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Sensitivity of $PM_{2.5}$ to NO_x emissions and meteorology in North China based on observations

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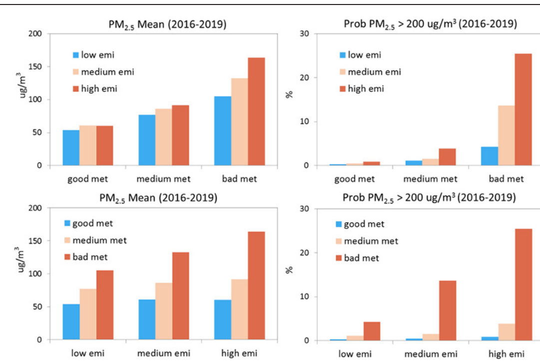
HIGHLIGHTS

- Daily $PM_{2.5}$ decreases from bad to good meteorological conditions in all North China cities in spite of their emission levels.
- Significant decrease in daily $PM_{2.5}$ is observed in bad meteorological conditions between high- and low-emission cities.
- During 2017-2019, the probability of high $PM_{2.5}$ decreased by 16% in high-emission cities in bad meteorological conditions.
- More attention still should be paid to high-emission cities in the formulation of emission regulation policies.

GRAPHICAL ABSTRACT



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ABSTRACT

This study examines the sensitivity of daily $PM_{2.5}$ to NO_x emissions and meteorology using in situ observations from main cities of North China (NC). NC cities are divided into low-, medium-, and high-emission groups by the ranking of their 4-year mean NO_2. For each emission group, daily NO_2 levels are used to divide the days into good-, medium-, and bad-meteorological conditions. Regardless of their emission levels, all cities reveal significant decreases (96%-172%) in daily $PM_{2.5}$ levels from bad to good meteorological conditions. The largest difference in $PM_{2.5}$ concentrations between the emissions groups is found under bad meteorological conditions, with 56% higher $PM_{2.5}$ in high-emission cities than low-emission cities, indicating $PM_{2.5}$ under bad meteorological conditions has the largest sensitivity to emissions. The high-emission, bad-meteorology group saw a 24% decrease in mean daily $PM_{2.5}$ levels from 2017, a high-emission year, to 2019, a low-emission year. However, under good meteorological conditions, the high-emissions group shows an increase of 8.8 $\mu g/m^3$ in mean daily $PM_{2.5}$ from 2017 to 2019 with a 2.6% increase in the possibility of high $PM_{2.5}$. These results suggest the current emission reduction measures are more effective in controlling $PM_{2.5}$ in high-emission cities under bad meteorological conditions than under other meteorological conditions.

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1. Introduction

Due to the rapid growth in economy and energy consumption, emissions of particulate matters and their precursors have increased significantly since the 2000s till recently, leading to a growing number of wintertime haze days (Chen and Wang, 2015; Li et al., 2016). Air pollution over North China (NC) has received massive attention, especially after January 2013 when persistent and severe haze events were first recorded by the national monitoring network newly established then (Yang et al., 2013; He et al., 2014; Wang et al., 2014; Zhang et al., 2014; An et al., 2019). High loadings of fine particulate matters ($PM_{2.5}$) during haze events pose great threats to human health by causing cardiovascular and respiratory diseases (von Klot et al., 2005; Dominici et al., 2006; Lipsett et al., 2011; Thurston et al., 2016; Gao et al., 2018; Yuan et al., 2019). These aerosols influence significantly also on regional weather and global climate change (Rotstayn and Lohmann, 2002; Chung et al., 2005; Qian et al., 2006, 2007; Choudhury et al., 2019; Kang et al., 2019).

In response to severe $PM_{2.5}$ pollution over NC, the Chinese government implemented various emission reduction measures during the 11th (2006–2010) and 12th (2011–2015) five-year plan, as well as the Air Pollution Prevention and Control Action Plan in 2013. As a result of these policies, SO_2 concentrations were found to start decreasing after around 2007 (Nickolay et al., 2016), while NO_x and $PM_{2.5}$ were observed to decrease after 2013 (Nickolay et al., 2016; Ronald et al., 2017; Li et al., 2017; Jia et al., 2018; Lin et al., 2019). Such persistent, longer-term decline in air pollution levels in China is mostly likely to be caused by the reduction in emissions, whereas meteorology such as large-scale circulation patterns and local conditions would play a more important role in modulating the interannual to daily variations (e.g. Jia et al., 2015; W. Cai et al., 2017; Zou et al., 2017). On the interannual time scale, the variability of NC winter haze is reported to be related to the strength of East Asia winter monsoon, the position of the Siberian High, El Nino, and Arctic Oscillation (Gao and Li, 2015; Jia et al., 2015; Zhang et al., 2019). During the haze period, weakened northerly winds, increased relative humidity, suppressed mixing layer are conducive to high $PM_{2.5}$ concentrations (Wang et al., 2014; W. Cai et al., 2017; S. Cai et al., 2017; Huang et al., 2014).

Many studies analyzed the roles that emission and meteorology play in the variations of wintertime $PM_{2.5}$ over NC in the recent years (S. Cai et al., 2017; Zhang et al., 2018; Zhai et al., 2019; Gao et al., 2020), especially when the anthropogenic emissions and $PM_{2.5}$ concentrations started to decrease. Sensitivity simulations by a chemical transport model, GEOS-Chem, reveal that only 10% decrease of NC winter $PM_{2.5}$ from 2012 to 2016 was due to emission reduction, while extremely polluted events were predominantly affected by meteorological conditions (Zhang et al., 2018). This study implies that emission control after 2013 may have little effects on the number of extremely polluted days. Gao et al. (2020) simulated $PM_{2.5}$ concentrations in Beijing over the winters of 2002–2016 using WRF-Chem and found emission control measures led to a 21% decrease in mean mass concentrations of $PM_{2.5}$ in Beijing from 2011 to 2016. S. Cai et al. (2017) reported that a 28.3% decrease of $PM_{2.5}$ over Beijing-Tianjin-Hebei region during 2012–2017 was owing to emission control by WRF-Chem simulations. The aforementioned studies mainly used atmospheric chemistry transport models to examine the response of NC winter $PM_{2.5}$ to emissions vs. meteorology, and the discrepancies between the results may lie in different emission inventories applied in the model simulations as well as different mechanisms of aerosol formation in models. For example, in both haze and clean periods, GEOS-Chem over-estimated the fraction of secondary inorganic aerosols (Wang et al., 2014), and this bias can cause the model to over-estimate the sensitivity of $PM_{2.5}$ to variability in meteorology.

To avoid errors introduced by models, in this study we use in-situ observations to examine the sensitivity of $PM_{2.5}$ to meteorology and NO_x emissions changes over NC during 2016–2019. Specifically, we use long-term mean in-situ NO_2 concentrations as a proxy of NO_x emissions and use the day-to-day variations in NO_2 concentrations to define the effect of meteorology variability on primary pollutants.

Observations of NO_2 are a good indicator to the geographical locations of air pollution because the residence time of NO_2 in the lower atmosphere is relatively short and thus NO_2 is concentrated near its sources (Richter, 2009). In the top-down inventories, satellite measured NO_2 are often applied to quantify NO_x emissions (Beirle et al., 2004; Beirle et al., 2011; Liu et al., 2016). The paper is organized as follows. Section 2 describes the data used in this study and the methods applied to define meteorological and emission conditions. In Section 3, we first analyze the distribution of daily $PM_{2.5}$ and the possibility of extremely high $PM_{2.5}$ (> 200 μg/m^3) conditions in different meteorological and emission conditions. Then we compare the change of $PM_{2.5}$ between 2017 and 2019 to study the response of $PM_{2.5}$ to NO_x emission control in different conditions. The sensitivity of the results to different assumptions is presented in Section 4. The conclusions are given in Section 5.

2. Data and method

2.1. In-situ NO_2 and $PM_{2.5}$

The in-situ observations of daily NO_2 and $PM_{2.5}$ are from the Ministry of Ecology and Environment of the People's Republic of China and can be obtained at http://beijingair.sinaapp.com/ from May 2014 to the present. Since in 2014, observations in only 190 main cities in China are available, the study period is defined as the winters of 2016–2019, which includes 367 main cities. In this study, the winter indicates the months of December–January. For instance, the winter during 2016 refers to December 2015 through February 2016, and the other winters are defined the same way. Fig. 1 illustrates the distribution of the 4-year mean winter $PM_{2.5}$ and NO_2 in 367 cities. The NC region is delineated by longitude from 110° E to 120° E, and by latitude from 30° N to 40° N, and 97 cities with observations are in NC. Despite the decline in concentrations of NO_2 and $PM_{2.5}$ after the implementation of the Air Pollution Prevention and Control Action Plan in 2013 (Nickolay et al., 2016; Ronald et al., 2017; Li et al., 2017; Jia et al., 2018), mean concentrations of NO_2 and $PM_{2.5}$ exhibit still highest over NC (Fig. 1).

2.2. Definition of meteorological and emission conditions

Since the current study focuses only on pollution over NC, which is relatively a small region compared to the spatial scale of East Asian winter monsoon in winter (Chang and Lu, 2012), the influence of large-scale circulation patterns over NC should be spatially homogeneous. Moreover, the lifetime of near surface NO_2 is few hours (Richter, 2009), so measurements of NO_2 can be good indicators of the geographical locations of air pollution. Thus, we assume the spatial variability of the multi-winter mean NO_2 concentrations among the cities is driven primarily by emissions.

Fig. 2 depicts the cumulative frequency of 4-year mean winter NO_2 in 97 NC cities, and the 33th and 67th percentiles of winter mean NO_2 are 43.5 μg/m^3 and 51.0 μg/m^3 respectively. According to this frequency distribution, the 97 NC cities are divided evenly into three emission groups: 32 low-emission cities with 4-year winter mean NO_2 lower than 43.5 μg/m^3, 33 medium-emission cities with the mean NO_2 between 43.5 μg/m^3 and 51.0 μg/m^3, and 33 high-emission cities with the mean NO_2 higher than 51.0 μg/m^3. This division of sites avoids the errors introduced by the sample size to some extent. The locations of the cities in the three emission groups are shown in Fig. 3. Mean NO_2 in the three groups are 38.6 μg/m^3, 47.2 μg/m^3 and 56.5 μg/m^3, respectively, and the differences in the mean value between the neighboring groups are much higher than the std. within each group (Fig. 3). Therefore, the differences in the emission level among the cities in the same group are not considered in the following analysis.

While the multi-year mean NO_2 is a good indicator of emission levels, there are substantial day-to-day variations of NO_2 concentrations

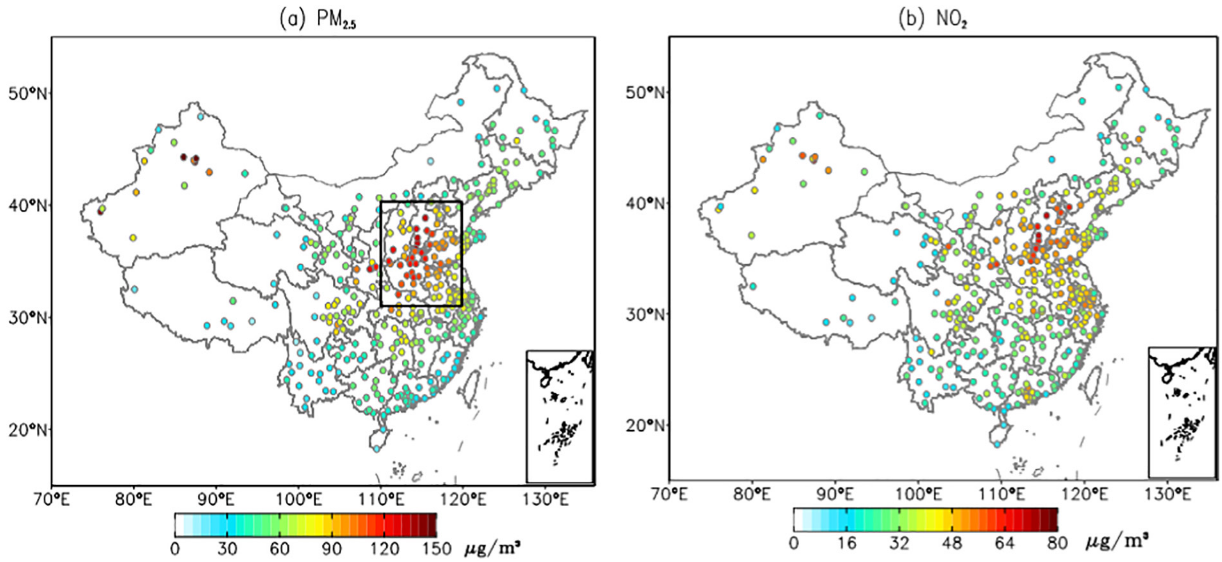

Fig. 1. 4-year mean winter in-situ (a) $PM_{2.5}$ and (b) NO_2 from 2016 to 2019. The black box outlines the NC region.

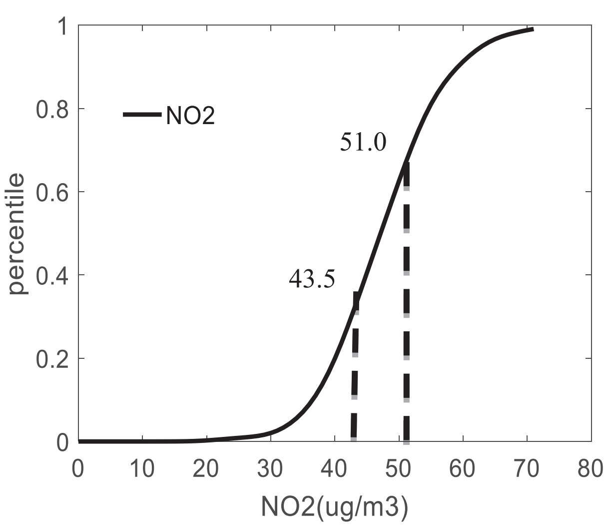

Fig. 2. Cumulative frequency of 4-year mean winter (2016–2019) NO_2 in 97 cities of NC.

in a city that are mainly influenced by meteorology such as wind speed (Song et al., 2011), planetary boundary layer height (Harkey et al., 2015), relative humidity (Song et al., 2011), and etc. Fig. 4 shows the probability distribution function of daily NO_2 for each emission group. Since the weekly pattern of traffic flows might also affect daily variations of NO_2 (Shutters and Balling, 2006), only NO_2 concentrations during weekdays are used in the Figure and the following analysis. The mean daily NO_2 concentration on weekdays of each emission group is 37.1 μg/m^3, 47.7 μg/m^3 and 60.6 μg/m^3, and the std. is 13.5 μg/m^3, 18.5 μg/m^3 and 25.1 μg/m^3, respectively (Fig. 4). Since emissions are not expected to drive such a large variability in NO_2, we assume the day-to-day variations of NO_2 concentration on the weekdays in the same emission group are mainly due to local meteorology. Thus, the day-to-day variability in NO_2 is considered as a proxy for the variability in the effect of meteorology on primary pollutants. Then good, medium, and bad meteorological conditions are defined using daily NO_2 concentrations in each emission group. Similar to the previous step, the 33th and 67th percentiles of daily NO_2 in each group are calculated (Table 1), and the days with NO_2 lower than the 33th percentile, between the 33th and 67th percentile, higher than the 67th percentile are defined as good, medium, and bad meteorology days in each group. Combining both emission and meteorological conditions, the daily records of NO_2 can be divided evenly into nine groups, high/median/low emission by good/medium/bad meteorology. The daily observations of NO_2 concentration in 2017, 2018, and 2019 are also divided following the same procedure, and data in the nine groups during 2016–2019 are combined in the following analysis.

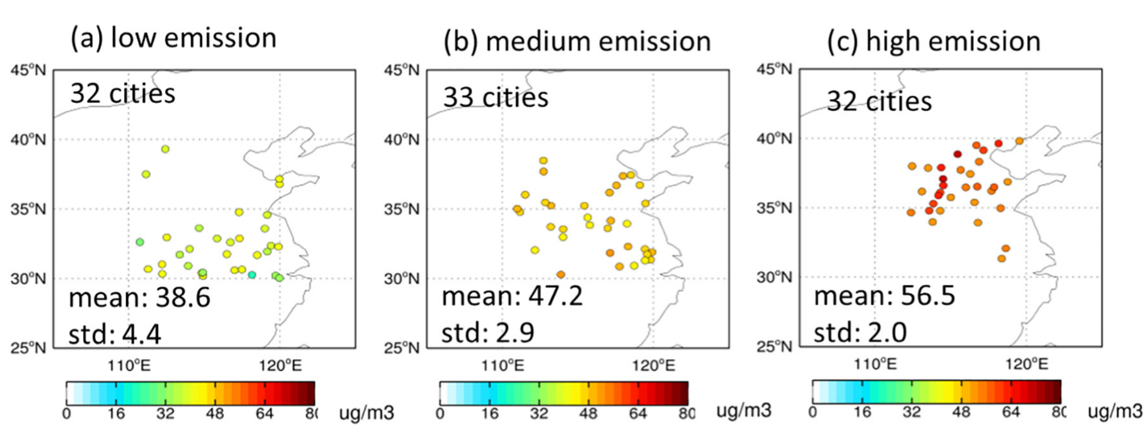

Fig. 3. 4-year mean winter NO_2 concentrations in cities in the three emission groups, and the mean and standard deviation (std) in each emission group (unit: μg/m^3).

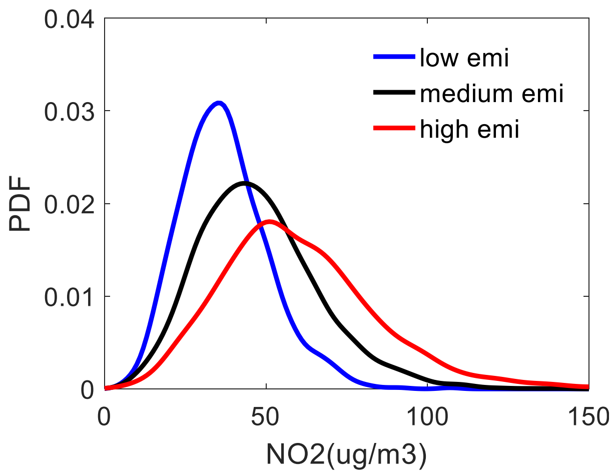

Fig. 4. Probability distribution function (PDF) of daily NO_2 in 2016 in three emission groups.

Table 1
The 33th and 67th percentiles of daily NO_2 in three emission groups (unit: μg/m^3).

	Low emission	Medium emission	High emission
33th	31	38	48
67th	42	54	68

3. Results

Daily $PM_{2.5}$ records in the nine groups during 2016–2019 are analyzed in this section to assess the sensitivity of $PM_{2.5}$ to meteorology and NO_x emissions. The PDFs of daily $PM_{2.5}$ in low-, medium-, and high-emission cities under various meteorological conditions are displayed in Fig. 5. In all the three emission groups, the PDF curves shift to higher $PM_{2.5}$ concentrations from good to bad meteorology, indicating the mean daily $PM_{2.5}$ and the probability of extremely high daily $PM_{2.5}$ both increase when the meteorology gets worse. To characterize the distribution of $PM_{2.5}$ in different groups quantitatively, three metrics of $PM_{2.5}$ including the mean and mode of daily $PM_{2.5}$, together with the probability of extremely high daily $PM_{2.5}$ (> 200 μg/m^3) are calculated (Tables S1-S3, Figs. 6, and S1).

Under good and medium meteorology, mean daily $PM_{2.5}$ and $PM_{2.5}$ mode do not show significant increase from low-emission to high-emission group (Fig. 6). In good and medium meteorological conditions, the large capacity of air pollution in the boundary layer makes $PM_{2.5}$ less sensitive to NO_x emissions due to the enhanced vertical and horizontal mixing. However, under bad meteorology, mean daily $PM_{2.5}$ increases significantly from 105 μg/m^3 to 164 μg/m^3 (56%). Similarly, the most significant increase of $PM_{2.5}$ mode and probability of high $PM_{2.5}$ are also observed under bad meteorology from low-emission group to high-emission group. Especially, the possibility that daily $PM_{2.5}$ exceeding 200 μg/m^3 under bad meteorological conditions increases from 4% in low-emission cities to 26% in high-emission cities. Therefore, under bad meteorological conditions, daily $PM_{2.5}$ records show the largest sensitivity to the changes of NO_x emissions, suggesting measurements to control NO_x emissions under bad meteorological conditions would be most effective to reduce mean $PM_{2.5}$ and the occurrence of extremely high $PM_{2.5}$ events.

This study also compares the changes of daily $PM_{2.5}$ to meteorology under different emissions (Fig. S1). Unlike in Fig. 6 that $PM_{2.5}$ is most sensitive to emissions under bad meteorology, the three metrics of $PM_{2.5}$ increase from good to bad meteorology in all emission groups. The largest enhancements are observed in high-emission group. The mean $PM_{2.5}$ increases by 2 times and the probability of high $PM_{2.5}$ increases by 27 times from good to bad meteorological conditions. Thus, daily $PM_{2.5}$ is sensitive to meteorology in all emission groups, and the largest sensitivity occurs in high-emission group.

After the publication and implementation of emission reduction measures, Chinese anthropogenic emissions have been decreased significantly, and evidence has been drawn from both in-situ and satellite data that NO_2 concentrations have shown large decrease since 2013 (Nickolay et al., 2016; Li et al., 2017). As illustrated in Fig. S2, wintertime NO_2 over NC decreased by 7.0 μg/m^3 (14%) from 2017 to 2019. To assess the influence of NO_x emission reduction on $PM_{2.5}$ in different groups, this study compares the mean daily $PM_{2.5}$ and the probability of extremely high $PM_{2.5}$ conditions between 2017, the year with the highest NO_2 concentrations over NC during 2016–2019, and 2019, the year with the lowest NO_2 concentrations (Tables S4-S5). Fig. 7 presents the changes of $PM_{2.5}$ metrics between 2017 and 2019 under good and bad meteorology in low- and high-emission groups. From 2017 to 2019, mean daily $PM_{2.5}$ and the possibility of extremely high $PM_{2.5}$ conditions display significant decrease in high-emission cities under bad meteorology, indicating the current emission reduction measures are successful in controlling $PM_{2.5}$ concentrations in high-emission cities under bad meteorology. However, under good meteorological conditions, the mean daily $PM_{2.5}$ increased by 3.4 μg/m^3 and the possibility of high $PM_{2.5}$ conditions increased by 1.5% in high-emission cities. Thus, the extremely high $PM_{2.5}$ conditions may occur more frequently under good meteorology in high-emission cities according to the current emission reduction policy. To test the robustness of these results, we analyze the uncertainties to different assumptions of meteorology in Section 4.

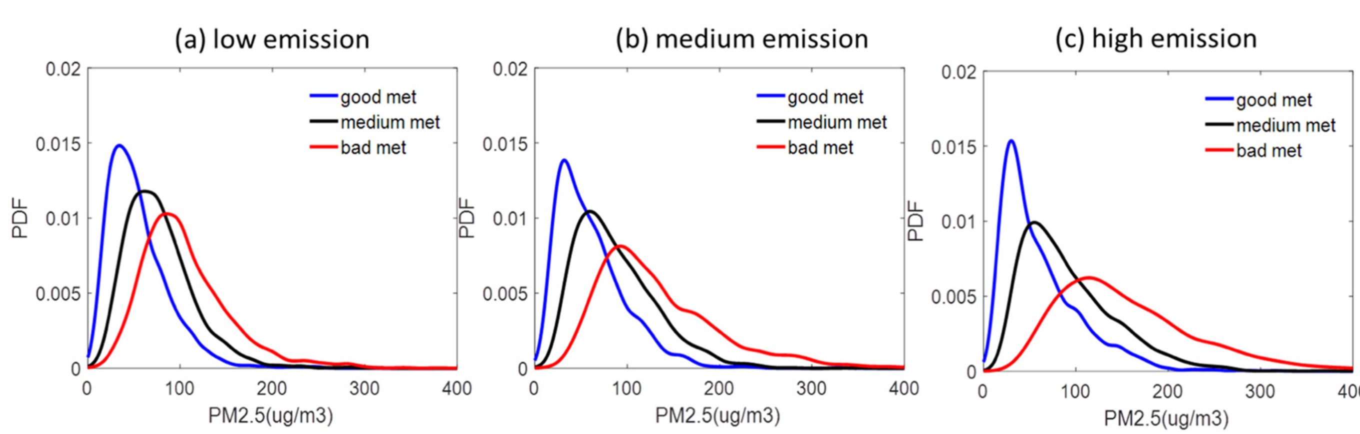

Fig. 5. PDF of daily $PM_{2.5}$ in (a) low-, (b) medium- and (c) high-emission groups under different meteorological conditions.

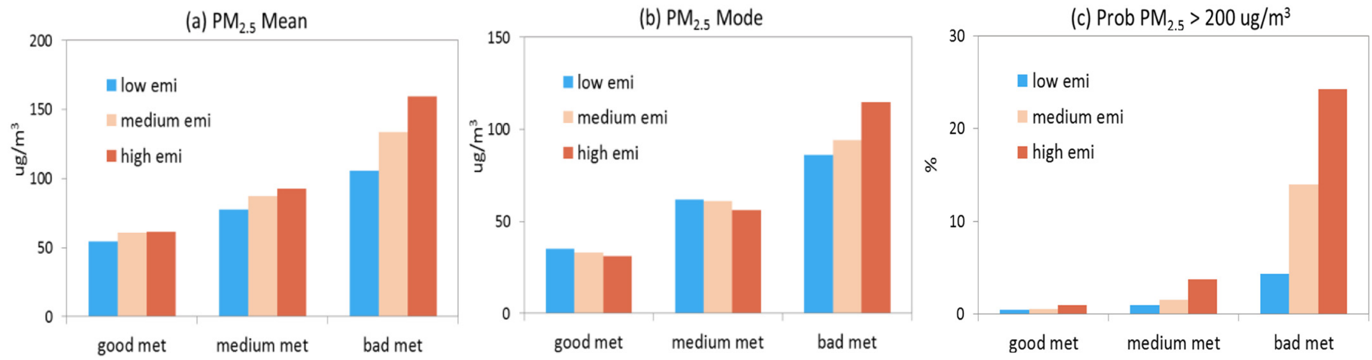

Fig. 6. Changes of (a) $PM_{2.5}$ mean, (b) $PM_{2.5}$ mode and (c) probability of $PM_{2.5} > 200\ \mu g/m^3$ under three meteorological conditions in different emission groups.

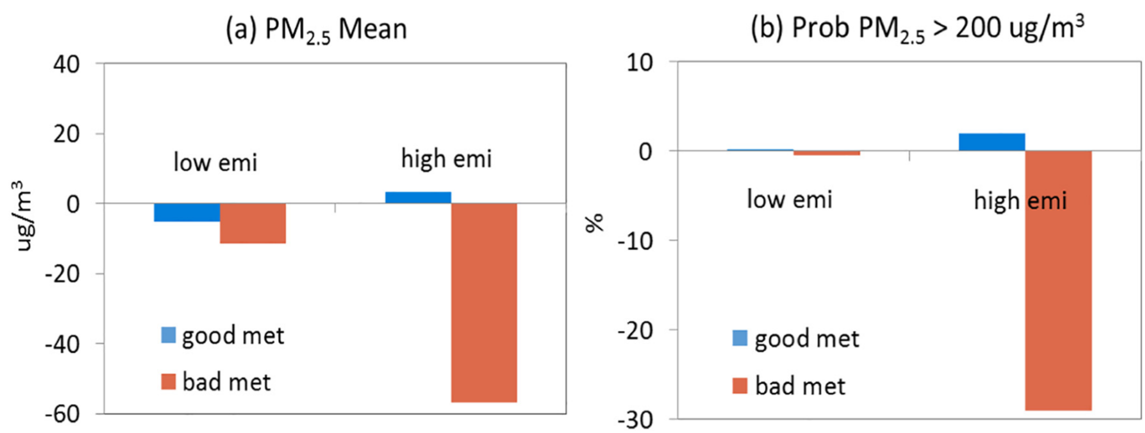

Fig. 7. Changes of (a) $PM_{2.5}$ mean, and (b) probability of $PM_{2.5} > 200\ \mu g/m^3$ in different groups (2019 minus 2017).

4. Uncertainty

In Section 2, the meteorological conditions are defined using the 33th and 67th percentiles of daily NO_2 in each emission group, and there are around 660 samples in each emission group under each meteorology. To avoid the influence of data noise on the results and examine the sensitivity to the definition of meteorological conditions, we expand the screening of good and bad meteorological conditions in 2017 and 2019 to the 40th and 60th percentiles of daily NO_2 in each emission group. Then there are around 780 samples in each emission level and meteorological condition, and we randomly select 85% of the samples (around 660) to do the comparison between 2017 and 2019. The random selection of the samples is repeated 50 times for 2017 and 2019 respectively, and the results are illustrated in Figs. 8–9.

The results of the random selections reveal that in low-emission cities, mean daily $PM_{2.5}$ and the probability of high $PM_{2.5}$ decrease from 2017 to 2019 under both good and bad meteorology, and more distinct decrease is observed under bad meteorology. In low-emission cities, the

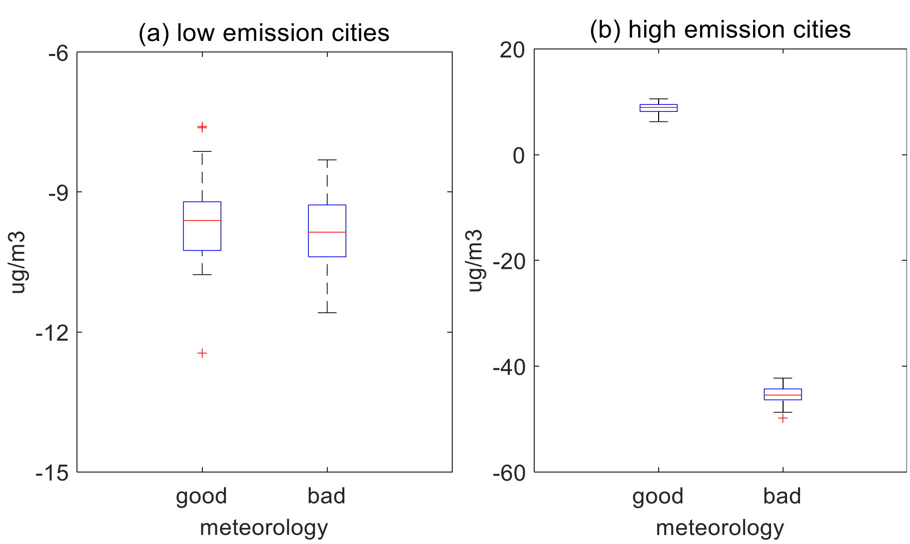

Fig. 8. Distribution of the changes in mean daily $PM_{2.5}$ in different groups (2019 minus 2017).

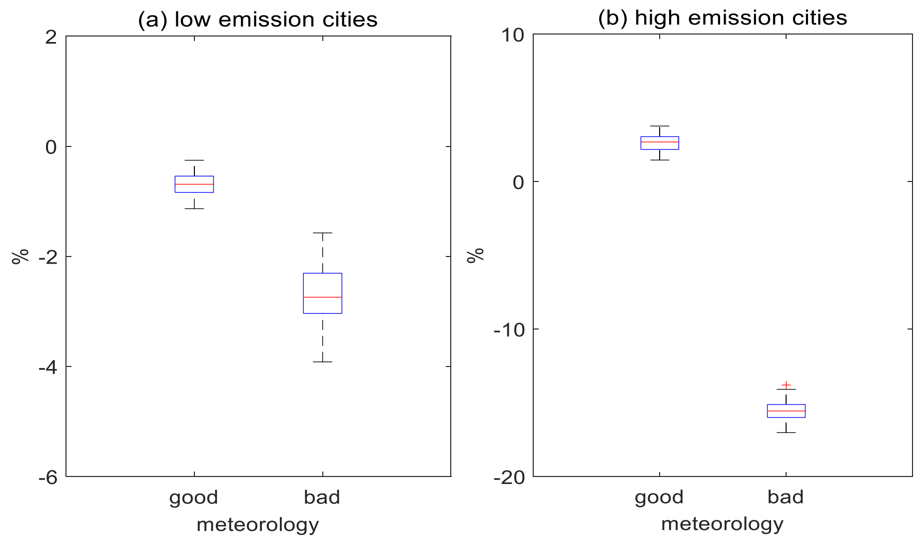

Fig. 9. Distribution of the changes in probability of $PM_{2.5}$ > 200 μg/m^3 in different groups (2019 minus 2017).

emission reduction measures lead to 10.0 μg/m^3 (7.3 μg/m^3–12.6 μg/m^3) decrease in mean $PM_{2.5}$ and 2.9% (2.0%–3.8%) decrease in the occurrence of extremely high $PM_{2.5}$ events on average from 2017 to 2019 under bad meteorology. In high-emission cities, mean $PM_{2.5}$ and the probability of high $PM_{2.5}$ decrease by 45.7 μg/m^3 (42.6 μg/m^3–50.2 μg/m^3) and 15.9% (14.1%–17.6%) respectively under bad meteorology. However, under good meteorology, mean daily $PM_{2.5}$ increases by 8.8 μg/m^3 (7.0 μg/m^3–11.6 μg/m^3), and the probability of high $PM_{2.5}$ increases by 2.6% (1.6%–3.9%) from 2017 to 2019 in high-emission cities. Xing et al. (2018) found that in January, Beijing-Tianjin-Hebei region is under VOC-limited condition, which will lead to a disbenefit of NO_x reduction for $PM_{2.5}$ by enhancing atmospheric oxidation ability and facilitating the formation of secondary aerosols. This may explain the increase of mean daily $PM_{2.5}$ and the probability of high $PM_{2.5}$ from 2017 to 2019 under good meteorology in high-emission cities.

5. Conclusion

In this study, we investigated the response of daily $PM_{2.5}$ to NO_x emissions in different cities and meteorological conditions using in-situ $PM_{2.5}$ and NO_2 data over NC. 97 cities in NC were divided into three emission groups according to their 4-year mean NO_2 concentrations. In each emission group, we defined good, medium and bad meteorological conditions according to daily NO_2 concentrations, and analyzed the daily $PM_{2.5}$ in different emission groups and meteorological conditions. In all the three emission groups, the rightward shifts of PDF curves of $PM_{2.5}$ with changes in meteorology indicate daily $PM_{2.5}$ is sensitive to meteorology under all emission levels, and the largest sensitivity to meteorology occurs in high-emission group. Mean daily $PM_{2.5}$ decreased by 172% in the high-emission group from bad to good meteorological conditions. However, daily $PM_{2.5}$ records only reveal significant sensitivity to emissions in bad meteorological conditions; mean $PM_{2.5}$ decrease by 59.1 μg/m^3 (56%), and the probability of high $PM_{2.5}$ decrease by 21% from high-emission group to low-emission group. This explains the temporary measures including shutting down high-emission factories and traffic restrictions under bad meteorology are effective in reducing daily $PM_{2.5}$.

Due to the implementation of emission reduction measures, NC wintertime NO_2 concentration has shown notable decrease, especially from 2017 to 2019. We then compared daily $PM_{2.5}$ in different groups of emission and meteorology to quantify the response of $PM_{2.5}$ to current polity. The mean daily $PM_{2.5}$ decreased by 10.0 μg/m^3 and 45.7 μg/m^3 in low-emission cities and high-emission cities respectively under bad meteorology, indicating the current polity is quite successful in controlling daily $PM_{2.5}$ in most cases. However, in cities with high NO_x emissions, mean $PM_{2.5}$ and the possibility of high $PM_{2.5}$ increase under good meteorology as consequences of the current emission reduction measures.

CRediT authorship contribution statement

Beixi Jia: Methodology, Data curation, Investigation, Writing - original draft. **Yuxuan Wang:** Methodology, Writing - review & editing, Supervision. **Chuanhui Wang:** Data curation. **Qianqian Zhang:** Writing - review & editing, Funding acquisition. **Meng Gao:** Writing - review & editing, Supervision, Funding acquisition. **Ken Kin Lam Yung:** Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.142275.

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