$PM_{2.5}$ reductions in Chinese cities from 2013 to 2019 remain significant despite the inflating effects of meteorological conditions

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Summary

Air pollution is a major environmental issue in China and imposes severe health burdens on Chinese citizens. Consequently, China has deployed a series of control measures to mitigate fine particulate matter ($PM_{2.5}$). However, the extent to which these measures have been effective is obscured due to the existence of confounding meteorological effects. Here we use a newly developed reduced-form model – that can address emission-driven $PM_{2.5}$ trends and control for meteorological effects – to examine the level of $PM_{2.5}$ reduction across 367 cities since the introduction of the Air Pollution Prevention and Control Action Plan (the Plan) in 2013. Our findings show that, on average, the national annual mean level of $PM_{2.5}$ decreased by 34% between 2013-2019 after removing meteorological effects, about 10% less than the reduction level officially observed. Despite this difference, assuming current control efforts continue through 2035, the long-term air quality target of 35 μg/m^3 as determined by the recently updated Plan will be met.

Keywords

$PM_{2.5}$, emission, meteorology, air pollution mitigation, reduced-form model, future projection

Introduction

Air pollution is a global environmental issue of great concern[1]. Exposure to air pollutants was estimated to lead to more than one million premature deaths annually, thereby significantly contributing to the overall disease burden in China[2]. In response to this concern, the Air Pollution Prevention and Control Action Plan (the Plan hereafter) was promulgated in late 2013, and the Plan was followed by a series of specific pollution-control measures by the central and local governments[3]. These actions aimed to reduce the annual mean $PM_{2.5}$ (particulate matter with aerodynamic diameter less than or equal to 2.5 μm) on either a short-term or a long-term basis. For example, the Plan targets a 10% reduction in annual $PM_{2.5}$ from 2012 to 2017. A long-term goal of reaching the national standard of 35 $\mu g/m^3$ by 2035 was also proposed[4]. To meet the short-term goals, specific emission reduction schemes were developed and implemented by all provinces[5].

It is of interest to policymakers, the public, and scientists whether the actions were and will be sufficiently effective to achieve the claimed goals[6]. Although routine monitoring data were available and reductions in annual mean $PM_{2.5}$ concentrations were reported by all cities by 2019, direct comparisons between two consecutive years can lead to misunderstandings because air quality is strongly affected by meteorological conditions. For example, the unfavorable meteorological conditions in 2013 contributed significantly to the abnormally high $PM_{2.5}$ concentrations during that winter[7,8]. Simply judging the controlling effects by comparing the $PM_{2.5}$ in 2013 with that in the subsequent years would thereby overestimate the policy efficiency due to the improvement of meteorological conditions. Several recent studies have tried to quantify the overall meteorological effects mostly based on a linear composition simulation approach. The basic idea is to simulate the differences in $PM_{2.5}$ concentrations driven by fixed baseline emissions and varying meteorological conditions. **Table S1** summarizes the results from previous studies, showing a large variation in the estimated meteorological effects (see **Note S1**). Some studies even reported reversed meteorological effects using the same input data but different baseline emissions[9-11]. To date, there is no consensus on the meteorological effects on $PM_{2.5}$, leading to different evaluations of emission reduction. Conceptually, the linear composition simulation treated emission and meteorological effects in the same way, ignoring the fact that these two factors have very different effects on atmospheric $PM_{2.5}$. Treating the meteorological effects in the same way as emissions can only evaluate the relative meteorological status, which can be highly varied due to the strong fluctuation in meteorological conditions (see **Note S1**). Compared with previous studies that quantified the meteorological effects as fixed percentages, the frequency might provide a scientifically better solution in view of the fluctuating features of meteorological conditions. The idea of frequency is extensively adopted in meteorology and hydrology to describe the severity of many occasional phenomena such as floods, earthquakes, and extreme weather[12]. The corresponding frequency is one of the major concerns when formulating measurements to mitigate the adverse influences of these events[13]. Similarly, knowing the frequency of meteorological effects would also enable us to understand the severity of the problem, leading us to formulate more effective policies to control air pollution. Recently, a novel reduced-form model was developed to distinguish the influences of emissions and meteorology[14]. By using the model, emissions-associated $PM_{2.5}$ concentrations and meteorology-dependent variations in $PM_{2.5}$ could be quantified individually by regression and probabilistic models[14]. This method provides a unique tool for evaluating mitigation measures quantitatively without confounding meteorological effects.

Here, we present the results of a series of evaluations on the effectiveness of implementing the Plan with a special focus on the following research questions: 1) do the current (2013-2019) emission reductions reach the preclaimed or postreported values? 2) does the $PM_{2.5}$ concentration reduction achieve the targets of the Plan with meteorological confounding effects excluded? and 3) will the long-term goal of 35 μg/m^3 in 2035 be achieved if the current efforts continue generally at the same level? A total of 367 cities that regularly report routine monitoring data were evaluated. The contributions of emissions to $PM_{2.5}$ in current and future periods were characterized using a set of reduced-form models, where the meteorological effects were quantified using probabilistic models. A time-for-space approach was adopted to predict the decreasing pace of emission reduction in the future regarding the increasing difficulty of emission mitigation as the $PM_{2.5}$ concentration continues to decline. The meteorological effects in this study denoted only the overall impacts, and the influences of single meteorological parameters were out of our scope and are not discussed. The detailed methodology is provided in the **Experimental Procedures** section.

We show that the national annual mean $PM_{2.5}$ decreased by 34% from 2013 to 2019 through the exclusion of meteorological effects, which was smaller than the result of 44% taken from observations. The difference is largely due to the poor dispersion conditions in 2013. Large variations were found among cities. Specifically, 91% of the cities showed $PM_{2.5}$ reductions in the range of 0% to 79%, whereas 4% of the cities showed $PM_{2.5}$ increasing by more than 10%. The mitigation effort and emission reduction rate for individual cities was found dependent on both the initial $PM_{2.5}$ pollution level in 2013 and socioeconomic development. Future prediction by assuming that the current effort will continue by 2035 (in terms of political willingness and financial support) show that the national annual mean $PM_{2.5}$ concentration will further decrease by 36% to 24.2±6.6 μg/m^3, and 95% of cities will meet the 35 μg/m^3 national standard.

Results and Discussion

National annual mean of $PM_{2.5}$ from 2013 to 2019

Based on the routine monitoring data from the 367 cities, annual mean $PM_{2.5}$ concentrations with standard deviations from 2013 to 2019 were determined and are shown in **Fig. S1**. Because the sample sizes in the first two years (74 and 190) were less than the sample sizes in the other years (367)[15], the annual mean concentrations cannot be compared directly. In fact, the cities that started their monitoring schemes earlier were generally more populated and polluted[16]. Taking 2015 as an example, the annual mean $PM_{2.5}$ concentrations of the 74 (54.5±19.4 μg/m^3) and 116 cities (53.8±17.8 μg/m^3) that started monitoring programs in 2013 and 2014, respectively, were 20% and 19% higher than the annual mean $PM_{2.5}$ concentrations of the 177 cities that started monitoring in 2015 (45.2±17.2 μg/m^3), respectively. To correct the bias, the annual mean $PM_{2.5}$ concentrations in 2013 and 2014 were adjusted by estimating the missing data using linear regressions based on the available observations and satellite-inversion data (see **Experimental Procedures**) (**Fig. S1**). Despite the high standard deviations due to order-of-magnitude differences among cities[17], the observed annual mean $PM_{2.5}$ concentrations of all cities show a steady decreasing trend from 65.7±27.3 μg/m^3 in 2013 to 36.8±12.0 μg/m^3 in 2019, indicating that the Plan has worked well on a national scale, which has been reported by a number of studies[9,18]. Moreover, the annual decreasing rate has slowed down gradually from 14% to 6%, which is likely because mitigation actions always began with easier tasks. For example, the main efforts from 2013 to 2015 included the phasing out of high-emission processes such as small-scale pig iron and cement manufacturing, which contributed significantly to emissions and were relatively easy to shut down[19]. Reducing emissions from large-scale industry is much more difficult, and the mitigation costs often increase exponentially as the emission strength decreases[20]. Consequently, although the Plan has made promising progress to this stage, considerable work will be required to achieve the long-term national goal of 35 μg/m^3 by 2035[21,22].

Based on the reported emission inventories and emission reduction rates, annual mean $PM_{2.5}$

concentrations were calculated using the reduced-form model[14] to exclude confounding meteorological effects (see **Experimental Procedures**). The results are shown as the solid line in **Fig. 1**, which represents the multiyear trend of annual mean $PM_{2.5}$ concentrations with the average meteorological conditions[14]. In addition, 95% confidence intervals of the model uncertainty (yellow shaded area) and meteorology associated variation (50% and 95% variation intervals, dark and light blue shaded areas) derived from a probabilistic model are also shown. By using the reduced-form model inversely constrained by observations from 2015 to 2019, the best estimates of actual emission reductions for individual cities were quantified (see **Experimental Procedures**). SO_2 and NOx emissions were found to have been reduced by 53±31% and 33±26% nationwide from 2013 to 2019, equivalent to 9±5% and 6±4% annual reductions, respectively. This finding means that the pledged emission reduction target was 115% and 37% overachieved for SO_2 and NOx, respectively[5,23]. However, the post-reported emission reduction rates by most provinces, which were higher than those pledged, were too optimistic, and our estimated SO_2 and NOx emission reductions were 82±53% and 74±57% of those reported.

Consistent with the observations, the model-calculated annual mean $PM_{2.5}$ concentrations also show a general decreasing trend. On average, the annual mean $PM_{2.5}$ concentrations decreased 5.7% each year compared with 7.3% for the observations. Such a difference is due largely to the high observed decreasing rate of 14% from 2013 to 2014, which is partially caused by extremely unfavorable dispersion conditions in 2013[7,8]. By excluding data from 2013, the average annual reduction rates would be 5.4% from the model calculation, which is very close to the 5.8% from observations. Obviously, the observed annual mean $PM_{2.5}$ concentration in 2013 was an outlier from the general trend. Because the observations were affected by confounding effects of meteorological conditions, which were removed by the reduced-form modeling[14], the differences between the observed and modeled results are meteorology dependent. For the seven years studied, most data points fell within the 50% uncertainty interval with a single exception of 2013. The observed mean $PM_{2.5}$ concentration in 2013 fell near the edge of the upper limit of the 95% uncertainty interval, suggesting that severe pollution in 2013 was a once-in-a-two-decade phenomenon. In fact, unfavorable weather conditions in middle and eastern China in 2013 winter have been well documented in previous literature[24]. The meteorological conditions in the winter of 2013 were dominated by suppressed near-surface wind (-0.18 m/s, -5.7%), shallow boundary layer heights (-45 m, -10%), high temperature (+0.07 ℃, +10%), and high relative humidity (+2.1%, +1.3%) due to the weakened East Asian winter monsoon[25]. However, the observations in 2015 and 2018 were lower than the model calculations due to the better dispersion led by strong meridional circulation[26]. In 2018, for example, the stronger meridional circulation brought stronger wind speeds (+0.2 m/s or +6.8%) and more frequent cold-air events (-0.82 ℃ or -52%), which stimulated the dispersion of $PM_{2.5}$ and suppressed the formation of secondary $PM_{2.5}$[27].

Meteorological influences could be characterized by the previously developed probabilistic models[14], which resulted in distributions that deviated from the estimated mean values. Taking 2018 as an example, the model-simulated national annual mean $PM_{2.5}$ concentration was 40.8 μg/m^3 given that there was no meteorological influence. The potential meteorological influence is shown in **Fig. 2** as a normal distribution. Although 40.8 μg/m^3 is the best estimation, there would be a 50% chance that the concentration varies from 38.6 μg/m^3 to 43.0 μg/m^3 and a 95% chance that the concentration varies from 34.4 μg/m^3 to 47.2 μg/m^3 under various meteorological conditions. The observed value for that year was 39.2 μg/m^3, which is equivalent to a less than 62% probability.

Differences among the cities

As discussed above, significant reductions in both emissions and ambient $PM_{2.5}$ concentrations were demonstrated on a national scale. Although the majority of provincial and local governments have developed their own mitigation action plans in compliance with the national goal that is not differentiated among cities[5], the efforts and achievements have varied extensively across the country. By using the reduced-form model, the reduction of $PM_{2.5}$ concentration driven by emissions for the 367 individual cities from 2013 to 2019 was derived with the meteorological effects excluded. **Fig. S2** shows the frequency distribution of the reduction rates, which vary extensively from -42% to 79%. In line with the national average, there were general decreasing trends in the annual mean $PM_{2.5}$ concentrations of most cities. The mean $PM_{2.5}$ concentrations of 333 of the 367 cities in 2019 were lower than the mean $PM_{2.5}$ concentrations in 2013. The cities with greater decreases are often those with higher initial $PM_{2.5}$ concentrations, whereas those that decrease more slowly or even rebound are those with lower initial $PM_{2.5}$ concentrations. The dependence of the decreasing rates on the initial $PM_{2.5}$ concentrations is shown in **Fig. S3**. A positively significant correlation ($p = 7\times10^{-40}$) was revealed between the concentrations in 2013 and the reduction rates of individual cities. Such a correlation can be explained by the fact that the identified goal of 35 μg/m^3 in 2035 is the target of the national Plan[5], and the more polluted cities in 2013 have to devote more efforts to achieving the goal and therefore contributed more to the overall reduction. However, the cities with

$PM_{2.5}$ concentrations that were unchanged or increased during the past few years are mostly those situated in western China, which initially had relatively low $PM_{2.5}$ concentrations. The average concentration for the 34 cities with annual mean concentrations that increased since 2013 was 30±14 μg/m^3 in 2013, and most were close to the national standard of 35 μg/m^3 already and much lower than the other cities (60±23 μg/m^3). In practice, these cities are not important when considering a national strategy or local goals.

Spatial variations in the annual mean $PM_{2.5}$ concentration reduction rates are mapped in **Fig. 3**, and they show the reduction rates over the period from 2013 to 2019 (color shade scale) and initial annual mean concentrations in 2013 (proportional to the symbol size) for all cities. Several clusters with profound reductions are located in the North China Plain, Yangtze Plain, and Sichuan Basin. In general, these cities are among the most polluted and populated regions in China, confirming again that the initial pollution level is the key driving force. A similar map with the symbol colors as the quotients of the reduction rates divided by the initial concentrations (**Fig. 3b**) shows a different pattern. Although the cities in the North China Plain show the highest cumulative reduction in $PM_{2.5}$, cities in the Yangtze River Delta performed much better considering the corresponding pollution levels. Such disparity resulted mainly from different socioeconomic levels since more developed cities often had more ambitious goals and invested more to achieve the goal. For example, Shanghai planned to achieve the national goal of 35 μg/m^3 in 2022, 13 years ahead of national goal; thus, the government spending on environmental protection was over 3% of the total gross domestic product (GDP) for the past few years compared with the national average of 1.24% in 2016[28]. Reducing air pollution in North China still requires considerable work, even if the current control pace is definitely faster than the pace in most other cities around the world[21].

Because the inversely modeled concentration reductions are free of meteorological effects, the differences between the model calculation and observations were caused partially by spatial variations in meteorological conditions, which are averaged at the national level to a certain extent but stand out for individual cities. To illustrate the meteorological confounding effects on the annual mean $PM_{2.5}$ concentrations for individual cities, probabilistic functions developed in conjunction with the reduced-form model, were applied to calculate the 95% confidence intervals[14]. **Fig. S4** shows the frequency distribution of the meteorological influences presented as percentages of the concentration reductions. Not only the reduction rates but also the confidence intervals are correlated significantly with the initial concentrations in 2013 ($p < 0.05$). Specifically, the negative correlation between $PM_{2.5}$ concentration intervals suggested that less polluted cities are often more vulnerable to meteorological changes. The variations for individual cities are generally higher than the national average simply because the meteorological effects for individual cities can cancel each other out. Based on these results, it is not surprising to see that the annual mean $PM_{2.5}$ concentrations of some cities occasionally rebounded despite continuous mitigation efforts. On average, the rebound probability can be as high as 10.4±22.3%, and occasional rebounding has been reported[29].

Based on the observed $PM_{2.5}$ concentrations in all cities, emission reductions at the city level over the study period were derived inversely using reduced-form modeling (see **Experimental Procedures**). The emission reduction rates of the 367 cities varied extensively from -65% to 97%, indicating high variation among the cities. The frequency distributions of SO_2 and NOx emission reductions in these cities are shown in **Fig. S5**. The relatively fast emission reduction of SO_2 compared with NOx is primarily because the desulfurization effort started several years earlier than the denitration effort for power stations and industries in China[30]. The emission reduction at the city level was further compared with provincial reported data, as shown in **Fig. S6**. As discussed above, the reported emission reduction was overestimated for the national average, which also applied to the provincial data despite the significant correlation between the calculated and reported values ($p < 0.05$). The emission reduction rates in 25 of 31 provinces were overestimated. Again, most of the 38% of the cities with underestimated emission reduction rates were more polluted. In fact, the provincial reported data were estimated based on the top-down statistical system coordinated by the National Bureau of Statistics, which focused mainly on collecting data for large emitters; thus, the data were inevitably associated with large uncertainty[31]. The reduction rates derived in reverse based on a large volume of field monitoring data (hourly data for 1641 sites) with meteorological effects excluded are theoretically more reliable. Our results also suggested that accurate data on a finer spatial scale (e.g., county, town, etc.) are urgently needed, which also stimulated the promotion of the national pollution census campaign in China[32]. Similar to the $PM_{2.5}$ concentration reductions, a significantly positive correlation was also revealed between the emission reduction rates from 2013 to 2019 and the initial emissions of $PM_{2.5}$ in 2013 ($p = 5.0\times10^{-46}$ and 2.0×10^{-26} for SO_2 and NOx, respectively), which is shown in **Fig. S7**. This result suggests that the level of pollution is a major factor driving mitigation actions at the city level. The emissions of a small number of cities actually increased over this period. However, because the emission rates of these cities were all at the lower end, they contributed negligibly to the national average. In addition, a significantly positive correlation was also found between the per-capita GDP (GDP_{cap}) and $PM_{2.5}$ decline ($p < 0.01$), again indicating the importance of financial capability in pollution control (**Fig. S8**). To a certain extent, the GDP_{cap} could actually demonstrate the investment in environmental protection, which could directly affect air pollution control (**Fig. S9**).

Future decrease of $PM_{2.5}$ in China

The goal of the Plan launched in 2016 is to reduce the annual mean $PM_{2.5}$ concentration to 35 μg/m^3 for all cities in China by 2035[5]. Although the achievability of the goal is of interest to policymakers and scientists, such predictions are not an easy task because specific emission reduction schemes have not been developed, and these specific schemes are essential for a quantitative evaluation of fixed future goals. Although it is reasonable to expect that the current efforts to fight air pollution in China will continue in the future due to the strong willingness of both the public and policymakers[33], simply extrapolating current emission reduction rates linearly to the future is not practical. The pace of reduction will gradually decrease because easy-to-control sources were targeted first, and mitigation costs often increase exponentially[20]. As discussed in the previous section, the emission reduction rates of Chinese cities in the past were significantly correlated with initial levels of pollution ($PM_{2.5}$ concentrations) and economic development status (GDP_{cap}). Based on these two parameters, we predicted the $PM_{2.5}$ trend for each city through 2035 (see **Experimental Procedures**). **Fig. 4** shows the national annual mean $PM_{2.5}$ concentrations for the 367 cities as solid lines together with 95% confidence intervals of the emission reduction predictions (light red-shaded) and linear regression model (yellow-shaded) and the 50% and 95% uncertainty intervals associated with fluctuating changes in meteorological conditions (dark and light blue-shaded, respectively). According to the results, the national annual mean $PM_{2.5}$ concentrations of the 367 cities will be further reduced by 36±19% from 37.8±13.0 μg/m^3 in 2019 to 24.2±6.6 μg/m^3 in 2035, which will be much lower than the targeted 35 μg/m^3. The trend is generally optimistic, and the level would be even lower than the WHO Interim target-2 of 25 μg/m^3. Because this prediction was based on a statistical approach, all of the results are presented on a probabilistic basis. For the same reason, any detailed discussion of specific cities is meaningless.

The projected emission trends in China from 2020 to 2035 in this study were compared with the projected emission trends from previous studies, as illustrated in **Fig. S10**. Based on these emissions, future trends of $PM_{2.5}$ concentration were calculated using the reduced-form model, as shown in **Fig. 5**. Previous projections were generally developed based on detailed scenarios of fuel consumption, energy mix, and end-of-pipe control technologies[34]. Compared with most scenarios from RCPs, ECLIPSE, and SRES, which were widely used for future predictions, the reductions of both emissions and $PM_{2.5}$ concentration were much stronger in this study. The major reason for the disparities is that our projection was based on the recent controlling actions in China, whereas RCPs, ECLIPSE, and SRES were mostly developed over one decade ago driven by broad goals and moderate control strengths[35,36]. Such differences were also found in other studies[34]. In view of the $PM_{2.5}$ trends, our estimations were close to those from the RCP2.6 and SSP1-2.6 scenarios, both of which assumed ambitious reduction in fossil fuel usage and improvements in energy efficiency[37,38]. For example, the SSP1-2.6 scenario assumed a rapid replacement of fossil fuels by renewable energy together with rapidly falling pollutant emission factors along with the promotion of new controlling technologies, leading to a continuous decreasing trend in $PM_{2.5}$[38]. These assumptions were like the policies enacted by the Chinese government in recent years[19]. The decreasing trend of $PM_{2.5}$ was slightly weakened after 2025 in this study compared with the SSP1-26 scenario, mostly owing to our assumption that the absolute control strength would fall along with the decline of $PM_{2.5}$. Given that the baseline years of RCP2.6 and SSP1-26 are before 2019, this agreement suggests that our prediction models can capture the emission trends with the continuous control efforts in China. Moreover, we also compared the results with several studies that adopted similar assumptions. We found that our results agreed well with these studies considering the current mitigation efforts in China. For example, Cai et al. (2018) projected that the SO_2 emissions in 2030 would decrease by 25.6% and 48.7%, respectively, by assuming constant and strong additional policies in 2030 compared with the 2017 level (see the '*Cai2108-CLE*' and '*Cai2018-WAM*' in **Fig. S10**)[34]. Our corresponding reduction was estimated as 40.6% (25.9% ~ 52.8% as UI95), which fell within the former two scenarios. By assuming all the regulations in 2010 would continue until 2030, the reduction scenario developed by Wang et al. (2014) showed a reduction of $PM_{2.5}$ concentration by 12% (see '*Wang2014-BAU1*' in **Fig. S10**)[39]. However, the reduction would be 28% when end-of-pipe control strategies were fully adopted (see '*Wang2014-BAU2*' in **Fig. S10**), which was close to our estimations (28%). Even stronger reduction would be expected when new energy-saving policies were adopted (see '*Wang2014-PC2*' in **Fig. S10**)[39].

The trends vary among the cities and regions. **Fig. S11** shows the distributions of the model-predicted annual mean $PM_{2.5}$ concentrations of 367 cities in 2035, and a significant shift can be observed towards the low concentrations during this period. In 2019, only 46% of cities had annual mean $PM_{2.5}$ concentrations below the target, and the percentage will increase to 95% by 2035. Although this result indicates a great improvement, it is still likely that a small percentage of cities will not necessarily reach the target of 35 $\mu g/m^3$ without extra effort. The spatial distributions of the predicted annual mean $PM_{2.5}$ concentrations of these cities in 2013, 2019, and 2035 are mapped in **Fig. S12**. Based on current and future efforts, the $PM_{2.5}$ concentrations in all cities decreased and will continue to decrease. The spatial difference will decrease substantially as well. In fact, the current target of 35 $\mu g/m^3$ is rather conservative when considering health impacts[13]. Although the single target is realistic nationwide at this stage, classified targets for different regions should be a better choice for the next stage of the mitigation strategy in China. For example, WHO IT-2 (25 $\mu g/m^3$) or even IT-3 (15 $\mu g/m^3$) can be reasonably targeted for the rapidly developed eastern coastal region. Such uneven targets are expected to be better able to serve national environmental and health benefits and promote the efficiency of mitigation efforts. However, with the continuous decline of $PM_{2.5}$, the government will also face an increasing financial burden from controlling air pollution. It is important to propose future targets with caution by considering the costs, especially after reaching the 35 $\mu g/m^3$ target. Therefore, the future $PM_{2.5}$ trend would be altered by the government's actions to balance the benefits of controlling air pollution and the corresponding costs. On the other hand, the future projection in this study did not consider the changes in SOA (secondary organic aerosol), which is actually attracting a growing concern in China's government after a remarkable achievement of controlling other pollutants. Along with the continuation of current actions, extra efforts targeting NMVOCs (non-methane volatile organic compounds) would bring further reduction in both $PM_{2.5}$ and ozone, and the corresponding impacts on air quality need further investigation. Moreover, future climate change can potentially affect air quality[40]. Previous studies have demonstrated that the frequency of extreme events can play a significant role in determining air pollution in a changing climate[41,42]. Specifically, predicted $PM_{2.5}$ fluctuations have shown strong sensitivities to the occurrence of atmospheric stagnation, which can overwhelm atmospheric circulation and exacerbate pollution[41,43]. However, large uncertainties in recognizing the magnitude and even the sign of future changes in such extreme events still exist[41,43,44]. The uncertainties suggest that care should be taken when interpreting the impacts of climate change[45], and future work on narrowing down the uncertainties is warranted.

Conclusions

In this study, we present a quantitative evaluation of current and future $PM_{2.5}$ trends in China. With the exclusion of meteorological effects, the continuous controlling efforts in China have led to a 34% decrease in national annual mean $PM_{2.5}$ concentration from 2013 to 2019. Due to the meteorological variation, the observed reduction is greater than our estimation, which again confirms the necessity of distinguishing the meteorological effects for an objective policy evaluation. Driven by different initial $PM_{2.5}$ pollution levels and socioeconomic development, mitigation efforts show large variations among individual cities. The reduction is predicted to continue if the current effort is carried on in the future, and the 35 $\mu g/m^3$ national standard would be achieved by most cities by 2035. The continuous decline in $PM_{2.5}$ concentration would bring tremendous health benefits, especially in the regions with heavy pollution at the current stage.

It should be noted that the methodology is subject to some limitations that could introduce potential uncertainties to the results. For example, one of the limitations is that the reduced-form model cannot address decadal or interdecadal climate change due to the limited model training period (35 years). Therefore, the $PM_{2.5}$ trends altered by long-term climate change are not considered in this study. With the advance in distinguishing the climate change signals, such uncertainties can be narrowed down by combining the current results with improved atmospheric transport modeling. Details on additional constrains of the methodology are discussed in **Experimental Procedures**.

Despite the limitations, we have proved the utility of the results. By comparing with previous studies that adopted traditional dynamic and/or integrated assessment models, we have shown that the framework developed here achieves comparable capabilities in reconstructing the current $PM_{2.5}$ trend and projecting future trajectories. The method and results presented in this study can be extended to future research with reasonable accuracy, especially for cases in which extensive model simulations in air quality studies and predictions are demanded. In addition, discerning the impacts of emission reduction and meteorological fluctuations is essential from the perspective of policymaking. Governments need to address the trade-off between controlling air pollution and financial expenses. Indeed, extensive efforts to control air pollution can bring more benefits to human health, which inevitably increases the financial burden due to the exponentially increased controlling difficulty[20]. Therefore, the results of unmasking the meteorological effects in this study can provide useful information to formulate objectives and effective strategies. The flexibility of the current method also allows rapid assessments of the proposed policy scenarios in emission reduction, contributing to future policy adjustment. However, these assessments usually take much more computation time in a traditional dynamic model. Better still, the frequency distribution of meteorological effects can help policymakers propose strategies to address severe pollution events possibly induced by meteorological extremes.

Experimental Procedures

Resource Availability

Lead Contact

Further information and requests for resources and reagents should be directed to and will be fulfilled by the Lead Contact, Shu Tao (taos@pku.edu.cn) .

Materials Availability

This study did not generate new unique materials.

Data and Code Availability

The baseline emission dataset is taken from Peking University emission inventories[55] (PKUEI, freely available at http://inventory.pku.edu.cn/). The $PM_{2.5}$ concentrations of 367 cities calculated in this study are deposited at Mendeley Data (http://dx.doi.org/10.17632/snf8sjg23c.1).

Observation of $PM_{2.5}$ The study focused on 367 cities in mainland China, where the $PM_{2.5}$ reduction goals targeted by the Plan and all official routine air quality monitoring stations are located. **Fig. S13** shows the distribution of these cities in China, with the population density in the background. Routine monitoring schemes of ambient $PM_{2.5}$ in mainland China did not begin until 2013, when an ambitious Plan and a routine $PM_{2.5}$ monitoring program were launched[15]. The monitoring program covered 74 major cities in 2013 and expanded to 190 and 367 cities in 2014 and 2015, respectively. Currently, there are 1,641 stations in the 367 cities reporting $PM_{2.5}$ data on an hourly basis. To fill the data gap, we estimated the data for those cities missing in 2013 and 2014 based on linear regressions between available observations and satellite-retrieved surface $PM_{2.5}$ concentrations[46], as shown in **Fig. S14**.

$PM_{2.5}$ prediction and model validation In this study, $PM_{2.5}$ concentrations were quantified using our predeveloped reduced-form model[14]. This model is a combination of multivariate regressions and probability functions to separately quantify the influence of emissions and meteorological conditions, respectively. The multivariate regressions were developed from a 35-year global simulation driven by fixed meteorology. Under the long-term average meteorological status, the annual $PM_{2.5}$ concentrations were calculated based on the total emissions of four pollutants (i.e., primary $PM_{2.5}$, SO_2, NOx, and NH_3) as described by Equation (1):

$$\log(\mathrm{PM}_{2.5}) = \sum_{i}^{4} a_i \log(\mathrm{Emis}_i) + b \quad (1)$$

where Emis_i indicates the annual emissions of the i^{th} pollutant emission involved, and a_i and b were obtained from the regression and differed among grid cells. To eliminate the meteorological effects, the calculation was based on average meteorological conditions, which were represented by the meteorology in 2014 according to our previous study[14]. Meanwhile, the meteorological effects on annual $PM_{2.5}$ were addressed using probability functions, which were derived from a long-term simulation with constant emissions. The idea of frequency distribution can reflect the fluctuating features of meteorological effects. Since both the regression models and probability functions were developed at grid-cell levels with a spatial resolution of 0.1°, large differences were found among the cities focused on in this study[14].

The coupled models used to calculate the $PM_{2.5}$ concentrations have been validated previously on a global scale[14] and were further validated in this study against observations in the study domain, with the validation performed for the temporal trends for four cities (namely, Beijing, Shanghai, Guangzhou, Chengdu) with data available before 2014 and the consistency of the calculated value against annual observations for 74 and 190 cities that began to report $PM_{2.5}$ concentrations in 2013 and 2014. The validation is shown in **Fig. S15**.

In this study, the impacts of emission reduction on $PM_{2.5}$ concentration were obtained directly from the regression models. For meteorological effects, we presented two kinds of evaluations. By comparing the calculated $PM_{2.5}$ distribution from the reduced-form model and observations, we estimated the frequency of occurrence of the meteorological effects to understand the severity of the effects, which was a brand-new perspective. To compare our results with previous studies, we also evaluated the specific meteorological effects by subtracting the emission-driven changes from the observed $PM_{2.5}$. As a validation, both the emission and meteorological effects in this study were compared with previous studies, as summarized in **Tables S1** and **S2**. With the exclusion of meteorological effects, the $PM_{2.5}$ reductions due to emission mitigation in this study agreed well with previous studies despite the different approaches adopted, which generally confirmed the substantial progress in fighting $PM_{2.5}$ pollution in recent years in China. For meteorological effects, large disparities were found in previous studies. In comparison, the meteorological effects in this study were generally stronger, possibly due to the weakened meteorological signals from previous studies by choosing a baseline with low emission levels. A more detailed discussion can be found in **Note S1**.

The calculation of meteorological effects based on the probability functions assumed, in reality, that the chemical transport model could capture the $PM_{2.5}$ responses to meteorological fluctuations. This assumption was tested by comparing the sensitivities of modeled and measured $PM_{2.5}$ concentration to meteorological fluctuations. We regressed the detrended concentrations to key meteorological parameters and used the slopes to represent the sensitivities[47]. The comparisons between modeled and observed sensitivities are shown in **Fig. S16**. We discovered that the chemical transport model could largely reconstruct the observed response to fluctuations in the wind field and relative humidity but underestimated the sensitivities to air temperature and precipitation. The imperfect model representation of $PM_{2.5}$ sensitivities was also found in other chemical transport models[47,48]. A detailed discussion can be found in **Note S2**.

By using the reduced-form model, we decomposed the $PM_{2.5}$ variabilities to the emission-induced trends and the annual fluctuations induced by meteorological conditions. The meteorological effects included the impacts of interannual climate variations such as ENSO and NAO as the probability functions were developed based on 35-year simulations, which exceeded the standard period of "climate normal" as suggested by the WMO (World Meteorological Organization)[49]. In addition, $PM_{2.5}$ is also influenced by long-term climate cycles such as the Pacific Decadal Oscillation and other atmospheric teleconnection patterns[50,51]. These impacts from long-term changes in climate were not fully included in our reduced-form model, mostly due to the limited model training period (1980-2014), which was not sufficient to capture the long-term (decadal) climatic cycles well. For the same reason, climate change impacts were not considered in future predictions. Previous studies have suggested the long-term climate perturbations on $PM_{2.5}$ concentrations have smaller impacts than interannual variations and emission changes that have been addressed in our reduced-form model[24,52].

Historical and current emissions The emissions of four pollutants were used in the reduced-form model, and they are primary $PM_{2.5}$, SO_2, NOx, and NH_3, which are the most important contributors to both primary and secondary aerosols in air[53,54]. The inventories of the four pollutants from 1980 to 2014 were derived directly from Peking University emission inventories (PKUEI) [55] and used for historical modeling[14].

For the current period (2015-2019), when the PKUEI was not available, we utilized a trial-and-error approach to obtain the best estimates of emissions on the city level. Based on the baseline emissions in 2014, a total of 41 emission reduction scenarios from 0%, 5%, 10%, 15%, … to 200% of the provincially reported emission reductions were used to calculate the annual mean $PM_{2.5}$ concentrations of individual cities under average meteorological conditions. By comparing the results with field observations and choosing the scenario with the calculated $PM_{2.5}$ immediately above the observed $PM_{2.5}$ as a conservatively estimated emission reduction, the emissions for the current years were obtained, with this calculation repeated for all cities. Since the emission reductions of primary $PM_{2.5}$ and NH_3 were not reported officially, we assumed that they were the same as those for SO_2 and NOx, respectively. Such estimates of emissions are based on the similar meteorological conditions in 2015-2019 to the long-term average status as represented by 2014, which was confirmed in most cities (93%) by adopting a paired t-test on a daily basis (**Fig. S17**). The best emission estimations were then used to calculate the $PM_{2.5}$ concentration for the period 2015-2019 with meteorological confounding effects excluded. For the short-term evaluation, the influences from the reduction of NMVOC emissions were not considered because previous studies have shown that their emissions trends are quite constant, and they have hardly contributed to China's $PM_{2.5}$ reduction in recent years[19,56,57]. Given the much stronger uncertainties in the emissions of NMVOCs than other pollutants[58], involving NMVOCs would result in extra uncertainties to the overall evaluation.

Future prediction For long-term predictions from 2020 to 2035, a specific mitigation scheme was not available and needed to be quantitatively characterized. However, simply extrapolating the current emission reduction is unreasonable since the motivation and difficulties can alter along with the pace of air pollution control[20]. By assuming that the factors affecting the temporal change of emission reduction over time have a similar influence on the spatial variation, two regression models (Equation 2, 3) were adopted to estimate the future reduction rate (*R*e) of SO_2 and NOx based on the historical data of initial $PM_{2.5}$ concentration and GDP_{cap}:

$$Re\ (SO_2) = 0.15 \log(PM_{2.5}) + 1.0\times10^{-7}\ GDPcap - 0.19, \qquad R^2 = 0.55 \qquad (2)$$

$$Re\ (NOx) = 0.17 \log(PM_{2.5}) + 4.2\times10^{-7}\ GDPcap - 0.23, \qquad R^2 = 0.45 \qquad (3)$$

The first positive slopes of the models represent the difficulty of achieving emission reductions at low pollution levels, whereas the second slopes represent the financial capacities of local governments to promote mitigation efforts. The differences in emission reduction among individual cities can be quantified by equations. To confirm the rationality of the models, the model-predicted *R*e values are plotted in **Fig. S18**, and the results show generally acceptable trends, although only approximately half of the variation can be captured, and the models cannot be further improved at this stage. By assuming that the factors causing the differences among the cities can affect the temporal variations in a similar way, the regression models developed were applied to predict future emission reductions of SO_2 and NOx up to 2035. Without specific data available, the two equations were also applied to primary $PM_{2.5}$ and NH_3. Such a time-for-space substitution approach has been successfully used to predict energy consumption[59,60].

The regression models can be used based on the predicted GDP_{cap} and $PM_{2.5}$ for coming years. The basic logic behind this approach is that the mitigation efforts of all cities will generally be kept at the same level in terms of political willingness and financial investment up to 2035. Without a specific plan for mitigation measures, this approach is generalized. Because the concentrations and emissions depend on each other, e.g., *R*e is a function of log($PM_{2.5}$), whereas $PM_{2.5}$ concentrations are affected by the *R*e of the previous year, an iterative method was used. Specifically, as the first step, the *R*e values of SO_2 and NOx were predicted using the regression models, and the *R*e values of primary $PM_{2.5}$ and NH_3 were assumed to be similar to the *R*e values of SO_2 and NOx, respectively. Then, $PM_{2.5}$ concentrations were derived using the reduced-form model[14]. The two steps were repeated until the results converged to derive the annual mean $PM_{2.5}$ concentrations of all cities from 2020 to 2035 with confounding meteorological effects excluded. This emission-driven prediction operates under the assumption that China's efforts to fight air pollution will continue, which is actually a very possible case in the future[61]. Since the future $PM_{2.5}$ trends were generated by an iteration algorithm, the cumulative 95% confidence intervals (CI95) were used in this study and obtained by adopting the edge value of CI95 of $PM_{2.5}$ concentration in the previous year to calculate the limits of CI95 for the next year. Moreover, the fluctuating influences of meteorological conditions were quantified using probabilistic functions developed previously[14].

The future prediction aimed mainly at exploring the impacts of continuous emission mitigation in China up to 2035, while the effects of climate change in the near future were not considered. Therefore, the effects of meteorological fluctuations were directly characterized using the probability function. Such an assumption on climate variability was tested by comparing the fluctuation ranges of meteorological effects with previous studies focusing on the net impacts of future climate change on $PM_{2.5}$ in China, which are listed in **Table S3**. Existing studies focused on even larger time scales than up to 2035, when stronger climate changes were expected[62]. Nevertheless, we found that the climate changes from previous studies were mostly within the 95% uncertainty ranges from our estimation, suggesting that the near future climate change might not exceed the fluctuations of meteorological conditions that had been considered in this study.

Other analysis As shown in **Fig. S19**, the model-calculated $PM_{2.5}$ concentrations of 367 cities were log-normally distributed (KS-test, $p > 0.05$), and log-transformation was applied whenever necessary. SPSS 23.0 (International Business Machines Corporation, NY, USA) was used for the statistical analysis at a significance level of 0.05[63]. To simulate meteorology-induced variability, Monte Carlo simulations were conducted using MATLAB R2016b (The MathWorks, Inc., Natick, MA, USA)[64].

Limitations and constraints There are constraints in the methodology and uncertainties in the results. One limitation of the reduced-form model is that the reductions in different pollutants cannot be individually quantified. Unfortunately, this is not the case in reality. For example, SO_2 emissions are mostly associated with coal burning, while NOx emissions are strongly connected to motor vehicles and power generation[65,66]. As a result, SO_2 and NOx are often mitigated at different rates at different stages[61]. Actually, the reported average R_e of SO_2 was 1.25±15.4 times that of NOx for the 367 cities, showing more efforts in desulfurization and very large variation among the cities. In addition, primary $PM_{2.5}$ and NH_3 are not covered by the current report, but both are important in terms of $PM_{2.5}$ concentrations in the air[27]. For example, residential solid fuels are strongly associated with emissions of primary $PM_{2.5}$ but not SO_2 and NOx[67]. In this study, the simulation was based mainly on SO_2 and NOx by assuming that the $PM_{2.5}$ reduction is the same as the reduction in SO_2 and the NH_3 reduction is the same as the reduction in NOx, and the fractions of these pollutants were fixed for various scenarios. In addition, the reduced-form model, which can distinguish the influence of emissions and meteorology, may introduce additional uncertainty compared with chemical transport modeling[14]. The uncertainty was addressed using the CI95 of the regression. Moreover, the methodology in this study is subject to systematic uncertainties stemming from the chemical transport model that we used to develop the meteorological probability functions. The imperfect representation of $PM_{2.5}$ sensitivities to meteorological fluctuations in the transport model can induce extra uncertainties in our analysis. Other transport models may actually suffer from similar problems, and improvement in models could further assist the understanding of meteorological effects on $PM_{2.5}$[47,48].

Acknowledgements

This work was funded by the National Natural Science Foundation of China (Grant 41830641, 41991312, 41922057, and 41821005), the 111 Program (B14001), the Strategic Priority Research Program of the Chinese Academy of Sciences (XDA23010100), and the Undergraduate Student Research Training Program.

Author contributions

S.T. proposed the idea. S.T. and Q.Z. designed the modelling procedure and wrote the manuscript with input from D.G. Q.Z. performed the modelling. Q.Z., S.T., and J.M. conducted the data analysis with important input from J.L., H.S., G.S., D.G., X.Y., W.M., X.Y., H.C., D.Z., Y.W., and J.H.

Declaration of Interests: The authors declare no competing interests.

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Figure titles and captions

Fig. 1 **Comparison between the model-calculated and observed annual $PM_{2.5}$ concentrations in China.** The red dots show the observations of national annual mean $PM_{2.5}$ concentrations with 95% confidence intervals (error bars). The model-calculated result is shown as the black line. The 95% confidence intervals of the regression model (CI95) and 50% and 95% uncertainty intervals induced by meteorological effects (UI50 and UI95) are shown as yellow-, dark blue-, and light blue-shaded areas, respectively.

Fig. 2 **Probability distribution of meteorological effects on the national annual mean $PM_{2.5}$ concentration in 2018.** The probability from the reduced-form model is shown as the frequency distribution of national mean $PM_{2.5}$ concentration (yellow area). The dark and light blue-shaded ranges show UI50 and UI95, respectively. Actual observations in 2018 are shown by the red dashed line (39.2 μg/m^3).

Fig. 3 **Spatial distributions of cumulative $PM_{2.5}$ reductions from 2013 to 2019 for the 367 cities.** (A) The cumulative $PM_{2.5}$ reductions from 2013 to 2019; (B) The cumulative $PM_{2.5}$ reductions divided by $PM_{2.5}$ concentrations in 2013. The sizes of the circles are proportional to the $PM_{2.5}$ concentrations in 2013. The color of the circles refers to the left panel of the color bar for (A) and the right panel for (B).

Fig. 4 **Future projection of the national annual mean $PM_{2.5}$ concentrations of the 367 cities from 2020 to 2035.** The predicted trends are constituted by the model means (solid line), $PM_{2.5}$ prediction uncertainty (CI95 $PM_{2.5,}$ i.e., 95% confidence intervals of the regression models as yellow-shaded areas), emission reduction prediction uncertainty (CI95 $R_{e,}$ i.e., 95% confidence intervals of the emission reduction prediction as light red-shaded areas), and meteorology-associated variations (UI50 and UI95, i.e., 50% and 95% uncertainty intervals as dark and light blue-shaded area).

Fig. 5 **Comparisons of current (2016-2019) and projected future $PM_{2.5}$ trends (2020-2035) between previous literature and this study**. The trends were calculated based on the emission trends in **Fig. S10** using linear regression models. The results are illustrated as the $PM_{2.5}$ changes relative to the 2019 level. The shaded areas show the 95% confidence intervals from our estimation. A full list of the data sources can be found in **Fig. S10**.