RC1

This is a well-organized and clearly written manuscript on quantified the relative influences of anthropogenic emissions and meteorological conditions on measured $PM_{2.5}$ concentrations, an important topic in the field of atmospheric environment. It merits to be published after some revisions.

1) Figure 3a showed that the concentrations of $PM_{2.5}$ have been declining since 2012. However, many studies reported that winter of 2013 was suffered the most serious air pollution in recent years. Is the data used in this study different?

Reply: Yes, the winter of 2013 (January 2013) was the most seriously polluted period in recent years. The data used in this study are similar to others. In our definition, winter of 2012 include December of 2012, January of 2013 and February of 2013. We clarified this in lines 157:160, "Wintertime periods defined as the last month of the year and the following two months of the next year were simulated for years 2002-2016. For example, the winter of 2002 includes December of 2002, January of 2003 and February of 2003."

2) In line 220, revise "Over the entire study period" into "Over the entire study period, 2002-2016";

Reply: We have changed it.

3) in line 248, what are the reasons of RH decreases slightly (-5.3 %/decade) between 2002 and 2016 period (Fig. 4a);

Reply: As pointed out by Ding et al. (2014), the decrease of the relative humidity was partly due to the increase of surface temperature. We added this reason in the revised manuscript: "As suggested by Ding et al. (2014), the decrease of RH was partly caused by the increase of surface temperature."

Ding, Y. and Liu, Y.: Analysis of long-term variations of fog and haze in China in recent 50 years and their relations with atmospheric humidity, Science China Earth Sciences, 57(1), pp.36-46, 2014.

4) Is it significant or not that the correlation between wind speed and pressure difference in figure 4c?

Reply: The correlation between WRF simulated wind speeds and pressure difference is 0.49 ($p=0.06$), and the correlation between observed wind speeds and pressure difference is 0.51 ($p=0.05$). They are well correlated and p values are around the threshold of significance (0.05). As wind speeds are affected also by other factors including surface roughness, its correlation with pressure differences is not perfect.

5) in the introduction around line 107, I suggest indicating the increase northly wind could be the potential threaten to air quality over the Yangtze River Delta, China, downwind of north China plain (Kang et. al, 2019).

ref. to: Kang, H., Zhu, B., Gao, J., He, Y., Wang, H., Su, J., Pan, C., Zhu, T., and Yu, B.: Potential impacts of cold frontal passage on air quality over the Yangtze River Delta, China, Atmos. Chem. Phys., 19, 3673-3685, https://doi.org/10.5194/acp-19-3673-2019, 2019.

Reply: We added this reference in the introduction in the revised manuscript: "The variability of northly winds in North China has also been linked to air quality over the Yangtze River Delta, which is a downwind region of North China (Kang et al., 2019)".

RC2

This manuscript is to understand the effect of changes in meteorology and emission on wintertime $PM_{2.5}$ concentrations in Beijing. There are some concerns that need to be addressed.

1) There are more journal papers studying effect of meteorology and emissions on air quality in China. The introduction should be more comprehensive.

Reply: We added more references with similar topics in the revised manuscript: "The influences of anthropogenic emissions and meteorological conditions on air quality over shorter periods have been investigated extensively (Gao et al., 2017a; Y. Gao et al., 2011; Liu et al., 2017; Wang et al., 2016; Xing et al., 2011)."

Gao, M., Liu, Z., Wang, Y., Lu, X., Ji, D., Wang, L., Li, M., Wang, Z., Zhang, Q. and Carmichael, G.R.: Distinguishing the roles of meteorology, emission control measures, regional transport, and co-benefits of reduced aerosol feedbacks in "APEC Blue", Atmos. Env., https://doi.org/10.1016/j.atmosenv.2017.08.054, 2017.

Gao, Y., Liu, X., Zhao, C. and Zhang, M.: Emission controls versus meteorological conditions in determining aerosol concentrations in Beijing during the 2008 Olympic Games, Atmos. Chem. Phys., 11, 12437-12451, https://doi.org/10.5194/acp-11-12437-2011, 2011.

Liu, T., Gong, S., He, J., Yu, M., Wang, Q., Li, H., Liu, W., Zhang, J., Li, L., Wang, X. and Li, S.: Attributions of meteorological and emission factors to the 2015 winter severe haze pollution episodes in China's Jing-Jin-Ji area, Atmos. Chem. Phys., 17(4), https://doi.org/10.5194/acp-17-2971-2017, 2017.

Wang, Y., Zhang, Y., Schauer, J.J., de Foy, B., Guo, B., and Zhang, Y.: Relative impact of emissions controls and meteorology on air pollution mitigation associated with the Asia-Pacific Economic Cooperation (APEC) conference in Beijing, China, Sci. Total Env., https://doi.org/10.1016/j.scitotenv.2016.06.215, 2016.

Xing, J., Zhang, Y., Wang, S., Liu, X., Cheng, S., Zhang, Q., Chen, Y., Streets, D.G., Jang, C., Hao, J. and Wang, W.: Modeling study on the air quality impacts from emission reductions and atypical meteorological conditions during the 2008 Beijing Olympics, Atmos. Env., https://doi.org/10.1016/j.atmosenv.2011.01.025, 2011.

2) Line 85: Peral -> Pearl.

Reply: Changed.

3) Did the model include dust emissions? If so, it is better to provide the information in the section 2.2.

Reply: Yes, we added in sect. 2.3: "Dust emissions and online sea-salt emissions were also calculated online."

4) Line 166: the second scenario had varying meteorological conditions and fixed anthropogenic emissions. According to my understanding, by comparing this scenario with the CTL scenario, the difference should reflect the effect of anthropogenic emissions. It is suggested to change the name of the second scenario as "Emis".

Reply: In the second scenario, emissions were fixed and only meteorological conditions will affect the simulated concentrations of air pollutants. Thus, we use this simulation to represent the effects of meteorological conditions (MET) on the variations of $PM_{2.5}$. In the presentation of the results, we use the values from CTL and MET simulations directly, instead of the differences between two simulations. We conducted MET simulation to answer the question: if emissions were not changing, what would be the effects of climate conditions on air pollution? To make it clearer, we added clarification in the revised manuscript: "The MET simulation can be used to answer the question: what the climate would have done to the variations of $PM_{2.5}$ if emissions were not changing? The CTL simulation contains the information of changes in both meteorological conditions and emissions."

5) Section 3.1: Emission inventory, and initial and boundary meteorological conditions, for different years varied, and thus the model performance for different years also varied. So, the difference of simulated air pollutant levels between years may not fully reflect the trend. In particular, the authors claimed that the $PM_{2.5}$ in Beijing showed a decreasing trend of 1.4 µg/m3 per year. This result is within the model error magnitude and uncertainty.

Reply: Thanks for pointing out this issue.

Yes, the performance of model varies with years. As seen from Fig. 2, model overestimates concentrations of $PM_{2.5}$ before 2012. Over 2012-2016, model shows reasonably good performance. Over the period 2012-2016, both model and observations show rapid declines. We agree that the decreasing trend of 1.4 µg/m3 might have been overstated given the overestimation of $PM_{2.5}$ in early years.

In the manuscript, we have already acknowledged this issue: "**Given the more serious overestimate of $PM_{2.5}$ in early periods (Fig. 2), the declining rates inferred from the CTL simulation might have been slightly overstated.**" To make it clearer, we added acknowledgement also in the abstract: "Given the overestimation of $PM_{2.5}$ by model, the effectiveness of stringent emission control measures might have been slightly overstated" in the revised manuscript.

However, this will not change our conclusion that unfavorable climate conditions would have led to more pollution in Beijing, and stringent emission control measures have changed the direction of changes. To make the conclusion more reasonable, we only used statistics drew between 2011 and 2016, during which model shows better performance: "Between the winters of 2011 and 2016, stringent emission control measures resulted in a 21% decrease in mean mass concentrations of $PM_{2.5}$ in Beijing, with 7 fewer haze days per winter on average. "

6) The study used 150μg/m^3 as a threshold to define a haze day. However, the model showed an overestimation of $PM_{2.5}$ that would affect the simulated of haze days.

Reply: Thanks for pointing out this issue. The overestimation of $PM_{2.5}$, especially during early years, would lead to overestimated haze days. In the revised manuscript, we also explored what would change if the threshold of 75 μg/m^3 was used. As shown in Fig. r1, the trends are similar to the those using 150 μg/m^3 as a threshold. We have added this discussion in the revised manuscript: "We explored if different thresholds of haze days would change the findings and found the variations are similar when a threshold of 75 μg/m^3 was used (Fig. S2)."

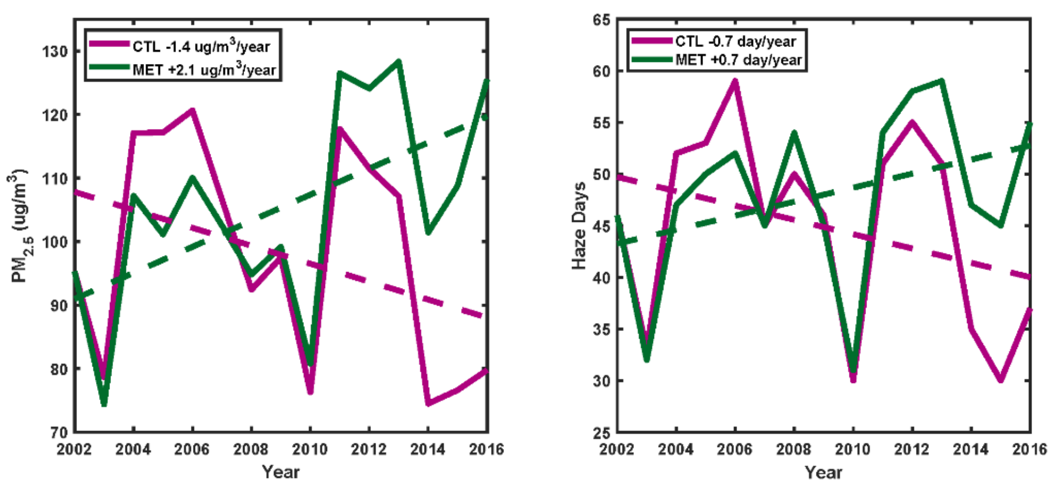

Fig. r1. Wintertime mean concentrations of $PM_{2.5}$ and number of haze days (defined with daily mean concentration above 75 μg/m^3) from the CTL and MET simulations.

It is true that notable overestimation of $PM_{2.5}$ in early years would make the trends of both $PM_{2.5}$ concentrations and haze days different. In the revised manuscript, we added the comparison between simulated and observed number of haze days (Fig. r2). Due to the overestimation of number of haze days by the model, the trend has been overestimated by the model. In fact, the observations show no significant declining trend over 2005-2016. However, this will not change our conclusion that the stringent emission control measures by government has changed the direction of $PM_{2.5}$, especially over 2012-2016 period. We calculated the declining rate of number of haze days inferred from model and observation, and the values are generally consistent (-4.8 day/year and -3.0 day /year). This further indicate that the emission inventory is more accurate over recent years because there were more data to constrain it.

In the revised manuscript, we added the following descriptions to make the presentation clearer: "Due to the overestimation of number of haze days by the model in early years, the declining rates of number of haze days inferred from the CTL simulation might have been overstated. As seen from Fig. S3, there is no notable declining trend in number of haze days inferred from observations over 2005-2016. However, it is consistent that both model and observations indicate rapid declines in number of haze days (-4.8 days and -3.0 days per winter, respectively). With fixed emissions, MET simulation suggests that unfavorable climate conditions would have led to more haze days, emphasizing the significance of emission control in recent years."

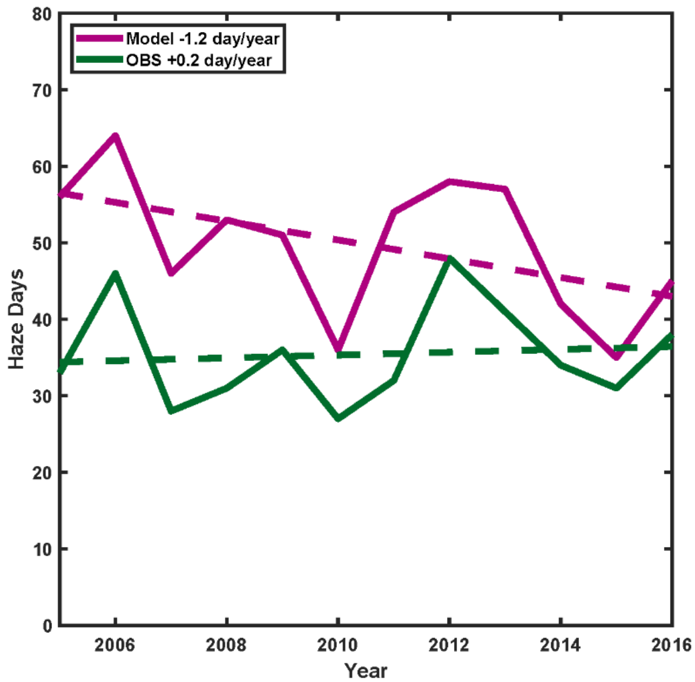

Fig. r2. Modeled and observed number of haze days (defined with daily mean concentration above 75 μg/m^3)

7) Line 254-260: the results are not clear. It is claimed that the RH is a good indicator, but it is mentioned that the variability of RH2 is unlikely to be the driver of enhanced $PM_{2.5}$ under changing conditions of climate.

Reply: RH can explain the variability, but not the increasing trend. RH is positively correlated with $PM_{2.5}$ as higher RH promotes more productions of $PM_{2.5}$. We observed that long-term meteorological conditions have led to higher $PM_{2.5}$, while RH declined over this period. As pointed out by Ding et al. (2014), the decrease of the relative humidity was partly due to the increase of surface temperature. We added this reason in the revised manuscript: "As suggested by Ding et al. (2014), the decrease of RH was partly caused by the increase of surface temperature."

Because the declining trend of RH contradicts with the enhancement of $PM_{2.5}$, we concluded that RH2 is unlikely to be the reason for higher $PM_{2.5}$. However, the declining wind speeds could explain the trend. To make it clearer, we added the following explanation in the revised manuscript. "RH2 has been found to explain the interannual variability of $PM_{2.5}$, but not the increasing trend."

Ding, Y. and Liu, Y.: Analysis of long-term variations of fog and haze in China in recent 50 years and their relations with atmospheric humidity, Science China Earth Sciences, 57(1), pp.36-46, 2014.

8) Line 291: It is believed that the effect of meteorology and emissions should not be linear. The study performed two simulations (CTL and MET); however, it is worth to investigate one more simulation with change emissions and fixed meteorology to confirm the results.

Reply: We agree that the effects of meteorology and emissions are not linear. We performed MET simulation to examine how climate/meteorological conditions will affect air pollution when emissions are not changing. We conducted CTL to explore how variations of emissions would have changed the influence of climate conditions. The purpose of current study is not to investigate how emission changes would change air quality under fixed meteorology as meteorological conditions is difficult to be fixed in reality.

The proposed simulation by the review with fixed meteorology and changing emissions were examined in our previous two studies. In Gao et al. (2016b), we investigated how emission changed over 1960-2010 will affect haze pollution. We found that from 1960 to 2010, the dramatic changes in emissions lead to +260 % increases in sulfate, +320 % increases in nitrate, +300 % increases in ammonium, +160 % increases in BC, and +50 % increases in OC. The responses of $PM_{2.5}$ to individual emission species indicate that the simultaneous increases in SO_2, NH_3, and NO_x emissions dominated the increases in $PM_{2.5}$ concentrations.

In a recent study just published online (Zhou et al., 2019), we conducted three sets of simulations to explore the changes over 2011/2012-2017/2018.

(1) M11E11: using meteorological data of year 2011/2012, and emission data of year 2011

(2) M17E11: using meteorological data of year 2017/2018 and emission data of year 2011;

(3) M17E17: using meteorological data of year 2017/2018, and emission data of year 2017;

The difference between M17E11 and M17E17 is what proposed by the reviewer. We found that over the period 2011-2018, emission reductions play a more important role than meteorological factors. In the revised manuscript, we added the conclusions in Zhou et al. (2019). “Zhou et al.

(2019) concluded that emission reductions play a more important than meteorological conditions in determining the declines in $PM_{2.5}$ over 2011/2012-2017/2018."

Gao, M., Carmichael, G. R., Saide, P. E., Lu, Z., Yu, M., Streets, D. G. and Wang, Z.: Response of winter fine particulate matter concentrations to emission and meteorology changes in North China, Atmos. Chem. Phys., 16, 11837-11851, https://doi.org/10.5194/acp-16-11837-2016, 2016b.

Zhou, W., Gao, M., He, Y., Wang, Q., Xie, C., Xu, W., Zhao, J., Du, W., Qiu, Y., Lei, L., Fu, P., Wang, Z., Worsnop, D.R., Zhang, Q. and Sun, Y.: Response of aerosol chemistry to clean air action in Beijing, China: Insights from two-year ACSM measurements and model simulations, Env. Pol., doi: https://doi.org/10.1016/j.envpol.2019.113345, 2019.

9) Table 1: it is surprised that only RH2 and WS10 had a high statistical significance. It is believed BLH should also be one of the major factors that affect air quality. Also, wind direction should play an important role too.

Reply: I agree that BLH is an important factor that affects the occurrence of air pollution episodes. As we showed in previous study (Gao et al., 2016, Fig. 9d) that the concentrations of $PM_{2.5}$ have an inverse relationship with BLH. In this study, we are looking at long-term trend, and only wintertime mean concentrations of $PM_{2.5}$ and meteorological factors are considered instead of hourly mean or daily mean. We found that long-term BLH is not as important as WS10 and RH2 in determining the long-term trend and interannual variability of $PM_{2.5}$.

Cai et al. (2017) also examined the correlation between BLH and long-term $PM_{2.5}$ and found they are not highly correlated.

Yes, wind direction is an important factor. As mentioned in our previous study (Gao et al., 2016) that haze episodes happen under continuous southerly winds and end under strong northerly winds. The wind directions affect air quality through two ways: (1) in wintertime, speeds of southerly winds are weaker than speeds of northerly winds; (2) southerly winds can bring humid air from the ocean, which can accelerate secondary aerosol formation. As shown in this study that pollution has a close connection with RH. Thus, the influences of wind direction have been contained in the influences of wind speeds and RH.

Gao, M., Carmichael, G.R., Wang, Y., Saide, P.E., Yu, M., Xin, J., Liu, Z. and Wang, Z., 2016. Modeling study of the 2010 regional haze event in the North China Plain. Atmospheric Chemistry and Physics, 16(3), p.1673.

Cai, W., Li, K., Liao, H., Wang, H. and Wu, L., 2017. Weather conditions conducive to Beijing severe haze more frequent under climate change. Nature Climate Change, 7(4), p.257.

10) Figs. 1 and 6: the aspect ratio of the maps seems to be incorrect.

Reply: We've updated Figs. 1 and 6 in the revised manuscript.

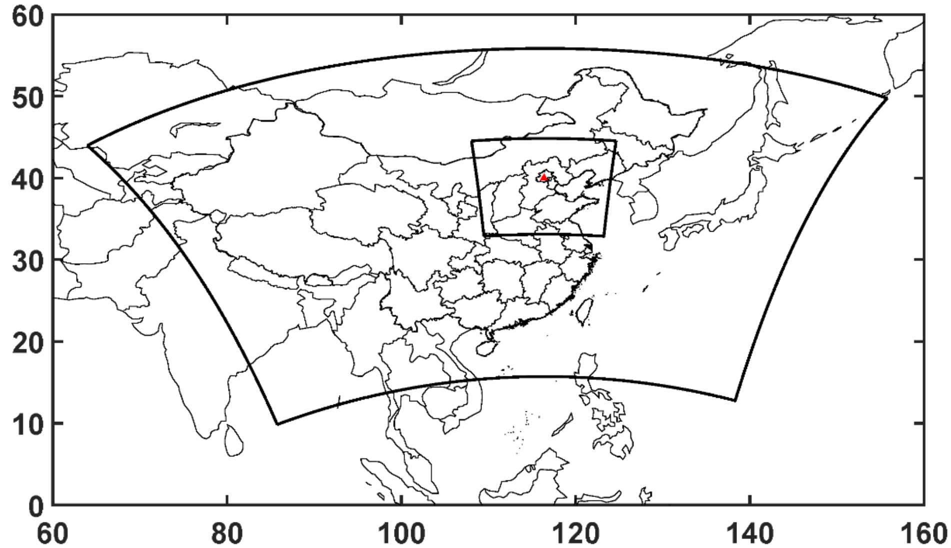

Fig.1. WRF-Chem modeling domain settings and locations of observations.

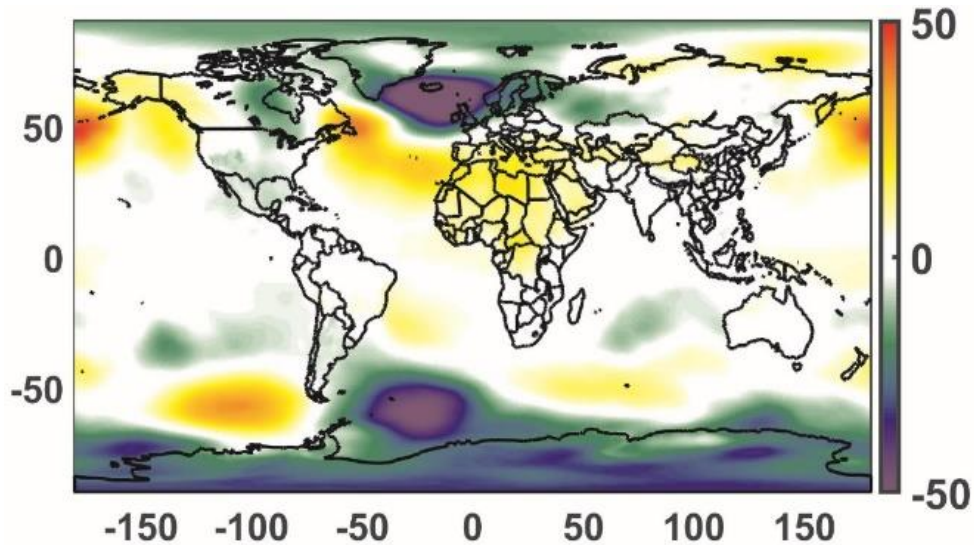

Fig. 6. Trends of winter sea level pressure during 2002-2016 period (unit: Pa/year).

China's Clean Air Action has suppressed unfavorable influences of climate on wintertime $PM_{2.5}$ concentrations in Beijing since 2002

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Abstract

Severe wintertime $PM_{2.5}$ pollution in Beijing has been receiving increasing worldwide attention, yet the decadal variations remain relatively unexplored. Combining field measurements and model simulations, we quantified the relative influences of anthropogenic emissions and meteorological conditions on $PM_{2.5}$ concentrations in Beijing over winters of 2002-2016. Between the winters of 2011 and 2016, stringent emission control measures resulted in a 21% decrease in mean mass concentrations of $PM_{2.5}$ in Beijing, with 7 fewer haze days per winter on average. Given the overestimation of $PM_{2.5}$ by model, the effectiveness of stringent emission control measures might have been slightly overstated. With fixed emissions, meteorological conditions over the study period would have led to an increase of haze in Beijing, but the strict emission control measures have suppressed the unfavorable influences of recent climate. The unfavorable meteorological conditions are attributed to the weakening of the East Asia Winter Monsoon associated particularly with an increase in pressure associated with the Aleutian low.

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1 Introduction

In recent years, persistent and severe haze episodes with high $PM_{2.5}$ concentrations occur frequently in China, attracting worldwide attention (*Cheng et al., 2016; Gao et al., 2016*). High aerosol concentrations during haze have been reported to cause traffic jams and flight cancelations (*Wu et al., 2005*), and have been linked to health damages (*Dockery et al., 1993*), climate change (*Ramanathan and Carmichael, 2008*) and ecosystem degradation (*Chameides et al., 1999*). The annual mean $PM_{2.5}$ concentrations in Beijing exceeded 90 μg/m^3 in 2013, nearly twice the China's National Ambient Air Quality Standard (NAAQS) of 35 μg/m^3 (*MEP, 2012*). January 2013 was reported as the haziest month over the past 60 years in Beijing, with maximum hourly and daily mean $PM_{2.5}$ concentrations exceeding 1000 μg/m^3 and 500 μg/m^3, respectively (*Wang et al., 2014*).

Since then, the State Council of China issued the Air Pollution Prevention and Control Action Plan (APPCAP, denoted as the Clean Air Action hereafter), which describes explicitly the pollution control measures, and proposed specific goals for concentrations by 2017 (*China State Council, 2013*). This action has been considered the most stringent air pollution control policy in Chinese history. The Jing-Jin-Ji, Yangtze River Delta, and ~~Peral~~ Pearl River Delta regions were required to reduce annual mean $PM_{2.5}$ concentrations by 15-25% compared with the concentrations in 2013, and the annual mean concentrations of $PM_{2.5}$ in Beijing should not exceed 60 μg/m^3 (*Cheng et al., 2018; China State Council, 2013*). Specific control measures included eliminating small coal-fired boilers, phasing out small, high-emitting factories, installing control facilities for emissions of VOCs (volatile organic compounds), and replacing residential coal burning with electricity and natural gas among others. (*Zheng et al., 2018*). With these rigorous control measures, China has made impressive progress, with annual mean $PM_{2.5}$ concentrations reduced in major metropolitan regions by 28-40% between 2013 and 2017 (*Zheng et al., 2018*).

A number of studies have used visibility as a surrogate to indicate trends of haze pollution in China over the past several decades (*Che et al., 2009; Chen and Wang, 2015; Ding and Liu,*

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2014; Wang and Chen, 2016). *Chen and Wang (2015)* reported that haze days increased rapidly in the 1970s and remained relatively stable up to present. However, *Che et al. (2009)* illustrated that there was a decreasing haze trend in winter for many cities over the interval 1981-2005. *Wang et al. (2019)* argued that visibility is impacted significantly by meteorological factors, especially relative humidity, and thus visibility does not accordingly reflect the real changes in air pollution. The influences of anthropogenic emissions and meteorological conditions on air quality over shorter periods have been investigated extensively (Gao et al., 2017a; Y. Gao et al., 2011; Liu et al., 2017; Wang et al., 2016; Xing et al., 2011). Due to the lack of long-term measurements of $PM_{2.5}$ in China, limited studies have been conducted exploring the roles of anthropogenic emissions and meteorological conditions for the long-term variations of $PM_{2.5}$ in China. *Yang et al. (2016)* used the GEOS-Chem model to simulate $PM_{2.5}$ in China between 1985 and 2005, and concluded that the increase of winter $PM_{2.5}$ was dominated over this period by the increase in anthropogenic emissions. They found that weakening of winds was the dominant meteorological factor. The variability of winds in North China has also been linked to air quality over the Yangtze River Delta, which is a downwind region of North China (Kang et al., 2019). While *Yang et al. (2016)* ~~this study~~ has explored the relative roles of emissions and meteorology, no model validation of $PM_{2.5}$ was provided, and the recent decades were not covered in the study period. Long-term measurements of $PM_{2.5}$ in Beijing reveal a slight decreasing trend of annual mean concentration over 2004-2012 (*Liu et al., 2015*). With the increase in availability of recent measurements, a closer reading of long-term variations is needed to better define the relative roles of anthropogenic emissions and meteorology.

In this study, we present a comprehensive analysis of the decadal trend of wintertime $PM_{2.5}$ in Beijing based on regional meteorology-chemistry modeling, a new decadal emission inventory, and long-term observations of $PM_{2.5}$, including their composition. We address the following questions: (1) the influences of decadal changes in anthropogenic emissions and meteorology on the variations of winter $PM_{2.5}$ in Beijing; and (2) the key driving factors for the decadal variation of meteorology. The descriptions of model, emissions, and numerical experiments are presented in Sect. 2. The two questions highlighted above are addressed in detail in Sect. 3; Sect. 4 provides an overall summary of the study.

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2 Model Description and Configurations

2.1 Meteorology-chemistry modeling

We used the WRF-Chem (Weather Research and Forecasting model coupled with chemistry) model version 3.6.1 to simulate meteorology and emissions, transport, mixing, and the chemical transformation of trace gases and aerosols. Two nested domains were applied with the outer domain covering East Asia and part of Southeast Asia, with the inner domain focusing on North China (Fig. 1). Horizontal resolutions of 81km and 27km were configured respectively for these two domains, and the model accounted for 27 vertical layers extending from the surface to 50 hPa. The gas phase chemical mechanism CBMZ (*Zaveri and Peters, 1999*) coupled with the 8-bin sectional MOSAIC model with aqueous chemistry (*Zaveri et al., 2008*) was adopted. The model treats all the important aerosol species, including sulfate, nitrate, chloride, ammonium, sodium, black carbon (BC), primary organic and inorganic material. The Fast-j radiation scheme (*Wild et al., 2000*) was selected to calculate photolysis rates. These configurations have been shown in previous studies (*Gao et al., 2016a, 2016b, 2017b*) to be capable of reproducing winter haze episodes in North China.

2.2 Emissions

The monthly Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/) covering the years 2002-2017 (*Zheng et al., 2018*) was used for anthropogenic emissions. This inventory considers emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOC), ammonia (NH_3), black carbon (BC), organic carbon (OC), $PM_{2.5}$, PM_{10}, and carbon dioxide (CO_2) associated with power generation, and the industrial, residential, transportation, and agricultural sectors. The trends of wintertime emissions of these species in Beijing-Tianjin-Hebei region over the interval 2002-2016 are displayed in Fig. S1. Emissions of SO_2 have decreased continuously since 2004, while NO_x emissions have declined since 2011. Emissions of all involved species have decreased rapidly since 2012. Biogenic emissions were calculated online using the MEGAN model (*Guenther et al., 2006*). Emissions from biomass burning were taken from the GFED v3 dataset (*Randerson et al., 2015*). Dust emissions and online sea-salt emissions were

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also calculated online.

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2.3 Numerical experiments

Meteorological initial and boundary conditions were obtained from the National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) dataset. Chemical initial and boundary conditions were taken from the climatological data provided by the NOAA Aeronomy Lab Regional Oxidant Model (NALROM). Wintertime periods defined as the last month of the year and the following two months of the next year were simulated for years 2002-2016. For example, the winter of 2002 includes December of 2002, January of 2003 and February of 2003. Nudging (assimilation) of the analyses was applied to produce realistic meteorological simulations. The simulations were conducted month by month (15 years × 3 month/year = 45 months). To overcome the impacts of initial conditions, five more days were simulated for each month and discarded as spin-up. Two sets of simulations were performed to elucidate the relative roles of changes in anthropogenic emissions and meteorological conditions: (1) CTL simulation, simulations of winter periods from 2002 to 2016 with varying meteorological conditions and anthropogenic emissions; and (2) MET simulation, simulations of winter periods from 2002 to 2016 with varying meteorological conditions only, with anthropogenic emissions fixed at levels that applied in 2002. The MET simulation can be used to answer the question: what the climate would have done to the variations of $PM_{2.5}$ if emissions were not changing? The CTL simulation contains the information of changes in both meteorological conditions and emissions.

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3 Results and Discussion

3.1 Model evaluation

Model evaluation was conducted in terms of both $PM_{2.5}$ concentrations and $PM_{2.5}$ chemical compositions, using measurements from urban Beijing (location is marked with red dot in Fig. 1). $PM_{2.5}$ was measured using the Two tapered Element Oscillating Microbalances (TEOM) system at the Institute of Atmospheric Physics site. More descriptions of the observations were archived in *Liu et al. (2015)*. Fig. 2 displays the variations of simulated and observed daily mean $PM_{2.5}$ concentrations over winters from 2002 to 2016. Temporal variations of simulated and observed $PM_{2.5}$ concentrations are generally consistent, with correlation coefficients ranging from 0.75 to 0.83 (Fig. 2). Notably, the model overestimates $PM_{2.5}$ concentrations for all periods. However, the overestimations decline gradually over time. Values of the mean bias decrease from 54.1 $\mu g/m^3$ over the 2004-2006 period to 15.9 $\mu g/m^3$ for recent winters. The broad ranges of errors in different periods reflect the changing uncertainty of emission inventories for different periods. In early times, documentations of emission sources were not as comprehensive as those available for recent years, leading to larger errors in early inventories.

According to results reported by *Zheng et al. (2018)*, SO_2 emissions decreased by 59%, NO_x emissions decreased by 21%, BC emissions decreased by 28%, and OC emissions decreased by 32% for China between 2013 and 2017. These remarkable changes in emissions are expected to lead to notable changes in both the abundance and composition of $PM_{2.5}$. As shown in Fig. 3, sulfate and OC exhibit the largest declines, ammonium and BC show slight decreases, while nitrate concentrations remain relatively stable over 2013-2017. These measured trends are captured generally well by the model, except that sulfate is still underestimated, and BC is overestimated in Beijing. The underestimate of sulfate by models and the overestimate of BC in Beijing have been well documented in previous studies (*Cheng et al., 206; Gao et al., 2016a, 2018a; Song et al., 2018*), attributed to missing reaction pathways and aging/deposition treatments in models (*Song et al., 2019*). Several heterogeneous reaction pathways for sulfate formation have been proposed, including the oxidation of SO_2 by NO_2, transition-meta-catalyzed O_2, or H_2O_2 in aerosol water, and by NO_2 or O_2 on aerosol surfaces (*Cheng et al., 2016; He et al., 2014; Hung et al., 2018; Li et al., 2018*). More recently, *Song et al. (2019)* proposed that the heterogeneous production of hydroxymethanesulfonate (HMS) from the reaction of SO_2 and formaldehyde could be an important chemical mechanism for wintertime haze in China. With the rapid declines in sulfate in Beijing, the relative importance of nitrate in $PM_{2.5}$ is enhanced, a circumstance worthy of special attention for future pollution control policy.

3.2 Influences of Anthropogenic Emissions and Meteorological Conditions on Haze in

Beijing

Fig. 4 illustrates the wintertime mean concentrations of $PM_{2.5}$ and the numbers of haze days from the CTL and MET simulations. Haze days are defined as occasions with daily mean concentrations exceeding 150 μg/m^3. With fixed anthropogenic emissions, wintertime averaged concentrations of $PM_{2.5}$ would have increased at a rate of 2.1 μg/m^3/year in Beijing (Fig. 4a). Due to the implementation of China's Clean Air Action, the wintertime averaged concentrations of $PM_{2.5}$ have been declining since 2012. Over 2002-2016 winters, mean concentrations of $PM_{2.5}$ in Beijing decreased at a rate of 1.4 μg/m^3/year. Compared to concentrations in the MET simulation, the mean mass concentrations of $PM_{2.5}$ decreased by 21% in Beijing over the winters of 2011-2016.

The effectiveness of China's Clean Air Action has been highlighted also in the decline in the number of haze days in Beijing. In the MET simulation, the total number of haze days over 2011-2016 winters amounted to 157 days, reduced by 44 days as a result of the emission controls implemented over this period (Fig. 4b). On average, China's Clean Air Action resulted in 7 fewer haze days per winter over 2011-2016. Over the entire study period 2002-2016, China's Clean Air Action altered the direction of changes in wintertime haze days, with rates changing from 0.8 day/year to -0.3 day/year. The differences in both mean concentrations of $PM_{2.5}$ and numbers of haze days from the two sets of simulations underscore the impressive success of China's Clean Air Action. Given the more serious overestimate of $PM_{2.5}$ in early periods (Fig. 2), the declining rates inferred from the CTL simulation might have been slightly overstated. With unchanged anthropogenic emissions, the increasing trend of haze pollution that would have occurred for Beijing highlights the unfavorable influences of recent changes in local meteorology. We explored if different thresholds of haze days would change the findings and found the variations are similar when a threshold of 75 μg/m^3 was used (Fig. S2). Due to the overestimation of number of haze days by the model in early years, the declining rates of number of haze days inferred from the CTL simulation might have been overstated. As seen from Fig. S3, there is no notable declining trend in number of haze days inferred from observations over 2005-2016. However, it is consistent that both model and observations indicate rapid declines in number of haze days (-4.8 days and -3.0 days per winter, respectively). With fixed emissions, MET simulation suggests that unfavorable climate conditions would

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have led to more haze days, emphasizing the significance of emission control in recent years. Using similar approach, *Cheng et al. (2019)* found that meteorological conditions explain 12.1% of the improved $PM_{2.5}$ air quality during 2013-2017, while large portions of the improvement are dominated by local (65.4%) and regional (22.5%) emission reductions. Zhou et al. (2019) concluded that emission reductions play a more important than meteorological conditions in determining the declines in PM2.5 over 2011/2012-2017/2018. The current study examines longer term trend since 2002, but the more favorable meteorological conditions mentioned in *Cheng et al. (2019)* are illustrated also in Fig. 4. Our findings highlight also the significance of emission reductions, especially after 2013, while the long-term trend of meteorological conditions since 2002 differs from it during 2013-2017.

3.3 Significance of Different Meteorological Variables

To identify the key meteorological variables for the unfavorable influences on air quality, we applied the stepwise linear regression model (SLR) to determine the relative significance of multiple meteorological variables in terms of their contributions to the variations of $PM_{2.5}$ concentrations. In a SLR model, the selected predictors are included in the regression equation one by one. The predictor that contributes the most to the model is included first, and the process is continued if the additional predictor can statistically improve the regression (*Bendel and Afifi, 1977*). Thus, the SLR model is widely used to select meaningful predictors. In this study, boundary layer heights (BLH), precipitation (PREC), near surface relative humidity (RH2), near surface temperature (T2) and near surface wind speeds (WS10) were selected as predictors for the SLR model. These variables were extracted from the WRF-Chem meteorological simulations with analyses nudging applied. Table 1 summarizes the p values for each predictor. RH2 and WS10 were selected as the most significant predictors for wintertime $PM_{2.5}$ in Beijing (p values < 0.05). The influence of WS10 is greater than that for RH2. *Shen et al. (2018)* reported also that RH and meridional wind speeds drive stagnation and chemical production of $PM_{2.5}$ in Beijing.

The impacts of RH on aerosol composition and processes in winter were examined using measurements in Beijing, and the largest impacts were found for the growth of sulfate and organic aerosols associated with coal combustion (*Sun et al., 2013; Wang et al., 2017*).

Although RH has been shown to be a good predictor for $PM_{2.5}$, it decreases slightly (-5.3 %/decade) between 2002 and 2016 period (Fig. 5a), which contradicts the predicted increasing trend of $PM_{2.5}$ under fixed emissions (Fig. 4a). RH2 has been found to explain the interannual variability of $PM_{2.5}$, but not the increasing trend. Thus, the variability of RH2 is unlikely to be the driver of enhanced $PM_{2.5}$ under changing conditions of climate. As suggested by Ding et al. (2014), the decrease of RH was partly caused by the increase of surface temperature.

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Between 2002 and 2016, simulated wintertime WS10 in Beijing declined gradually at a rate of 0.3 m/s/decade (Fig. 5b), in agreement with the declining trends inferred from observations (Fig. ~~S2~~S4). Weaker wintertime near surface wind speeds are associated with enhanced $PM_{2.5}$ concentrations and increasing numbers of haze days since 2002. In winter, the North China plain features northwesterly winds associated with the East Asian Winter Monsoon (EAWM), properties of which depend largely on the development of both the Siberian high and the Aleutian low (*Jhun and Lee, 2004*). We calculated the intensity of the EAWM index using the pressure difference between area mean sea level pressure (SLP, data were taken from MERRA-2 reanalysis) over the region 90º E-110º E, 40º N-50º N and the area mean SLP over the region 120º -170º E, 40º-60º N. These two regions represent the central focal areas of the Siberian high and the Aleutian low, respectively. Fig. 5(c) indicates that the variations of EAWM intensity has declined gradually over time, consistent with the variations of wind speeds in Beijing. The weakening of EAWM intensity is due partially to the increasing pressure in the regions of the Aleutian low. As shown in Fig. 6, there is no significant change in SLP over the regions of the Siberian high, but SLP of the Aleutian low intensity decreased significantly between 2002 and 2016 (more than 50 Pa/year). Although no significant trend was observed for the strength of the Siberian high, the changes in the position of the Siberian high has been linked to wintertime air quality in China over the past decades (*Jia et al., 2015*).

Yin et al. (2015) found a significant negative correlation between winter haze and the East Asia Winter Monsoon from 1986 to 2010. Deterioration of air quality in China has been linked also to a weaker East Asia Summer Monsoon (*Chin et al., 2012*). The slacking of winds is observed not only for China (*Sherman et al., 2017*), but also for other countries including India (*Gao et al., 2018b*). Analyses using climate projections suggest that wind speeds in continental regions in the Northern Hemisphere will continue to decline under a warming climate (*Karnauskas et al., 2018*), imposing greater pressure on measures for future control of air pollution.

4 Summary

Combining field measurements and model simulations, we quantified the relative influences on $PM_{2.5}$ concentrations in Beijing of anthropogenic emissions and meteorological conditions over winters of 2002-2016. China's Clean Air Action has been effective in reducing both mass concentrations of $PM_{2.5}$ and the number of haze days. With fixed emissions, meteorological conditions over the study period should have resulted in an increase in haze in Beijing, but the strict emission control measures implemented by the government have suppressed the unfavorable influences associated with recent climate. Using a statistical method, we concluded that RH2 and WS10 offer useful predictors for wintertime $PM_{2.5}$ in Beijing, with the variations of WS10 in particularly good agreement with the increasing trend of $PM_{2.5}$ concentrations with fixed emissions. The increasing trend of $PM_{2.5}$ under unfavorable meteorological conditions was attributed further to the weakening of Aleutian low and the EAWM. The variations of $PM_{2.5}$ concentrations were investigated in this study for the urban Beijing region, so were the changes in wind speeds. We do not exclude the possibility that the trend may have been influenced by other factors, including for example increases in surface roughness (*Vautard et al, 2010*).

4 figures are listed in the supplement.

Author contribution

M.G. and M.B.M designed the study; M.G. performed model simulations and analyzed the data with the help from S.S., P.S., and G.R.C.; B. Zheng and Q. Zhang provided the emission inventory; Z.W., Y.W., Z.L., D.J., J.X., and C.L. provided measurements. M.G. and M.B.M. wrote the paper with inputs from all other authors.

Data availability

The measurements and model simulations data can be accessed through contacting the corresponding authors.

Competing interests

The authors declare that they have no conflict of interests.

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Table 1. p-values for stepwise linear regression model for Beijing.

Meteorological Variables	p-values
BLH	0.38
PREC	0.64
RH2	0.02
T2	0.95
WS10	0.00

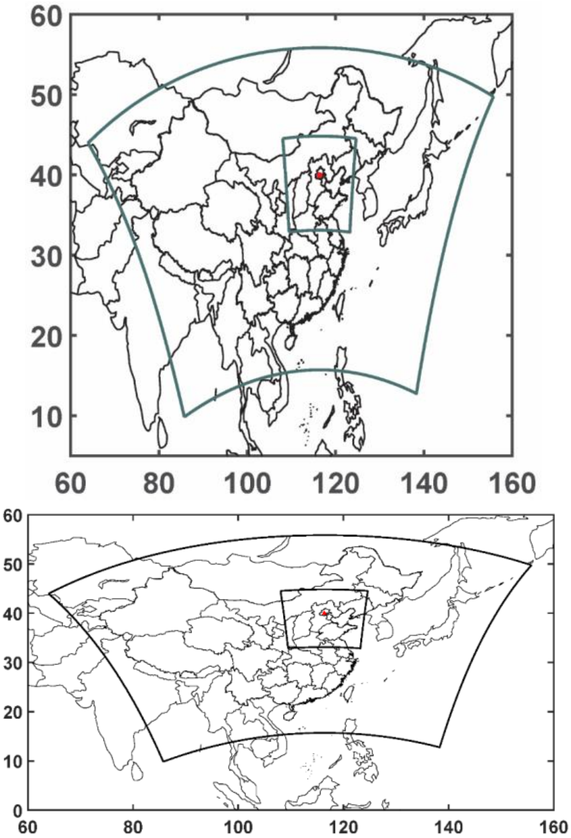

Fig.1. WRF-Chem modeling domain settings and locations of observations.

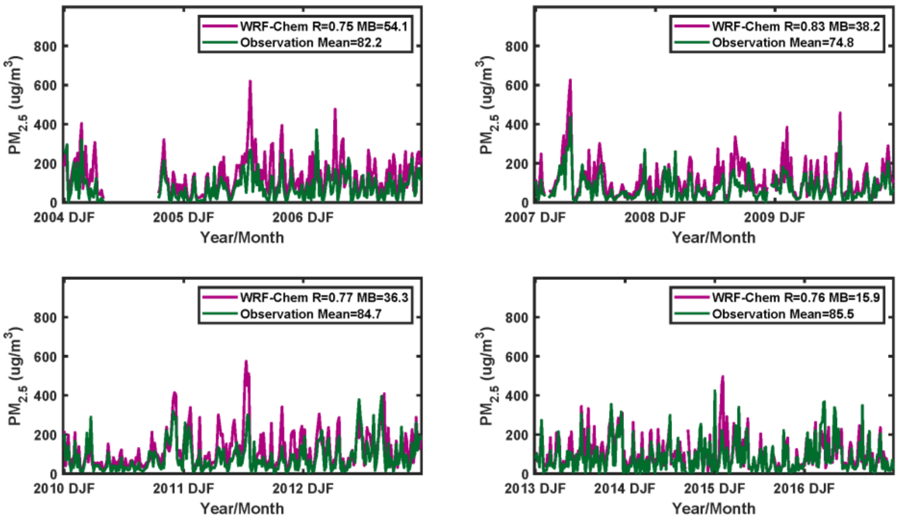

Fig. 2. Simulated and observed temporal variations of daily mean $PM_{2.5}$ concentrations in Beijing, with correlation coefficient and mean bias.

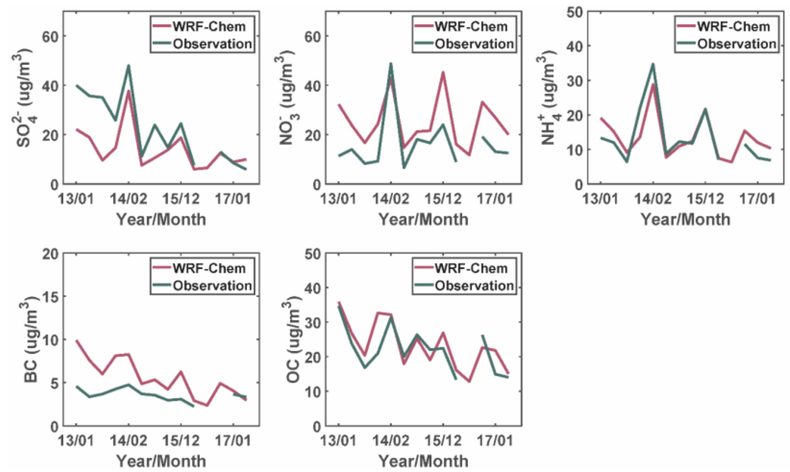

Fig. 3. Simulated and observed temporal variations of monthly mean concentrations of $PM_{2.5}$ chemical from 2013 to 2017.

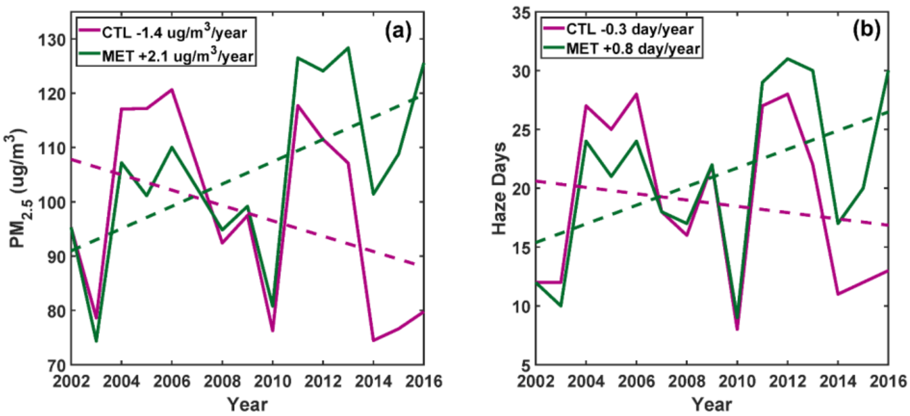

Fig. 4. Wintertime mean concentrations of $PM_{2.5}$ and number of haze days (defined with daily mean concentration above 150 μg/m^3) from the CTL and MET simulations.

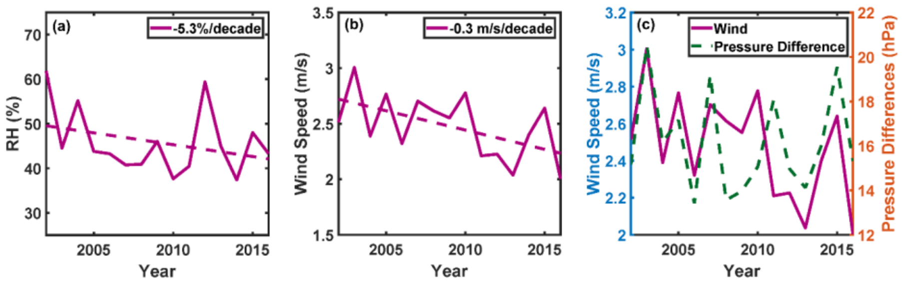

Fig. 5. Simulated winter mean RH, wind speeds in Beijing with declining rates, and pressure difference indicating the intensity of East Asia Winter Monsoon.

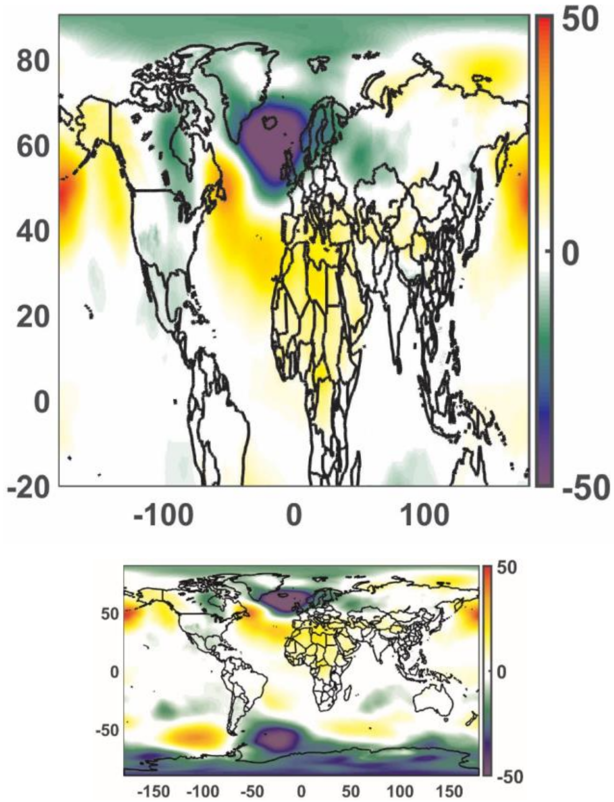

Fig. 6. Trends of winter sea level pressure during 2002-2016 period (unit: Pa/year).