

Effects of air pollution control policies on PM_{2.5} pollution improvement in China from 2005 to 2017: a satellite based perspective

Zongwei Ma ^{1,2}, Riyang Liu ¹, Yang Liu ³, Jun Bi ^{1,2,*}

¹ State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment,
5 Nanjing University, Nanjing, Jiangsu, China

² Jiangsu Collaborative Innovation Center of Atmospheric Environment and Equipment
Technology (CICAEET), Nanjing University of Information Science & Technology, Nanjing,
Jiangsu, China

³ Department of Environmental Health, Rollins School of Public Health, Emory University,
10 Atlanta, GA, USA

*** Correspondence to:**

Dr. Jun Bi, School of the Environment, Nanjing University, 163 Xianlin Avenue, Nanjing
210023, P. R. China. Tel.: +86 25 89681605. E-mail address: jbi@nju.edu.cn.

15

ABSTRACT

Understanding the effectiveness of air pollution control policies is important for future policy making. China implemented strict air pollution control policies since 11th Five Year Plan (FYP). There is still a lack of overall evaluation of the effects of air pollution control policies on PM_{2.5} pollution improvement in China since 11th FYP. In this study, we aimed to assess the effects of air pollution control policies from 2005 to 2017 on PM_{2.5} from the view of satellite remote sensing. We used the satellite derived PM_{2.5} of 2005-2013 from one of our previous studies. For the data of 2014-2017, we developed a two-stage statistical model to retrieve satellite PM_{2.5} data using the Moderate Resolution Imaging Spectroradiometer (MODIS) Collection 6 aerosol optical depth (AOD), assimilated meteorology, and land use data. The first-stage is a day-specific linear mixed effect (LME) model and second-stage is generalized additive model (GAM). Results show that the Energy Conservation and Emissions Reduction (ECER) policy, implemented in 11th FYP period and focused on SO₂ emissions control, had co-benefits on PM_{2.5} reductions. The increasing trends of PM_{2.5} pollution (1.88 and 3.14 $\mu\text{g}/\text{m}^3/\text{year}$ for entire China and Jingjinji Region in 2004-2007, $p < 0.005$) was suppressed after 2007. The overall PM_{2.5} trend for entire China was $-0.56 \mu\text{g}/\text{m}^3/\text{year}$ with marginal significance ($p = 0.053$) and PM_{2.5} concentrations in Pearl River Delta Region had a big drop ($-4.81 \mu\text{g}/\text{m}^3/\text{year}$, $p < 0.001$) in 2007-2010. The ECER policy during 12th FYP period were basically the extension of 11th FYP policy. PM_{2.5} is a kind of composite pollutant which comprises primary particles and secondary particles such as sulfate, nitrate, ammonium, organic carbon, elemental carbon, etc. Since ECER policy focused on single-pollutant control, it had shown great limitation for PM_{2.5} reductions. The PM_{2.5} concentrations did not decrease from 2010 to 2013 in polluted areas (p values of the trends were greater than 0.05). Therefore, China implemented two stricter policies: 12th FYP on Air Pollution Prevention and Control in Key Regions (APPC-KR) in 2012, and Action Plan of Air Pollution Prevention and Control (APPC-AP) in 2013. The goal of air quality improvement (especially PM_{2.5} concentration improvement) and measures for multi-pollutant control were proposed. These policies had led to dramatic decrease in PM_{2.5} after 2013 ($-4.27 \mu\text{g}/\text{m}^3/\text{year}$ for entire China in 2013-2017, $p < 0.001$).

1 Introduction

Fine particulate matter (PM_{2.5}, particulate matter with aerodynamic diameter less than 2.5 μm) is a major atmospheric pollutant, which has been shown to be strongly associated with adverse health effects (e.g., cardiovascular and respiratory morbidity and mortality) in many epidemiological studies (Crouse et al., 2012;Dominici et al., 2006;Pope et al., 2002). With the rapid economic development and industrialization in the past decades, PM_{2.5} pollution has gradually become a major environmental issue in China (Liu et al., 2017a). However, the Chinese government did not focus on the PM_{2.5} issues until 2012. Therefore, air pollution control policies implemented before 2012 mainly focus on SO₂, industrial dust and soot emission control. The air pollution control policies of China started to pay attention to PM_{2.5} since late 2012.

Understanding the effectiveness of air pollution controls policies is important for future air pollution control in China. Several studies have examined the historical air pollution control policies and their association with the trends of SO₂, NO₂, and PM₁₀ (Jin et al., 2016;Chen et al., 2011;Hu et al., 2010). Since the national PM_{2.5} monitoring network was established in late 2012, few studies have evaluated the effects of air pollution control policies on PM_{2.5} concentrations before 2013 due to the lack of historical ground monitoring data. Therefore, it is difficult to understand whether the air pollution control policies had synergistic effects on PM_{2.5} reductions before 2012.

In recent years, many studies have shown that satellite remote sensing provides a powerful tool to assess the spatiotemporal trends of air pollutions for both global and regional scales (Miyazaki et al., 2017;Itahashi et al., 2012;Krotkov et al., 2016). Estimating ground PM_{2.5} using satellite aerosol optical depth (AOD) data was also an effective way to fill the spatiotemporal PM_{2.5} gaps left by ground monitoring network (Liu, 2013, 2014;Hoff and Christopher, 2009). There are two major methods to estimate ground PM_{2.5} concentration using AOD data, i.e., the scaling method and statistical approach (Liu, 2014). The scaling method uses atmospheric chemistry models to simulate the association between AOD and PM_{2.5}, and then calculate the satellite-derived PM_{2.5} using the

equation: $Satellite\text{-}derived\ PM_{2.5} = \frac{Simulated\ PM_{2.5}}{Simulated\ AOD} \times Satellite\ AOD$ (Liu, 2014). Boys et al.

(2014) and van Donkelaar et al. (2015) estimated the global satellite $PM_{2.5}$ time series using the scaling method. Compared to the scaling method, statistical models have greater prediction accuracy but require large amount ground-measured $PM_{2.5}$ data to develop the models (Liu, 2014).

5 By taking advanced of the newly established ground $PM_{2.5}$ monitoring network, we developed a two-stage statistical model to estimate historical monthly mean $PM_{2.5}$ concentrations using Aqua Moderate Resolution Imaging Spectroradiometer (MODIS) Collection 6 AOD data in one of our previous studies (Ma et al., 2016). Validation results shows that this monthly $PM_{2.5}$ dataset has high prediction accuracy ($R^2 = 0.73$). This accurate historical $PM_{2.5}$ dataset from 2004 to 2013 allowed us
10 to examined the effects of pollution control policies on $PM_{2.5}$ concentrations. In this previous study (Ma et al., 2016), we preliminarily analyzed the effects of Energy Conservation and Emissions Reduction (ECER) policy in 11th five year plan (2006-2010). We found an inflection point around 2008, after which $PM_{2.5}$ concentration showed slight decreasing trend, showing the co-benefits of the ECER policy. From 2013 to 2017, China implemented the Action Plan of Air Pollution
15 Prevention and Control (APPC-AP), which focused on $PM_{2.5}$ pollution. Currently, there is still a lack of overall evaluation of the effects of air pollution control policies on $PM_{2.5}$ pollution improvement in China from 2005 to 2017.

In this study, we aimed to assess the effects of air pollution control policies from 2005 to 2017 on $PM_{2.5}$ from the view of satellite remote sensing. We used the satellite-derived $PM_{2.5}$ dataset
20 developed in our previous study (Ma et al., 2016). Since this dataset was from 2004 to 2013 and data after 2014 has been lacking, we extended the dataset to 2017 in the present work. To keep consistent with our previous satellite $PM_{2.5}$ dataset, we used the same method as described in our previous study (Ma et al., 2016).

2 Overview of air pollution control policies in China from 2005 to 2017

25 During 2005 to 2017, China implemented a series air pollution prevention and control policies, including 11th Five Year Plan (FYP) on Environmental Protection (2006-2010), ECER Policy

during 11th FYP period, 12th FYP on Environmental Protection (2011-2015), 12th FYP on ECER, The 12th FYP on Air Pollution Prevention and Control in Key Regions (APPC-KR), and APPC-AP (2013-2017). The base year, implementation period, major goals, and major measures are listed in Table 1.

5 During 11th FYP period, there was no specific air pollution control policy. Air pollution prevention and control measures were incorporated in the whole environmental protection plan or policy (i.e., 11th FYP on Environmental Protection and ECER policy). From Table 1 we can see that the air pollution policies during 11th FYP mainly focused on total emission reduction. In this period, environmental management in China was emission control oriented, that is, the indicators for local
10 governments' environmental performance assessment were emission reduction rates, not the environmental quality. The 12th FYP on Environmental Protection and ECER policy were basically the extension of the 11th FYP policies, which mainly focused on emission reduction.

The 12th FYP on APPC-KR is the first special plan for air pollution prevention and control. This plan proposed the idea of unification of total emission reduction and air quality improvement.
15 And it proposed the goals of air pollutant concentration control for the first time. PM_{2.5} pollution control was also incorporated in this plan. Although the implementation period of 12th FYP on APPC-KR is 2011-2015, it was issued in October 29, 2012. After that, China issued the APPC-AP (2013-2017) in September 10, 2013, which strengthened the air pollution control and the goals of air quality improvement. These policies indicated that the focus of air pollution control in China
20 began to focus on PM_{2.5} concentrations reductions.

3 Data and method

3.1 Satellite-based PM_{2.5} from 2004 to 2013

We estimated the monthly satellite-based PM_{2.5} data from 2004 to 2013 at 0.1 °resolution in our previous work (Ma et al., 2016). Briefly, we developed a two-stage statistical model using
25 MODIS Collection 6 AOD and assimilated meteorology, land use data, and ground monitored PM_{2.5} concentrations in 2013. The overall model cross-validation R² (coefficient of determination) was

0.79 (daily estimates) for the model year. Since ground monitor data before 2013 has been lacking and therefore it is unable to develop statistical models before 2013 to estimate historical PM_{2.5} concentrations. Thus, the historical PM_{2.5} concentrations (2004-2012) were then estimated using the model developed based on 2013 model. Two ways were used to validate the accuracy of historical estimates. First, we compared the historical estimates monitoring data from Hong Kong and Taiwan before 2013. Second, we estimated PM_{2.5} concentrations in the first half of 2014 using the 2013 model and compared them with the ground measurements to evaluate the accuracy of PM_{2.5} estimates beyond the model year, which can represent the accuracy of historical estimates. Validation results indicated that it accurately predicted PM_{2.5} concentrations with little bias at the monthly level ($R^2 = 0.73$, slope = 0.91).

For PM_{2.5} concentrations from 2004 to 2013, we directly used above-mentioned satellite-based PM_{2.5} dataset, which was estimated using the model developed in 2013. First, this dataset has been shown high accuracy and has been widely used in environmental epidemiological (Liu et al., 2016a; Wang et al., 2018a), health impact (Liu et al., 2017b; Wang et al., 2018b), and social economic impact (Chen and Jin, 2019; Yang and Zhang, 2018) studies in China. Second, a recent study has shown that the historical hindcast ability of the annual model decreased when hindcast year was long before the model year (Xiao et al., 2018). Therefore, we did not use the models of 2014 to 2017 to estimate the hindcast PM_{2.5}.

3.2 Satellite-based PM_{2.5} from 2014 to 2017

Unlike historical estimates from 2004 to 2012, we have sufficient ground monitored PM_{2.5} data to develop statistical models after 2013, which allowed us to estimate daily PM_{2.5} concentrations accurately. Therefore, we developed a separate PM_{2.5}-AOD statistical model for each year of 2014-2017 to estimate the spatially-resolved (0.1 °resolution) PM_{2.5} concentrations. To keep satellite PM_{2.5} estimates of 2014-2017 consistent with our previous satellite PM_{2.5} dataset, we used the same method as described in our previous study (Ma et al., 2016). The data, model development, and model validation are briefly introduced as follows.

The data used in this study include ground monitored PM_{2.5} concentrations (μg/m³), Aqua MODIS Collection 6 Dark Target (DT) AOD and Deep Blue (DB) AOD data, planetary boundary layer height (PBLH, 100 m), wind speed (WS, m/s) at 10 m above the ground, mean relative humidity in PBL (RH_PBLH, %), surface pressure (PS, hPa), precipitation of the previous day (Precip_Lag1; mm), MODIS active fire spots, urban cover (%), and forest cover (%). Ground monitored PM_{2.5} data were collected from China Environmental Monitoring Center (CEMC), environmental protection agencies of Hong Kong and Taiwan. Figure 1 shows the ground PM_{2.5} monitors used in this study. AOD were downloaded from the Level 1 and Atmospheric Archive and Distribution System (<https://ladsweb.modaps.eosdis.nasa.gov/>, accessed on Mar 29, 2019). Meteorological data were extracted from Goddard Earth Observing System Data Assimilation System GEOS-5 Forward Processing (GEOS 5-FP) meteorological data (<ftp://rain.ucis.dal.ca>, accessed on Mar 29, 2019). MODIS fire spots were from the NASA Fire Information for Resource Management System (<https://earthdata.nasa.gov/earth-observation-data/near-real-time/firms>, accessed on Mar 29, 2019). Land use information were downloaded from Resource and Environment Data Cloud Platform of Chinese Academy of Science (<http://www.resdc.cn/data.aspx?DATAID=184>, accessed on Mar 29, 2019).

Previous studies have shown the data quality issue of ground PM_{2.5} measurements from CEMC network (Liu et al., 2016b; Rohde and Muller, 2015). We performed the data screening procedure before model fitting. Abnormal values (extreme high or extreme low values for a site compared with its neighboring sites, repeated values for continuous hours, etc.) were deleted before model fitting. We required at least 20 hourly records to calculate the daily average PM_{2.5} concentrations. DT and DB AOD were combined using inverse variance weighting method to improve the spatial coverage of AOD data (Ma et al., 2016). This combined AOD data has been shown good consistency ($R^2=0.8$, mean bias=0.07) with ground AOD measurements from Aerosol Robotic Network (AERONET) (Ma et al., 2016). All data were assigned to a predefined 0.1 ° grid. Then all of the variables were matched by grid cell and day-of-year (DOY) for model fitting.

A two-stage statistical model was developed for each year separately from 2014 to 2017. The

first-stage linear mixed-effects (LME) model included day-specific random intercepts and slopes for AOD, season-specific random slopes for meteorological variables, and fixed slope for precipitation and fire spots. The model structure of first-stage model was shown as follows:

$$\begin{aligned}
 PM_{2.5,st} = & (\mu + \mu') + (\beta_1 + \beta_1')AOD_{st} + (\beta_2 + \beta_2')WS_{st} + (\beta_3 + \beta_3')PBLH_{st} + (\beta_4 + \beta_4')PS_{st} + (\beta_5 + \\
 5 \quad & \beta_5')RH_PBLH_{st} + \beta_6Precip_Lag1_{st} + \beta_7Fire_spots_{st} + \varepsilon_{1,st}(\mu' \beta_1') \sim N[(0,0), \Psi_1] + \\
 & \varepsilon_{2,sj}(\beta_2' \beta_3' \beta_4' \beta_5') \sim N[(0,0,0,0), \Psi_2] \tag{1}
 \end{aligned}$$

where $PM_{2.5,st}$ is ground $PM_{2.5}$ measurements at grid cell s on DOY t ; AOD_{st} is DT-DB merged AOD; WS_{st} , $PBLH_{st}$, PS_{st} , RH_PBLH_{st} , $Precip_Lag1_{st}$ are meteorological variables; $Fire_spots_{st}$ is the fire count; μ and μ' are the fixed and day-specific random intercepts, respectively; β_1 - β_7 are
 10 fixed slopes; β_1' is the day-specific random slope for AOD; β_2' - β_5' are the season-specific random slopes for meteorological variables; $\varepsilon_{1,st}$ is the error term at grid cell s on DOY t ; $\varepsilon_{2,sj}$ is the error term at grid cell s in season j ; Ψ_1 and Ψ_2 are the variance-covariance matrices for the day- and season-specific random effects, respectively. The first-stage model was fitted for each province separately. We created a buffer zone for each province to include data with at least 3,000 data
 15 records and at least 300 days. We averaged overlapped predictions from neighboring provinces to generate a smooth national $PM_{2.5}$ concentration surface.

The second-stage generalized additive model (GAM) established the relationship between the residuals of the first-stage model and smooth terms of geographical coordinates, forest and urban cover.

$$20 \quad PM_{2.5_resid_{st}} = \mu_0 + s(X, Y)_s + s(ForestCover)_s + s(UrbanCover)_s + \varepsilon_{st} \tag{2}$$

where $PM_{2.5_resid_{st}}$ is the residual of first-stage model at grid cell s on DOY t ; μ_0 is the intercept; $s(X, Y)_s$ is the smooth term of the coordinates of the centroid of grid cell s ; $s(ForestCover)_s$ and $s(UrbanCover)_s$ are the smooth functions of forest cover and urban area for grid cell s ; and ε_{st} is the error term.

25 10-fold cross validation (CV) was used to evaluate the model over-fitting, that is, the model could have better prediction performance in the model fitting dataset than the data which are not

included model fitting. In 10-fold CV, all samples in the model dataset are randomly and equally divided into ten subsets. One subset was used as testing samples and the rest subsets are used to fit the model. This process was repeated for 10 rounds until each subset was used for testing for once. Statistical indicators of coefficient of determination (R^2), mean prediction error (MPE), and root mean squared prediction error (RMSE) were calculated and compared between model fitting and CV to assess model performance and over-fitting.

3.3 Time series analysis

Monthly mean $PM_{2.5}$ concentrations for each grid cell were calculated to perform the time series analysis. Following our previous study (Ma et al., 2016), we required at least six daily $PM_{2.5}$ predictions in each month to calculate the monthly mean $PM_{2.5}$. We deseasonalized the monthly $PM_{2.5}$ time series by calculating the monthly $PM_{2.5}$ anomaly time series for each grid cell to remove the seasonal effect. $PM_{2.5}$ trend for each grid cell was calculated using least squares regression (Weatherhead et al., 1998):

$$(PM_{2.5})_{anomaly, s, m} = (PM_{2.5})_{s, m} - \overline{(PM_{2.5})_{s, j}} \quad m = 1, 2, 3, \dots, M \quad j = 1, 2, 3, \dots, 12 \quad (3)$$

$$(PM_{2.5})_{anomaly, s, m} = \mu + \beta \times m + \varepsilon, \quad m = 1, 2, 3, \dots, M \quad (4)$$

where $(PM_{2.5})_{anomaly, s, m}$ is the $PM_{2.5}$ anomaly at grid cell s for month m during the calculating period; $(PM_{2.5})_{s, m}$ is the estimated $PM_{2.5}$ concentration at grid cell s for month m ; m is the month index and M is the total number of months during the calculating period (2004-2017, $M=168$); $\overline{(PM_{2.5})_{s, j}}$ is the 14-year average $PM_{2.5}$ concentration of the month to which month m belongs ($j = 1$ for January, $j = 2$ for February, ..., etc.); μ is the intercept; β is the slope, which is also the trend of $PM_{2.5}$ ($\mu\text{g}/\text{m}^3/\text{month}$); ε is the error term. The annual $PM_{2.5}$ trend ($\mu\text{g}/\text{m}^3/\text{year}$) = $12 \times \beta$. The method of t test was used to obtain the statistical significance of the trends. This method has been successfully applied to trend analyses of monthly mean $PM_{2.5}$ and AOD anomaly time-series data (Hsu et al., 2012;Boys et al., 2014;Zhang and Reid, 2010;Xue et al., 2019).We analyzed the $PM_{2.5}$ trend for different periods to examine the effects of air pollution control policies on $PM_{2.5}$ pollution improvement.

4. Results and discussion

4.1 Validation of satellite-based PM_{2.5} concentrations from 2014 to 2017

Table S1 (Supplemental Materials, SM) summarized the statistics of all variables for the modeling dataset from 2014 to 2017. Overall, there are 95 649, 110 805, 113 490, and 123 652 matchups for the model fitting datasets for years of 2014, 2015, 2016, and 2017, respectively. The average PM_{2.5} concentration decreases year by year, from 65.66 µg/m³ in 2014 to 48.32 µg/m³ in 2017. Correspondingly, the average AOD also shows a decreasing trend from 0.67 in 2014 to 0.50 in 2017.

Figure 2 shows the model fitting and cross validation results for each year's model. The model fitting R² ranges from 0.75 (2015) to 0.80 (2017) and CV R² ranges from 0.72 (2015) to 0.77 (2017), which is similar to the 2013 model (0.82 for model fitting and 0.79 for CV) developed in our previous study (Ma et al., 2016). The model prediction accuracy is different among years, which is consistent with previous studies. Hu et al. (2014) studied the 10-year spatial and temporal trends of PM_{2.5} concentrations in the southeastern US from 2001 to 2010. They developed a separate two-stage statistical model for each year and found the CV R² ranged from 0.62 in 2009 to 0.78 in 2005 and 2006. Kloog et al. conducted two studies in Northeast US and also found that the validation R² varied among years (Kloog et al., 2011; Kloog et al., 2012). Compared to the model fitting R², the CV R² only decreases 0.02 in 2016 and 0.03 in 2014, 2015, and 2017, showing that our models were not substantially over-fitted. For the monthly mean concentrations calculated from at least six daily PM_{2.5} predictions, the validation R² values ranges from 0.75 to 0.81 (Figure 3). The results show that the overall prediction accuracy of the models from 2014 to 2017 is satisfying.

The fixed effects, model fitting, and CV results of the first-stage LME model for each province are shown in Tables S2-S5 (SM). AOD is the only variable that was statistically significant in all provincial models for all years ($p < 0.05$). Wind speed, relative humidity, precipitation, and fire spots were significant in most provincial models. The CV R² varies for different province and different year. The CV R² values range from 0.61 in Xinjiang to 0.77 in Heilongjiang for 2014,

from 0.34 in Xinjiang to 0.76 in Hebei for 2015, from 0.44 in Tibet to 0.77 in Jiangsu for 2016, and from 0.38 in Xinjiang to 0.79 in Sichuan for 2017. We also fitted a first-stage LME model for entire China. Results show that the overall CV R^2 values for first-stage LME model dropped to 0.57, 0.52, 0.56, and 0.54, for 2014, 2015, 2016, and 2017, respectively. Therefore, fitting the first-stage model for each province separately can greatly improve the prediction accuracy.

A potential source of uncertainties of statistical models is the uneven spatial distribution of ground $PM_{2.5}$ monitors. The CEMC air quality network mainly covers large urban centers with very limited sites coverage in rural areas, especially in western part of the country. Since it requires large amount ground-measured $PM_{2.5}$ data to develop satellite-based statistical model, this bias cannot be avoided. Despite this limitation, high model performances have been achieved in this study and previous similar studies (Zheng et al., 2016;Huang et al., 2018;Xue et al., 2019), which are much better than the scaling method. For example, Geng et al. (2015) estimated long-term $PM_{2.5}$ concentrations in China using scaling method and found the validation R^2 of $PM_{2.5}$ predictions was 0.72 compared to the five-month averaged ground $PM_{2.5}$ concentrations for Jan-May, 2013. A global study of $PM_{2.5}$ estimates combining scaling and statistical methods shows that their validation R^2 of long-term average $PM_{2.5}$ was 0.67 for their first-stage scaling method (van Donkelaar et al., 2016).

4.2 Overall spatial and temporal trend of $PM_{2.5}$ concentrations in China from 2004 to 2017

Figure 4 shows that spatial distribution characteristics of annual mean $PM_{2.5}$ concentrations are similar among the years from 2004 to 2017. The most polluted area was North China Plain (including south of Jingjinji Region, Henan, and Shandong Provinces), which was also the largest polluted area. The Sichuan Basin (including east of Sichuan and western Chongqing) is another polluted area. The cleanest areas were mainly located in Tibet, Hainan, Taiwan, Yunnan, and the north of Inner Mongolia. The spatial distributions of satellite-derived $PM_{2.5}$ concentrations from 2013 to 2017 are consistent with the spatial characteristics of ground monitored $PM_{2.5}$ (Figure S2, SM)

Figure 5 shows the spatial distributions of PM_{2.5} trends and significance levels in China from 2004 to 2017. Over all, the PM_{2.5} pollution level of most area in China showed a decreasing trend ($p < 0.05$). Figure 6 and Table 2 shows that the overall trends of 2004-2017 for entire China, Jingjinji, Yangtze River Delta (YRD), Pearl River Delta (PRD) Regions were -1.27, -1.55, -1.60, and -1.27 $\mu\text{g}/\text{m}^3/\text{year}$ (all $p < 0.001$), respectively. Back to Figure 4, we can see that the decrease of PM_{2.5} mainly happened after 2013. PM_{2.5} concentrations had an obvious increase from 2004 to 2007. The area with PM_{2.5} concentrations higher than 100 $\mu\text{g}/\text{m}^3$ continuously expanded during this period. From 2008 to 2013, the pollution levels plateaued in most areas. After 2013, the PM_{2.5} concentrations obviously decreased.

10 4.3 Effect of ECER policy during 11th Five Year Plan period

To assess the effect of ECER policy during 11th FYP, we calculated the trends of PM_{2.5} for 2005-2010, 2004-2007, and 2007-2010 for each grid cell (Figure 7).

15 Compared to the base year (2005) of the 11th FYP period, the overall PM_{2.5} pollution of 2010 did not have obvious change. Some of the area had decreasing trends (Figure 7(a)) but the trends were insignificant (Figure 7(b)). Some regions (Shandong, Henan, Jiangsu Provinces, and Northeast China) had slight increasing trend ($\sim 1\text{-}2 \mu\text{g}/\text{m}^3/\text{year}$, $p < 0.001$). Overall, the trends of entire China, Jingjinji, YRD, and PRD Regions were all insignificant (0.41, 0.26, 0.61, and -1.26 $\mu\text{g}/\text{m}^3/\text{year}$, and all $p > 0.1$) during 11th FYP period.

20 However, when separating this period into two periods, we can see that before 2007, the PM_{2.5} concentrations generally had significant increasing trends (Figure 7(c, d)), especially in South of Jingjinji Region, Henan, Shandong, and Hubei Provinces. The overall trends of entire China and Jingjinji Region are 1.88 ($p < 0.001$) and 3.14 ($p < 0.005$) $\mu\text{g}/\text{m}^3/\text{year}$ (Table 2). The trends of YRD and PRD Regions are insignificant. During the 10th YFP period, China missed the emission control goals. The emission of sulfur dioxide increased by $\sim 28\%$ (Xue et al., 2014; Schreifels et al., 2012).
25 The 11th FYP for National Economic and Social Development of China released in 2006 proposed the ECER goals. However, China did not achieve the annual goal in 2006. These could explain the

increasing trend of PM_{2.5} during 2004-2007.

After that, China released the Comprehensive Working Plan on ECER (http://www.gov.cn/zwggk/2007-06/03/content_634545.htm, accessed on Mar 29, 2019) in 2007 to strengthen the ECER measures. Major control measures included (Schreifels et al., 2012):

5 implementing flue gas desulphurization for coal-fired power plant, closing inefficient and backward production capacity, implementing energy conservation projects, increasing pollution levy for SO₂ emission, recommending baghouse dust filter for industrial soot and dust emission control etc. As a result, great achievements had been made at the end of 11th FYP (Schreifels et al., 2012; Zhou et al., 2015): total emission of SO₂ decreased by ~14% compared to the level of 1995; approximate 86%
10 of the power plant were installed with desulphurization facilities in 2010 compared to 14% in 2005; nearly 80 GW of small coal-fired power units were closed during 2006-2010; soot emission of coal-fired power plants in 2010 was reduced by 55.6% compared with that in 2005, etc.

Due to these control measures, the increasing trend of PM_{2.5} pollution was suppressed after 2007. PM_{2.5} concentrations of Central and South China decreased significantly, with highest trend
15 of around -9 µg/m³/year (Figure 7(e, f), $p < 0.01$). For south of Jingjinji Region, Henan, Shandong, and Hubei Provinces, which had significantly increased before 2007, showed insignificant trends (Figure 7(f), $p > 0.05$). Table 2 shows that the overall PM_{2.5} trend for entire China was -0.56 µg/m³/year with marginal significance ($p = 0.053$). Overall trends of Jingjinji and YRD Regions were not significant during the latter half of 11th FYP period. And PM_{2.5} concentrations in PRD Region
20 had a big drop (-4.81 µg/m³/year, $p < 0.001$). Results show that although air pollution control policies of 11th FYP were not designed for PM_{2.5} prevention and control, they still had co-benefits on PM_{2.5} pollution control. There were two main reasons. First, SO₂ is the precursor gas of sulfate. Previous studies have shown that sulfate was the major component of PM_{2.5} during 11th FYP period (Li et al., 2009; Li et al., 2010; Pathak et al., 2009). The reduction of SO₂ could therefore contribute to the
25 suppression of increasing PM_{2.5} pollution. Second, the control of industrial dust and soot, which include a portion of primary PM_{2.5} (Yao et al., 2009), also contributed to the PM_{2.5} pollution reduction.

4.4 Effect of air pollution control policies in 12th Five-Year Plan period (2011–2015)

Figure 8(a) and (b) show that most of the areas of China show significant decreasing trend during 12th FYP period. PM_{2.5} concentrations of entire China, Jingjinji, and YRD had dropped by 2.89, 3.63, and 3.33 $\mu\text{g}/\text{m}^3/\text{year}$ ($p < 0.001$). When considering the years from 2010 to 2013, although overall trend of entire China was $-1.03 \mu\text{g}/\text{m}^3/\text{year}$ ($p < 0.05$, Table 2), the decreasing trend mainly happened in Xinjiang and Central Inner Mongolia. The deserts in Xinjiang and Inner Mongolia are the major sources of dust pollution in China. A recent study showed that dust is the largest contributor to PM_{2.5} over this region (Philip et al., 2014). The change in natural dust in desert areas may be the major contributor to the decreasing trend of PM_{2.5} during 2010-2013. Most of the polluted area in China did not had obvious change (Figure 8(c) and (d)). As we mentioned above, The ECER policy during 12th FYP period was basically the extension of the policy in 11th FYP, which mainly focused on emissions reduction. As the further development of social economic, the ECER policy had shown its limitation for PM_{2.5} reductions. PM_{2.5} is a kind of composite pollutant and its constituents includes primary particles and secondary particles such as sulfate, nitrate, ammonium, organic carbon, elemental carbon, etc. With the deepening of SO₂ and industrial dust/soot emission reduction, their contributions to PM_{2.5} pollution control would reduce. Although 12th FYP on Environmental Protection also proposed 10% reduction of NO_x from 2010 to 2015. However, along with economic growth in China, the benefits of emission control for single-pollutant could be offset by increased energy usage. Considering the complicated PM_{2.5} compositions, comprehensive and coordinated control measures for multiple pollutants were urgently needed.

Therefore, China issued the 12th FYP on APPC-KR in late 2012, which is the first special plan for air pollution prevention and control and focused on air quality improvement. APPC-KR proposed a series of key projects which included 477 SO₂ treatment projects, 755 NO_x treatment projects, 10 073 industrial soot and dust treatment projects, 1 311 VOCs treatment projects in key industrial sectors, 281 vapor recovery projects for oil and gas. 188 yellow-sticker vehicle elimination projects, 192 fugitive dust comprehensive treatment projects, and 122 capacity building

projects. An English translation version of APPC-KR and its key projects has been prepared by Clean Air Alliance of China (CAAC) and can be found elsewhere (<http://www.cleanairchina.org/product/6347.html>, accessed on Mar 29, 2019) (CAAC, 2013c, a).

In addition, in 2012, China issued a new air quality standard, i.e., the *National Ambient Air Quality Standard of China* (NAAQS) (GB 3095-2012). Compared with the former NAAQS (GB 3095-1996) issued in 1996, this new standard incorporated PM_{2.5} as a major control pollutant. According to GB 3095-2012, the Level 1 annual mean standard of PM_{2.5} is 15 µg/m³, which is assigned for protecting the air quality of natural reserves and scenic areas and is equivalent to the World Health Organization (WHO) Air Quality Interim Target-3 (IT-3) Level. The Level 2 standard of 35 µg/m³ is designated for residential, cultural, industrial, and commercial areas, which is equivalent to WHO Air Quality Interim Target-1 (IT-1) Level. Meanwhile, a comprehensive real-time air quality monitoring network covering 74 major Chinese cities was established in late 2012.

The implementation of APPC-KR, together with the implementation of APPC-AP starting from 2013 (shown in the following section), had led to dramatic drops in PM_{2.5} concentrations in China after 2013. Table 3 shows PM_{2.5} concentration improvement goals and final accomplishments for key regions (see Figure S1, SM) of 12th FYP on APPC-KR calculated from satellite PM_{2.5}. Results show that all key regions had accomplished the goals except for Yinchuan. The changes in population weighted averages also show similar results. Overall, the 12th FYP on APPC-KR accomplished its air pollution control goals. And the decrease of PM_{2.5} concentrations was mainly attributable to the decrease after 2013.

4.5 Effect of Action Plan for Air Pollution Prevention and Control (2013-2017)

China issued the APPC-AP (2013-2017) in late 2013, which further strengthened the air pollution control measures and air quality improvement goals. The air pollution control measures included ten categories:

- Increase effort for comprehensive pollution control, reduce emissions of multi-pollutants;
- Optimize industrial structure, promote industrial restructuring;

- Accelerate technology transformation, improve innovation capability;
- Adjust energy structure, increase clean energy supply;
- Strengthen environmental thresholds, optimize industrial layout;
- Promote the role of market mechanism, improve environmental economic policies;
- 5 • Improve law and regulation system, carry on supervision and management based on law;
- Establish regional coordination mechanism and integrated regional environmental management;
- Establish monitoring and warning system, cope with heavy pollution episodes;
- Clarify responsibilities of government, enterprise and society, mobilize public participation
- 10

Detailed measures in the APPC-AP can be found in its English translation version

(<http://www.cleanairchina.org/product/6349.html>, accessed on Mar 29, 2019) (CAAC, 2013b). To ensure that APPC-AP goals could be accomplished, China adopted a temporary measure in 2017, i.e., the intensified supervision for air pollution control in Jinjinji and around area

15 (http://www.gov.cn/hudong/2017-07/14/content_5210588.htm, accessed on Mar 29, 2019). There had been great achievements at the end of 2017. For examples (Zheng et al., 2018): 71% of the power plants met the ultralow emission levels; average efficiency of coal fired power units reduced from 321 gce/kWh in 2013 to 309 gce/kWh in 2017; Non-methane volatile organic compounds (NMVOC) emissions were cut down by 30% through the implementation of leak detection and repair (LDAR) program for petrochemical industry; all coal boilers smaller than 7MW in urban areas were shut down; all “yellow label” vehicles (referring to which gasoline and diesel vehicles that fail to meet Euro 1 and Euro 3 standards, respectively) were eliminated by the end of 2017, etc.

25 The implementation of APPC-AP, together with 12th FYP on APPC-KR, had led to dramatic drop in PM_{2.5} concentrations from 2013 to 2017 (Figure 8(e) and (f)). PM_{2.5} trends of 2013-2017 for entire China, Jingjinji, YRD, and PRD Regions were -4.27, -6.77, -6.36, and -2.11 µg/m³/year (all $p < 0.05$), respectively (Table 2). This is comparable to a recent study (Silver et al., 2018), which found that median trend in annual mean PM_{2.5} concentration across all ground air pollution

monitoring stations is $-3.4 \mu\text{g}/\text{m}^3/\text{year}$ from 2015 to 2017. Table 4 shows $\text{PM}_{2.5}$ concentration improvement goals and final accomplishments for APPC-AP. The goals required $\text{PM}_{2.5}$ concentrations in Jingjinji, YRD, and PRD Regions in 2017 should be decreased by 25%, 20%, and 15% compared to 2012, and the annual mean $\text{PM}_{2.5}$ of Beijing should reach at around $60 \mu\text{g}/\text{m}^3$.

5 Since there were no ground measurements in 2012, the Ministry of Ecology and Environment (MEE) of China used 2013 as the base year to assess the performance of APPC-AP (http://www.mee.gov.cn/gkml/sthjbgw/stbgth/201806/t20180601_442262.htm, accessed on Mar 29, 2019). To maintain consistency with the official performance assessment, we also used 2013 as the base year. Results show that the arithmetic average of satellite $\text{PM}_{2.5}$ concentrations for Jingjinji, 10 YRD, and PRD Regions were decreased by 36.9%, 37.1%, and 14.0%, respectively. And annual mean $\text{PM}_{2.5}$ of Beijing was $44.67 \mu\text{g}/\text{m}^3$ in 2017. From the view of satellite, Jingjinji, YRD, and Beijing had accomplished their goals, and PRD was very close to the goal. However, the pollution level was still higher than WHO Air Quality IT-1 level and NAAQS (GB 3095-2012) Level 2 annual $\text{PM}_{2.5}$ standard (both $35 \mu\text{g}/\text{m}^3$).

15 According to the official results of APPC-AP performance assessment (Table 4), $\text{PM}_{2.5}$ of Jingjinji, YRD, and PRD Regions were decreased by 39.6%, 34.3%, and 27.7%, respectively. And annual mean $\text{PM}_{2.5}$ of Beijing was $58 \mu\text{g}/\text{m}^3$ in 2017. Compared to the arithmetic average satellite $\text{PM}_{2.5}$, the population weighted average results (Table 4) are more closed to the official results. The main reason is that official performance assessment used ground measurements. However, the 20 spatial distribution of ground monitors is uneven. Most of the sites are distributed in populated urban areas and only a few are located in rural areas. Compared to ground monitors, satellite remote sensing has more comprehensive spatial coverage. Figure S3 shows the spatial distribution of satellite and ground $\text{PM}_{2.5}$ concentrations of 2017 in Beijing. It can be seen that the ground monitors are clustered in polluted urban centers. The cleaner north and northwest of Beijing have few sites. 25 Thus the population weighted results of satellite $\text{PM}_{2.5}$ are closer to the official results, but still have differences. Since satellite $\text{PM}_{2.5}$ have better spatial coverage than ground monitors, satellite $\text{PM}_{2.5}$ can better represent the spatial variation of $\text{PM}_{2.5}$ pollution. The population weighted average

satellite PM_{2.5} can better represent the health impact of PM_{2.5} pollution. When using ground monitors to calculate the regional mean concentrations, the weights of area and population for each site should be considered.

5 Discussion and Conclusions

5 Xue et al. (2019) developed a machine learning method to estimate PM_{2.5} concentrations in China from 2000–2016. They reported that overall trends of PM_{2.5} in China were 2.097 µg/m³/year ($p < 0.001$), 0.299 µg/m³/year ($p > 0.05$), -4.511 µg/m³/year ($p < 0.001$) in 2000-2007, 2008-2013, and 2013-2016, respectively. Lin et al. (2018) estimated high-resolution PM_{2.5} in annual scale in China from 2001 to 2015, and found nation-scale trends of 0.04 µg/m³/year, -0.65 µg/m³/year, -2.33
10 µg/m³/year in 2001-2005, 2005-2010, and 2011-2015, respectively. Overall, our satellite-based PM_{2.5} trends are consistent with these two recent studies, except that we found no significant trend from 2005 to 2010 (0.41 µg/m³/year but $p > 0.05$), which is different from the study of Lin et al. (2018). The main reason could be that they did not include western China in their study area. And statistical significance levels were not reported in their study, which could not allow us to know
15 whether the trend was significant or not.

Although there have been several studies have studied the historical trends of PM_{2.5} in China, few has study the relations between the trends and air pollution control policies. This paper reviewed the air pollution control policies from 2005 to 2017. And for the first time we gave an overall evaluation of the effects of these policies on PM_{2.5} pollution improvement in China from the
20 perspective of satellite remote sensing. Results show that our satellite PM_{2.5} dataset is a good source to evaluate the performance of air pollution policies. The trends of satellite-derived PM_{2.5} concentrations is consistent with the implementation of air pollution control policies in different periods.

The ECER policy implemented in 11th FYP period (see Table 1 and Section 4.3) had co-
25 benefits on PM_{2.5} pollution control. The overall PM_{2.5} pollution had certain decrease (-0.56 µg/m³/year for entire China, $p = 0.053$) after 2007, but the effects were limited. The Environmental

Protection Plan and ECER policy during 12th FYP period were basically the extension of 11th FYP policy, with additional total emission control on NO_x. However, the total emission control oriented policy had shown its limitation. The PM_{2.5} concentrations of polluted areas did not decrease from 2010 to 2013 (e.g., -0.45 μg/m³/year for Jingjinji Region, $p=0.783$).

5 To address the PM_{2.5} pollution issue, China implemented two strict policies: the 12th FYP on APPC-KR in 2012 and APPC-AP in 2013. The goal of air quality improvement was proposed for the first time. Besides, China incorporated PM_{2.5} as a major control pollutant into the National Ambient Air Quality Standard (GB 3095-2012). All these polices (details can be found in Table 1 and Sections 4.4 and 4.5) had led to dramatic decreases of PM_{2.5} after 2013 (-4.27 μg/m³/year for
10 entire China, $p<0.001$). And the implementation of these policies was also an important mark that environmental management in China began to change from total emission control oriented mode to environmental quality improvement oriented mode.

It should be noted that inter-annual variation in meteorology has also contributed to the changes in PM_{2.5}. A recent study shows that meteorological conditions contributed approximately
15 20% of the PM_{2.5} reduction in Beijing from 2013 to 2017, while the control of anthropogenic emissions contributed 80% (Chen et al., 2019). In addition, the slowdown of economic development after financial crisis in 2008 might contribute to the PM_{2.5} emissions reduction. According to China Statistical Yearbook (NBS, 2018), the gross domestic products (GDP) growth rate decreased from
20 14.2% in 2007 to 6.9% in 2017. However, the GDP growth rates are still relatively high at current stage (6%~7%). Contrarily, the PM_{2.5} concentrations have decreased dramatically. Without effective air pollution control policies, the PM_{2.5} pollution level would not decrease rapidly. Therefore, effective air pollution control policy was the main reason for PM_{2.5} pollution reduction after 2013. Meteorological conditions also contributed a small portion of PM_{2.5} reductions.

The trends in PM_{2.5} concentrations in China also showed spatial heterogeneity. Multiple
25 reasons may explain the regional differences, e.g., the pollution levels of base year, the regional differences of industrial structures, the spatial heterogeneity of anthropogenic and natural emissions, economic and industry development differences, variations of regional policies, and variations of

meteorological conditions, etc.

Currently, China has achieved great success in PM_{2.5} pollution control. However, PM_{2.5} concentrations in many areas are still much higher than Level 2 annual PM_{2.5} standard of 35 µg/m³ of GB 3095-2012, which is corresponding to WHO Air Quality IT-1 level. China has implemented
5 a new air pollution control policy from 2018, i.e., the Three-year Action Plan to Win Battle for Blue Skies (2018-2020). China's air quality is expected to be further improved in the next three years.

This study extended the satellite PM_{2.5} dataset in our previous study (Ma et al., 2016) to the year of 2017 and obtained longer time series of satellite PM_{2.5} data, which can provide more spatially-resolved and high accurate PM_{2.5} data for epidemiological, health impact assessment, and
10 social economic impact studies in China.

Authors contributions

J. B. conceived and designed the study. R. L. collected and processed the data. Z. M. and Y. L. performed statistical modeling for satellite PM_{2.5} predictions. Z. M. analyzed the spatiotemporal trends of PM_{2.5} concentrations. J. B. prepared and analyzed the air pollution control policies. Z. M.
15 prepared the manuscript with contributions from all co-authors.

Acknowledgment

This work was supported by the National Natural Science Foundation of China (91644220, 71433007, and 41601546), and the Fundamental Research Funds for the Central Universities of
20 China (0211-14380078).

References

Boys, B., Martin, R., van Donkelaar, A., MacDonell, R., Hsu, C., Cooper, M., Yantosca, R., Lu, Z., Streets, D. G., Zhang, Q., and Wang, S.: Fifteen-year global time series of satellite-derived fine particulate matter, *Environ. Sci. Technol.*, 48, 11109-11118, 2014.
25 CAAC: Key Projects of "Twelfth Five-Year Plan" on Air Pollution Prevention and Control in Key Regions-Sample English Translation, Clean Air Alliance of China, 2013a.

- CAAC: English Translation of Air Pollution Prevention and Control Action Plan, Clean Air Alliance of China, 2013b.
- CAAC: English Translation of “Twelfth Five-Year Plan” on Air Pollution Prevention and Control in Key Regions, Clean Air Alliance of China, 2013c.
- 5 Chen, B., Kan, H., Chen, R., Jiang, S., and Hong, C.: Air pollution and health studies in China—policy implications, *J. Air Waste Manag. Assoc.*, 61, 1292-1299, 2011.
- Chen, S., and Jin, H.: Pricing for the clean air: Evidence from Chinese housing market, *J. Clean Prod.*, 206, 297-306, 2019.
- 10 Chen, Z., Chen, D., Kwan, M., Chen, B., Cheng, N., Gao, B., Zhuang, Y., Li, R., and Xu, B.: The control of anthropogenic emissions contributed to 80 % of the decrease in PM_{2.5} concentrations in Beijing from 2013 to 2017, *Atmos. Chem. Phys. Discuss.*, 2019, 1-25, 2019.
- Crouse, D. L., Peters, P. A., van Donkelaar, A., Goldberg, M. S., Villeneuve, P. J., Brion, O., Khan, S., Atari, D. O., Jerrett, M., and Pope, C.: Risk of nonaccidental and cardiovascular mortality in relation to long-term exposure to low concentrations of fine particulate matter: a Canadian national-level cohort study, *Environ. Health Perspect.*, 120, 708-714, 2012.
- 15 Dominici, F., Peng, R. D., Bell, M. L., Pham, L., McDermott, A., Zeger, S. L., and Samet, J. M.: Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases, *JAMA-J. Am. Med. Assoc.*, 295, 1127-1134, 2006.
- Geng, G., Zhang, Q., Martin, R. V., van Donkelaar, A., Huo, H., Che, H., Lin, J., and He, K.: 20 Estimating long-term PM_{2.5} concentrations in China using satellite-based aerosol optical depth and a chemical transport model, *Remote Sens. Environ.*, 166, 262-270, 2015.
- Hoff, R. M., and Christopher, S. A.: Remote sensing of particulate pollution from space: have we reached the promised land?, *J. Air Waste Manag. Assoc.*, 59, 645-675, 2009.
- 25 Hsu, N. C., Gautam, R., Sayer, A. M., Bettenhausen, C., Li, C., Jeong, M. J., Tsay, S. C., and Holben, B. N.: Global and regional trends of aerosol optical depth over land and ocean using SeaWiFS measurements from 1997 to 2010, *Atmos. Chem. Phys.*, 12, 8037-8053, 2012.
- Hu, H., Yang, Q., Lu, X., Wang, W., Wang, S., and Fan, M.: Air Pollution and Control in Different Areas of China, *Crit. Rev. Env. Sci. Technol.*, 40, 452-518, 2010.
- 30 Hu, X., Waller, L., Lyapustin, A., Wang, Y., and Liu, Y.: 10-year spatial and temporal trends of PM_{2.5} concentrations in the southeastern US estimated using high-resolution satellite data, *Atmos. Chem. Phys.*, 14, 6301-6314, 2014.
- Huang, K., Xiao, Q., Meng, X., Geng, G., Wang, Y., Lyapustin, A., Gu, D., and Liu, Y.: Predicting monthly high-resolution PM_{2.5} concentrations with random forest model in the North China Plain, *Environ. Pollut.*, 242, 675-683, 2018.
- 35 Itahashi, S., Uno, I., Yumimoto, K., Irie, H., Osada, K., Ogata, K., Fukushima, H., Wang, Z., and Ohara, T.: Interannual variation in the fine-mode MODIS aerosol optical depth and its relationship to the changes in sulfur dioxide emissions in China between 2000 and 2010, *Atmos. Chem. Phys.*, 12, 2631-2640, 2012.
- Jin, Y., Andersson, H., and Zhang, S.: Air pollution control policies in China: a retrospective and 40 prospects, *Int. J. Environ. Res. Public Health*, 13, 1219, 2016.
- Kloog, I., Koutrakis, P., Coull, B. A., Lee, H. J., and Schwartz, J.: Assessing temporally and spatially resolved PM_{2.5} exposures for epidemiological studies using satellite aerosol optical depth measurements, *Atmos. Environ.*, 45, 6267-6275, 2011.
- 45 Kloog, I., Nordio, F., Coull, B. A., and Schwartz, J.: Incorporating local land use regression and satellite aerosol optical depth in a hybrid model of spatiotemporal PM_{2.5} exposures in the Mid-Atlantic states, *Environ. Sci. Technol.*, 46, 11913-11921, 2012.
- Krotkov, N. A., McLinden, C. A., Li, C., Lamsal, L. N., Celarier, E. A., Marchenko, S. V., Swartz, W. H., Bucsela, E. J., Joiner, J., Duncan, B. N., Boersma, K. F., Veefkind, J. P., Levelt, P. F., Fioletov, V. E., Dickerson, R. R., He, H., Lu, Z., and Streets, D. G.: Aura OMI observations of 50 regional SO₂ and NO₂ pollution changes from 2005 to 2015, *Atmos. Chem. Phys.*, 16, 4605-4629, 2016.
- Li, L., Wang, W., Feng, J., Zhang, D., Li, H., Gu, Z., Wang, B., Sheng, G., and Fu, J.: Composition, source, mass closure of PM_{2.5} aerosols for four forests in eastern China, *Journal of Environmental Sciences*, 22, 405-412, 2010.

- Li, W., Bai, Z., Liu, A., Chen, J., and Chen, L.: Characteristics of Major PM_{2.5} Components during Winter in Tianjin, China, *Aerosol Air Qual. Res.*, 9, 105-119, 2009.
- Lin, C., Liu, G., Lau, A. K. H., Li, Y., Li, C. C., Fung, J. C. H., and Lao, X. Q.: High-resolution satellite remote sensing of provincial PM_{2.5} trends in China from 2001 to 2015, *Atmos. Environ.*, 180, 110-116, 2018.
- 5 Liu, C., Yang, C., Zhao, Y., Ma, Z., Bi, J., Liu, Y., Meng, X., Wang, Y., Cai, J., and Kan, H.: Associations between long-term exposure to ambient particulate air pollution and type 2 diabetes prevalence, blood glucose and glycosylated hemoglobin levels in China, *Environ. Int.*, 92, 416-421, 2016a.
- 10 Liu, J., Li, W., and Li, J.: Quality screening for air quality monitoring data in China, *Environ. Pollut.*, 216, 720-723, 2016b.
- Liu, M., Bi, J., and Ma, Z.: Visibility-based PM_{2.5} concentrations in China: 1957–1964 and 1973–2014, *Environ. Sci. Technol.*, 51, 13161-13169, 2017a.
- 15 Liu, M., Huang, Y., Ma, Z., Jin, Z., Liu, X., Wang, H., Liu, Y., Wang, J., Jantunen, M., Bi, J., and Kinney, P. L.: Spatial and temporal trends in the mortality burden of air pollution in China: 2004–2012, *Environ. Int.*, 98, 75-81, 2017b.
- Liu, Y.: New Directions: Satellite driven PM_{2.5} exposure models to support targeted particle pollution health effects research, *Atmos. Environ.*, 68, 52-53, 2013.
- 20 Liu, Y.: Monitoring PM_{2.5} from space for health: past, present, and future directions, *Environ. Manager*, 2, 6-10, 2014.
- Ma, Z., Hu, X., Sayer, A. M., Levy, R., Zhang, Q., Xue, Y., Tong, S., Bi, J., Huang, L., and Liu, Y.: Satellite-Based Spatiotemporal Trends in PM_{2.5} Concentrations: China, 2004–2013, *Environ. Health Perspect.*, 124 184-192, 2016.
- 25 Miyazaki, K., Eskes, H., Sudo, K., Boersma, K. F., Bowman, K., and Kanaya, Y.: Decadal changes in global surface NO_x emissions from multi-constituent satellite data assimilation, *Atmos. Chem. Phys.*, 17, 807-837, 2017.
- NBS: China Statistical Yearbook, National Bureau of Statistics of China, 2018.
- Pathak, R. K., Wu, W. S., and Wang, T.: Summertime PM_{_{2.5}} ionic species in four major cities of China: nitrate formation in an ammonia-deficient atmosphere, *Atmos. Chem. Phys.*, 9, 1711-1722, 2009.
- 30 Philip, S., Martin, R. V., van Donkelaar, A., Lo, J. W.-H., Wang, Y., Chen, D., Zhang, L., Kasibhatla, P. S., Wang, S., Zhang, Q., Lu, Z., Streets, D. G., Bittman, S., and Macdonald, D. J.: Global Chemical Composition of Ambient Fine Particulate Matter for Exposure Assessment, *Environ. Sci. Technol.*, 48, 13060-13068, 2014.
- 35 Pope, C. A., Burnett, R. T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K., and Thurston, G. D.: Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution, *JAMA-J. Am. Med. Assoc.*, 287, 1132-1141, 2002.
- Rohde, R. A., and Muller, R. A.: Air Pollution in China: Mapping of Concentrations and Sources, *PLoS One*, 10, e0135749, 2015.
- 40 Schreifels, J. J., Fu, Y., and Wilson, E. J.: Sulfur dioxide control in China: policy evolution during the 10th and 11th Five-year Plans and lessons for the future, *Energy Policy*, 48, 779-789, 2012.
- Silver, B., Reddington, C. L., Arnold, S. R., and Spracklen, D. V.: Substantial changes in air pollution across China during 2015–2017, *Environ. Res. Lett.*, 13, 114012, 2018.
- 45 van Donkelaar, A., Martin, R. V., Brauer, M., and Boys, B. L.: Use of satellite observations for long-term exposure assessment of global concentrations of fine particulate matter, *Environ. Health Perspect.*, 123, 135-143, 2015.
- van Donkelaar, A., Martin, R. V., Brauer, M., Hsu, N. C., Kahn, R. A., Levy, R. C., Lyapustin, A., Sayer, A. M., and Winker, D. M.: Global Estimates of Fine Particulate Matter using a Combined Geophysical-Statistical Method with Information from Satellites, Models, and Monitors, *Environ. Sci. Technol.*, 50, 3762-3772, 2016.
- 50 Wang, C., Xu, J., Yang, L., Xu, Y., Zhang, X., Bai, C., Kang, J., Ran, P., Shen, H., and Wen, F.: Prevalence and risk factors of chronic obstructive pulmonary disease in China (the China Pulmonary Health [CPH] study): a national cross-sectional study, *The Lancet*, 391, 1706-1717, 2018a.

- Wang, Q., Wang, J., He, M. Z., Kinney, P. L., and Li, T.: A county-level estimate of PM_{2.5} related chronic mortality risk in China based on multi-model exposure data, *Environ. Int.*, 110, 105-112, 2018b.
- 5 Weatherhead, E. C., Reinsel, G. C., Tiao, G. C., Meng, X. L., Choi, D., Cheang, W. K., Keller, T., DeLuisi, J., Wuebbles, D. J., and Kerr, J. B.: Factors affecting the detection of trends: Statistical considerations and applications to environmental data, *J. Geophys. Res. Atmos.*, 103, 17149-17161, 1998.
- 10 Xiao, Q., Chang, H. H., Geng, G., and Liu, Y.: An ensemble machine-learning model to predict historical PM_{2.5} concentrations in China from satellite data, *Environ. Sci. Technol.*, 52, 13260-13269, 2018.
- Xue, B., Mitchell, B., Geng, Y., Ren, W., Müller, K., Ma, Z., Puppim de Oliveira, J. A., Fujita, T., and Tobias, M.: A review on China's pollutant emissions reduction assessment, *Ecol. Indic.*, 38, 272-278, 2014.
- 15 Xue, T., Zheng, Y., Tong, D., Zheng, B., Li, X., Zhu, T., and Zhang, Q.: Spatiotemporal continuous estimates of PM_{2.5} concentrations in China, 2000–2016: A machine learning method with inputs from satellites, chemical transport model, and ground observations, *Environ. Int.*, 123, 345-357, 2019.
- Yang, J., and Zhang, B.: Air pollution and healthcare expenditure: Implication for the benefit of air pollution control in China, *Environ. Int.*, 120, 443-455, 2018.
- 20 Yao, Q., Li, S. Q., Xu, H. W., Zhuo, J. K., and Song, Q.: Studies on formation and control of combustion particulate matter in China: A review, *Energy*, 34, 1296-1309, 2009.
- Zhang, J., and Reid, J. S.: A decadal regional and global trend analysis of the aerosol optical depth using a data-assimilation grade over-water MODIS and Level 2 MISR aerosol products, *Atmos. Chem. Phys.*, 10, 10949-10963, 2010.
- 25 Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, *Atmos. Chem. Phys.*, 18, 14095-14111, 2018.
- 30 Zheng, Y., Zhang, Q., Liu, Y., Geng, G., and He, K.: Estimating ground-level PM_{2.5} concentrations over three megalopolises in China using satellite-derived aerosol optical depth measurements, *Atmos. Environ.*, 124, 232–242, 2016.
- Zhou, K., Yang, S., Shen, C., Ding, S., and Sun, C.: Energy conservation and emission reduction of China's electric power industry, *Renew. Sustain. Energ. Rev.*, 45, 10-19, 2015.

35

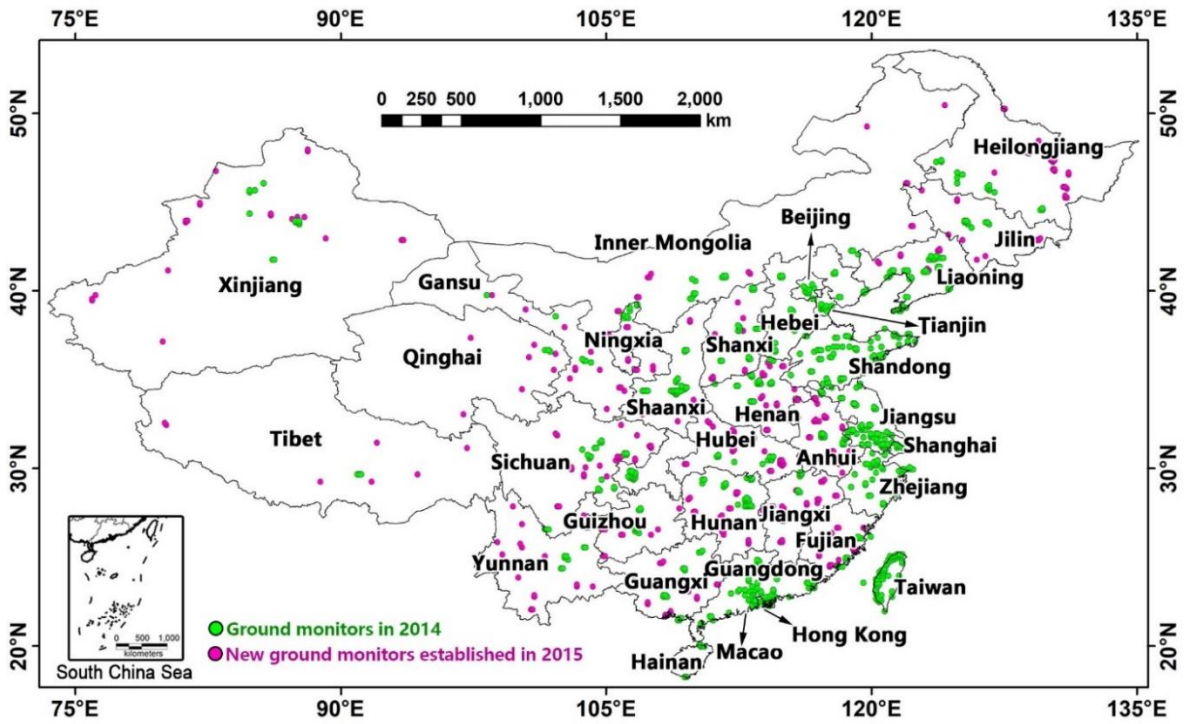


Figure 1. Spatial distributions of ground PM_{2.5} monitors involved in model fitting and validation. Red circles denote the ground monitor in 2014. Pink circles denote new ground monitors established in 2015.

5

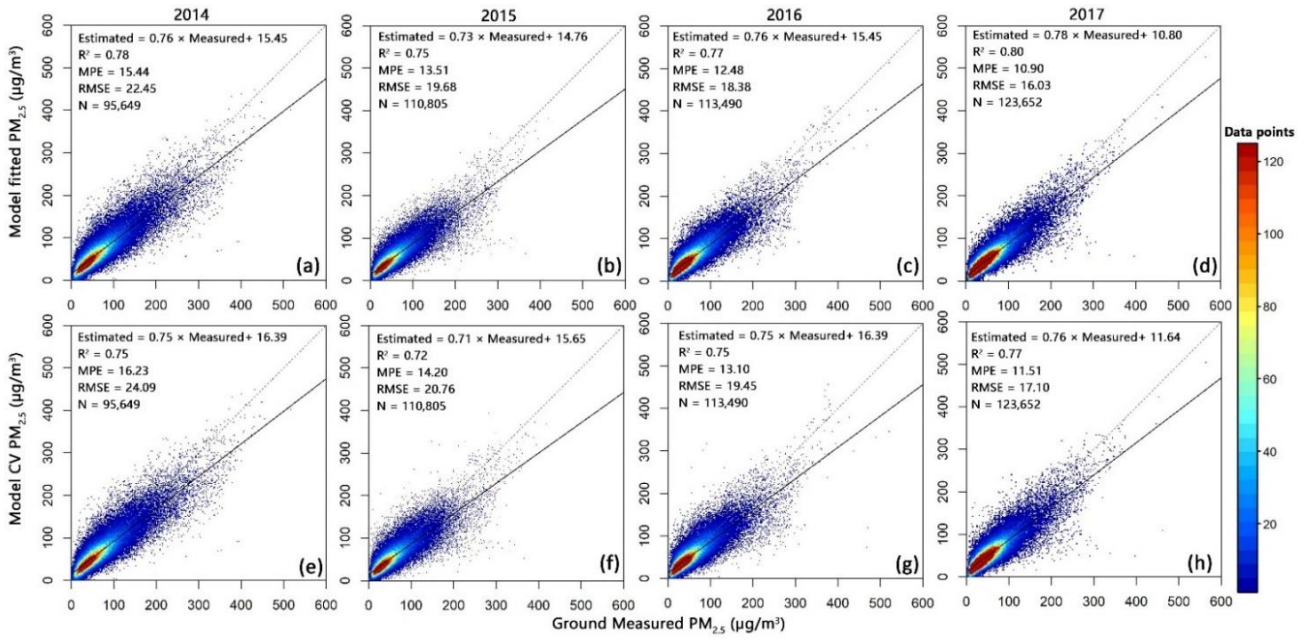


Figure 2. Model fitting (upper row) and cross validation (CV, lower row) results for satellite PM_{2.5} prediction models from 2014 to 2017.

5

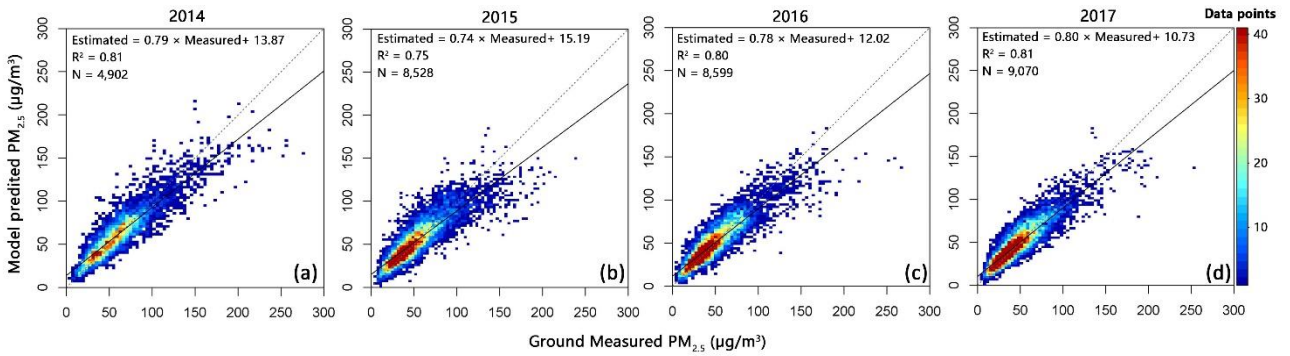


Figure 3. Validation of monthly mean PM_{2.5} predictions from 2014 to 2017.

10

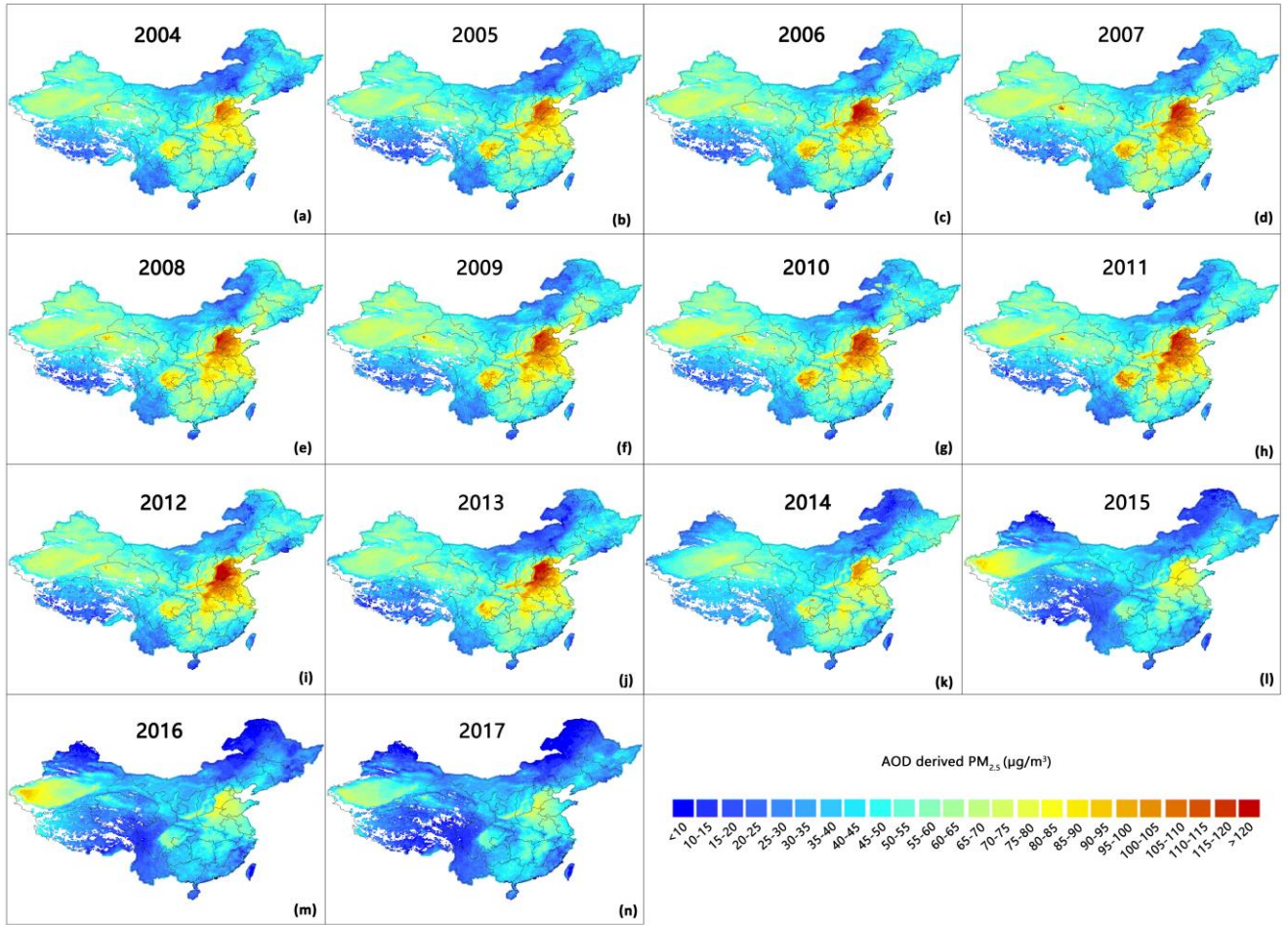
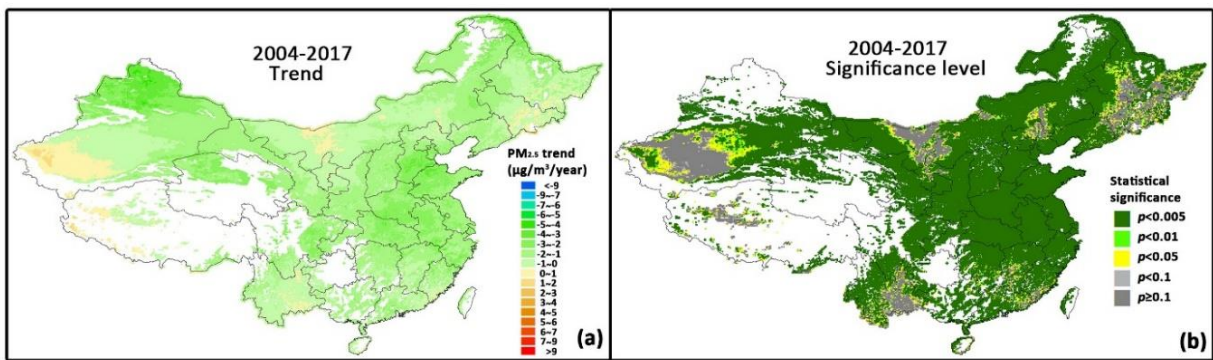


Figure 4. Spatial distributions of annual mean satellite-derived $PM_{2.5}$ concentrations from 2004 to 2017.



5

Figure 5. Spatial distributions of $PM_{2.5}$ trends and significance levels in China from 2004 to 2017.

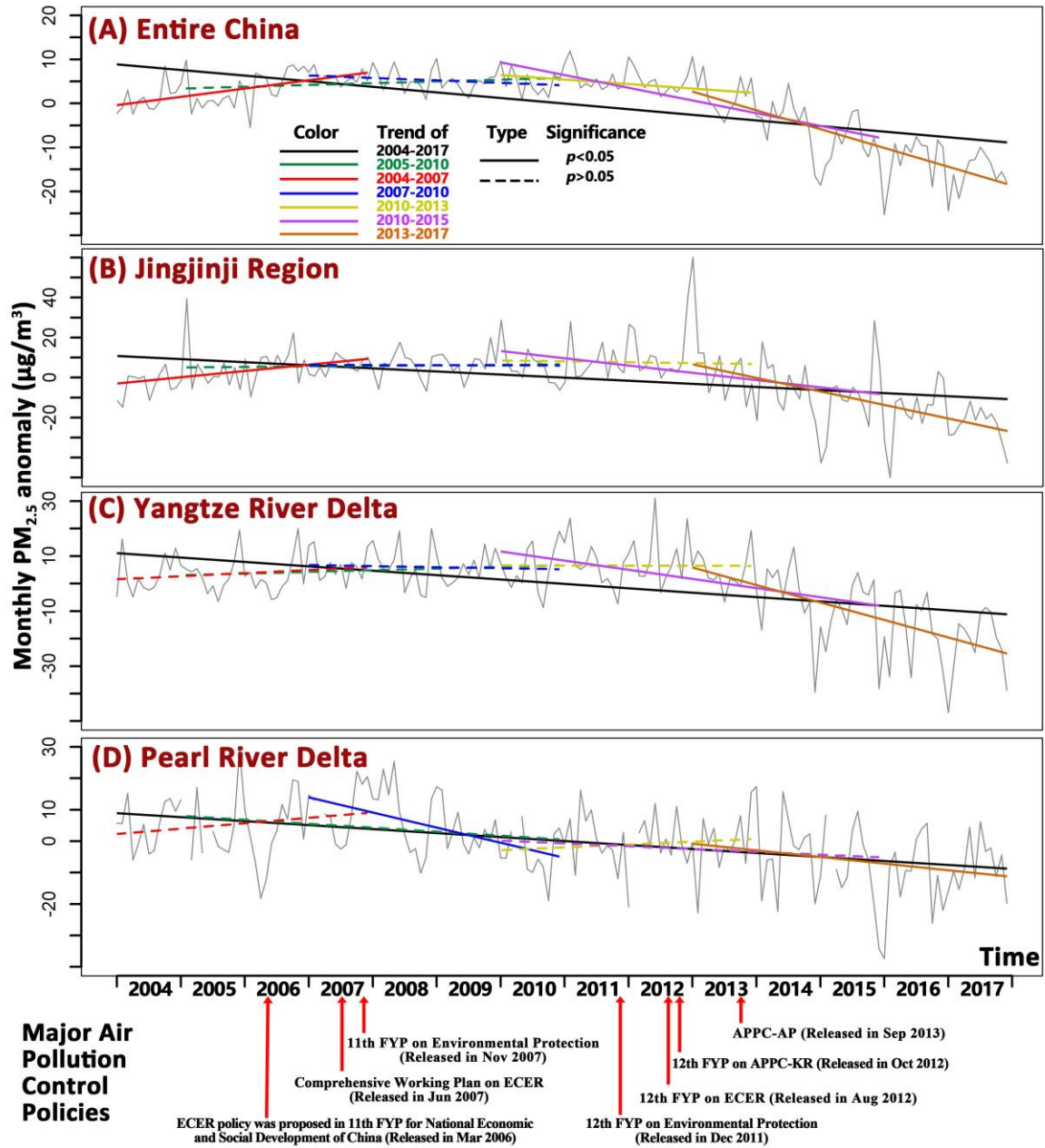


Figure 6. $PM_{2.5}$ trends of entire China, Jingjinji, Yangtze River Delta (YRD), and Pearl River Delta (PRD) Regions from 2004 to 2017, and corresponding air pollution control policies

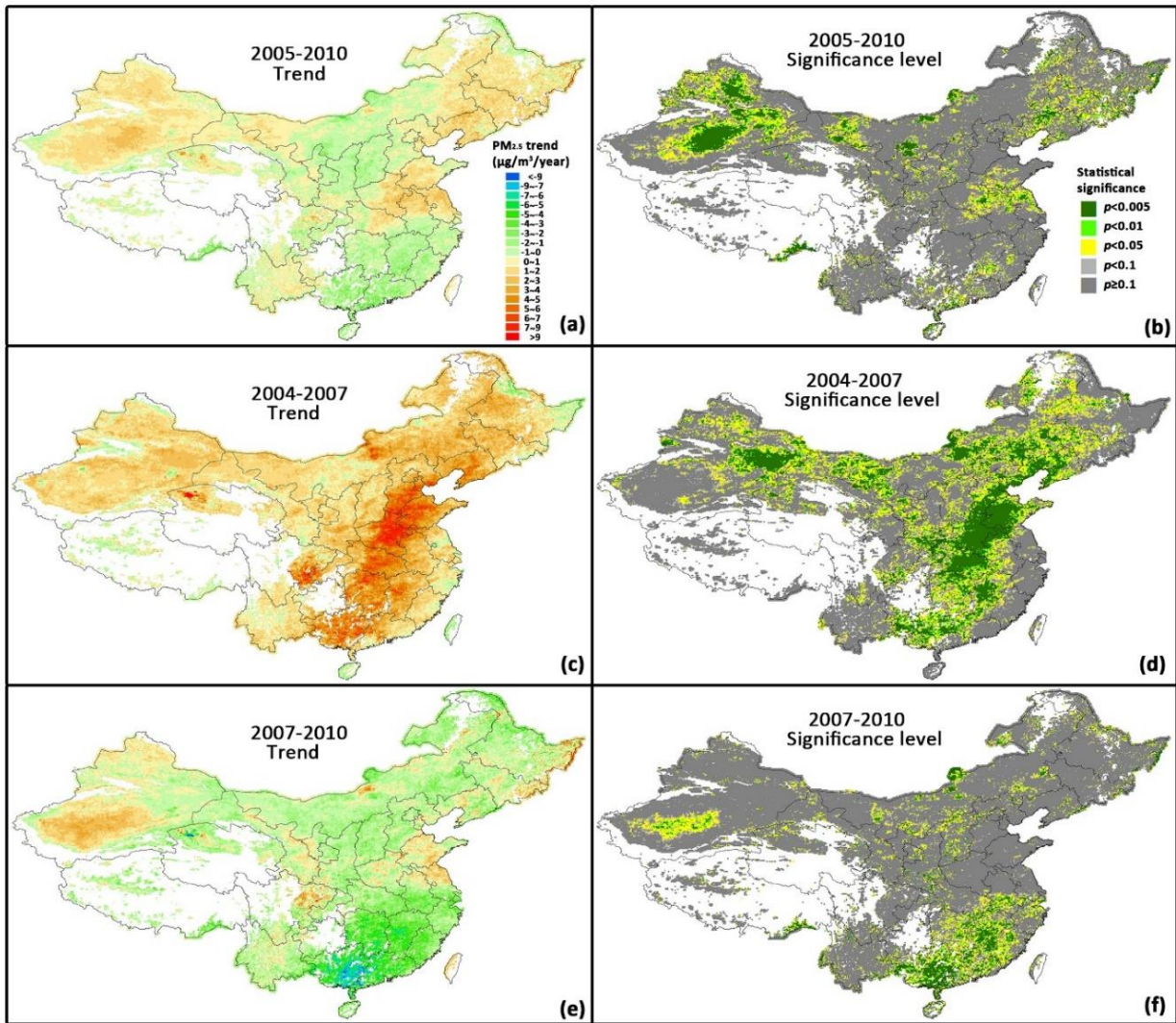


Figure 7. Spatial distributions of PM_{2.5} trends and significance levels in China from 2005 to 2010.

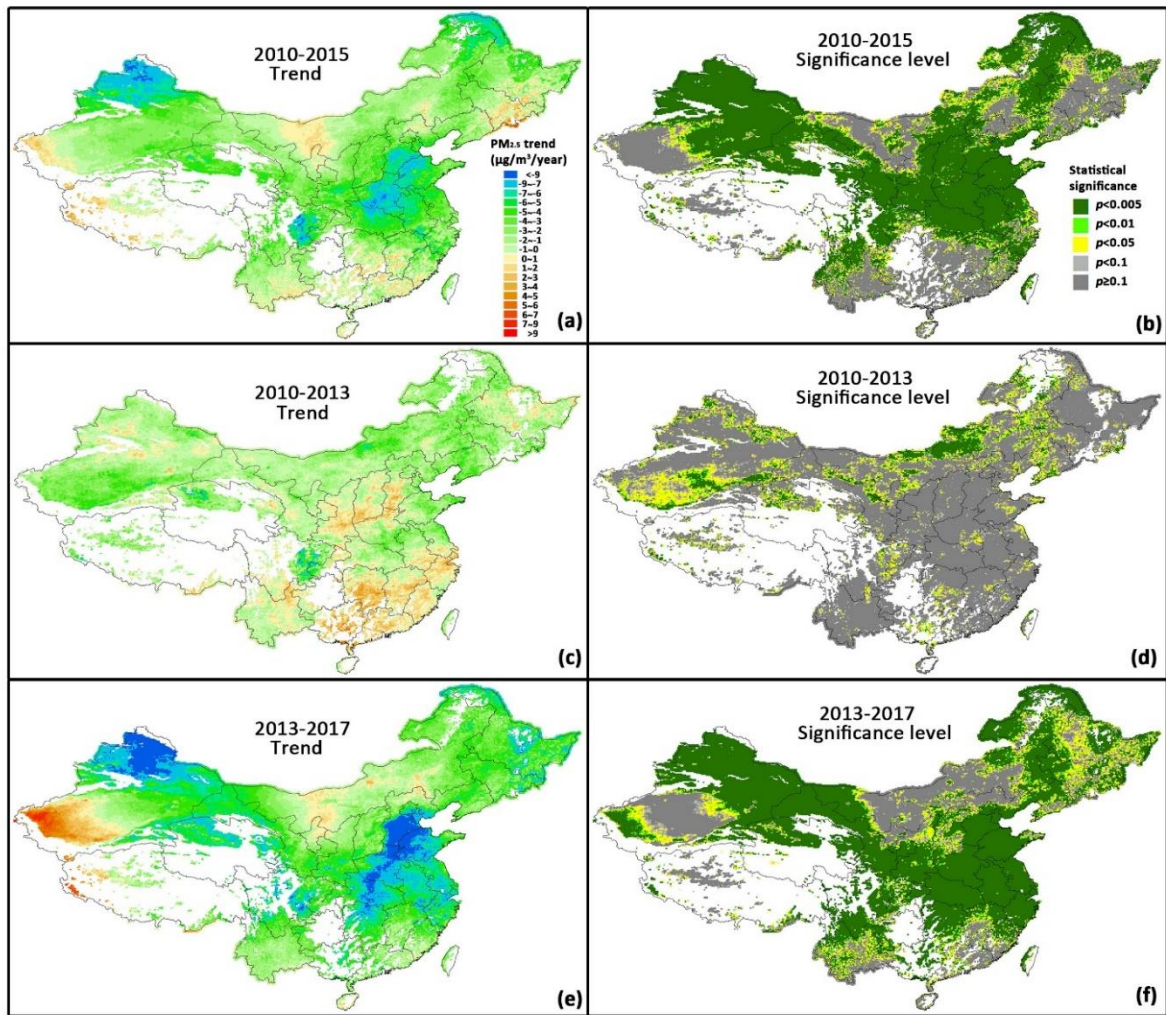


Figure 8. Spatial distributions of PM_{2.5} trends and significance levels in China from 2010 to 2017.

Table 1. Overview of major air pollution control policies in China from 2005 to 2017

Policy ^a	Base year	Implementation period	Major goals (Compared to base year)	Major measures
11 th FYP on Environmental Protection	2005	2006-2010	<ul style="list-style-type: none"> SO₂ emission should reduce by 10% 	<ul style="list-style-type: none"> Implement of desulphurization projects of coal-fired power plants Prevent and control urban PM₁₀ pollution, relocate pollution industrial plants in urban areas, control construction and road dust Implement total emission control policy for key industrial pollution sources, control emission of sulfur dioxide and soot (dust) Strengthen vehicle pollution prevention and control, improve quality and efficiency of gasoline
ECER during 11 th FYP	2005	2006-2010	<ul style="list-style-type: none"> Energy consumption per GDP capita should decrease by 20% SO₂ emission should reduce by 10% 	<ul style="list-style-type: none"> Promote industrial and energy structure adjustment, restrain the development of industries with high energy consumption and pollution, eliminate backward production capacity, promote production capacity with low energy consumption and low pollution Implement ten major energy conservation projects, implement desulphurization projects of coal-fired power plants
12 th FYP on Environmental Protection	2010	2011-2015	<ul style="list-style-type: none"> SO₂ emission should reduce by 8% NO_x emission should reduce by 10% 	<ul style="list-style-type: none"> Implement desulphurization and denitration facilities for coal-fired power sector and major industrial sectors Control NO_x emissions of vehicles and ships Deepen PM and VOCs pollution control Promote urban air pollution prevention and control, implement coordinated control of various pollutants in key areas, monitor PM_{2.5} and O₃ in Jingjinji, Yangtze River Delta, and Pearl River Delta regions
ECER during 12 th FYP	2010	2011-2015	<ul style="list-style-type: none"> Energy consumption per GDP capita should decrease by 16% SO₂ emission should reduce by 8% NO_x emission should reduce by 10% 	<ul style="list-style-type: none"> Adjust and optimize industrial structure, control the development of industries with high energy consumption and pollution, eliminate backward production capacity Adjust energy consumption structure, strengthen energy conservation for industrial, building, transportation, commercial and civil areas, etc. Strengthen emissions reduction in key industrial sectors, promote desulphurization and denitration, control emissions of vehicles, promote the control of PM_{2.5}
The 12 th FYP on APPC-KR ^b	2010	2011-2015	<ul style="list-style-type: none"> Emission of the SO₂, NO_x, and industrial PM should decrease 12%, 13%, and 10%, respectively The annual average concentration of PM₁₀, SO₂, NO₂ and PM_{2.5} should decrease by 10%, 10%, 7%, and 5%, respectively 	<ul style="list-style-type: none"> Identify the key regions and implement regional specific management Strictly control high energy consumption and high pollution projects, control new pollutants emissions, implement strict emission standard, and enhance control requirements of VOCs in key regions Strengthen elimination of backward production capacity, optimize industrial layout Optimize energy consumption structure, develop clean energy, control total coal consumption, establish restricted zones for high polluting fuels, eliminate small coal boilers, promote clean and efficient utilization of coal Comprehensively implement co-control of multiple pollutants (SO₂, NO_x, PM, VOCs), strengthen vehicle pollution prevention and control Innovate regional management mechanism, establish joint regional prevention and control coordination mechanism, establish and perfect ground monitoring network
APPC-AP	2012	2013-2017	<ul style="list-style-type: none"> PM_{2.5} concentrations of Jingjinji, Yangtze River Delta, and Pearl River Delta regions should reduce by 25%, 20%, and 15% respectively PM_{2.5} concentrations of Beijing should be controlled at around 60 µg/m³ 	<ul style="list-style-type: none"> Enhance comprehensive air pollution control on industrial enterprises, deepen non-point source control, strengthen vehicle pollution control Adjust, optimize, and upgrade industrial structure, strictly control new capacity with high energy consumption and high pollution, accelerate elimination of backward production capacity, reduce excess capacity Accelerate energy structure adjustment, accelerate utilization of clean energy, control total coal consumption, promote clean utilization of coal, improve energy efficiency Optimize industrial layout Utilize the market mechanism, improve the pricing and tax policy, establish regional coordination mechanism Establish monitoring, early warning, and emergency system for heavy pollution episodes

^a Abbreviations: FYP: Five Year Plan; ECER: Energy Conservation and Emissions Reduction; APPC-KR: Air Pollution Prevention and Control in Key Regions; APPC-AP: Action Plan of Air Pollution Prevention and Control

^b The key regions are shown in Figure S1 (Supplemental Materials)

Table 2. Trends and 95% confidence intervals (CI) of PM_{2.5} concentrations for entire China, Jingjinji, Yangtze River Delta, and Pearl River Delta Regions from 2004 to 2017

Period	Trend	Entire China	Jingjinji Region	Yangtze River Delta	Pearl River Delta
2004-2017	Trend ($\mu\text{g}/\text{m}^3/\text{year}$)	-1.27	-1.55	-1.60	-1.27
	95% CI ($\mu\text{g}/\text{m}^3/\text{year}$)	(-1.50, -1.04)	(-2.06, -1.03)	(-2.02, -1.18)	(-1.66, -0.88)
	Significance	$p<0.001$	$p<0.001$	$p<0.001$	$p<0.001$
2005-2010	Trend ($\mu\text{g}/\text{m}^3/\text{year}$)	0.41	0.26	0.61	-1.26
	95% CI ($\mu\text{g}/\text{m}^3/\text{year}$)	(-0.01, 0.82)	(-0.83, 1.36)	(-0.31, 1.54)	(-2.73, 0.21)
	Significance	$p=0.055$	$p=0.633$	$p=0.191$	$p=0.091$
2004-2007	Trend ($\mu\text{g}/\text{m}^3/\text{year}$)	1.88	3.14	1.12	1.72
	95% CI ($\mu\text{g}/\text{m}^3/\text{year}$)	(1.12, 2.64)	(1.07, 5.22)	(-0.51, 2.74)	(-0.79, 4.23)
	Significance	$p<0.001$	$p<0.005$	$p=0.174$	$p=0.174$
2007-2010	Trend ($\mu\text{g}/\text{m}^3/\text{year}$)	-0.56	-0.08	-0.37	-4.81
	95% CI ($\mu\text{g}/\text{m}^3/\text{year}$)	(-1.12, 0.01)	(-1.80, 1.64)	(-2.10, 1.35)	(-7.06, -2.55)
	Significance	$p=0.053$	$p=0.927$	$p=0.664$	$p<0.001$
2010-2013	Trend ($\mu\text{g}/\text{m}^3/\text{year}$)	-1.03	-0.45	-0.04	0.89
	95% CI ($\mu\text{g}/\text{m}^3/\text{year}$)	(-1.84, -0.21)	(-3.73, 2.83)	(-2.16, 2.08)	(-1.34, 3.13)
	Significance	$p<0.050$	$p=0.783$	$p=0.970$	$p=0.425$
2010-2015	Trend ($\mu\text{g}/\text{m}^3/\text{year}$)	-2.89	-3.63	-3.33	-0.90
	95% CI ($\mu\text{g}/\text{m}^3/\text{year}$)	(-3.50, -2.28)	(-5.59, -1.68)	(-4.76, -1.89)	(-2.34, 0.54)
	Significance	$p<0.001$	$p<0.001$	$p<0.001$	$p=0.219$
2013-2017	Trend ($\mu\text{g}/\text{m}^3/\text{year}$)	-4.27	-6.77	-6.36	-2.11
	95% CI ($\mu\text{g}/\text{m}^3/\text{year}$)	(-5.20, -3.34)	(-9.46, -4.07)	(-8.38, -4.34)	(-4.14, -0.09)
	Significance	$p<0.001$	$p<0.001$	$p<0.001$	$p<0.050$

5

10

Table 3. Goals accomplishments for key regions of 12th FYP on APPC-KR

Region	Goal (Decreased by)	Average satellite PM _{2.5} concentrations			Population weighted average satellite PM _{2.5} concentrations		
		2010 (µg/m ³)	2015 (µg/m ³)	Decreased by	2010 (µg/m ³)	2015 (µg/m ³)	Decreased by
Beijing	15%	68.75	58.47	14.9%	83.41	70.61	15.3%
Tianjin	6%	97.17	75.17	22.6%	96.13	76.09	20.8%
Hebei	6%	74.72	58.19	22.1%	101.25	75.15	25.8%
Shanghai	6%	66.41	58.83	11.4%	64.30	60.67	5.7%
Jiangsu	7%	81.23	62.24	23.4%	82.18	63.19	23.1%
Zhejiang	5%	52.85	38.73	26.7%	58.68	47.37	19.3%
Pearl River Delta	5%	45.00	37.97	15.6%	50.07	40.99	18.1%
Central Liaoning	6%	58.10	53.00	8.8%	64.97	58.40	10.1%
Shandong	7%	94.57	71.83	24.0%	97.83	73.76	24.6%
Wuhan Region	5%	75.02	55.41	26.1%	79.86	58.62	26.6%
Changzhutan Region	5%	64.81	52.75	18.6%	72.32	60.19	16.8%
Chongqing	6%	65.89	47.48	27.9%	77.36	52.71	31.9%
Chengdu Region	5%	83.55	52.22	37.5%	92.22	57.40	37.8%
Fujian	4%	37.42	28.02	25.1%	34.48	29.22	15.3%
Central and Northern Shanxi	4%	53.76	40.05	25.5%	63.05	46.78	25.8%
Guanzhong	4%	65.91	45.33	31.2%	79.54	53.91	32.2%
Lanzhou Region	4%	55.42	45.31	18.2%	62.47	47.77	23.5%
Yinchuan	5%	42.81	48.14	-12.4%	46.51	51.81	-11.4%
Urumqi Region	4%	60.26	27.83	53.8%	65.80	36.05	45.2%

Table 4. Goal accomplishments of APPC-AP (2013-2017)

Region	Goal (Decreased by)	Official assessment results ^a	Average satellite PM _{2.5} concentrations			Population weighted average satellite PM _{2.5} concentrations		
			2013 ($\mu\text{g}/\text{m}^3$)	2017 ($\mu\text{g}/\text{m}^3$)	Decreased by	2013 ($\mu\text{g}/\text{m}^3$)	2017 ($\mu\text{g}/\text{m}^3$)	Decreased by
Jingjinji	25%	39.6%	76.01	47.98	36.9%	100.91	60.97	39.6%
Yangtze River Delta	20%	34.3%	66.60	41.87	37.1%	71.98	46.45	35.5%
Pearl River Delta	15%	27.7%	45.15	38.84	14.0%	49.96	40.37	19.2%
Beijing	Be controlled at around 60 $\mu\text{g}/\text{m}^3$	58 $\mu\text{g}/\text{m}^3$	68.20	44.67	34.5%	82.69	55.07	33.4%

^a See http://www.mee.gov.cn/gkml/sthjbgw/stbgth/201806/t20180601_442262.htm, accessed on Mar 29, 2019