based methods (like CMB and PMF), trajectory-based methods (like PSCF and EEI), source-oriented methods (like CAMx-PSAT and CMAQ-ISAM) (Li et al, 2015), they can hardly consider the meteorology and emission changes simultaneously. Therefore, the zero-out approach might be a better choice to attribute the contribution of local and regional emission control as well as meteorology changes under one complete decomposition framework. The zero-out method is also widely used in estimating the contribution of air pollution sources (Lelieveld et al., 2015; Han et al., 2016; Baker et al., 2016; Zhang et al., 2017; Zhang et al., 2017; Ni et al., 2018). However, the response of PM<sub>2.5</sub> formulation is not linear to the meteorology and emission changes; thus, the zero-out approach would introduce extra bias in research. The non-linear effects of the analyse period of 2013-2017 could be evaluated by the following equation (Zhang et al., 2017).

$$\text{Bias} = (\text{SCon}(M) + \text{SCon}(S) + \sum_{i=1}^7 \text{SCon}(pi)) - (\text{SPM}_{2.5}(E_{L13S13}M_{13}) - \text{SPM}_{2.5}(E_{L17S17}M_{17})) \quad (7)$$

Where  $\text{SPM}_{2.5}(E_{L13S13}M_{13})$  and  $\text{SPM}_{2.5}(E_{L17S17}M_{17})$  represent the direct simulated PM<sub>2.5</sub> concentration of base case in 2013 and 2017. The balance of their values is the actual PM<sub>2.5</sub> decrement during 2013-2017 under the mixed impacts of meteorology change, regional and local emission reductions. The sum of  $\text{SCon}(M)$ ,  $\text{SCon}(S)$  and  $\sum_{i=1}^7 \text{SCon}(pi)$  represents a linear result of all contributors during this period. The extra bias can be estimated as the difference between the linear addition and the actual decrement. According to equation (7), we estimated biases in the analyse of 2013-2017 were 1.4  $\mu\text{g m}^{-3}$ , accounting for 4.3%. Similarly, the absolute and relative biases in the analysis of 2016-2017 were estimated as -0.6  $\mu\text{g m}^{-3}$  and -3.6%. Both indicated the non-linear effects are relatively small and acceptable.

#### 3.5.2 Uncertainties in emission estimates

The incomplete research and investigation of activity rates, emission factors, and removal efficiencies would introduce uncertainties in estimating the emission trends and magnitudes, as well as the measure-based emission reductions.

Based on our previous work, the uncertainty of MEIC was estimated to be  $\pm 12\%$  for  $\text{SO}_2$ ,  $\pm 31\%$  for  $\text{NO}_x$ ,  $\pm 70\%$  for  $\text{CO}$ ,  $\pm 68\%$  for VOCs,  $\pm 130\%$  for  $\text{PM}_{2.5}$ ,  $\pm 208\%$  for BC and  $\pm 258\%$  for OC (Zhang et al., 2009). The larger uncertainty of BC and OC are mostly because their major emitters are much more scattered and harder to investigate or quantify, such as heavy diesel vehicle and residential burning. The uncertainty of updated emission inventory of China during 2010-2017 was evaluated in Zheng's work by comparing the emissions with observations (Zheng et al., 2018), and reported a good agreement. Similarly, we discussed the uncertainty of emission trends and relative change ratios in BJ-EI. Ground-based, satellite-based observational data and the estimated emissions from BJ-EI were all normalized to the base year, 2013. We compared the observational concentration trends of major pollutants with their precursor emissions (SI, Figure S5). A good agreement was found in the trends of  $\text{SO}_2$  emission, OMI  $\text{SO}_2$  column and surface observational  $\text{SO}_2$  concentration, with a decreasing ratio of 83.6%, 77.6%, 69.8% respectively (SI, Figure S5(a)), indicating a relatively small uncertainty in  $\text{SO}_2$  emission estimation. Surface concentration trend during 2016-2017 became flatter than OMI  $\text{SO}_2$  columns trend, which might be partly owing to the relatively less surface  $\text{SO}_2$  emission reductions while a more significant high-stack emission reductions in local and surrounding areas. The  $\text{NO}_x$  emission trend was basically consistent with the variation in  $\text{NO}_2$  tropospheric vertical column, decreasing 43.2% and 40.3% respectively, but both lower than the surface  $\text{NO}_2$  concentration trend (SI, Figure S5(b)). This phenomenon might be caused by the meteorology impacts (Uno et al., 2007), chemical reactions of nitrous oxides (Valin et al., 2011), and the overestimation of surface  $\text{NO}_2$  observations (Lamsal et al., 2010). Satellite-derived and surface observational  $\text{PM}_{2.5}$  concentrations decreased 36.0% and 35.2% respectively during 2013-2017 in Beijing, both agreed with the trends of primary  $\text{PM}_{2.5}$  and precursor emissions (SI, Figure S5(c)). Among all precursors, the decreasing rate of  $\text{SO}_2$  was more significant and rapid than that of  $\text{NO}_x$  and  $\text{HN}_3$ , which was consistent with the simulation results that the proportion of sulfate in  $\text{PM}_{2.5}$  decreased obviously while the contributions of nitrate and ammonium increased (SI, Figure S4). The decrement of VOCs emissions contributed to  $\text{PM}_{2.5}$  abatements by decreasing OM. Similar phenomenon was also reported in previous researches (Shao et al., 2018). In general, the relatively good coherence of emission and observation variations indicated that the BJ-EI basically well quantified the actual emission trends and variations in Beijing during 2013-2017.

Estimation of measure-based emission reductions is another major aspect of introducing uncertainty. However, the uncertainty of this aspect is hard to quantify. Emission control measures can have independent or integrated impacts on activity rate, emission factor, technology evolution, and end-of-pipe removal efficiency, which are all sensitive to emission calculation and remaining large uncertainty. *Coal-fired boilers control* was the most explicit control policy to quantify, for the detailed and accurate information of unit-based power plants and facility-based boilers. The amount of eliminated coal, newly increased other clean energy, the evolutions of emission factor and removal efficiency of each boiler were collected sufficiently, which can largely lower the uncertainty of the reduction estimation. Similar to *Coal-fired boilers control*, *Improve end-of-pipe control* mainly focused on the heavy-polluted manufacturing factories and gas/oil-fired boilers. The facility-based information and the accurate eliminated capacities make the uncertainty relatively small. As for the policy of *Clean fuels in residential sector*, the reduced residential coal use, the eliminated traditional biofuels were collected at a county-level, thus the uncertainty of activity rate estimation was relatively small. However, along with the promotion of coal quality (such as the lower sulfur and

ash content) and the evolution of domestic burning equipment, the improvement of emission factors was hard to estimate, especially in the rural areas, which would introduce large uncertainty. Major limitations of estimating *Optimize industrial structure* reductions came from the elimination of small, clustered and polluting factories. Different from high-polluted enterprises, the specific information of these scattered factories was hard to investigate. Although we knew a total of 11,000 such factories were phased out during 2013-2017, the activity rate, emission factor or end-of-pipe control information were ambiguous, which would easily lead to the underestimation or overestimation of reductions from this sub-measure. *Integrated treatment of VOCs* included the VOCs control of chemical industry and solvent use sectors. Yan Shan company is the only chemical factory in Beijing, and the specific information of this factory made the estimation more reasonable. However, investigations for the amounts of solvent used were limited; meanwhile, the lack of emission factor measurements for various solvent-use-related sources also introduced large uncertainty (Li et al., 2017a). Reductions from *fugitive dust control* might be the most difficult one to estimate, with the larger uncertainty. This policy type contained the control of road dust and construction dust. Due to the lack of measurements for the real-time traffic flow, threshold friction velocities, surface roughness length, the efficiency of road cleaning process and other key parameters for emission calculation, we estimated the emission reductions from this sub-measure by the improvements of cleaning process adopted ratio and the various road areas, which might not totally reflect the actual emission change of road dust. For the construction dust estimation, although we collected the information of each construction site and stock yard in Beijing, the indefinite emission process and factors would also create uncertainties for estimation. In general, emission reductions from the policies which focused on non-point and scattered emission sources, such as road dust, small and clustered factories, various solvent use sources, are more difficult to quantify, and would cause larger uncertainties. Additional detailed information and real-world measurements might help to lower these uncertainties.

### 3.5.3 Other uncertainties

The decomposed analysis was also affected by the inherent WRF-CMAQ uncertainties, as Sect.2.4.2 discussed. The missing mechanism and insufficient simulation of secondary organic aerosols (SOA) formulations in CMAQ model might lead to the underestimation of the OM, as well as the contribution of VOCs emission control. The rough vertical layers, the underestimation of nature source emissions, the defect of upper boundary simulation in regional model and the uncertainties of VOCs emission inventories might all lead to the deficiency of O<sub>3</sub> underestimation. The missing of construction dust and the underestimation of in-line windblown dust model might result in the lower PM<sub>10</sub> simulation in other regions. And the open biomass burning was not included in this study, which would also introduce certain uncertainties. Given that we focused more on the decomposed attribution of anthropogenic emission changes and meteorology impacts, and the simulations of PM<sub>2.5</sub> concentrations, compositions basically captured the temporal and spatial variations, the uncertainties originated from this aspect were relatively small. However, further studies and efforts should be made to improve the model simulations.

#### 4 Concluding Remarks

The remarkable decreases in the annual average  $PM_{2.5}$  concentrations in Beijing from 2013-2017 and 2016-2017 were the combined results of various factors. In this study, based on a series of numerical simulation experiments and a decomposed attribution analysis, local air pollution control policies, surrounding emission reductions and favourable meteorological conditions were estimated to contribute 65.4% ( $20.6 \mu\text{g m}^{-3}$ ), 22.5% ( $7.1 \mu\text{g m}^{-3}$ ) and 12.1% ( $3.8 \mu\text{g m}^{-3}$ ), respectively, of the total  $PM_{2.5}$  abatements in Beijing ( $31.5 \mu\text{g m}^{-3}$ ) from 2013-2017; and 53.7% ( $8.0 \mu\text{g m}^{-3}$ ), 16.8% ( $2.5 \mu\text{g m}^{-3}$ ), 29.5% ( $4.4 \mu\text{g m}^{-3}$ ), respectively, of the total  $PM_{2.5}$  abatements ( $14.9 \mu\text{g m}^{-3}$ ) from 2016-2017. During 2013-2017, air pollution control policies had the most dominant effect on  $PM_{2.5}$  abatements, accounting for nearly 88%, but the meteorological impacts have been considerable since 2016, especially in the winter of 2016-2017 and the autumn of 2017.

Under the *Beijing Action Plan*, anthropogenic emissions were reduced by 83.6% for  $SO_2$ , 42.9% for  $NO_x$ , 42.4% for VOCs, and 54.7% for  $PM_{2.5}$  compared with the 2013 level. Under the *APPCAP*, the areas surrounding Beijing also reduced their pollutant emissions by 59.5% for  $SO_2$ , 22.9% for  $NO_x$ , and 36.6% for  $PM_{2.5}$ . A measure-by-measure analysis showed that *Coal-fired boiler control*, *Clean fuels in the residential sector* and *Optimize industrial structure* were the most effective control measures in general for Beijing during 2013-2017 and 2016-2017, both in terms of emission reductions and  $PM_{2.5}$  pollution mitigation.

The results indicated several options for future air pollution control in Beijing. The most notable effect of the *Beijing Action Plan* mainly came from the control of combustion, which suggests that power plants, coal-fired boilers, and residential burning have account for the majority of air pollution sources for a long time. Consequently, Beijing should continue to optimize the city's energy structure to achieve a qualitative improvement in energy consumption. However, with the progress on air pollution control in Beijing, the contributions of combustion and industry emission control, of which the sectors and sources are relatively easy to identify and manage, have gradually decreased, and there is less room for further improvement. Pollutant emissions from domestic living, such as transportation, restaurant fumes and residential solvent use, have increasingly accounted for larger proportions. Vehicles, VOCs emission sources and fugitive dust have gradually become the major and most difficult challenges for Beijing's future air pollution control. On the one hand, the government should further apply stronger and more effective management of non-point pollution sources arising from the demands for city development, such as catering enterprises, vehicles, off-road transportation, construction sites, and the use of solvents and coatings. More resources and investment, more accurate identification and refined management strategies are needed for these diffuse pollution sources. On the other hand, the support and innovation of science and technology should be enhanced further, including not only high-technology strategies of pollutant removal and equipment renovation but also the understanding of pollution mechanisms and the identification of pollution sources. For instance, the scientific source apportionment of atmospheric particulates, the dynamic update of emission inventories, the application of widespread observation systems, the construction of pollution forecasts and warning systems, etc. should be developed further. A support system for air quality

analysis, decision making, implementation, assessment and optimization should be established in the future to make qualitative leaps in environmental protection.

### Author contributions

Q.Z., J.L. and K.H. conceived the study; J.S., T.C. and F.S. developed bottom-up emission inventory over Beijing; X.L., X.D.,  
5 and Y.Y. collected pollution control policies over Beijing; D.T. and Y.Z. estimated regional emission reductions; Y.L collected  
and handled with the satellite data; J.C. estimated emission reductions over Beijing and performed CMAQ experiments; J.C.  
and Q.Z. prepared the manuscript with contributions from all co-authors.

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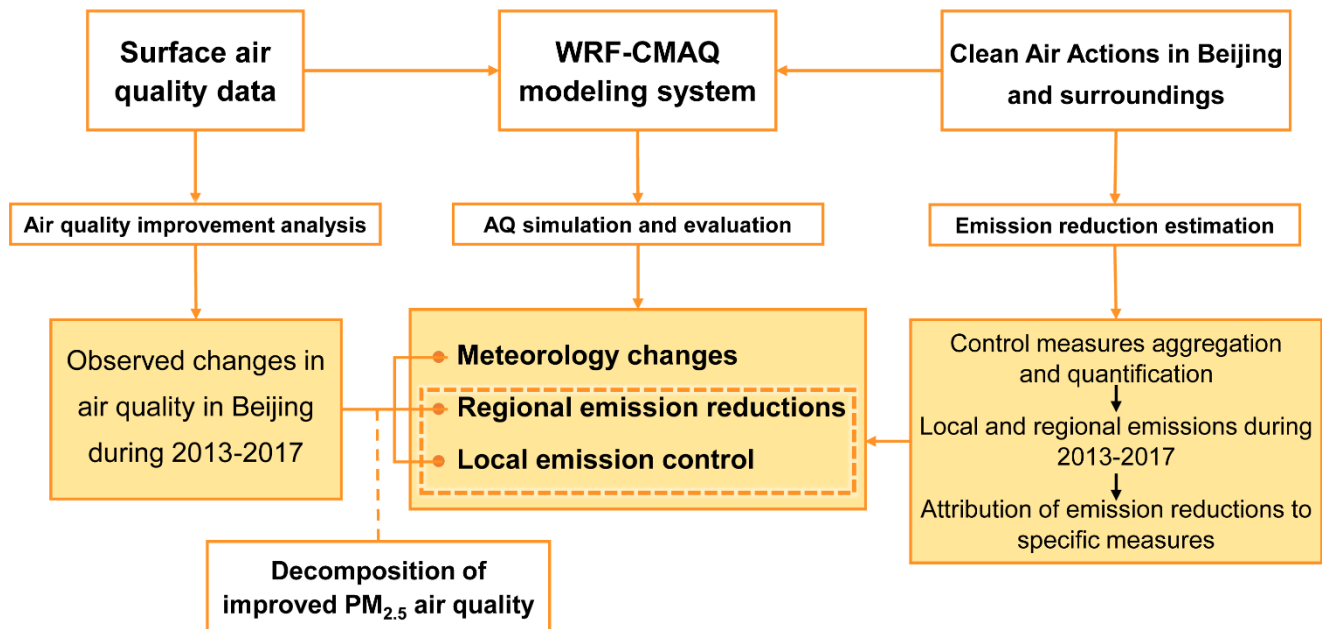


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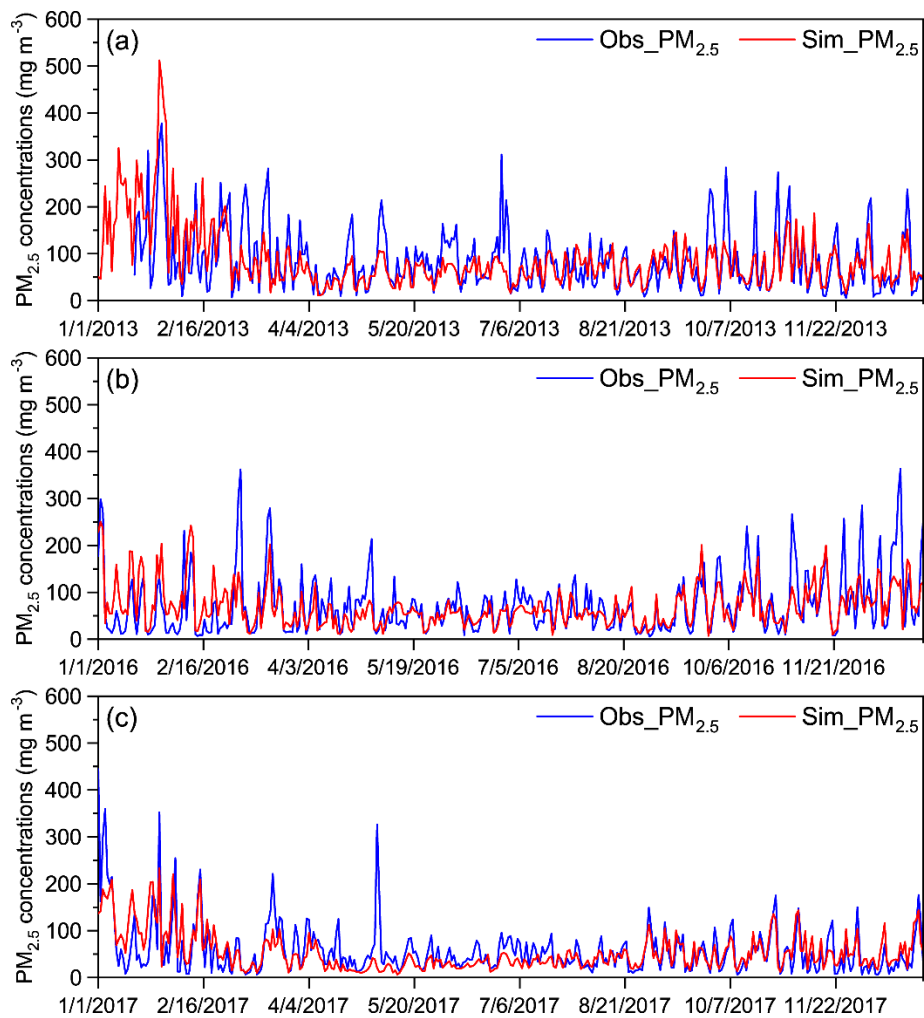
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5 **Figure 1: Methodological framework for the decomposition analysis of improved PM<sub>2.5</sub>-related air quality in Beijing during 2013-2017.**



**Figure 2: Comparison of observed (blue) and CMAQ-simulated (red) daily mean PM<sub>2.5</sub> concentrations over Beijing in 2013 (a), 2016 (b) and 2017 (c). Observation data was obtained and averaged from 12 national observation stations in Beijing. Simulated concentrations were extracted from the grids corresponding to the station locations.**

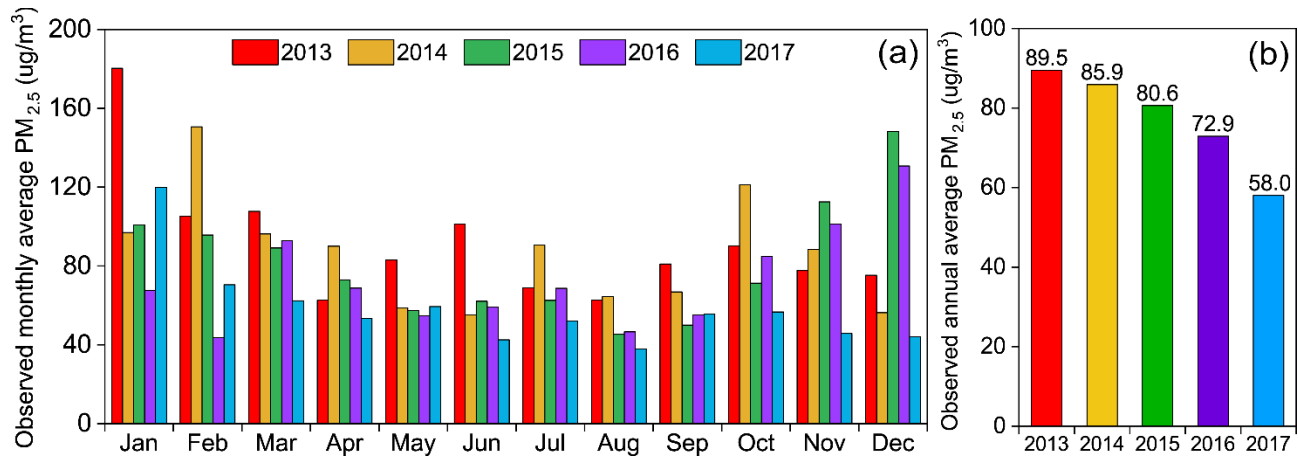


Figure 3: Observed monthly (a) and annual (b) average PM<sub>2.5</sub> concentrations in Beijing during 2013-2017.

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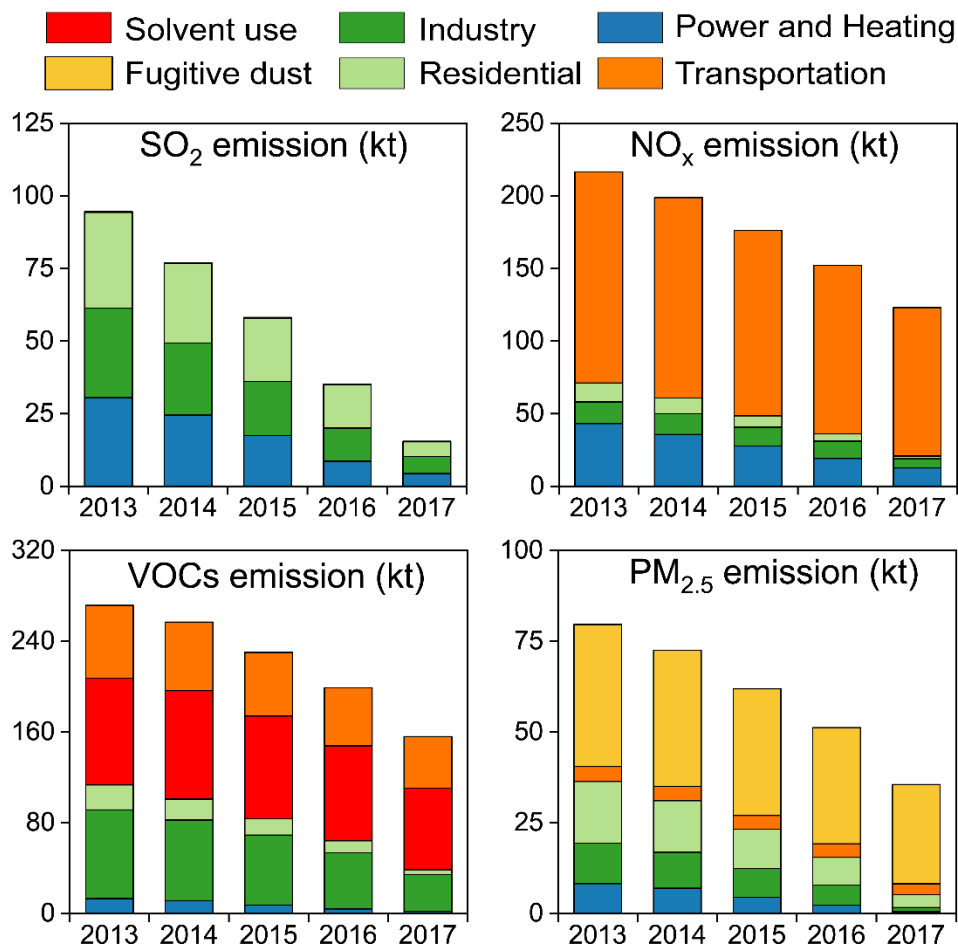


Figure 4: Changes in anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, VOCs and primary PM<sub>2.5</sub> in Beijing during 2013-2017.



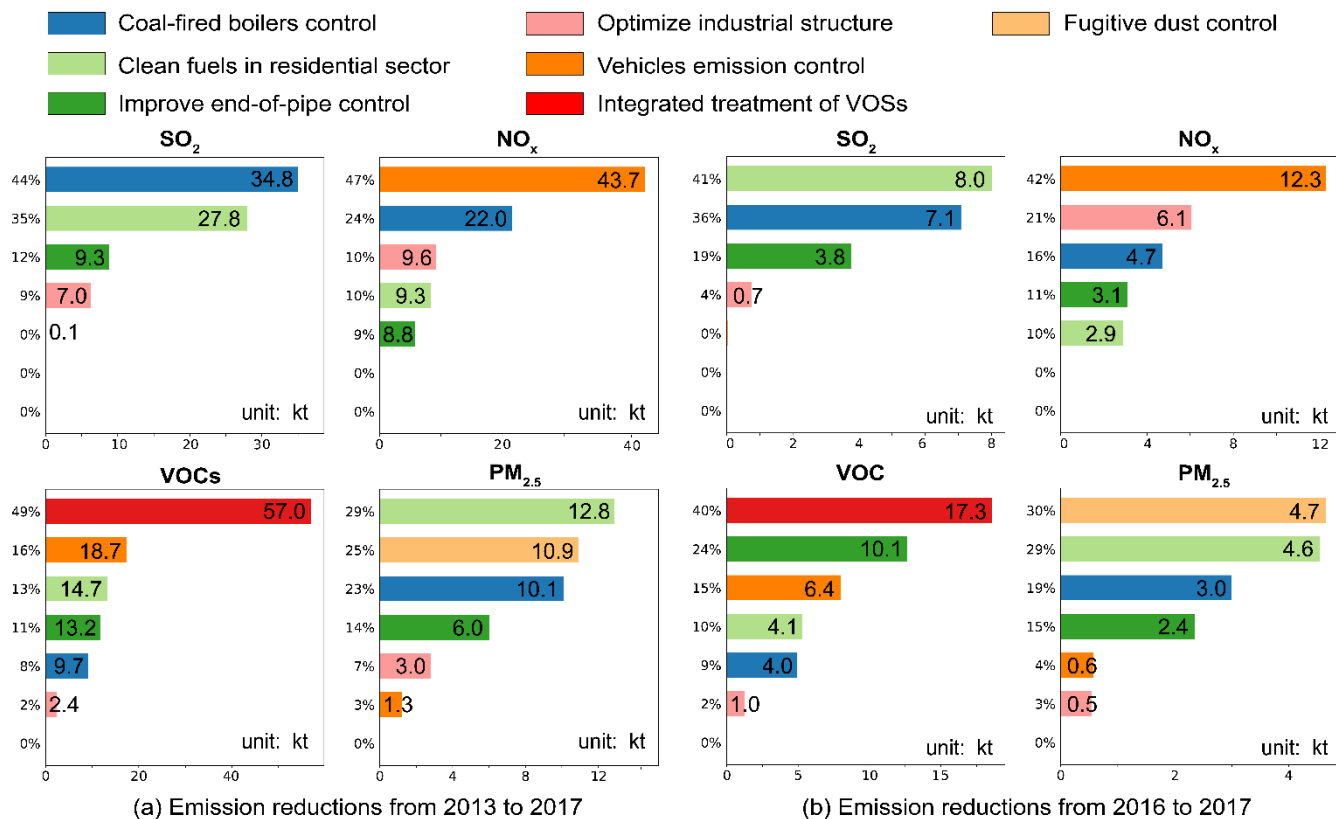


Figure 5: Emission reductions of SO<sub>2</sub>, NO<sub>x</sub>, VOCs and primary PM<sub>2.5</sub> attributed to seven types of control policies in Beijing for the periods of 2013-2017 (a) and 2016-2017 (b). The relative contribution of each policy to the total emission reduction is presented on the Y-axis. The number on each bar represents the absolute emission reductions by the relevant control policy.

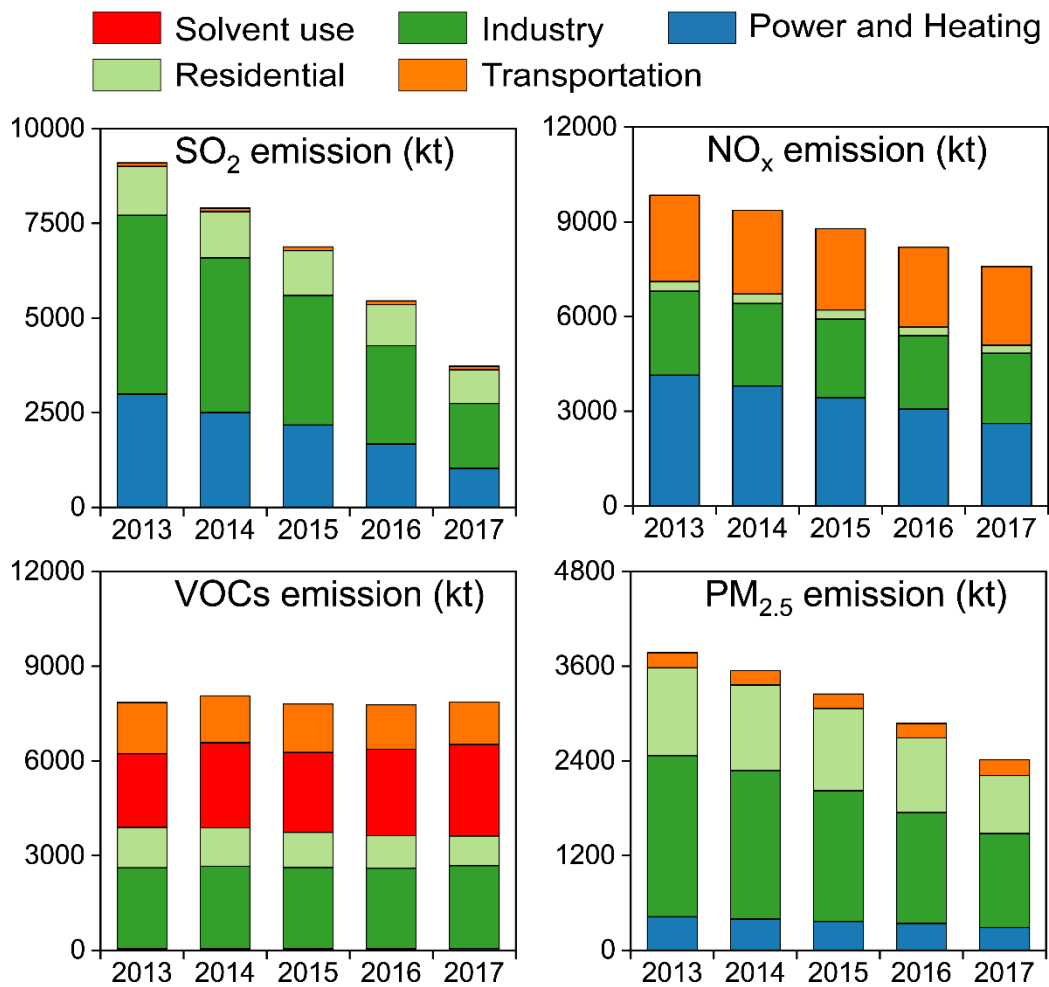
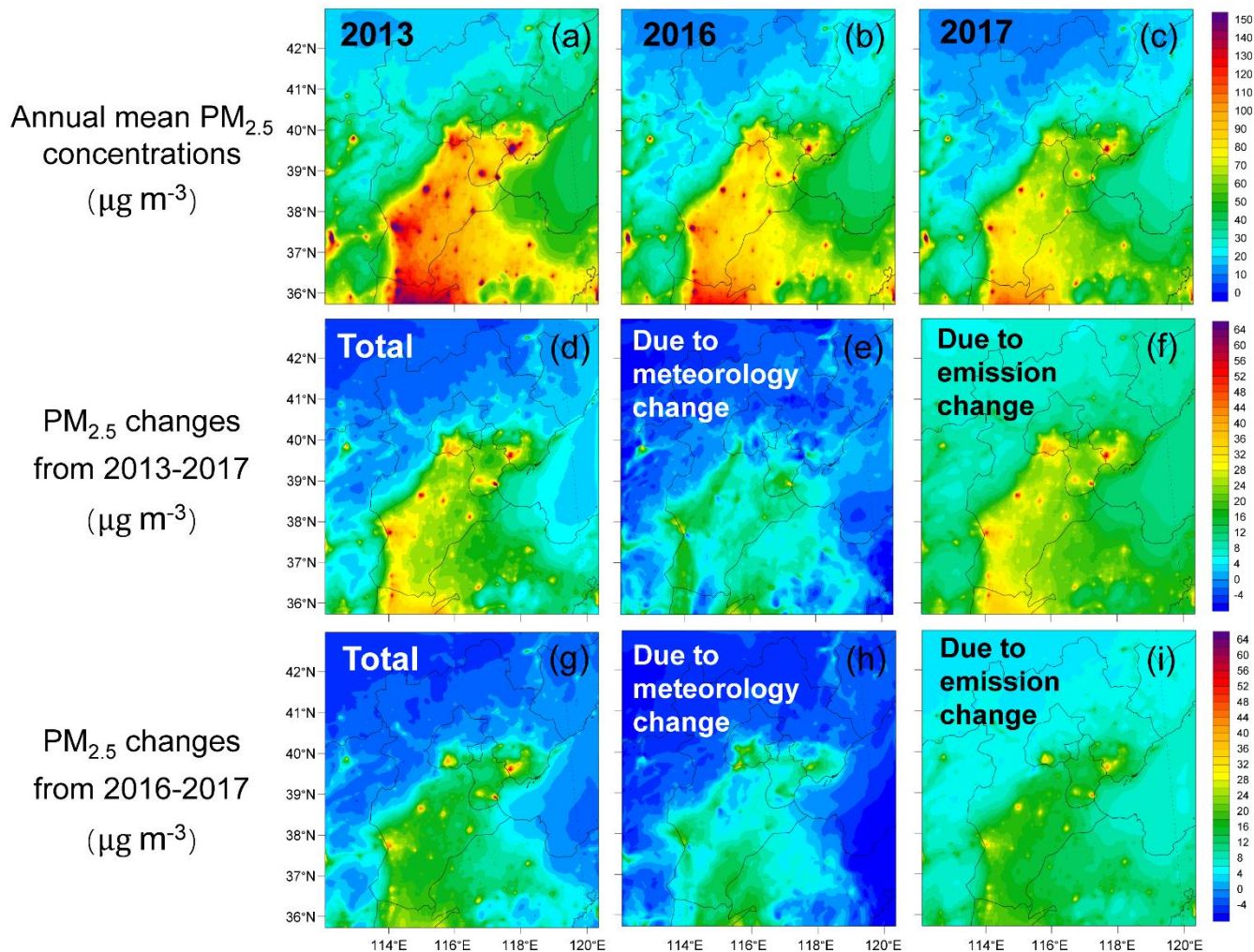
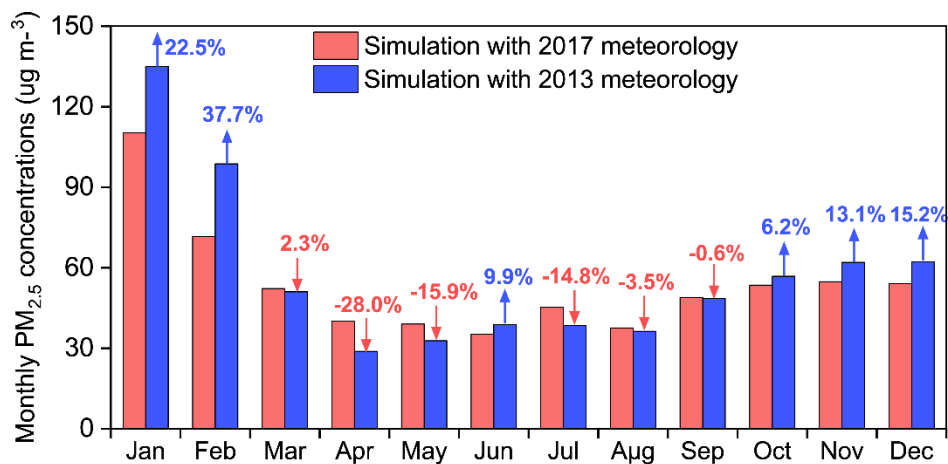


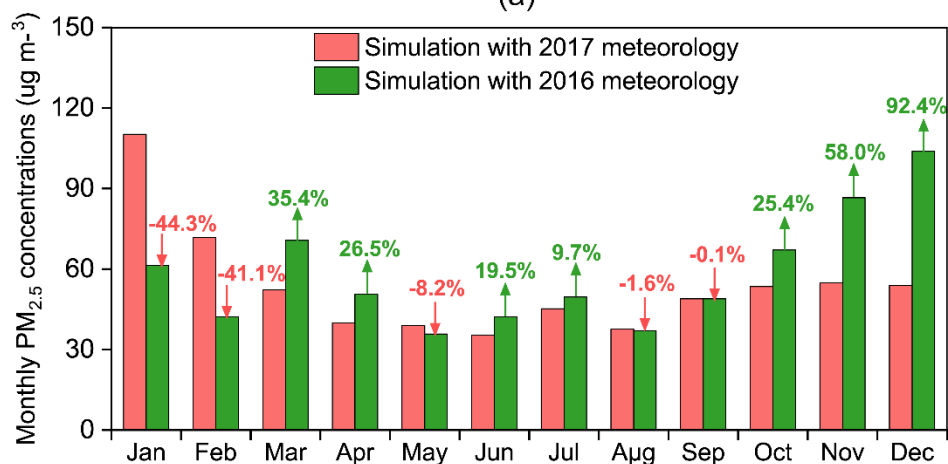
Figure 6: Changes in anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, VOCs and primary PM<sub>2.5</sub> in the areas surrounding Beijing during 2013-2017. The regions include Tianjin, Hebei, Henan, Shandong, Shanxi and Inner Mongolia.



**Figure 7: Changes in CMAQ-simulated annual mean PM<sub>2.5</sub> concentrations. (a)-(c): base simulations of 2013, 2016, and 2017; (d-f) total PM<sub>2.5</sub> changes and the changes attributed to meteorology, emission variations during 2013-2017; (g-i): total PM<sub>2.5</sub> changes and the changes attributed to meteorology, emission variations during 2016-2017.**

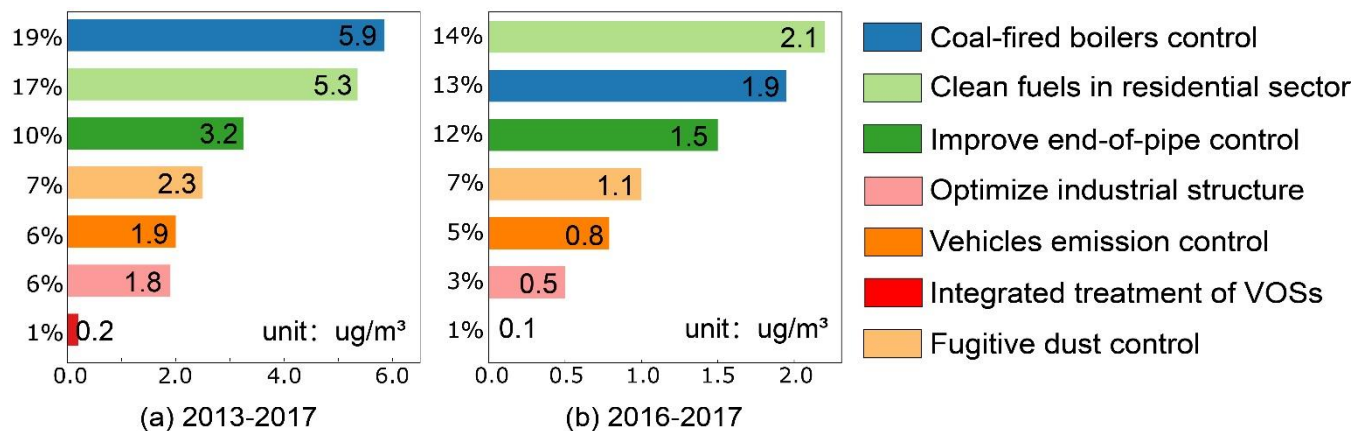


(a)



(b)

5 **Figure 8: CMAQ-simulated monthly PM<sub>2.5</sub> concentrations in Beijing under different meteorological conditions. The numbers shown in each panel represent the monthly relative change rates of fixed-emission simulation results compared with the base simulation results.**



**Figure 9: Contributions of the seven types of control policies to the PM<sub>2.5</sub> abatements in Beijing for the periods of 2013-2017 (a) and 2016-2017 (b). The relative contribution rate of each policy to the total PM<sub>2.5</sub> abatements is presented on the Y-axis. The number on each bar represents the normalized absolute PM<sub>2.5</sub> abatement by the relevant control policy.**

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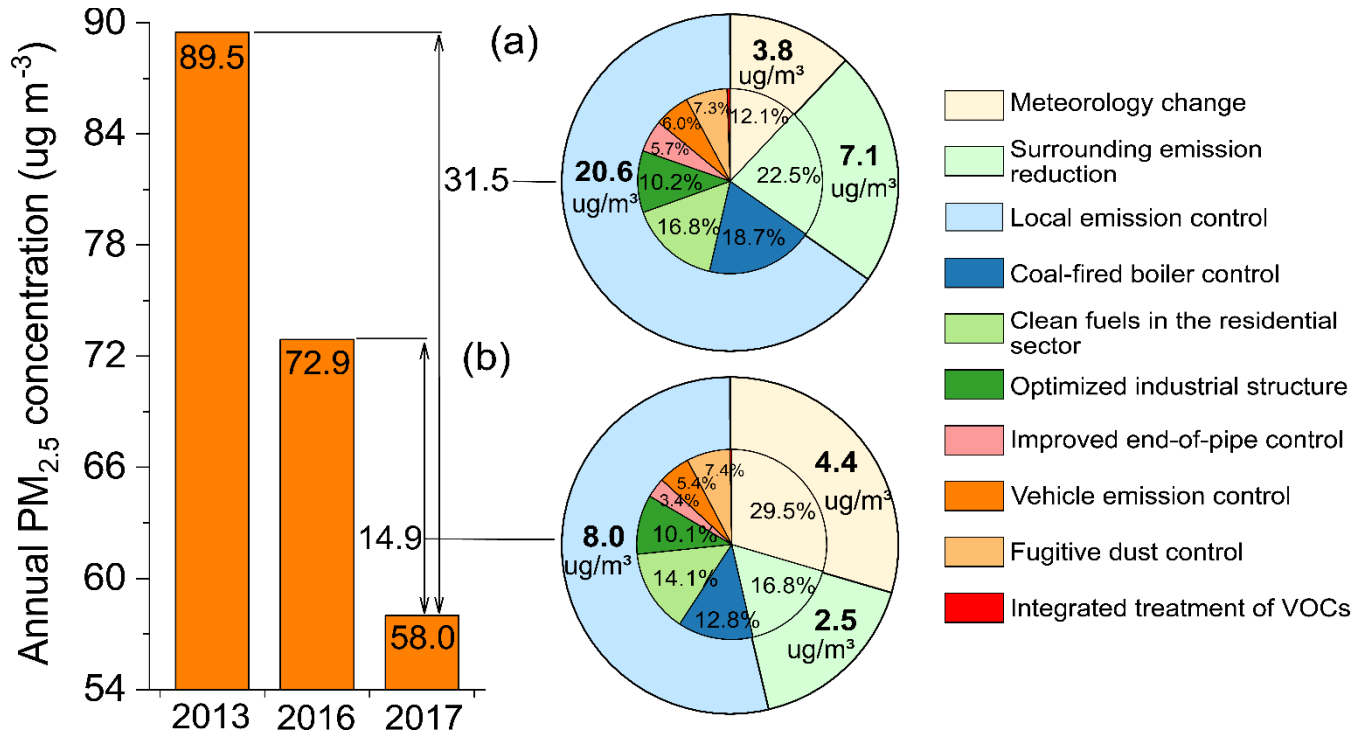


Figure 10: Decomposition of improved PM<sub>2.5</sub> air quality in Beijing during (a) 2013-2017 and (b) 2016-2017.

**Table 1.** Summary of emission control measures implemented in the *Beijing Action Plan (2013-2017)*.

Policy type	Measure ID	Specific control measures	Sectors
1. Coal-fired boiler control	1-1	By the end of 2017, Beijing had closed the four major coal-fired power plants and constructed four great natural gas thermal-power cogeneration centres instead, which reduced coal use by nearly 8.6 million tons in total.	Power and Heating
	1-2	Beijing made great efforts to renovate the coal-fired facilities with capacities of less than 25 MW in urban districts and 7 MW in the whole city. A total capacity of 390 thousand MW of coal-fired boilers were eliminated or replaced by clean fuels such as natural gas and electricity. Coal use was reduced by more than 8.5 million tons during this process.	Industry
2. Clean fuels in the residential sector	2-1	Through cutting down nonpeak household power prices and establishing new gas heating systems, approximately 900,000 households in Beijing were converted from using coal to using gas or electricity, and coal use was reduced by a total of 2.1 million tons.	Residential
	2-2	The burning of biomass, such as wood and crops, was thoroughly forbidden by the end of 2016.	Residential
3. Optimize industrial structure	3-1	During 2013 to 2017, Beijing phased out a total of 1,992 large high-pollution enterprises in the chemical engineering, furniture manufacturing, printing, and non-metal mineral product industries; furthermore, three-quarters of the cement industry was eliminated.	Industry
	3-2	In the last 5 years, especially since 2016, Beijing has made great efforts to eliminate the small, clustered and polluting factories that cannot meet efficiency, environmental and safety standards, and a total of 11,000 such factories were managed or eliminated.	Industry
4. Improved end-of-pipe control	4-1	Since 2013, a total of 468 projects involving cleaner production and technological upgrades for high-pollution industrial sectors were carried out by the government. During this process, Beijing has gradually retrofitted all cement factories to achieve thorough denitrification and degassing and has enhanced the desulfurization retrofitting of the non-metal mineral product and chemical engineering industries.	Industry
	4-2	Beijing first promoted low-nitrogen-burning (LNB) combustion for all industrial sectors in 2013, and the LNB transformation of nearly 21,000 MW of gas-fired boilers and oil-fired boilers has been completed.	Industry

5. Vehicle emission control	5-1	1) During 2013 to 2017, Beijing retired a total of 2,167,000 old vehicles, and all “yellow-labelled” cars (gasoline and diesel cars that failed to meet Euro I and Euro III standards) were eliminated completely by 2017. 2) In Mar 2017, Beijing first implemented the latest China VI emission standards, which are one of the most tightened emission standards in the world. 3) A total of 51,000 taxis completed the replacement of three-way catalytic converters, and 17,000 heavy-duty diesel vehicles were equipped with wall flow particle traps. 4) In Sep 2017, all out-of-city diesel vehicles with lower emission standards than China III were forbidden to travel within the sixth ring road.	On-road transportation
	5-2	1) By the end of 2016, all off-road vehicles were required to comply with “ <i>Limits and measurement methods for exhaust pollutants from diesel engines of non-road mobile machinery (China III, IV)</i> ”. 2) Since 2017, the use of heavily polluting off-road vehicles has been restricted in the six major urban districts and Tongzhou district.	Off-road transportation
6. Integrated treatment of VOCs	6-1	Beijing started to eliminate organic solvent coatings, bituminous waterproof materials and organic painted furniture manufacturing in 2013. Meanwhile, Beijing promoted the use of high-solids and waterborne paints, which contain much fewer organic chemicals, in machine manufacturing, printing, coating and automobile repair sectors.	Solvent use
	6-2	During 2013 to 2017, the Yanshan company, the only petrochemical industry enterprise in Beijing, completed seven extensive VOCs control projects, such as the innovation of sealing and defocusing technology, the detection and repair of leakage points, and the specialized management of refined oil production and storage areas.	Industry
7. Fugitive dust control	7-1	Beijing increased the quality and frequency of the road cleaning process. By the end of 2017, a mechanized cleaning process were adopted in an area of 90,580,000 square metres, accounting for 88% of the total urban road area.	Fugitive dust
	7-2	Beijing shut down a total of 310 concrete mixing plants and updated over 20 thousand cinderblock transporters. Additionally, more than 1,200 construction sites were equipped with video monitoring system at the exits and entrances.	Fugitive dust
	7-3	By the end of 2015, Beijing completed an afforestation project in advance and afforested nearly 700 square kilometres in nearby plain areas.	Fugitive dust

**Table 2. CMAQ simulations conducted in this study for the decomposition analysis. The seven types of emission control policies include *Coal-fired boiler control (policy1), Clean fuels in the residential sector (policy2), Optimize industrial structure (policy3), Improve end-of-pipe control (policy4), Vehicle emission***



*control (policy5), Integrated treatment of VOCs (policy6) and Fugitive dust control (policy7). The parenthesis on rightmost column indicate the annual mean observational PM<sub>2.5</sub> concentrations of 2013 (89.5), 2016 (72.9) and 2017 (58.0).*

Case label	Year of meteorological data	Year of emission data in Beijing	Year of emission data in surrounding regions	Purpose of the simulation	Simulated PM <sub>2.5</sub> (μg m <sup>-3</sup> )
E <sub>L13S13</sub> M <sub>13</sub>	2013	2013	2013	reproduce air quality in 2013	86.3 (89.5)
E <sub>L16S16</sub> M <sub>16</sub>	2016	2016	2016	reproduce air quality in 2016	70.1 (72.9)
E <sub>L17S17</sub> M <sub>17</sub>	2017	2017	2017	reproduce air quality in 2017	53.5 (58.0)
E <sub>L17S17</sub> M <sub>13</sub>	2013	2017	2017	quantify the impact of meteorology compared with 2013	57.4
E <sub>L17S17</sub> M <sub>16</sub>	2016	2017	2017	quantify the impact of meteorology compared with 2016	57.8
E <sub>L17S13</sub> M <sub>17</sub>	2017	2017	2013	quantify the contribution from the emission reduction of surroundings during 2013-2017	60.9
E <sub>L17S16</sub> M <sub>17</sub>	2017	2017	2016	quantify the contribution from the emission reduction of surroundings during 2016-2017	55.9
E <sub>L<sub>pi</sub>S17</sub> M <sub>17</sub> (i = 1 - 7)	2017	2017 (excluding the implementation of each control policy since 2013)	2017	quantify the contribution of each control policy during 2013-2017	(SI)
E <sub>L<sub>qi</sub>S17</sub> M <sub>17</sub> (i = 1 - 7)	2017	2017 (excluding the implementation of each control policy since 2016)	2017	quantify the contribution of each control policy during 2016-2017	(SI)