Comments to the Author:

You have not adequately addressed the comments made by the reviewers.

To be able to have this manuscript published I would like you to take into consideration the following remarks:

To Editor:

Thanks so much for providing us a chance to revise this manuscript again. We have carefully checked this manuscript and revised it fully according to your comments. Please feel free to contact us if further revisions are required.

In the Introduction, please make sure that you site all the relevant work that has been devoted to looking at trends of $PM_{2.5}$ over Beijing. In addition, the last 3-4 sentences of the introduction should summarize the sections that you develop in the paper.

R: Thanks so much for this point. We have added more relevant works concerning trends of $PM_{2.5}$ over Beijing. Meanwhile, the introduction section has been revised according to your suggestions.

We have carefully searched relevant publications that looking at trends of $PM_{2.5}$ variations from 2013 to 2017, which is a specific period for evaluation. Since the completion of this period just passed for one year, not many relevant papers found. In the revised manuscript, we included another five papers that mentioned $PM_{2.5}$ variations in Beijing from 2013 to 2017. These papers mainly discussed the spatial-temporal variations of $PM_{2.5}$ variations in Beijing from 2013 to 2017. Several of them employed some field-collected $PM_{2.5}$ sample to analyze the source of $PM_{2.5}$ component during short-term pollution episodes. Therefore, they are not highly correlated with the major aim of this research, the meteorological influences on $PM_{2.5}$ variations from 2013 to 2017.

Shao, P., Tian, H., Sun, Y., Liu, H., Wu, B., Liu, S., Liu, X., Wu, Y., Liang, W., Wang, Y., Gao, J., Xue, Y., Bai, X., Liu, W., Lin, S., Hu, G.: Characterizing remarkable changes of severe haze events and chemical compositions in multi-size airborne particles (PM_1, PM2.5 and PM10) from January 2013 to 2016–2017 winter in Beijing, China. Atmospheric environment, 189, 133-144, 2018.

Xu, H, Xiao Z, Chen K, Tang M, Zheng N, Li P, Yang N, Yang W, Deng X.: Spatial and temporal distribution, chemical characteristics, and sources of ambient particulate matter in the

Beijing-Tianjin-Hebei region. Science of The Total Environment, 658, 280-293, 2019.

Wang T, Du Z, Tan T, Xu N, Hu M, Hu J, Guo S.: Measurement of aerosol optical properties and their potential source origin in urban Beijing from 2013-2017. Atmospheric Environment, 206, 293-302, 2019.

Liang, L., Cai, Y., Barratt, B., Lyu, B., Chan, Q., Hansell, A.L., Xie, W., Zhang, D., Kelly, F.J., Tong, Z.: Associations between daily air quality and hospitalisations for acute exacerbation of chronic obstructive pulmonary disease in Beijing, 2013–17: an ecological analysis. The Lancet Planetary Health, 3(6), 270-279, 2019.

Sun, J., Gong, J., Zhou, J., Liu, J., Liang, J..: Analysis of $PM_{2.5}$ pollution episodes in Beijing from 2014 to 2017: Classification, interannual variations and associations with meteorological features. Atmospheric Environment,213, 384-394, 2019.

Zhai S, Jacob D J, Wang X, Shen L, Li K, Zhang Y, Gui K, Zhao T, Liao H.: Fine particulate matter (PM 2.5) trends in China, 2013–2018: separating contributions from anthropogenic emissions and meteorology. Atmospheric Chemistry and Physics, 19(16), 11031-11041 ,2019.

So we gave a general introduction of these studies concerning the specific trends of $PM_{2.5}$ over Beijing during the Clean Air Action period

"The notable decrease of $PM_{2.5}$ concentrations attracted nationwide attentions and growing studies have been conducted to understand spatio-temporal characteristics (Shao et al., 2018; Sun et al., 2019; Wang et al., 2019), sources (Chen et al., 2019; Xu et al., 2019; Cheng et al., 2019) and health effects (Liang et al., 2019) of $PM_{2.5}$ variations in Beijing from 2013 to 2017. These studies revealed that air quality in Beijing was improved significantly in 2017 in terms of annual mean $PM_{2.5}$ concentrations, polluted days and pollution durations. Furthermore, despite different outputs, both source apportionment during pollution episodes based on collected samples (Shao et al., 2019; Xu et al., 2019; Chen et al., 2019) and long-term model simulation based on regional and local emission inventories (Cheng et al., 2019) suggested that local and regional anthropogenic emissions (e.g. coal combustion and vehicle emissions) were the major

influencing factors for long-term and short-term $PM_{2.5}$ variations in Beijing."

We also added some introduction for a recently published paper concerning $PM_{2.5}$-meteorology relationship across China from 2013 to 2018

"Based on a stepwise multiple linear regression (MLR) model, Zhai et al. (2019) quantified the relative contribution of meteorology to $PM_{2.5}$ variations from 2013 to 2018 in Beijing-Tianjin-Hebei region, Yangtze River Delta, Pearl River Delta and Sichuan Basin and Fenwei plain was 14%, 3%, 19%, 27% and 23% respectively."

According to your comment, we added a short introduction of the structure of this manuscript at the end of the introduction section as follows:

"This manuscript is structured as follows: Firstly, major data sources, including $PM_{2.5}$ and meteorological data, and emission inventories, employed for this research are briefly introduced. Secondly, the principle and parameter setting of two models, KZ filtering and WRF-CMAQ, and model verification are explained. In the result section, the relative contribution of meteorological conditions and anthropogenic emissions to $PM_{2.5}$ variations in Beijing from 2013 to 2017 calculated using both models is presented. In the discussion and conclusion part, implementations of this research and suggestions for further improving air quality in Beijing are given."

Thanks again for this valuable comments.

Remarks to be addressed concerning reviewer 1:

I propose that you include the following Table with the relevant explanations from your answers to comment 3 from Reviewer 1 in the manuscript:

The comparison of The local environmental statistical data used for this research and other official statistical data in 2017 (unit: 10k tons)

SO2 NOx CO VOC NH3 PM10 PM2.5 BC OC

Statistical data for this research 1.38 10.15 49.54 13.47 3.20 14.74 3.92 0.17 0.44

National Environmental Statistics Bulletin 1.38 12.16 52.03 24.24 3.26 14.68 3.91 0.22 0.41
“2+26” center for air pollution prevention and control 0.89 9.24 48.98 13.93 3.16 13.82 3.72 0.19 0.46

R: Thanks so much for this comment. We have added this table and the following text to the revised manuscript.

“**As shown in table 1, it is highly consistent with other official statistical data, such as the Annual report from National Environmental Statistics Bulletin (http://www.mee.gov.cn/gzfw_13107/hjtj/qghjtjgb/) and “2+26” Center for Air Pollution Prevention and Control, and 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).**”

Where in the revised version do you indicate the following results that appears in your answer to Comment 4 of reviewer 1?
“This means the extracted seasonal component and short-term component made a significant contribution to seasonal and short-term variations of original PM2.5 concentrations in Beijing from 2013 to 2017, indicating a satisfactory KZ filtering result. »

R: Thanks so much for pointing this out. There are one major approach to verify the efficiency of KZ. If the total variations of long-term, seasonal and short-term component was close to 1, it suggests that a majority of meteorological influences has been considered and effectively removed. Specifically, the variation of seasonal (ranging from 9%-23.8%) and short-term component (ranging from 66.8%-83.8%) was much larger than that of long-term component (ranging from 1.2%-3.5%).

So in the revised manuscript, we included the following text

“**The sum of the long-term, seasonal and short-term component contributed to more than 93.6~95.3% of the total variance in different stations respectively. The larger the total variance, the three components are more independent to each other. The total variance close to 100% suggests that a majority of**

meteorological influences has been considered and effectively removed. As shown in Table 3, the large value of the total variation in all stations indicated a satisfactory output from the KZ filtering.

Specifically, the relative contribution of the seasonal component (ranging from 9%-23.8%) and short-term component (ranging from 66.8%-83.8%) was much larger than that of the long-term component (ranging from 1.2%-3.5%), suggesting that seasonal and short-term variations of meteorological and emission factors exerted a major influence on the rapid change of $PM_{2.5}$ concentrations in Beijing."

In addition to the statistical results, according to the comment 4 from reviewer 1, we added a Figure 2 to present the decomposed long-term, seasonal and short-term components using KZ filter. According to Figure 2, we can see that the long-term component demonstrates a smooth curve whilst the trend of season component and short-term component is highly consistent with that of the original $PM_{2.5}$ time series, especially for some simultaneous peaks. Therefore, the seasonal and short-term variations of $PM_{2.5}$ concentrations were effectively extracted as indicative seasonal component and short-term component. In the revised manuscript, we employed the following text to explain this

"According to Fig 2, the notable peaks of decomposed seasonal and short-term component were highly consistent with the peaks of $PM_{2.5}$ concentrations in the original time-series, which further proved the dominant influence of seasonal and short-term variations of meteorological and anthropogenic factors on the temporal changes of PM2.5 concentrations in Beijing."

You do not answer adequately the comment 5 of reviewer 1.

"Q5. Section 3.2.2. Model evaluation is the key point in this paper. If the model data is not consistent with observation, contribution of emission control is out of the question. It seems that lots of data are far from the observation especially during the heavy air pollution days. So it is better to convert Fig 2 to time series plots, which can tell us more detailed information about the model evaluation."

Please draft an adequate answer and modify the manuscript accordingly.

R: Thanks so much for pointing this out. According to this comment, firstly, we have converted this Figure (Fig 3 in revised manuscript) to time series plots and included other three urban stations, which presented detailed information about the model evaluation.

As we acknowledged in the revised manuscript

"According to Fig 3, the general trend of the simulated $PM_{2.5}$ concentrations was consistent with that of the observed $PM_{2.5}$ concentrations. For six 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 0.63~0.91, -6%~6%, 26%~40%, -5%~7%, and 27%~46% respectively, indicating a satisfactory simulation output (EPA，2005; Boylan et al., 2006). However，as shown in Figure 3, WRF-CMAQ may notably underestimate $PM_{2.5}$ concentrations during heavy pollution episodes due to unified parameter setting for long-term simulation, the uncertainty in emission inventories, and especially insufficient chemical reaction mechanisms, which is a common challenge for CTM-based $PM_{2.5}$ simulation (Li et al., 2011)."

The general accuracy of model simulation was satisfactory in terms of R, NMB, NME, MFB and MFE. Meanwhile, the long-term trend of simulated $PM_{2.5}$ concentrations was consistent with that of observed $PM_{2.5}$ concentrations. However, as the reviewer pointed out, WRF-CAMQ could lead to large variations during heavy pollution episodes, especially for long-term simulation with unified parameters (Li et al., 2011). We explained some underlying reasons for this common and unsolved challenge, the uncertainty in emission inventories, and especially insufficient chemical reaction mechanisms. We gave an example of this issue and its potential solution in the revised manuscript. More finer-scale emission inventories and better descriptions of reaction mechanisms in WRF-CAMQ can further improve simulation accuracy.

"For instance, without considering heterogeneous/aqueous reactions between multiple precursors, CTMs failed to approach the maximum $PM_{2.5}$ concentrations during severe haze episodes and the simulation accuracy was dramatically improved by including proper descriptions of heterogeneous/aqueous reactions into CTMs (Chen, D. et al. 2016). With more finer-scale emission inventories and better descriptions of reaction mechanisms between precursors, the accuracy of $PM_{2.5}$ simulation can be improved significantly"

In your revised manuscript 2 Tables are referenced as Table 3. You should have picked up this mistake.

R: We are very sorry for this mistake. We have checked the manuscript carefully and revised this and other typos.

In the new Figure 3 you have 6 plots of timeseries and 3 scatterplots. I want a descriptive Figure caption for ALL of them.

In the new Figure 4 you have 6 plots of timeseries and 2 plots with stacked lines. Please write a Figure caption for ALL of them.

R: I think there is some misunderstanding here. It may be attributed to the change-track version of the manuscript and deleted figures may appeared as part of the new Figures. Actually, in the clean version of the revised manuscript, according to the reviewer 1's comments, for Figure 3, we simply have 6 plots of time series images. As follows:

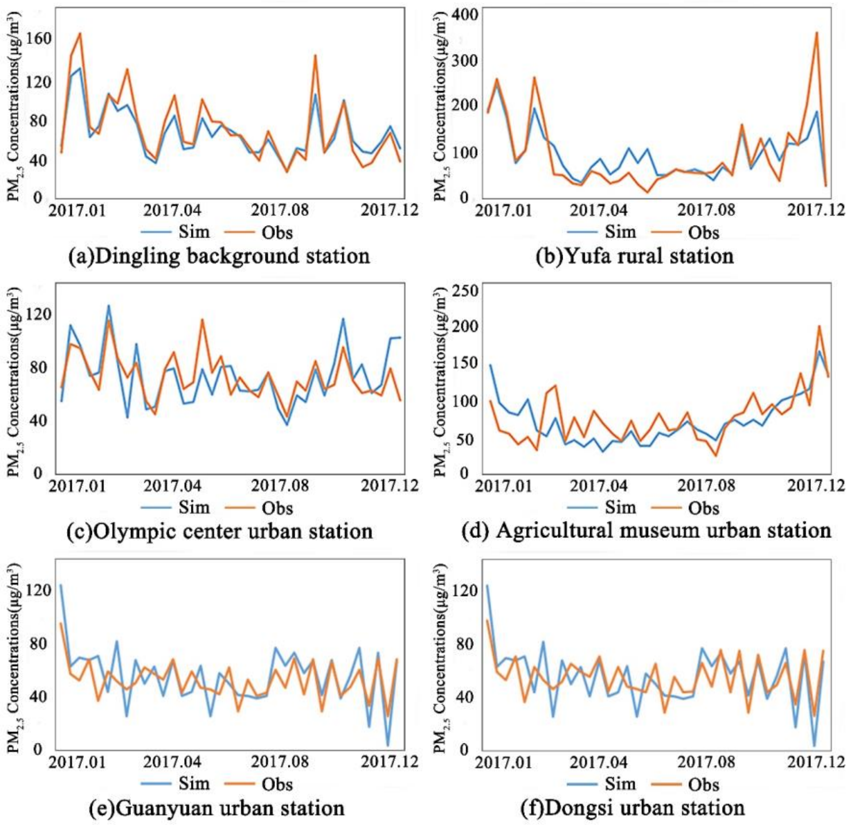

Fig 3. The comparison between observed and WRF-CMAQ simulated $PM_{2.5}$ concentrations in 2017 in six stations across Beijing

For figure 4, we simply have 6 plots of time series. As follows:

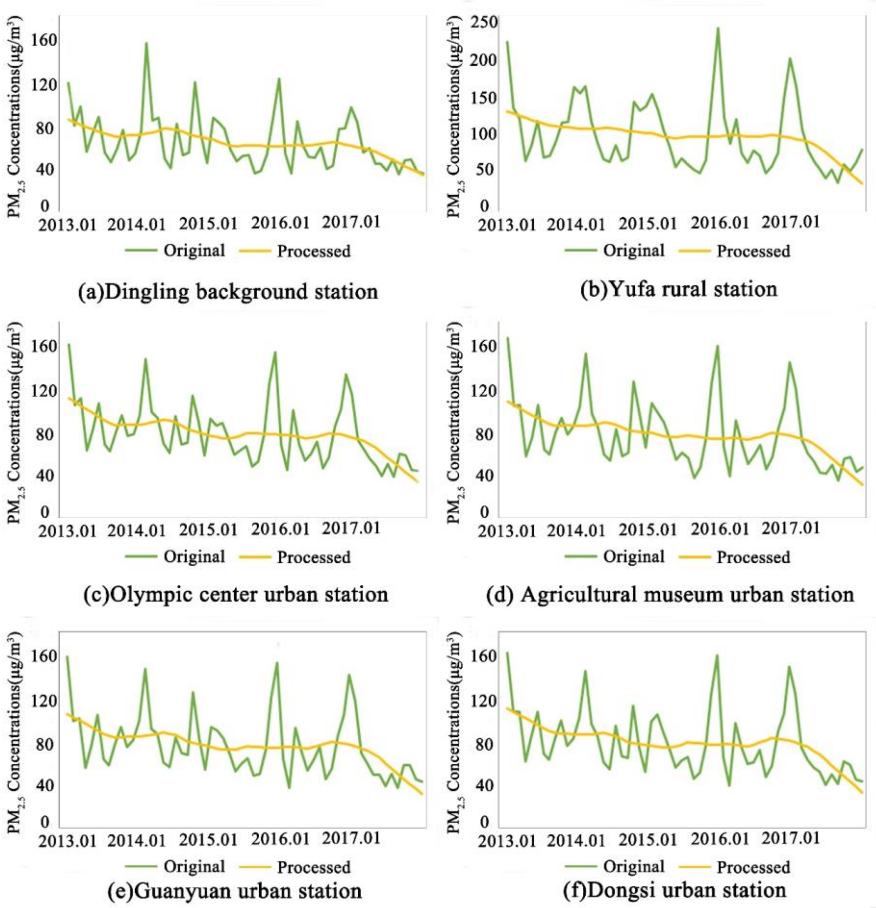

Fig 4. The comparison of original and KZ processed time series of $PM_{2.5}$ concentrations in six stations from 2013 to 2017

How did you change the text to reflect Question 7 of reviewer 1:

"L339. How did you get the conclusion "KZ filtering provides a more reliable method"? Just because the KZ filtering was station-based and WRF-CMAQ model was a grid-based? The averaged relative contribution of meteorological variations to PM2.5 reduction using the WRF-CMAQ model was very similar to that using KZ filtering. Verification is very important » ?

R: Thanks so much for this. In the revised manuscript, we included the text as follows to explain why KZ filter was a more reliable method to quantify the relative contribution

of meteorological conditions and anthropogenic emissions.

"It is worth mentioning that WRF-CMAQ is 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 multiple stations. Furthermore, WRF-CMAQ simply considered the differences between meteorological conditions in 2013 and 2017 without considering their variations during the five-year period while the KZ filtering analyzed the entire time series of $PM_{2.5}$ and meteorological data from 2013 to 2017."

" Since KZ filtering is fully based on observed data, and simply considers the influence of time-series meteorology data on $PM_{2.5}$ time series, less uncertainty is involved. The accuracy of KZ filtering is influenced mainly by the variations of $PM_{2.5}$-meteorology interactions in different areas and seasons. On the other hand, CTMs, e.g. WRF-CMAQ or WRF-CAMx consider both meteorological conditions (mainly large-scale meteorological data for model simulation, not as accurate as local observed meteorological data) and anthropogenic emissions for estimating $PM_{2.5}$ concentrations under different emission scenarios. The accuracy of these models are not only decided by proper understanding of $PM_{2.5}$-meteorology interactions, but also the reliability of emission inventories and proper descriptions of reaction mechanisms for $PM_{2.5}$ production, especially during heavy pollution episodes, which is a major challenge for current model simulation. Consequently, KZ filtering provides a more reliable method for researchers and decision makers to understand the relative importance of emission-reduction and meteorological conditions in recent $PM_{2.5}$ reduction in Beijing. Meanwhile, similar outputs from 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."

How did you change the text to reflect the comment "Question 8" of Reviewer 1?
"L398-399. Supplement the correlation coefficient between wind speed and PM2.5. And how about the influence of the other meteorological parameters (such as T, RH, wind direction) on PM2.5?

R: Thanks so much for pointing this out. In the revised manuscript, we changed the following text to reflect the comment.

"**We examined correlations between seasonal $PM_{2.5}$ concentrations in Beijing and a series of meteorological factors, including temperature, wind speed, wind direction, precipitation, relative humidity, solar radiation, evaporation and air pressure. Due to limited space, detailed correlations between $PM_{2.5}$ concentrations and individual meteorological factors in Beijing are not presented here and readers can refer to previous studies for more information (Chen et al., 2017; 2018). The correlation analysis revealed that wind speed, relative humidity, temperature and solar radiation were strongly and significantly correlated with $PM_{2.5}$ concentrations in Beijing (as shown in Table 2), which was consistent with findings from other studies (Sun et al., 2013; Wang et al., 2018).**

Table 2. Major meteorological factors strongly correlated with seasonal $PM_{2.5}$ concentrations in Beijing (Chen et al., 2017)

Spring	**Summer**	**Autumn**	**Winter**
	RHU**(0.648)	RHU**(0.587)	RHU**(0.738)
RHU**(0.532)	SSD**(−0.447)	SSD**(−0.509)	SSD**(−0.715)
	TEM**(0.554)	WIN**(−0.468)	WIN**(−0.558)

****Correlation is significant at the 0.01 level (2 tailed);**

RHU: Relative humidity; SSD: Sunshine Duration; TEM: Temperature; WIN: Wind speed"

Finally all your co-authors should read and approve of your answers. Is it the case?

R: Yes, all co-authors read the comments carefully and we discussed together to make revisions and write responses. All co-authors approved our answers.

Sincerely,

Yves Balkanski

Non-public comments to the Author:

I confirm that this is not of adequate quality to be published in ACP. So make a big effort if you want it to be published.

R: Dear Professor Yves Balkanski

Thanks so much for giving us a chance to resubmit this manuscript. We realized that the previous manuscript should be improved significantly by adding more necessary technical details, providing better figures, citing more relevant works, better structuring and careful wording. Thanks again for all the valuable comments from reviewers and you. Please feel free to let us know if further revisions are required. We are more than willing to conduct all necessary revisions until this manuscript meet the requirement of ACP publications.

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 $PM_{2.5}$ concentration 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 were the major driver for such rapid decrease in $PM_{2.5}$ concentrations. To comprehensively evaluate this five-year plan, we employed Kolmogorov-Zurbenko (KZ) filter and WRF-CMAQ 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 to the decrease of $PM_{2.5}$ concentrations calculated by KZ filtering and WRF-CMAQ was 80.6% and 78.6% respectively. KZ filtering suggested that short-term variations of meteorological and emission conditions contributed majorly to rapid changes of $PM_{2.5}$ concentrations in Beijing.

WRF-CMAQ revealed that the relative contribution of local and regional emission-reduction to $PM_{2.5}$ decrease in Beijing was 53.7% and 24.9% respectively. For local emission-reduction measures, the regulation of coal boilers, increasing use of clean fuels for residential use and industrial restructuring contributed to 20.1 %, 17.4% and 10.8% of $PM_{2.5}$ reduction respectively. Both models suggested that the control of anthropogenic emissions accounted for around 80% of the $PM_{2.5}$ reduction in Beijing, indicating that emission-reduction was crucial for air quality enhancement in Beijing from 2013 to 2017. Consequently, such long-term air quality clean plan should be continued in the following years to further reduce $PM_{2.5}$ concentrations in Beijing.

Keywords: $PM_{2.5}$, anthropogenic emissions, meteorological conditions, Kolmogorov-Zurbenko (KZ) filtering, WRF-CMAQ

1 Introduction

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In January 2013, persistent haze episodes occurred in Beijing, during which the highest hourly $PM_{2.5}$ concentration once reached 886 μ g/m^3, a historic high record. High-concentration $PM_{2.5}$ led to long-lasting black and thick fogs, which not only significantly influenced people's daily life (low-visibility induced traffic jam), but also posed a severe threat to 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 been observed in Beijing and other regions across 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. Consequently, a national network for monitoring hourly $PM_{2.5}$ concentrations has been established gradually, including 35 ground observation stations in Beijing, which provide important support for better understanding and managing $PM_{2.5}$ concentrations. To effectively mitigate $PM_{2.5}$ pollution, Beijing Municipal Government released "Beijing Five-year Clean Air Action Plan (2013-2017)" with a series of long-term emission-reduction measures, including shutting down heavily polluting factories, restricting traffic emissions and replacing coal fuels with clean energies, and "Heavy Air Pollution Contingency Plan" with a series of contingent emission-reduction measures during heavy pollution episodes. By the end of 2017, these long-term and contingent emission-reduction measures worked jointly to reduce the annually mean $PM_{2.5}$ concentration in Beijing from 89.5 μg/m^3 in 2013 to 58.0 μg/m^3 in 2017, indicating a great success of $PM_{2.5}$ management during the past five years. The notable decrease of $PM_{2.5}$ concentrations attracted nationwide attentions and growing studies have been conducted to understand spatio-temporal characteristics (Shao et al., 2018; Sun et al., 2019; Wang et al., 2019), sources (Chen et al., 2019; Xu et al., 2019; Cheng, J. et al., 2019) and health effects (Liang et al., 2019) of $PM_{2.5}$ variations in Beijing from 2013 to 2017. These studies revealed that air quality in Beijing was improved significantly in 2017 in terms of annual mean $PM_{2.5}$ concentrations, polluted days and pollution durations. Furthermore, despite different outputs, both source apportionment during pollution episodes based on collected samples (Shao et al., 2019; Xu et al., 2019; Chen et al., 2019) and long-term model simulation based on regional and local emission inventories (Cheng, J. et al., 2019) suggested that local and regional anthropogenic emissions (e.g. coal combustion and vehicle emissions) were the major influencing factors for long-term and short-term $PM_{2.5}$ variations in Beijing.

In addition to anthropogenic emissions, the strong meteorological influences on $PM_{2.5}$ concentrations in Beijing have been widely acknowledged (Zhao et al., 2013; Wang et al., 2014; UNEP, 2016; Cheng et al., 2017; Chen et al., 2017; Sun et al., 2019~~Cheng et al., 2017; Chen et al., 2017, 2018; UNEP, 2016; Wang et al., 2014; Zhao et al., 2013~~). For instance, for 2014, more than 180 days in Beijing experienced a dramatic daily AQI (Air Quality Index) change (△AQI>50) (Chen, Z. et al., 2016). Considering that anthropogenic emissions for a mega city unlikely changed significantly on a daily basis, rapid variations of meteorological conditions were one major driver for the dramatic change of daily air quality in Beijing. In winter 2017, strong northwest winds led to favorable meteorological conditions for $PM_{2.5}$ diffusion and low $PM_{2.5}$ concentrations in Beijing. This raised the controversy that meteorological conditions, instead of emission-reduction，accounted for the remarkable $PM_{2.5}$ reduction in Beijing~~ from 2013 to 2017~~. In this case, with the completion of the five-year plan, it is highly necessary to quantify the relative contribution of meteorological conditions and emission-reduction to the notable decrease in $PM_{2.5}$ concentrations in Beijing from 2013 to 2017.

In recent years, growing studies have been conducted to investigate meteorological and anthropogenic influences on long-term $PM_{2.5}$ variations. Based on Goddard Earth Observing System (GEOS) chemical transport model (GEOS-Chem), Yang et al (2016) revealed that the relative contribution of meteorological conditions to $PM_{2.5}$ variations in Eastern China from 1985 to 2005 was 12%. Based on a multiple general linear model (GLM), Gui et al. (2019) quantified that meteorological conditions accounted for 48% of $PM_{2.5}$ variations in Eastern China from 1998 to 2016. Based on a stepwise multiple linear regression (MLR) model, Zhai et al. (2019) quantified the relative contribution of meteorology to $PM_{2.5}$ variations from 2013 to 2018 in Beijing-Tianjin-Hebei region, Yangtze River Delta, Pearl River Delta and Sichuan Basin and Fenwei plain was 14%, 3%, 19%, 27% and 23% respectively. ~~–~~Through a two-stage hierarchical clustering method, Zhang et al. (2018) calculated that the relative contribution of meteorological conditions to heavy pollution episodes within the Beijing-Tianjin-Hebei region was larger than 50% from 2013 to 2017. These studies quantified the overall meteorological influences on long-term $PM_{2.5}$ variations using different statistical models and chemical transport models (CTMs). However, due to strong interactions between individual meteorological factors, traditional statistical methods such as correlation analysis and linear regression may be biased significantly when quantifying meteorological influences on $PM_{2.5}$ concentrations (Chen et al., 2017). On the other hand, the accuracy of CTMs can be influenced largely by the uncertainty in emission inventories (Xu et al., 2016) and deficiency of heterogeneous/aqueous processes (Li et al., 2011). Therefore, multiple advanced models should be comprehensively considered to better quantify meteorological influences on $PM_{2.5}$ concentrations (Pearce et al., 2011).

To evaluate this five-year clean-air plan, we employ an advanced statistical model, Kolmogorov-Zurbenko (KZ) filtering, which is advantageous of filtering meteorological influences on long-term time series of airborne pollutants, and a CTM model, WRF-CMAQ, which is advantageous of quantifying the relative contribution of different emission sources, to comprehensively investigate the relative contribution of meteorological conditions and emission-reduction to $PM_{2.5}$ reduction in Beijing from 2013 to 2017 respectively. 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.

This manuscript is structured as follows: Firstly, major data sources, including $PM_{2.5}$ and meteorological data, and emission inventories, employed for this research are briefly introduced. Secondly, the principle and parameter setting of two models, KZ filtering and WRF-CMAQ, and model verification are explained. In the result section, the relative contribution of meteorological conditions and anthropogenic emissions to $PM_{2.5}$ variations in Beijing from 2013 to 2017 calculated using both models is presented. In the discussion and conclusion part, implementations of this research and suggestions for further improving air quality in Beijing are given.

2 Data Sources

2.1 $PM_{2.5}$ and meteorological data

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In this study, hourly $PM_{2.5}$ concentration data were acquired from the website PM25.in (www.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 spatio-temporal variations of $PM_{2.5}$ concentrations and quantify their influencing factors. In addition to these urban stations, we 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) that is far away from central Beijing for the following analysis. The DingLing and MiYun Reservoir Station 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. The Qianmen transportation station received more influences from vehicle emissions. Long-term variations of $PM_{2.5}$ concentrations in different type of stations provide a useful reference for comprehensively understanding the effects of emission-reduction measures on $PM_{2.5}$ decrease in Beijing from 2013 to 2017. Meteorological data for this research were collected from the Guanxiangtai Station (GXT,54511, 116.46° E, 39.80° N), Beijing and downloaded from the Department of Atmospheric Science, College of Engineering, University of Wyoming (http://weather.uwyo.edu/upperair/sounding.html). Both $PM_{2.5}$ and meteorological data were collected from January 1st, 2013 to December 31st, 2017. The locations of these selected stations are shown in Fig 1.

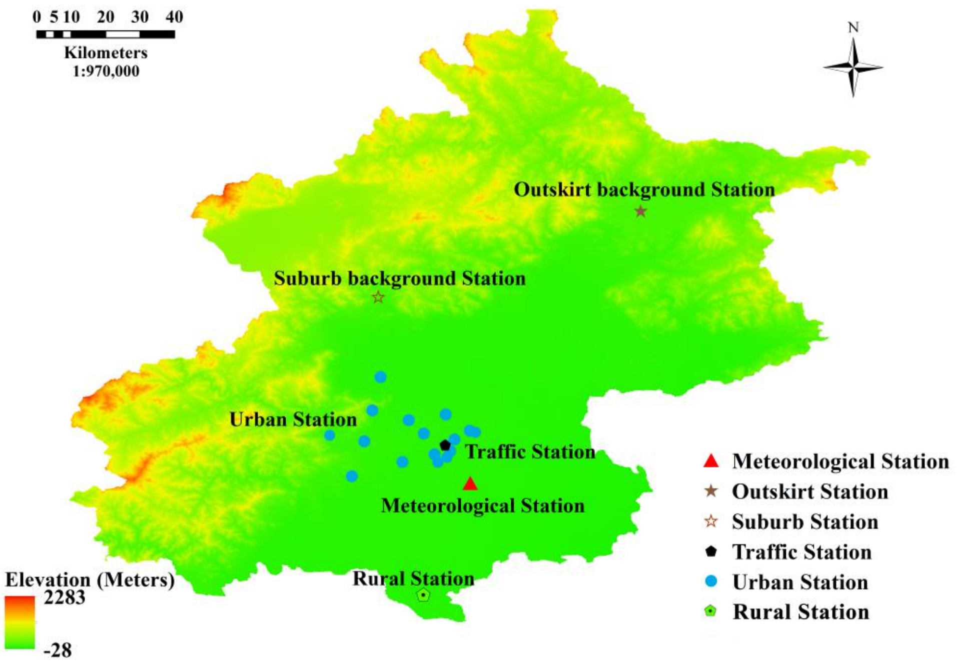

Fig 1. Locations of different ground monitoring stations.

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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.). For simulating five-year $PM_{2.5}$ concentrations, MEIC from 2013 to 2017 are required. Since official MEIC 2017 has not been available yet, we employed a strategy from previous studies (Chen et al., 2019; etc) and updated MEIC 2016 for simulating emission-reduction scenarios and $PM_{2.5}$ concentrations in 2017 by considering official 2017 emission-reduction plans (e.g. the target of coal combustion reduction) required by the local government.

Different from regional emission inventories, local emission inventories are usually produced independently by local institutions. The Beijing local-emission inventory employed for this research was 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 Beijing-Tianjin-Hebei region. This Beijing local-emission inventory from 2013 to 2017 was produced by synthesizing local environmental statistical data and reported emission data, carrying out field investigations and conducting a series of estimation according to Beijing Five-year Clean Air Action Plan. As shown in table 1, i~~I~~t is highly consistent with other official statistical data, such as the Annual report from National Environmental Statistics Bulletin (**http://www.mee.gov.cn/gzfw_13107/hjtj/qghjtjgb/**) and "2+26" Center for Air Pollution Prevention and Control, and 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).

Table 1. The comparison of local environmental statistical data used for this research and other official statistical data in 2017 (unit: 10k tons)

	SO_2	NO_x	CO	VOC	NH_3	PM_{10}	$PM_{2.5}$	BC	OC
Statistical data for this research	1.38	10.15	49.54	13.47	3.20	14.74	3.92	0.17	0.44
National Environmental Statistics Bulletin	1.38	12.16	52.03	24.24	3.26	14.68	3.91	0.22	0.41
"2+26" center for air pollution prevention and control	0.89	9.24	48.98	13.93	3.16	13.82	3.72	0.19	0.46

3 Methods

A key step for quantifying the relative contribution of anthropogenic emissions to $PM_{2.5}$ variations 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 CTM to comprehensively evaluate the role of anthropogenic emissions and meteorological conditions in the decrease of $PM_{2.5}$ concentrations in Beijing from 2013 to 2017.

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 produces data sets suitable for understanding the trend of $PM_{2.5}$ concentrations mainly influenced by anthropogenic factors (Eskridge et al., 1997). To better analyze the trend of time series data without the disturbances from other major influencing variables, a statistical method Kolmogorov-Zurbenko (KZ) filtering was proposed by Rao et al. (1994). The KZ filter is advantageous of removing high-frequency variations in data sets through 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 KZ achieved higher confidence in detecting long-term trend than other models. Due to its reliable performance in trend detection in complicated ecosystems, the KZ filter has been

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increasingly 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; Ma et al., 2016; Cheng, N et al., 2019). One potential limitation of the KZ filter is that iterative moving average (*m*) may impose an influence on detecting abrupt variations. Therefore, Zurbenko et al.（1996）proposed an enhanced KZ filter that employed a dynamic variable *m* that decreased with the increase in changing rate. For this research, we employed this dynamic m to produce an adjusted time-series of $PM_{2.5}$ concentrations in Beijing by removing large inter-annual and seasonal variations in meteorological conditions. The principle of the KZ filter is briefly introduced as follows.

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

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

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

$$E(t) = KZ_{365,3}(X) \qquad (3)$$

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

$$W(t) = X(t) - KZ_{15,5}(X) \qquad (5)$$

Where X (*t*) is the original time series of airborne pollutants, E(*t*) is the long-term trend component, S(*t*) is the seasonal component, W(*t*) is the short-term (synoptic-scale) component or residue. $KZ_{i,j}(X)$ indicates 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 and seasonal component, presenting steady trend variation. E(*t)* is mainly affected by long-term anthropogenic emission and climate change. S(*t*) is mainly influenced by the seasonal variation of emission and meteorological conditions. 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 meteorological factors strongly correlated with airborne pollutants.

We examined correlations between seasonal $PM_{2.5}$ concentrations in Beijing and a series of meteorological factors, including temperature, wind speed, wind direction, precipitation, relative humidity, solar radiation, evaporation and air pressure. Due to limited space, detailed correlations between $PM_{2.5}$ concentrations and individual meteorological factors in Beijing are not presented here and readers can refer to previous studies for more information (Chen et al., 2017; 2018). The correlation analysis revealed that wind speed, relative humidity, temperature and solar radiation were strongly and significantly correlated with $PM_{2.5}$ concentrations in Beijing (as shown in Table 2), which was consistent with findings from other studies (Sun et al., 2013; Wang et al., 2018).

Table 2. Major meteorological factors strongly correlated with seasonal $PM_{2.5}$ concentrations in Beijing (Chen et al., 2017)

Spring	**Summer**	**Autumn**	**Winter**
	RHU**(0.648)	RHU**(0.587)	RHU**(0.738)
RHU**(0.532)	SSD**(−0.447)	SSD**(−0.509)	SSD**(−0.715)
	TEM**(0.554)	WIN**(−0.468)	WIN**(−0.558)

****Correlation is significant at the 0.01 level (2 tailed);**

RHU: Relative humidity; SSD: Sunshine Duration; TEM: Temperature; WIN: Wind speed

–Therefore, we further established multiple linear regression equations between $PM_{2.5}$ concentrations and wind speed, relative humidity, temperature and solar radiation as follows.

$$W(t) = \alpha_0 + \sum \alpha_i w_i(t) + \varepsilon_w(t) \quad (6)$$

$$X_b(t) = b_0 + \sum b_i x_i(t) + \varepsilon_b(t) \quad (7)$$

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

Where $w_i(t)$ and $x_i(t)$ stand for the different short-term and baseline component of the ith meteorological factor. ε_w and ε_b is the regression residue of the short-term and baseline component. $\varepsilon(t)$ indicates the total residue, including the short-term influence of local emission and meteorological factors neglected during the regression process and other noises.

Next, KZ filtering was conducted on the $\varepsilon(t)$ 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), $\overline{E(t)}$.

$$X_{LT}(t) = \overline{E(t)} + \varepsilon_E(t) \quad (9)$$

After KZ filtering, the relative contribution of meteorological conditions to $PM_{2.5}$ variations

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can be calculated as follows:

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

Where $P_{contrib}$ is the relative contribution of meteorological conditions to $PM_{2.5}$ variations 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 with filtered influences from meteorological variations.

3.2 WRF-CMAQ model

We employed WRF-CMAQ for simulating the effects of emission-reduction on the decrease of $PM_{2.5}$ concentrations. WRF-CMAQ 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, Shandong and Henan Province). The vertical layer was divided into 20 unequal layers, eight of which were of a less-than-1km distance to the ground for better featuring the structure of atmospheric boundary. The height of the ground layer was 35m.

We employed ARW-WRF3.2 to simulate the meteorological field. The setting of the center and the bidirectional nest for WRF and CMAQ was similar. There were 35 vertical layers for 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 outputs from WRF were interpolated to the region and grid of CMAQ using the Meteorology-Chemistry Interface Processor (MCIP, https://www.cmascenter.org/mcip). The meteorological factors used for this model included temperature, air pressure, humidity, geopotential height, zonal wind, meridional wind,

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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. 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/) provided anthropogenic emission data. 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 of $PM_{2.5}$ concentrations.

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

Where $P_{contrib}$, C and C_{base} are the contribution rate of emission-reduction to $PM_{2.5}$ concentrations, 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 four 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 to estimate the relative contribution of meteorological conditions 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 concentrations to $PM_{2.5}$ reduction in Beijing from 2013 to 2017. Since the WRF-CMAQ simulation simply considers $PM_{2.5}$ 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 $PM_{2.5}$ reduction in Beijing. However, the output from this sensitivity experiment serves as a useful reference for cross-verifying the output from the KZ filtering. For the remaining three 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, and thus quantified the relative contribution of one specific emission-reduction measure to $PM_{2.5}$ reduction in Beijing from 2013 to 2017. Consequently, we quantified the relative contribution of three major emission-reduction measures to $PM_{2.5}$ reduction in Beijing (Table ~~1~~3).

Table ~~1~~3. The design and materials for two baseline and four sensitivity experiments using WRF-CMAQ

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 in Beijing 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 in Beijing 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 in Beijing 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 in Beijing 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. MEIC 2017 was acquired based on our update of MEIC 2016 according to official 2017 emission-reduction targets required by the local government.

3.3 Model verification

3.3.1 Verification of 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, seasonal and short-term component to the total variance is shown as Table ~~2~~4. The sum of the long-term, seasonal and short-term component contributed to more than 93.6~95.3% of the total variance in different stations respectively. The larger the total variance, the three components are more independent to each other. The total variance close to 100% suggests that a majority of meteorological influences has been considered and effectively removed. As shown in Table ~~2~~4, the large value of the total variation in all stations indicated a satisfactory output from the KZ filtering.

Specifically, t~~T~~he relative contribution of the seasonal component (ranging from 9%-23.8%) and short-term component (ranging from 66.8%-83.8%) was much larger than that of the long-term component (ranging from 1.2%-3.5%)~~the short-term component was much larger than that of the seasonal and long-term component~~, suggesting that seasonal and short-term variations of meteorological and emission factors exerted a major influence on the rapid change of $PM_{2.5}$ concentrations in Beijing. The decomposed long-term, seasonal and short-term component from the original time series of mean urban $PM_{2.5}$ concentrations in Beijing from 2013 to 2017 ~~is~~ are demonstrated as Fig 2. According to Fig 2, the notable peaks of decomposed seasonal and short-term component were highly consistent with the peaks of $PM_{2.5}$ concentrations in the original time-series, which further proved the dominant influence of seasonal and short-term variations of meteorological and anthropogenic factors on the temporal changes of $PM_{2.5}$ concentrations in Beijing.

Table ~~2~~4. 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 component (%)	Seasonal component (%)	Short-term component (%)	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

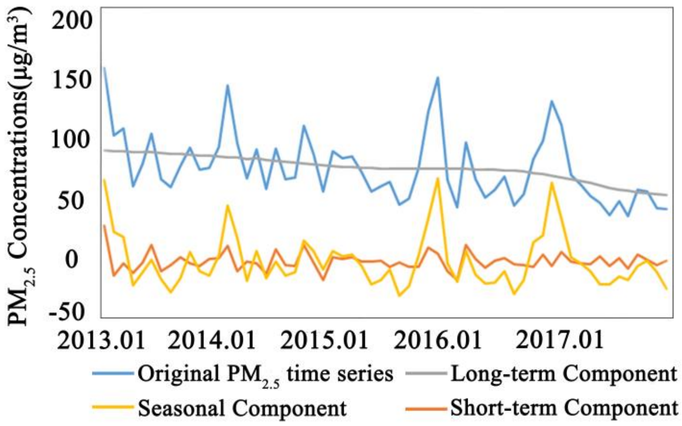

Fig 2. The long-term, seasonal and short-term component extracted from the original time series of mean urban $PM_{2.5}$ concentrations in Beijing from 2013 to 2017

3.3.2 Verification of WRF-CMAQ

We employed the emission inventory and meteorological data for 2017 to verify the accuracy of WRF-CMAQ simulation. For six stations of different types (DingLing background station, Yufa rural station, Olympic Center urban station, Guanyuan urban station, Dongsi urban station and Agricultural museum urban station), we compared the observed and estimated $PM_{2.5}$ concentrations and presented the comparison result as Fig 3. According to Fig 3, the general trend of the simulated $PM_{2.5}$ concentrations was consistent with that of the observed $PM_{2.5}$ concentrations. For six 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 0.63~0.91, -6%~6%, 26%~40%, -5%~7%, and 27%~46% respectively, indicating a satisfactory simulation output (EPA， 2005; Boylan et al., 2006). However， as shown in Figure 3, WRF-CMAQ may notably underestimate $PM_{2.5}$ concentrations during heavy pollution episodes due to unified parameter setting for long-term simulation, the uncertainty in emission inventories, and especially insufficient chemical reaction mechanisms, which is a common challenge for CTM-based $PM_{2.5}$ simulation (Li et al., 2011). For instance, without considering heterogeneous/aqueous reactions between multiple precursors, CTMs failed to approach the maximum $PM_{2.5}$ concentrations during severe haze episodes and the simulation accuracy was dramatically improved by including proper descriptions of heterogeneous/aqueous reactions into CTMs (Chen, D. et al. 2016). With more finer-scale emission inventories and better descriptions of reaction mechanisms between precursors, the accuracy of $PM_{2.5}$ simulation can be improved significantly.

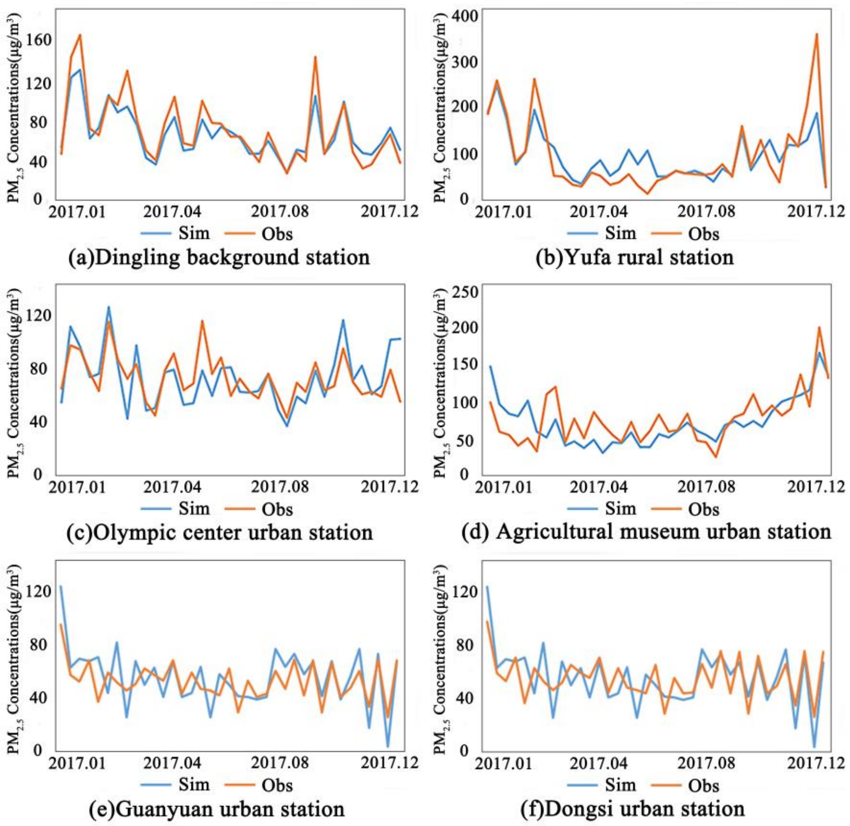

Fig 3. The comparison between observed and WRF-CMAQ simulated $PM_{2.5}$ concentrations in 2017 in six stations across Beijing

4 Results

4.1 The relative contribution of emission-reduction and meteorological variations to the decrease of $PM_{2.5}$ concentrations in Beijing from 2013 to 2017

4.1.1 Estimation based on KZ filtering

Through KZ filtering, the adjusted time-series of $PM_{2.5}$ concentrations with filtered meteorological variations was acquired. Next, for each station, the actual $PM_{2.5}$ variations and adjusted $PM_{2.5}$ variations without the disturbance of meteorological variations from 2013 to 2017 were calculated respectively (as shown in Table ~~3~~5). Based on this, the relative contribution of emission-reduction and meteorological conditions to $PM_{2.5}$ reduction in Beijing from 2013 to 2017 can be quantified.

The original and KZ-processed time series of $PM_{2.5}$ concentrations were illustrated using one background station, one rural station and four urban stations (Fig 4). As shown in Fig 4, most abrupt variations in the original time series of $PM_{2.5}$ concentrations have been smoothed through KZ filtering and the generally decreasing trend of $PM_{2.5}$ variations from 2013 to 2017 caused by anthropogenic emissions can be clearly presented.

Table ~~3~~5. 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.

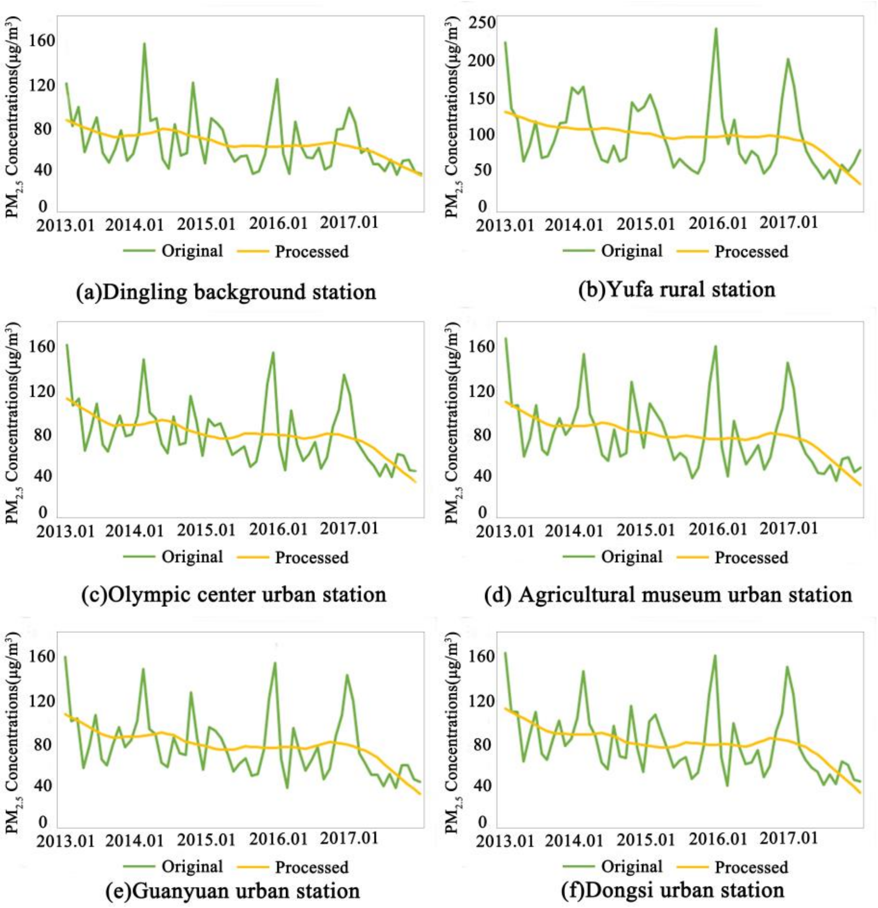

Fig 4. The comparison of original and KZ processed time series of $PM_{2.5}$ concentrations in six stations from 2013 to 2017

According to Table ~~3~~5, the annual mean $PM_{2.5}$ concentration in Beijing in 2017 was 35.6% lower than that in 2013. By filtering the influence of meteorological variations, the adjusted annual mean $PM_{2.5}$ concentration in Beijing in 2017 decreased by 31.7% when compared to that in 2013, indicating that the variation in meteorological conditions exerted a moderate influence on $PM_{2.5}$ reduction from 2013 to 2017. Meteorological conditions in Beijing were generally favorable for $PM_{2.5}$ dispersion during the five-year period, 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 mainly attributed to the favorable meteorological conditions or emission-reduction. Table ~~3~~ 5 suggests that the control of anthropogenic emissions contributed to 75.2%~85.0% of $PM_{2.5}$ decrease in the five-year period, indicating that emission-reduction worked effectively in all rural, urban and background stations. On average, the relative contribution of emission-reduction 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 from 2013 to 2017.

4.1.2 Estimation based on WRF-CMAQ

In addition to the KZ filter, we also employed WRF-CMAQ to estimate the relative contribution of emission-reduction and meteorological conditions to the decrease of $PM_{2.5}$ concentrations in Beijing. The result is shown in Table ~~4~~6.

Table ~~4~~6. Estimated relative contribution of emission-reduction and meteorological variations to $PM_{2.5}$ reduction in Beijing from 2013 to 2017 using WRF-CMAQ

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

Based on WRF-CMAQ, the relative contribution of meteorological variations to the decrease in $PM_{2.5}$ concentrations in Beijing ranged from 20.3% to 22.2% in different stations, whilst emission-reduction accounted for about four-fifths of $PM_{2.5}$ reduction from 2013 to 2017. It is worth mentioning that WRF-CMAQ is 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 multiple stations. Furthermore, WRF-CMAQ simply considered the differences between meteorological conditions in 2013 and 2017 without considering their variations during the five-year period 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 WRF-CMAQ was 21.4%, very similar to the 19.4% calculated using KZ filtering. The slightly larger meteorological contribution calculated using WRF-CMAQ might be attributed to that WRF-CMAQ simply considered the favorable meteorological conditions in 2017 whilst KZ fully considered the long-term meteorological variations from 2013 to 2017.

Since KZ filtering is fully based on observed data, and simply considers the influence of time-series meteorology data on $PM_{2.5}$ time series, less uncertainty is involved. The accuracy of KZ filtering is influenced mainly by the variations of $PM_{2.5}$-meteorology interactions in different areas and seasons. On the other hand, CTMs (e.g. WRF-CMAQ or WRF-CAMx) consider both meteorological conditions (mainly large-scale meteorological data for model simulation, not as accurate as local observed meteorological data) and anthropogenic emissions for estimating $PM_{2.5}$ concentrations under different emission scenarios. The accuracy of these models are not only decided by proper understanding of $PM_{2.5}$-meteorology interactions, but also the reliability of emission inventories and proper descriptions of reaction mechanisms for $PM_{2.5}$ production, especially during heavy pollution episodes, which is a major challenge for current model simulation.~~When KZ filtering is an advanced statistical model solely based on observed meteorological and PM2.5 time series data whilst CTMs involved meteorological data, PM2.5 data, a diversity of reaction mechanisms and emission inventories, CTMs are influenced by more types of data and mechanism uncertainties.~~ Consequently, KZ filtering provides a more reliable method for researchers and decision makers to understand the relative importance of emission-reduction and meteorological conditions in recent $PM_{2.5}$ reduction in Beijing. ~~However~~ Meanwhile, similar outputs from 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 addition to the combined effects of all emission-reduction measures, we further employed WRF-CMAQ to quantify the relative contribution of different

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emission-reduction measures to the decrease in $PM_{2.5}$ concentrations in Beijing from 2013 to 2017.

4.2 The relative contribution of different emission-reduction measures to the decrease in $PM_{2.5}$ concentrations in Beijing

The observed annual average $PM_{2.5}$ concentration in Beijing in 2017 was 58 mg/m^3, compared with 89.5 μg/m^3 in 2013. Based on WRF-CMAQ simulation, meteorological conditions contributed 6.7 μg/m^3 whilst the control of anthropogenic emissions contribute contributed 24.7 μg/m^3 to the total $PM_{2.5}$ reduction of 31.5 μg/m^3 in Beijing from 2013 to 2017. Specifically, local and regional emission-reduction accounted for 16.9 μg/m^3 and 7.8 μg/m^3 of $PM_{2.5}$ reduction. Local emissions and regional transport took up 68.4% and 31.6% of total anthropogenic emissions in Beijing. This result is consistent with our recent study (Chen et al., 2019). Chen et al. (2019) investigated four pollution episodes in Beijing in 2013, 2016, 2017 and 2018 respectively and found that local emissions accounted for 69.3%, 76.8%, 49.5% and 88.4% of total emissions in Beijing respectively. Except for the moderate pollution episode in 2017, local emissions caused more than two thirds of anthropogenic emissions in Beijing. Therefore, local emissions played a dominant role for $PM_{2.5}$ variations in Beijing in both long-term run and heavy pollution episodes. According to three emission-reduction scenarios designed, 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 μg/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 μg/m^3 and 3.4 μg/m^3 respectively. The three major strategies accounted for around half of the total effects of emission-reduction on $PM_{2.5}$ variations in Beijing.

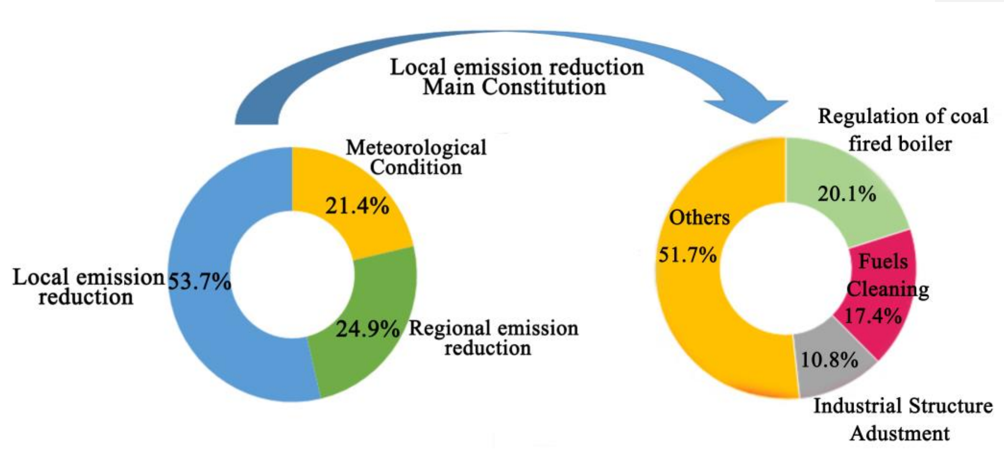

Fig 5. The relative contribution of different influencing factors to the decrease of $PM_{2.5}$ concentrations in Beijing from 2013 to 2017

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 $PM_{2.5}$ concentration to less than 60 μg/m^3. Meanwhile, in November 2017, strong northerly winds in Beijing resulted in the cleanest winter in the past five years, raising arguments whether the favorable meteorological conditions were primarily responsible for $PM_{2.5}$ reduction or whether the significant improvement in air quality in Beijing was mainly attributed to the control of anthropogenic emissions. In this case, a quantitative comparison between the influence of meteorological conditions and emission-reduction on $PM_{2.5}$ reduction is necessary for comprehensively evaluating the Five-year Clean Air Action Plan. Based on two different approaches, this research revealed that the control of anthropogenic emissions contributed to around 80% of $PM_{2.5}$ reductions in Beijing from 2013 to 2017, indicating that the Five-Year Clean Air Plan exerted a dominant influence on air quality enhancement in Beijing. 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 major emission-reduction strategies 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 WRF-CMAQ, the regulation of coal boilers and increasing use of clean fuels for residential use jointly 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 caused 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 long-term clean air plan should be further designed and implemented in Beijing and other mega cities with heavy $PM_{2.5}$ pollution.

Recently, with growing attention to the completion of the Five-year Clean Air Action Plan, some other studies have also been conducted to evaluate this five-year plan. Cheng, J. et al. (2019) employed a finer-scale and more detailed local emission-inventory and quantified the relative contribution of multiple emission-reduction strategies, including the control of coal-fired boilers, increasing use of clean fuels, optimization of industrial structure, fugitive dust control, vehicle emission control, improved end-of-pipe control, and integrated treatment of VOCs. The relative contribution of these emission-reduction measures to $PM_{2.5}$ reduction in Beijing from 2013 to 2017 was 18.7%, 16.8%, 10.2%, 7.3%, 6.0%, 5.7% and 0.6% respectively. By contrast, our research revealed that three major emission-reduction measures (the regulation of coal-fired boiler, increasing use of clean fuels and industrial restructuring) contributed 20.1%, 17.4% and 10.8% of total $PM_{2.5}$ reduction in Beijing from 2013 to 2017, which was very close to Cheng, J et al. (2019)'s findings. Based on finer-scale local emission-inventories with more field-collected emission data, Cheng, J et al. (2019) provided a comprehensive and reliable understanding of the effects of multiple emission-reduction measures on $PM_{2.5}$ reduction in Beijing. The similar outputs from the two studies further proved the reliability of WRF-CMAQ simulation. Meanwhile, Cheng, J et al. (2019) and UNEP (2019) jointly quantified that the total amount of reduction in SO_2, NO_x, VOCs and direct $PM_{2.5}$ induced by the control of anthropogenic emissions was 79420t, 93522t, 115752t and 44307t respectively, which was the major driver for the notable $PM_{2.5}$

reduction in Beijing from 2013 to 2017.

Although the “2+26” regional strategy for air quality improvement in Beijing has become a hotly debated issue and growing emphasis has been placed on the proper design and implementation of regional emission-reduction strategies in Beijing and its surrounding cities，previous studies (Chen et al., 2019; Cheng, J. et al., 2019) and this research proved that local emissions played a dominant role in affecting $PM_{2.5}$ concentrations in Beijing. Specifically, Chen et al. (2019) pointed out that with intensive reduction of coal-fired boilers in Beijing-Tianjin-Hebei region, the relative contribution of vehicle emissions to $PM_{2.5}$ concentrations in Beijing, especially during heavy pollution episodes, could be up to 50%. To further improve air quality in Beijing, stricter regulations on local vehicle emissions, including contingent strategies during pollution episodes (e.g. odd-even license plate policy) and long-term policies (e.g. increasing availability of public transit systems and electric cars) should be a major priority for the next stage clean-air actions.

Based on KZ filtering, Cheng, N et al. (2019) and Ma et al. (2016) suggested the seasonal component contributed dominantly to O_3 variations in Beijing. By comparison, this research revealed that the short-term component contributed dominantly to $PM_{2.5}$ variations in Beijing. These findings well explained the phenomenon that ground ozone pollution in Beijing, controlled by seasonal variations of emission and meteorological conditions (especially high-temperature and low-humidity), simply occurred in summer, whilst $PM_{2.5}$ pollution in Beijing, controlled by short-term variations of meteorological and emission factors, might occur in all seasons. Consequently, contingent emission-reduction measures during heavy pollution episodes are an effective approach to offset the short-term deterioration of meteorological conditions and improve local air quality.

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 balancedly. In addition to the control of anthropogenic emissions, $PM_{2.5}$ 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 accumulation or dispersion of $PM_{2.5}$ in Beijing. Meanwhile, previous studies (Chen et al., 2017) suggested that increasing wind speeds led 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, 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 long-term favorable meteorological influences on $PM_{2.5}$ reduction in Beijing.

6 Conclusions

To comprehensively evaluate the effect of the Beijing Five-year Clean Air Action Plan (2013-2017), we quantified the relative contribution of meteorological conditions and the control of anthropogenic emissions to the notable decrease in $PM_{2.5}$ concentrations in Beijing from 2013 to 2017. Based on KZ filtering, we found that meteorological conditions and emission-reduction accounted for 19.4% and 80.6% of the $PM_{2.5}$ reduction in Beijing, respectively. The large short-term component suggested that short-term variations of meteorological and emission factors exerted a dominant influence on the rapid variation of $PM_{2.5}$ concentrations in Beijing. Meanwhile, WRF-CAMQ revealed that meteorological conditions and emission-reduction contributed to 21.4% and 78.6% of $PM_{2.5}$ variations. Specifically, local and regional emission-reduction measures contributed to 53.7% and 24.9% of $PM_{2.5}$ reduction. For three major emission-reduction measures, the regulation of coal boilers, increasing use of clean fuels for residential use and industrial restructuring contributed to 20.1 %, 17.4% and 10.8% of $PM_{2.5}$ reduction, respectively. Similar outputs from two models suggested that the control of anthropogenic emissions contributed to around 80% of the total decrease in $PM_{2.5}$ concentrations in Beijing from 2013 to 2017, indicating that the Five-year Clean Air Plan worked effectively and such long-term clean air plan should be continued in the following years to further reduce $PM_{2.5}$ concentrations in Beijing.

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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, Gao, B and Li, R. conducted data analysis. Chen, D and Zhuang, Y. produced the figures. Kwan, M., and Chen, B helped revise this manuscript.

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