Meteorological Parameters and Gaseous Pollutant Concentrations as Predictors of Ground-level PM_{2.5} Concentrations in the Beijing-Tianjin-Hebei Region, China

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ABSTRACT

Ground-level PM_{2.5} concentrations—especially those during episodes of heavy pollution—are severely underestimated by mixed-effects models that ignore the effects of primary pollutant emissions and secondary pollutant conversion. In this work, meteorological parameters and NO₂, SO₂, CO, and O₃ concentrations are introduced as predictors to a mixed-effects model to improve the estimated concentration of PM_{2.5}, which is based on the Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol optical depth (AOD). The Beijing-Tianjin-Hebei (JingJinJi) region is used as the study area. The model provides an overall cross-validation (CV) R² of 0.84 and root-mean-square prediction error (RMSE) of 33.91 μg m⁻³. The CV R² and RMSE of the proposed model are higher by 0.11 and lower by 9.16 μg m⁻³, respectively, than those of a model lacking gaseous pollutants as predictors. The R² and RMSE of the proposed model increases and decreases by 0.14 and 13.37 μg m⁻³, respectively, when PM_{2.5} concentrations exceed 75 μg m⁻³. The high values predicted for the PM_{2.5} concentration indicate a drastic improvement in the estimation, and the spatial distribution generated by the model for periods of heavy pollution is highly consistent with that inferred from monitoring data. Thus, the proposed model can be used to generate highly accurate maps of the PM_{2.5} distribution for long-term and short-term exposure studies and to correctly classify exposure in heavily polluted areas.

Keywords: PM_{2.5}; Aerosol optical depth; Gaseous pollutant; Heavy pollution; Mixed-effects model.

INTRODUCTION

Epidemiological studies have illustrated that PM_{2.5}, defined as particulate matter with an aerodynamic diameter of < 2.5 μm, has adverse effects on human health (Song *et al.*, 2014). Long-term exposure studies have shown that the risks of ischemic heart disease (Crouse *et al.*, 2012), lung cancer (Pope III *et al.*, 2002), and cardiovascular mortality (Pope III *et al.*, 2004) increase by 16%, 8%, and 10%, respectively, with every 10 μg m⁻³ increment in the PM_{2.5} concentration. Short-term PM_{2.5} exposure studies have emphasized that every 10 μg m⁻³ elevation in the 2-day mean PM_{2.5} concentration increases the incidence of respiratory disease (Kloog *et al.*, 2012) and myocardial infarction (Zanobetti *et al.*, 2009) by 0.7% and 2.25%, respectively. Each 10 μg m⁻³ increment in the daily PM_{2.5} concentration is associated with elevated hospital admission rates and increases the incidence of heart failure (Dominici *et al.*, 2006), acute coronary syndrome (Belleudi

et al., 2010), and pediatric asthma (Strickland *et al.*, 2015) by 1.28%, 2.3%, and 1.3%, respectively. The accurate estimation of PM_{2.5} concentration is a prerequisite of studies on the effects of long-term and short-term PM_{2.5} exposure on human health.

The PM_{2.5} exposure level of a certain population has traditionally been estimated on the basis of PM_{2.5} concentration data acquired through ground monitoring over a certain distance (Laden *et al.*, 2006). However, the sparseness and uneven spatial distribution of PM_{2.5} monitoring sites will introduce uncertainty to the estimation of PM_{2.5} exposure and result in the underestimation of health risks (Hu *et al.*, 2014). Remotely sensed data for aerosol optical depth (AOD) have been widely used to estimate PM_{2.5} concentrations because of their spatial and temporal continuity (Donkelaar *et al.*, 2011). Different statistical models, such as linear regression models (Gupta *et al.*, 2006), generalized linear regression models (Liu *et al.*, 2007; You *et al.*, 2015), linear mixed models (Lee *et al.*, 2011; Xie *et al.*, 2015), geographically weighted regression models (Hu *et al.*, 2014; You *et al.*, 2016), generalized additive models (Paciorek *et al.*, 2008; Strawa *et al.*, 2013), and Bayesian statistical models (Chang *et al.*, 2014; Lv *et al.*, 2016), have been developed for the estimation of PM_{2.5} concentration from satellite-derived AOD data. Nevertheless,

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AOD-based methods severely underestimate high PM_{2.5} concentrations. To illustrate, PM_{2.5} concentrations exceeding 40 $\mu\text{g m}^{-3}$ in the United States and 60 $\mu\text{g m}^{-3}$ in China are severely underestimated by these methods (Liu *et al.*, 2007; Gupta and Christopher, 2009a; Ma *et al.*, 2014; Li *et al.*, 2017).

The underestimation of PM_{2.5} concentration will introduce uncertainty to long-term and short-term PM_{2.5} exposure studies. The Beijing-Tianjin-Hebei region (JingJinJi) has a large urban scale and human population and is characterized by high energy consumption (Wang *et al.*, 2016). Thus, residents of this region are exposed to high concentrations of pollutant emissions. In recent years, several incidences of extremely high PM_{2.5} concentrations that exceeded 500 $\mu\text{g m}^{-3}$ and that persisted for several days have been reported in the North China Plain, which is represented by the JingJinJi region (Andersson *et al.*, 2015). The underestimation of high PM_{2.5} concentrations in heavily polluted areas will increase the exposure risk associated with PM_{2.5} and result in the severe miscalculation of the effects of long-term and short-term PM_{2.5} exposure on public health. Therefore, improving the accuracy of PM_{2.5} estimation is crucial for reducing the misclassification of PM_{2.5} exposure levels and promoting epidemiological research in heavily polluted areas.

Early studies used AOD as the sole predictor of the surface PM_{2.5} concentration. AOD is the extinction coefficient of light that originates from particle scattering over the entirety of a vertical column. The PM_{2.5} concentration is defined as the mass concentration of dry particles measured near the surface of the column. Thus, AOD values and PM_{2.5} concentration are not strictly correlated (Chudnovsky *et al.*, 2013; Li *et al.*, 2015). Therefore, the AOD-based estimation of the PM_{2.5} concentration may be inaccurate (Saunders *et al.*, 2014). Meteorological parameters (MET), such as wind direction, wind speed, temperature, humidity, and boundary layer height, are used as predictors to improve the accuracy of surface PM_{2.5} concentration estimation (Paciorek *et al.*, 2008; Liu *et al.*, 2009). The characteristics of surface weather, however, are not the only determinants of air pollution development in the JingJinJi region (Miao *et al.*, 2015). In addition to unfavorable weather conditions, the excessive emission of primary pollutants and the secondary conversion of pollutants to particles are the main causes of heavy-pollution incidences in the JingJinJi region (Sun *et al.*, 2014; Wang *et al.*, 2014). Thus, additional predictors should be introduced to resolve the underestimation of high PM_{2.5} concentrations.

PM_{2.5} is a complex mixture of particles that is primarily composed of SO₄²⁻, NO₃⁻, NH₄⁺, elemental C, and organic C. The source emission of atmospheric pollutants and the conversion of gas pollutants are important sources of PM_{2.5} (Kloog *et al.*, 2012; Meng *et al.*, 2014). Numerous scholars have improved the accuracy of PM_{2.5} concentration estimation by introducing point emission and area-source emission data for North America to estimation models (Kloog *et al.*, 2012; Strawa *et al.*, 2013; Lee *et al.*, 2016). Nevertheless, the PM_{2.5} concentration in North America is lower than that in the JingJinJi region. Thus, the effects of

source data on the estimation of PM_{2.5} concentration have to be determined. The gaseous precursors of water-soluble inorganic salts include NO₂ and SO₂. These pollutants are homologous to primary pollutant emissions and can be oxidized into SO₄²⁻ and NO₃⁻ in the atmosphere. Atmospheric SO₄²⁻ and NO₃⁻, in turn, are the main sources of fine particulates in the JingJinJi region (Zhang *et al.*, 2007). Song *et al.* (2015) developed a statistical model for the PM_{2.5} concentration in Xi'an, China. Their model indicated that the PM_{2.5} concentration is strongly correlated with the concentrations of the gaseous pollutants (GASs) NO₂, SO₂, CO, and O₃. Thus, these pollutants can be used as auxiliary variables for PM_{2.5} prediction. Zheng *et al.* (2016) corrected the model for the estimation of the annual average PM_{2.5} concentration in the JingJinJi, Yangtze River, and Pearl River Delta regions by introducing the values for the annual average NO₂. Nevertheless, their approach is suitable only for the estimation of the long-term effects of PM_{2.5} exposure.

The mixed-effects model proposed by Lee *et al.* (2011) has greatly improved the accuracy of PM_{2.5} estimation by accounting for the temporal heterogeneity of PM_{2.5} concentrations and AOD values. In this work, we used AOD, MET, and GASs as predictive variables in the construction of a linear mixed model to resolve the underestimation of high PM_{2.5} concentrations. We take the JingJinJi region as the study area. Our approach increases the accuracy of PM_{2.5} concentration estimation and reduces PM_{2.5} exposure misclassification by accounting for the spatial and temporal distribution of PM_{2.5} concentrations. The rest of this paper is organized as follows: The study area, input datasets, and model structure are described in "Materials and Methods." The results for the model fitting, cross-validation (CV), and PM_{2.5} spatiotemporal distribution, and a comparison with other models are presented in "Results and Discussion."

MATERIALS AND METHODS

Study Area

The JingJinJi region is located north of the North China Plain, east of Taihang Mountain, and south of Yanshan. Its terrain is elevated in the northwest and depressed in the southeast (Fig. 1(b)). The JingJinJi region has an area of 218,000 km² and a population of 110,000,000. It is a core economic area in northern China. Energy consumption has gradually intensified in the region with the development of the national economy. The region has experienced several incidences of persistent heavy-pollution weather during which PM_{2.5} concentrations exceeded 500 $\mu\text{g m}^{-3}$.

Input Data

MODIS AOD Data

MODIS satellite data were retrieved from Terra and Aqua, the earth observation system satellites of the National Aeronautics and Space Administration (NASA). Terra and Aqua cross the equator at approximately 10:30 and 13:30 local time, respectively. Collection 6 (C6) is the latest version of MODIS Aerosol Data as of 2014. The Deep Blue algorithm (DB) of C6 has better AOD coverage and higher data quality than Collection 5.1 (Lee *et al.*, 2016). Therefore,

Fig. 1. Study area and the locations of PM_{2.5} monitoring sites and meteorological stations. (a) Location of JingJinJi in China, (b) Locations of meteorological stations and elevation of JingJinJi, and (c) Locations of PM_{2.5} monitoring sites.

the AOD data collected by Aqua C6 DB over the period of January 1, 2014 to December 31, 2014, were used in this work. The data had a resolution of 10 km (MODIS parameter name: Deep_Blue_Aerosol_Optical_Depth_550_land).

Ground Monitoring Data

The ground monitoring data used in this work included data for PM_{2.5}, NO₂, SO₂, CO, and O₃ concentrations. The hourly ground monitoring data collected from January 1, 2014 to December 31, 2014, were retrieved from the official website of the China Environmental Monitoring Center (CEMC; <http://113.108.142.147:20035/emcpublish/>) and the Beijing Environmental Monitoring Center (BJEMC;

<http://zx.bjmemc.com.cn/>). CEMC had 80 monitoring sites during the period of January 1 to April 31, 2014. After May 1, 23 monitoring sites were added in Beijing. Thus, CMEC and BJEMC had 103 monitoring sites in 2014. The distribution map of the PM_{2.5} monitoring sites is shown in Fig. 1(c).

PM_{2.5}, NO₂, SO₂, O₃, and CO data collected at 13:00 and 14:00 were extracted to match the satellite transit time and averaged as the data for PM_{2.5} prediction.

Meteorological Data

Hourly MET data collected over the period of January 1, 2014, to December 31, 2014, by 171 meteorological

monitoring stations (Fig. 1(b)) were retrieved from the Public Weather Service Center of the China Meteorological Administration. The data included wind direction, wind speed, pressure, temperature, and humidity. Daily 13:00 and 14:00 meteorological data were extracted to match the satellite transit time and averaged as the data for PM_{2.5} prediction.

Data Processing and Integration

Grid cells covering the whole JingJinJi region were first constructed with a resolution of 10 km for data integration. The 103 PM_{2.5} monitoring sites were matched to the corresponding grid cells on the basis of latitude and longitude, and the grid wherein PM_{2.5} monitoring sites were located was designated as the grid monitoring site. If multiple PM_{2.5} monitoring sites were located in the same grid, all monitoring site data located in that grid were averaged as the grid monitoring result. A total of 65 PM_{2.5} grid monitoring sites were finally included. The value of AOD that greater than 0 over a window size of 3 × 3 pixels centered at a given PM_{2.5} monitoring site was averaged as the matching result when PM_{2.5} and AOD data were matched. Meteorological monitoring data were assigned to each grid through the nearest-neighbor method to represent the meteorological conditions of the grid monitoring site. NO₂, SO₂, CO, and O₃ were interpolated onto the 10 km grid data through the Kriging method. DEM data with a resolution of 30 m were resampled into data with a resolution of 10 km and matched with the PM_{2.5} data of the grid monitoring site. In addition, the days with fewer than three matched data records were discarded during the data match process.

Method

Given the temporal heterogeneity of PM_{2.5} concentrations, AOD values, and GAS concentrations, a linear mixed model was developed to account for the daily variability in the relationship of PM_{2.5} with AOD and GASs. Furthermore, given the limited geographical area of the JingJinJi region, PM_{2.5}–(AOD, GAS) relationships were assumed to exhibit minimal spatial variations on a given day, and spatial nonstationary variations on a regional scale were ignored (Lee *et al.*, 2011; Zheng *et al.*, 2016). The linear mixed model that used AOD, GASs, and MET as predictors was designated the “AGM model” and is described below:

$$PM_{i,j} = (\beta_0 + \beta_{0,j}) + (\beta_1 + \beta_{1,j})AOD_{i,j} + (\beta_2 + \beta_{2,j})NO_{2i,j} + (\beta_3 + \beta_{3,j})SO_{2i,j} + (\beta_4 + \beta_{4,j})CO_{i,j} + (\beta_5 + \beta_{5,j})O_{3i,j} + \beta_6 PRS_{i,j} + \beta_7 WD_{i,j} + \beta_8 WS_{i,j} + \beta_9 TMP_{i,j} + \beta_{10} RH_{i,j} + \beta_{11} ALT_{i,j} + \varepsilon_{i,j}(\beta_{0,j}\beta_{1,j}\beta_{2,j}\beta_{3,j}\beta_{4,j}\beta_{5,j}) \sim N[(000000), \Psi] \quad (1)$$

where PM_{*i,j*} is the PM_{2.5} concentration at grid monitoring site *i* on day *j*; AOD_{*i,j*}, NO_{2*i,j*}, SO_{2*i,j*}, CO_{*i,j*}, O_{3*i,j*}, PRS_{*i,j*}, WD_{*i,j*}, WS_{*i,j*}, TMP_{*i,j*}, RH_{*i,j*}, and ALT_{*i,j*} are the AOD, NO₂, SO₂, CO, O₃, pressure, wind direction, wind speed, temperature, humidity, and elevation value at grid *i* on day *j*, respectively; and β₀ and β_{0,*j*} are the fixed and random intercepts, respectively. β_{*k*} (*k* = 1, 2, ..., 5) and the β_{*k,j*} (*k* = 1, 2, ..., 5) are the fixed and day-specific random slopes for the

predictors, respectively; ε_{*i,j*} is the error term at grid *i* on day *j*; and Ψ is the variance-covariance matrix for day-specific random effects. Fixed effects represent the averaged effects of the predictor on PM_{2.5} over the entirety of the study period, whereas random effects account for the daily variability in relationships between independent and dependent variables (Hu *et al.*, 2014; Zheng *et al.*, 2016). The model was fitted with AOD and MET (“AM model”) parameters to assess the improvements resulting from the introduction of GASs as predictors.

Model Validation

A 10-fold CV based on grid monitoring sites was performed to evaluate the performance of the model with the final goal of predicting the grid PM_{2.5} concentration. During verification, data from 90% of the grid monitoring sites were selected as the training data, and data from the remaining 10% of the grid monitoring sites were used as the validation data. This process was repeated 10 times, and all grid monitoring sites were tested. In addition, the spatial interpolation of GASs has been conducted based on training data. That is, the GASs in the validation data are interpolated from training data. The RMSE, coefficient of determination (R²), slope, and intercept were used to estimate the CV performance of the model.

RESULTS

Descriptive Statistics

Table 1 shows the descriptive statistics of the independent and dependent variables used in model fitting and validation. In 2014, the maximum PM_{2.5} concentration in the JingJinJi region was 858 μg m⁻³ with an annual mean value of 77.3 μg m⁻³ and a standard deviation of 83.54. The average value and standard deviation of AOD were 0.82 and 0.87, respectively. The average PM_{2.5} concentration at each monitoring site and the annual mean AOD are shown in Fig. S2. The average PM_{2.5} concentration in the JingJinJi region increased from the northwest to the southeast under the influence of topography and land-use type. The lowest and highest PM_{2.5} values of 29 and 139 μg m⁻³, respectively, were recorded at Zhangjiakou in the northwest and at Xingtai in the south, respectively. The spatial distribution of AOD in the JingJinJi region was consistent with that of PM_{2.5}. Specifically, PM_{2.5} concentrations were high in areas with high AOD values and vice versa. However, the spatial distribution of PM_{2.5} and AOD during different seasons contradicted the above situation (Fig. S2). The variable seasonal spatial distribution of PM_{2.5} and AOD indicates that the relationship between PM_{2.5} and AOD changed with time.

The results of the Pearson’s correlation analysis for the PM_{2.5} and independent variables are shown in Table 2. Correlation analysis revealed that NO₂ and CO had the strongest correlations with PM_{2.5} concentrations, with correlation coefficients of 0.748 and 0.697, respectively. AOD and SO₂ had the next strongest correlations with PM_{2.5} concentrations, with correlation coefficients of 0.6 and 0.565, respectively. The results of correlation analysis

Table 1. Descriptive statistics of dependent and independent variables (N = 12,955).

Variable	Unit	Min.	Max.	Mean	Median	Std. Dev.
PM _{2.5}	μg m ⁻³	1	858	77.3	49	83.54
AOD	N/A	0.014	3.5	0.82	0.51	0.87
NO ₂	μg m ⁻³	1	253	35.06	25	30.82
SO ₂	μg m ⁻³	1	596	42.2	24	51.49
O ₃	μg m ⁻³	2	362	97.05	83	58.59
CO	mg m ⁻³	0.0015	18.42	1.15	0.86	1.08
PRS	hPa	875.1	1038.5	1002.85	1008.4	26.92
WD	°	0.6	358	164.51	158.8	69.64
WS	m s ⁻¹	0.1	9.4	1.61	1.3	1
TMP	°C	-17.5	32.8	12.09	13.5	11.41
RH	%	9	99.9	55.73	56.9	19.84
ALT	m	7	908	124.7	39	207.06

Table 2. Variable correlation statistics (N = 12,955).

	PM _{2.5}	NO ₂	SO ₂	O ₃	CO	AOD	PRS	WD	WS	TMP	RH	ALT
PM _{2.5}	1	0.748	0.565	-0.12	0.697	0.6	0.177	-0.08	-0.07	-0.07	0.358	-0.15
NO ₂		1	0.683	-0.4	0.748	0.313	0.224	-0.08	-0.11	-0.29	-0.222	-0.14
SO ₂			1	-0.32	0.705	0.249	0.088	-0.01	-0.03	-0.34	0.126	-0.03
O ₃				1	-0.25	0.124	-0.24	-0.09	0.03	0.763	0.104	0.007
CO					1	0.358	0.16	-0.03	-0.04	-0.24	0.242	-0.11
AOD						1	0.121	-0.133	-0.072	0.193	0.524	-0.186
PRS							1	-0.06	0.063	-0.18	-0.01	-0.81
WD								1	0.33	-0.163	-0.282	0.116
WS									1	-0.031	-0.389	-0.035
TMP										1	0.247	-0.107
RH											1	-0.111
ALT												1

illustrate the feasibility of introducing GASs to PM_{2.5} prediction models as predictors. The multiple collinearity problems that exist among GASs were ignored given that NO₂, SO₂, CO, and O₃ represent different pollutant constituents (Zhang *et al.*, 2007) and PM_{2.5} concentration prediction is the focus of this study (Reid *et al.*, 2015).

Model Fitting and Validation

Fig. 2 shows the scatter plots for the model fitting and CV of the AGM and AM models. The CV R² values for the AM and the AGM models were 0.73 and 0.84, respectively. The CV RMSE values for the AM and AGM models were 43.07 and 33.91 μg m⁻³, respectively. The CV R² and RMSE obtained by the models were lower and higher than those obtained through model fitting, respectively. These results indicate that the proposed model was slightly overfitting. However, when GASs were introduced, the R² of model fitting and CV increased by 0.12 and 0.11, respectively, and the RMSE of model fitting and CV decreased by 12.54 μg m⁻³ and 9.16 μg m⁻³, respectively. These results indicate that the performance of both models was greatly improved. In addition, the scatter plot shows that when the PM_{2.5} concentrations exceeded 300 μg m⁻³, the AM model severely underestimated PM_{2.5} concentrations, whereas the AGM model greatly improved in PM_{2.5} concentration estimation. The improvement in estimation of high values was reflected by the increase in slope from

0.74 to 0.86 and the reduction in intercept value from 20.11 to 12.15.

The performance of the AM and the AGM models in the estimation of high PM_{2.5} concentrations is compared in Table 3. When the observed PM_{2.5} concentrations exceeded 0, 35, 75, and 100 μg m⁻³, the R² of the AGM model was higher than that of the AM model by 0.11, 0.12, 0.14, and 0.15, respectively, whereas the RMSE of the AGM model was lower than that of the AM model by 9.16, 11.17, 13.37, and 15.8 μg m⁻³, respectively. High PM_{2.5} concentrations were associated with high increments in R² and considerable decrements in RMSE. These associations indicate that high PM_{2.5} concentrations were associated with the drastic improvement in the performance of the AGM model.

Seasonal performance statistics are shown in Table 4. The performance of the AGM model for all seasons was better than that of the AM model. The performance improvement was the greatest for the heavily polluted winter, with RMSE reduced by 15.28 μg m⁻³ and R² increased by 0.14. The performance improvement was the least for summer, with RMSE reduced by 4.3 μg m⁻³ and R² increased by 0.09.

Signs and Magnitudes of the Model Coefficients

Table S2 and Fig. S1 show the fixed effects and daily variations of slopes and intercepts of the AGM model. The fixed coefficients of AOD and GASs are positive. However,

Fig. 2. Results of model fitting and CV. The dash line is the 1:1 line as a reference. (a) and (c) are fitting results of the AM and AGM models, respectively. (b) and (d) are CV results of the AM and AGM models, respectively.

Table 3. Comparison of the performance of the two models at different $\text{PM}_{2.5}$ concentrations.

Observed $\text{PM}_{2.5}$ ($\mu\text{g m}^{-3}$)	AM_RMSE	AGM_RMSE	AM_R ²	AGM_R ²	RMSE_Residual	R ² _Residual
> 0	43.07	33.91	0.73	0.84	−9.16	0.11
> 35	52.95	41.78	0.65	0.77	−11.17	0.12
> 75	62.63	49.26	0.55	0.69	−13.37	0.14
> 100	70.07	54.27	0.49	0.64	−15.8	0.15

Note: AM_RMSE and AGM_RMSE are the RMSE values for the AM and AGM models, respectively; AM_R² and AGM_R² are the R² values for the AM and AGM models, respectively.

Table 4. Performance statistics of the two models in different seasons.

	AGM_RMSE	AM_RMSE	AGM_R ²	AM_R ²
Spring	26.44	31.33	0.81	0.74
Summer	24.03	28.33	0.76	0.67
Autumn	33.62	41.92	0.88	0.81
Winter	46.21	61.49	0.81	0.67

Note: AM_RMSE and AGM_RMSE are the RMSE values for the AM and AGM models, respectively; AM_R² and AGM_R² are the R² values for the AM and AGM models, respectively.

as can be seen from Fig. S1, the slope of these variables varies daily. The slope of AOD varies the most, from -41.35 to 118.41 , followed by that of CO, which varies from -15.23 to 77.26 . It should be noted that the slopes of the variables are positive on most days, indicating that $PM_{2.5}$ is positively correlated with these variables most of the time. The daily variation in slope and intercept shows that the relationship between $PM_{2.5}$ and the predictor varies with time, indicating the importance of temporal heterogeneity

in $PM_{2.5}$ concentration prediction.

Variation in the Spatial Distribution of $PM_{2.5}$ during a Typical Heavy Pollution Period

Fig. 3 shows the variation in the spatial distribution of $PM_{2.5}$ during a typical heavy pollution period from October 6, 2014 to October 12, 2014. The spatial distribution variation of $PM_{2.5}$ during the heavy pollution process is described in Text S1. This typical heavy pollution process

Fig. 3. Grid site monitoring values and prediction results of the AGM model for a period of heavy pollution from October 6 to October 12, 2014. (a1)–(g1) Prediction results of the AGM model. (a2)–(g2) Grid site monitoring results. Given the lack of AOD data for October 12 and the similarity between site monitoring data for October 12 and October 13, the data from October 13 were used instead of the October 12 data for prediction.

indicates that the spatial distribution of $PM_{2.5}$ predicted by the AGM model was consistent with that inferred from monitoring data. Thus, the AGM model can accurately reflect the occurrence, diffusion, and disappearance of $PM_{2.5}$ and provides strong data support for studies on the diffusion of $PM_{2.5}$ during periods of heavy pollution.

Prediction of Seasonal and Annual Average $PM_{2.5}$ Concentrations

The spatial distribution of seasonal and annual average $PM_{2.5}$ concentration is shown in Fig. 4. The JingJinJi region experienced the heaviest pollution during winter. The average concentration of $PM_{2.5}$ in the JingJinJi region exceeded $120 \mu g m^{-3}$ during winter. Average $PM_{2.5}$ concentrations in the southeast of Handan and the east of Shijiazhuang, Xingtai, and Handan were higher than those in other areas. In addition to industrial emissions and motor vehicle exhaust emissions, residential coal-fired heating contributes to the high $PM_{2.5}$ concentrations in these areas. During autumn, heavily polluted areas were mainly located in the south of Beijing; the southeast of Baoding; central

Shijiazhuang, Xingtai, and Handan; and other areas. The average $PM_{2.5}$ concentrations were $90\text{--}120 \mu g m^{-3}$ and may exceed $120 \mu g m^{-3}$ in some areas. The lowest average $PM_{2.5}$ concentration in the JingJinJi region was recorded during summer and was less than $75 \mu g m^{-3}$ in most areas. The average $PM_{2.5}$ concentration in most areas of Zhangjiakou was less than $30 \mu g m^{-3}$, which meets the secondary $PM_{2.5}$ concentration standard set by the Ministry of Environmental Protection.

The average annual $PM_{2.5}$ concentration in most of the JingJinJi region exceeded the secondary $PM_{2.5}$ concentration standard. $PM_{2.5}$ concentrations in Baoding, Shijiazhuang, Xingtai, Handan, and other areas may even exceed $90 \mu g m^{-3}$. Therefore, the overall environmental situation in the JingJinJi region is not optimistic.

DISCUSSION

In this study, gaseous pollutant data were introduced into a mixed effects model to improve the underestimation of $PM_{2.5}$ in JingJinJi based on AOD and meteorological

Fig. 4. Comparisons of seasonal and annual average predicted and grid monitoring $PM_{2.5}$ concentrations. (a)–(e) Prediction results of the AGM model and (f)–(j) Grid site monitoring results.

data. There may be many causes for the underestimation of $PM_{2.5}$. One of the reasons may be that there are fewer matching data at high concentrations, as discussed by Liu *et al.* (2005), while the study by Gupta and Christopher (2009b) showed that a small range of high $PM_{2.5}$ mass concentrations corresponding to a large range of AOT values may result in an underestimation of high $PM_{2.5}$ concentrations. For further analysis, the correlation statistics between $PM_{2.5}$ and predictors at different $PM_{2.5}$ concentrations are shown in Table S1. The correlation between $PM_{2.5}$ and AOD decreases as the concentration of $PM_{2.5}$ increases. Therefore, AOD cannot adequately represent the correlation between high $PM_{2.5}$ concentrations and independent variables (Gupta and Christopher, 2009b; Liu *et al.*, 2005), resulting in an underestimation of high $PM_{2.5}$ concentrations.

The development of heavy pollution in the JingJinJi region is closely related to atmospheric pollutant concentrations, atmospheric oxidation, and MET (Wang *et al.*, 2015). Atmospheric pollutants include primary pollutant emissions and converted secondary pollutants. Primary pollutants mainly originate from coal fires and traffic emissions, and GASES such as NO_2 and SO_2 , being homologous to the primary pollutants, can represent primary pollutant emissions to a certain extent (Wang *et al.*, 2014). Secondary pollutants are mainly derived from secondary inorganic salts (SO_4^{2-} and NO_3^-) in the particulate state (Sun *et al.*, 2014). As an important component of $PM_{2.5}$, SO_4^{2-} and NO_3^- are mainly oxidized by SO_2 and NO_2 in the atmosphere. The oxidation efficiency of SO_2 and NO_2 is related to the atmospheric oxidant O_x ($NO_2 + O_3$). As an indicator of atmospheric oxidation capacity, a high O_x concentration can promote the secondary conversion of SO_2 and NO_2 to SO_4^{2-} and NO_3^- , thereby increasing $PM_{2.5}$ concentrations. We can draw the conclusion that NO_2 , SO_2 , CO , and O_3 not only represent the emissions of primary pollutants during heavy pollution formation but are also the precursors and oxidants of secondary pollutant transformation. In addition, GASES have a good correlation with high $PM_{2.5}$ concentrations, as shown in Table S1, which shows that they can more adequately represent the correlation between high $PM_{2.5}$ concentrations and independent variables. And the results in Table 3 show that high $PM_{2.5}$ concentrations are associated with high increments in R^2 and considerable decrements in RMSE. Therefore, NO_2 , SO_2 , CO , and O_3 can be used as auxiliary variables to resolve the underestimation of high $PM_{2.5}$ concentration and improve the accuracy of AOD-based $PM_{2.5}$ estimation.

Comparing the RMSE differences between seasons for the AGM and AM models reveals that the performance of the AGM model exhibited the greatest improvement for winter, with an RMSE difference of $15.28 \mu g m^{-3}$. Nevertheless, the AGM model showed negligible improvement in its performance in the prediction of summer $PM_{2.5}$ concentrations and presented an RMSE difference of $4.3 \mu g m^{-3}$. During winter, the increased prevalence of coal-fired heating increases the emission of $PM_{2.5}$ and its precursors, such as NO_2 and SO_2 . The formation of fine particulates through the secondary conversion of NO_2 and SO_2 to SO_4^{2-} and NO_3^- is the main cause of heavy pollution in the JingJinJi region during winter (Wang *et al.*, 2016). The introduction of NO_2 and other GASES as independent variables enabled the AGM model to fully reflect the correlation between high $PM_{2.5}$ concentrations and independent variables during winter and improved the performance of the AGM model in the estimation of the winter $PM_{2.5}$ concentration.

By introducing GASES, the AGM model gives higher CV R^2 (0.84) than previously published studies in JingJinJi, e.g., the mixed effects model for Beijing ($R^2 = 0.79$) (Xie *et al.*, 2015) and JingJinJi ($R^2 = 0.77$) (Zheng *et al.*, 2016) and the Bayesian model for northern China ($R^2 = 0.68$) (Lv *et al.*, 2016). However, the RMSE in this paper is higher than the values in those studies, which may be due to the reason that the concentration of $PM_{2.5}$ was lower in previous studies. The small range of $PM_{2.5}$ concentrations indicates that the concentrations of $PM_{2.5}$ are relatively concentrated, which results in the smaller RMSE value. For comparison, we also experimented with the most commonly used machine learning models, and the results are shown in Table 5. The performance of all the models was greatly improved by introducing GASES, which shows the feasibility of introducing GASES as predictors. After introducing GASES, the gradient boosting decision tree (GBDT) performed the best among the three machine learning models, with the CV R^2 and RMSE being 0.79 and $38.08 \mu g m^{-3}$, respectively. However, the R^2 and RMSE were lower by 0.05 and higher by $6.49 \mu g m^{-3}$, respectively, than for the AGM model, which may be due to the GBDT ignoring the temporal heterogeneity between $PM_{2.5}$ and the predictors.

Although our model demonstrated drastically improved $PM_{2.5}$ prediction performance, it continued to exhibit a certain degree of underestimation for the following reasons: First, we used Kriging interpolation to obtain the spatial distribution of GASES in the JingJinJi region; interpolation, however, is associated with a certain degree of uncertainty

Table 5. Performance comparison of the AGM with machine learning models.

Model ^a	R^2 gas	R^2 nogas	RMSE gas	RMSE nogas
RF	0.77	0.63	39.54	50.7
GBDT	0.79	0.65	38.08	49.56
DNN	0.79	0.65	38.32	49.7
This Study	0.84	0.73	33.91	43.07

^aRF: Random Forest Model; GBDT: Gradient Boosting Decision Tree Model; DNN: Deep Neural Network Model. R^2 _gas and R^2 _nogas represent the R^2 of models with or without GASES, respectively. RMSE_gas and RMSE_nogas are the RMSE of models with or without GASES, respectively.

(Oliver and Webster 1990). Second, a complex nonlinear relationship exists between high PM_{2.5} concentrations and AOD (Liu *et al.*, 2005; Ma *et al.*, 2014; Zheng *et al.*, 2016). The linear mixed model presupposes that a linear relationship exists between PM_{2.5} and predictive variables (Lee *et al.*, 2011). This assumption, however, oversimplifies the complex relationship between PM_{2.5} and the predictors (Reid *et al.*, 2015). Furthermore, in addition to meteorological conditions, primary pollutant discharge, secondary pollutant conversion, dust, and building dust are sources of PM_{2.5} in the JingJinJi region. However, we ignored the effects of these factors on the development of heavy pollution. Finally, we only predicted PM_{2.5} concentrations for available gridded AOD values. AOD data are often missing because of the effects of clouds, high surface reflection, and high PM_{2.5} concentrations (Levy *et al.*, 2010; Tao *et al.*, 2012), and if the missing values are related to high PM_{2.5} concentrations, the PM_{2.5} concentration may be underestimated if only the retrieved AOD fields are used (Lv *et al.*, 2016).

CONCLUSION

The AOD and MET cannot fully represent the correlation between high PM_{2.5} concentrations and predictive variables. Thus, estimation models that use the AOD and MET as predictors may significantly underestimate the PM_{2.5} concentration. We improved the accuracy of AOD-based PM_{2.5} prediction by introducing MET parameters and NO₂, SO₂, CO, and O₃ concentrations as predictors to a mixed-effects model. Our model provided 10-fold CV R² and RMSE values of 0.84 and 33.91 µg m⁻³, respectively. The PM_{2.5} concentrations mapped by our model for heavy-pollution weather and seasons are consistent with those inferred from monitoring data, and our results accurately reflect the appearance, diffusion, and disappearance of PM_{2.5} during such periods. Therefore, introducing GASs as independent variables improves the prediction of PM_{2.5} spatial distribution.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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