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## A case study of development and application of a streamlined control and response modeling system for PM<sub>2.5</sub> attainment assessment in China

Shicheng Long<sup>1</sup>, Yun Zhu<sup>1,\*</sup>, Carey Jang<sup>2</sup>, Che-Jen Lin<sup>1,3</sup>, Shuxiao Wang<sup>4</sup>, Bin Zhao<sup>4</sup>, Jian Gao<sup>5</sup>, Shuang Deng<sup>5</sup>, Junping Xie<sup>1</sup>, Xuezhen Qiu<sup>1</sup>

1. School of Environment and Energy, South China University of Technology, Guangzhou Higher Education Mega Center, Guangzhou 510006, China

2. USEPA/Office of Air Quality Planning & Standards, RTP, NC 27711, USA

3. Department of Civil Engineering, Lamar University, Beaumont, TX 77710-0024, USA

4. State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China

5. Chinese Research Academy of Environmental Sciences, Beijing 100012, China

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### ABSTRACT

This article describes the development and application of a streamlined air control and response modeling system with a novel response surface modeling-linear coupled fitting method and a new module to provide streamlined model data for PM<sub>2.5</sub> attainment assessment in China. This method is capable of significantly reducing the dimensions required to establish a response surface model, as well as capturing more realistic response of PM<sub>2.5</sub> to emission changes with a limited number of model simulations. The newly developed module establishes a data link between the system and the Software for Model Attainment Test—Community Edition (SMAT-CE), and has the ability to rapidly provide model responses to emission control scenarios for SMAT-CE using a simple interface. The performance of this streamlined system is demonstrated through a case study of the Yangtze River Delta (YRD) in China. Our results show that this system is capable of reproducing the Community Multi-Scale Air Quality (CMAQ) model simulation results with maximum mean normalized error < 3.5%. It is also demonstrated that primary emissions make a major contribution to ambient levels of PM<sub>2.5</sub> in January and August (e.g., more than 50% contributed by primary emissions in Shanghai), and Shanghai needs to have regional emission control both locally and in its neighboring provinces to meet China's annual PM<sub>2.5</sub> National Ambient Air Quality Standard. The streamlined system provides a real-time control/response assessment to identify the contributions of major emission sources to ambient PM<sub>2.5</sub> (and potentially O<sub>3</sub> as well) and streamline air quality data for SMAT-CE to perform attainment assessments.

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\* Corresponding author. E-mail: [zhuyun@scut.edu.cn](mailto:zhuyun@scut.edu.cn) (Yun Zhu).

## Introduction

Fine particulate matter pollution ( $PM_{2.5}$ ) has been one of the most important environmental pollution issues in China recently due to its adverse influences on regional haze (Sun et al., 2014; Zhao et al., 2013b), public health and global climate change (Buonocore et al., 2014; Tai et al., 2010). According to the air quality reports of the China National Environmental Monitoring Centre (CNEMC), the monitored annual average  $PM_{2.5}$  of most Chinese cities (China National Environmental Monitoring Centre (CNEMC), 2013) substantially exceeds the Grade II China National Ambient Air Quality Standard ( $35 \mu\text{g}/\text{m}^3$ ) (Ministry of Environmental Protection of the People's Republic of China, MEP, 2012). A large portion of  $PM_{2.5}$  in China is attributable to anthropogenic emissions such as gaseous precursors and primary PM emissions from power plants, industrial & domestic sectors, and transportation (Dong et al., 2014; Fu et al., 2013; Wang and Hao, 2012), especially for primary PM emissions (Xing, 2011; Zhao et al., 2013a). To estimate the response of  $PM_{2.5}$  to anthropogenic emissions under various control strategies, air quality models are an effective and widely used tool for attainment assessments in air quality management. The Community Multi-scale Air Quality (CMAQ) model, as a powerful air quality model tool, has been widely employed to compare the efficacy of various emission control strategies in China (Streets et al., 2007; Wang et al., 2010; Zhao et al., 2013b) and other countries (Appel et al., 2012; Byun and Schere, 2006; Chemel et al., 2014). However, it is extremely time-consuming for CMAQ to estimate a large number of control measures due to the high computational costs and the complicated nature of the required emission/ meteorology inputs and processing (Fann et al., 2012; Foley et al., 2014; Xing et al., 2011). Therefore, a software package called Response Surface Model—Visualization Analysis Tool (RSM-VAT) has been developed to address this issue.

RSM-VAT is an innovative reduced form of air quality model, which has been continually improved and applied successfully in testing and evaluation for  $PM_{2.5}$  (US EPA, 2006b; Zhao et al., 2014; Zhu et al., 2015) and ozone (US EPA, 2006a; Xing et al., 2011). However, there are two limitations in RSM-VAT. One is that the errors of RSM prediction performances sharply increase with the increase of variables since the sample space is greatly increased with the increase of dimensions (Xing et al., 2011). It requires many thousands of CMAQ model runs (samples) to build a reliable RSM prediction when the number of variables is more than a certain threshold (e.g., 56), which is unrealistic for CMAQ (Appel et al., 2011). There is also an assumption in RSM-VAT that all control variables involved in building the response surface model have a nonlinear response to pollutants (e.g.,  $PM_{2.5}$ ). This assumption is not appropriate for primary  $PM_{2.5}$ .  $PM_{2.5}$  is a mixture consisting of nonlinear contributions from gaseous precursors (e.g.,  $\text{SO}_2$ ,  $\text{NH}_3$  and  $\text{NO}_x$ ) and linear contributions from primary/direct  $PM_{2.5}$  emission sources (Dong et al., 2014; Li et al., 2014; Xing, 2011). In China, especially in the YRD region, the proportion of contributions from total primary  $PM_{2.5}$  emissions can be as high as 46% (Zhao et al., 2014). To overcome these limitations, we improve the previous version of RSM-VAT with a new RSM-Linear coupled fitting method. The updated response modeling system significantly reduces the dimensions required to establish a response

surface model and captures the realistic response of fine particles to both primary and secondary source changes with a reasonable number of model simulations.

The Software for Model Attainment Test—Community Edition (SMAT-CE) is a powerful regulatory model tool to demonstrate the air quality attainment for emission control strategies by combining monitoring data with modeled data from CMAQ or from the Comprehensive Air Quality Model with extensions (CAMx) (US EPA, 2007; Wang et al., 2015). However, the model output format cannot be directly used in SMAT-CE for efficient attainment assessment. The RSM-VAT can address this challenge, but these two software tools are operated in a stand-alone fashion and the SMAT-CE is unable to identify the gridded model data from RSM-VAT due to their different data formats. As such, a module for outputting gridded model data to SMAT-CE in the updated response modeling tool (RSM-VAT) was developed. The module establishes a data link between this updated RSM tool and SMAT-CE, which provides streamlined model data for  $PM_{2.5}$  and  $\text{O}_3$  attainment assessment. Finally, a case study on  $PM_{2.5}$  in the Yangtze River Delta (YRD) was conducted for attainment assessments in this paper. The streamlined modeling tool in this case study demonstrates an innovative approach that could provide real-time control/response assessment to identify the contributions of major emission sources to ambient  $PM_{2.5}$  (and potentially  $\text{O}_3$  as well) as well as streamline air quality data for SMAT-CE to perform  $PM_{2.5}$  and  $\text{O}_3$  attainment assessments.

## 1. Methodology

Fig. 1 schematically shows the operation process of creating RSM, validation, and outputting SMAT-CE formatted data for  $PM_{2.5}$  attainment test assessment. Our previous study (Zhu et al., 2015) has reported the creation of RSM with the method of high-dimensional Kriging and functional design of RSM-VAT in detail. Compared to the reported RSM-VAT (Zhu et al., 2015), the main improvements of this version include (1) the development of a RSM-Linear coupled fitting method for creating a  $PM_{2.5}$  response surface model and (2) a new module for saving gridded model data for SMAT-CE.

### 1.1. Control matrix design

Response surface modeling is a type of reduced-form modeling using statistical techniques to relate a response variable (e.g.,  $PM_{2.5}$ ) to a set of control variables that are of interest through the design of complex multi-dimensional experiments. A control matrix defines the multi-dimensional experiments consisting of a set of emission control scenarios parameterized by control variables. Here we select  $PM_{2.5}$  as our target pollutant. The emissions of  $PM_{2.5}$  are categorized into 56 control variables based on emission type and source category, including 32 gaseous  $PM_{2.5}$  precursor control variables and 24 primary  $PM_{2.5}$  control variables (Table 1). The matrix is created by sampling the 56 control variables in the design space. The sample values of these 56 control variables are set from 0 to 1.5. A control variable value (emission ratio) of 1.5 means the amount of the emission source/factor increases by 50% compared to the level of the base year. Fig. 2 shows an example of a final control matrix. The first N

samples are generated by using Latin hypercube sampling (LHS) (Hirabayashi et al., 2011) with the gaseous PM<sub>2.5</sub> precursor control variables. The size of *N* is determined by the balance between the validation error of the created RSM and the computation time of CMAQ simulations (Foley et al., 2014; Zhu et al., 2015). The next *n* single-fixed-factor samples are targeted to the primary PM<sub>2.5</sub> control variables, which only reduce the emission of one primary PM<sub>2.5</sub> control variable to 0.25 at a time while keeping the others unchanged. The control variable values in the final matrix are applied to the base year emission inventory to form (*N* + *n*) emission control scenarios for CMAQ simulations. These emission inventory variables, together with the meteorology and other necessary data, are applied in CMAQ simulations to predict the PM<sub>2.5</sub> concentration under these control scenarios. The CMAQ simulations were run for two months, January and August in 2010, due to their representativeness of typical winter and summer climatology conditions (Zhang et al., 2012). More information about CMAQ configuration, geophysical projection, meteorological, emission, and initial and boundary condition inputs used for this analysis are described in detail by Zhao et al. (2014). The Yangtze River Delta (YRD) region (Fig. 3) was selected as the study domain because it is the most populous and economically vigorous region in China, with extensive air emissions (Fu et al., 2014; Li et al., 2012).

1.2. RSM-Linear coupled fitting method

The steps of building an RSM for the non-linear response of PM<sub>2.5</sub>, using the linear method to build up the response of

PM<sub>2.5</sub> to primary emission changes, and results validation, are described below.

Step 1: Establish the response surface of PM<sub>2.5</sub> to precursor emission changes. A high-dimensional Kriging algorithm is employed to establish the nonlinear response, as reported in a previous publication (Zhu et al., 2015). The nonlinear response of PM<sub>2.5</sub> can be estimated efficiently by the created RSM using *N* (=254) scenarios (Fig. 2). The RSM-predicted value of PM<sub>2.5</sub> ambient concentration can be expressed by Eq. (1):

$$C_{\text{prec}}(i, j) = \text{RSM}^{\text{PM}_{2.5}}(X_1, X_2, \dots, X_F) \tag{1}$$

where (*X*<sub>1</sub>, *X*<sub>2</sub>, ..., *X*<sub>*F*</sub>) represents the *F* precursor emission ratios, e.g., *X*<sub>1</sub> is the first precursor control variable emission ratio; RSM<sup>PM<sub>2.5</sub></sup> represents the response surface of PM<sub>2.5</sub> to precursor emissions; and *C*<sub>prec</sub>(*i*, *j*) represents the RSM-predicted value of the PM<sub>2.5</sub> ambient concentration resulting from precursor emission changes at grid cell (*i*, *j*) associated with the scenario (*X*<sub>1</sub>, *X*<sub>2</sub>, ..., *X*<sub>*F*</sub>).

Step 2: Establish the response of PM<sub>2.5</sub> to primary PM<sub>2.5</sub> emission changes. The changes in the ambient concentration of PM<sub>2.5</sub> are directly affected by the emission changes of one primary PM<sub>2.5</sub> variable. For an additional scenario where only one primary PM<sub>2.5</sub> is disturbed and the others stay the same as for the base case, the changed variable is regarded as the *l* (1 ≤ *l* ≤ *f*) primary control variable. Its value changes from 1 to *X*<sub>*l*</sub>. According to the linear response of PM<sub>2.5</sub> concentration to primary PM<sub>2.5</sub> emissions (Zhao et al., 2014), the contribution of the *l* primary PM<sub>2.5</sub> control variable to PM<sub>2.5</sub> can be expressed as:

$$\Delta C_l(i, j) = (C_{N+l}(i, j) - C_{\text{base}}(i, j)) \times \frac{1 - X_l}{1 - X_{N+l}} \tag{2}$$

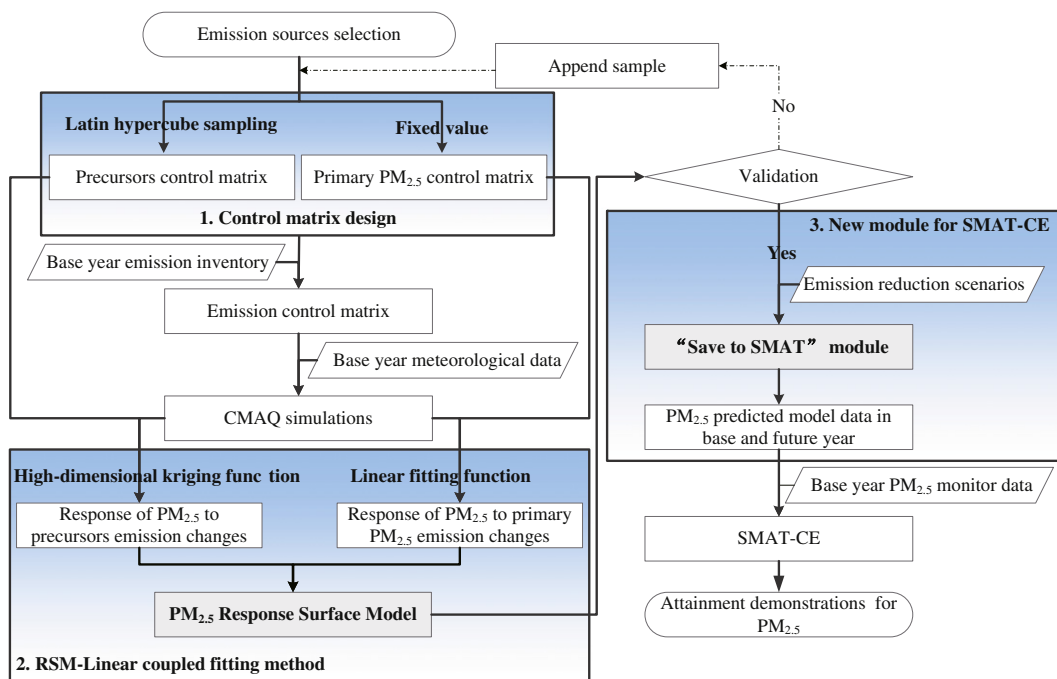


Fig. 1 – Key operation process for development and application of a streamlined control and response modeling system with a response surface modeling (RSM)-linear coupled fitting method for PM<sub>2.5</sub> attainment assessment. Colored boxes represent the major improvements compared to the previous version of response surface model. SMAT-CE: the Software for Model Attainment Test—Community Edition; CMAQ: Community Multi-Scale Air Quality.

**Table 1 – Emissions control variables selected for this study.**

Variable type	Control variable	Variable description	Variable details
Gaseous precursor variables	NO <sub>x</sub> _PP	NO <sub>x</sub> power plant source emissions	Eight variables in each of the 4 regions (e.g., Shanghai, Jiangsu, Zhejiang and other regions in the Yangtze River Delta (YRD)). There are 8 × 4 = 32 gaseous precursor control variables in total.
	NO <sub>x</sub> _IN&DO	NO <sub>x</sub> industrial and domestic source emissions	
	NO <sub>x</sub> _TR	NO <sub>x</sub> transportation source emissions	
	SO <sub>2</sub> _PP	SO <sub>2</sub> power plant source emissions	
	SO <sub>2</sub> _IN&DO	SO <sub>2</sub> industrial and domestic source emissions	
	VOC_IN&DO	Volatile organic carbon industrial and domestic source emissions	
Primary PM <sub>2.5</sub> variables	VOC_TR	Volatile organic carbon transportation source emissions	Six variables in each of the 4 regions. There are 6 × 4 = 24 primary PM <sub>2.5</sub> control variables in total.
	NH <sub>3</sub> _IN&DO	NH <sub>3</sub> industrial & domestic source emissions	
	PM <sub>2.5</sub> _PP	PM <sub>2.5</sub> directed power plant source emissions	
	PM <sub>2.5</sub> _IN&DO	PM <sub>2.5</sub> directed industrial and domestic source emissions	
	PM <sub>2.5</sub> _TR	PM <sub>2.5</sub> directed transportation source emissions	
	PMC_PP	PM coarse directed power plant source emissions	
PMC_IN&DO	PM coarse directed industrial and domestic source emissions		
PMC_TR	PM coarse directed transportation source emissions		

PP: power plants; IN&DO: industry and domestic; TR: transportation.

where  $\Delta C_l(i,j)$  is the change of PM<sub>2.5</sub> concentration at grid cell  $(i,j)$  resulting from the changes of the  $l$  primary control variable;  $C_{N+1}(i,j)$  is the CMAQ simulation value of the  $(N+1)$  scenario (Fig. 2) at grid cell  $(i,j)$ ;  $C_{base}(i,j)$  is the CMAQ simulation value of the baseline scenario at grid cell  $(i,j)$ ;  $X_l$  is the  $l$  primary PM<sub>2.5</sub> control variable emission, and  $X_{N+1}$  is a constant (equal to 0.25) of the  $l$  primary PM<sub>2.5</sub> control variable emission in the  $(N+1)$  scenario.

When the emissions of all primary PM<sub>2.5</sub> variables vary simultaneously, the changes of PM<sub>2.5</sub> are calculated according to the linear relationship between PM<sub>2.5</sub> and primary PM<sub>2.5</sub>:

$$\Delta C(i,j) = \sum_{l=1}^f \Delta C_l(i,j). \quad (3)$$

Step 3: Establish and validate the PM<sub>2.5</sub> response surface model. The PM<sub>2.5</sub> concentrations in the target region can be thought as the contributions of gaseous precursors and primary

		$F$						$f$					
$N$	1	1	1	1	...	1	1	1	1	1	...	1	
	0.129	0.677	0.074	0.915	...	0.529	1	1	1	1	...	1	
	0.836	0.250	0.802	0.871	...	1.127	1	1	1	1	...	1	
	0.182	1.452	0.153	0.732	...	0.170	1	1	1	1	...	1	
	...	...	...	...	...	...	...	...	...	...	...	...	...
	0.388	0.521	0.195	0.262	...	0.977	1	1	1	1	...	1	
$n$	1	1	1	1	...	1	0.25	1	1	1	...	1	
	1	1	1	1	...	1	1	0.25	1	1	...	1	
	1	1	1	1	...	1	1	1	0.25	1	...	1	
	1	1	1	1	...	1	1	1	1	0.25	...	1	
	...	...	...	...	...	...	...	...	...	...	...	...	
	1	1	1	1	...	1	1	1	1	1	...	0.25	

**Fig. 2 – Design of control matrix.**  $F$  ( $=32$ ) and  $f$  ( $=24$ ) represent the number of gaseous precursors and primary PM<sub>2.5</sub> control variables, respectively;  $N$  ( $=254$ ) stands for one CMAQ base case (the first row) and  $(N-1)$  samples generated by Latin hypercube sampling (LHS) method; and  $n$  ( $=24$ ) represents the sample number of single-fixed-factor scenarios in the primary emission control matrix. Each sample represents an emission control scenario.

PM<sub>2.5</sub> emissions. Therefore, based on Eqs. (1) and (3), the final RSM PM<sub>2.5</sub> predicted value at grid cell  $(i,j)$  affected by the changes of both precursors and primary PM<sub>2.5</sub> emissions can be expressed as:

$$C_{pred}(i,j) = \text{RSM}^{\text{PM}_{2.5}}(X_1, X_2, \dots, X_f) + \sum_{l=1}^f \Delta C_l(i,j). \quad (4)$$

Based on Eq. (4), the PM<sub>2.5</sub> response surface model is built. This model is validated through out-of-sample validation (OOS) and cross validation (CV). The RSM-predicted values are compared with CMAQ “true” values and a standard set of model performance evaluation metrics (e.g., bias, error) are computed. The above CMAQ “true” values of PM<sub>2.5</sub> and chemical components



**Fig. 3 – Study domain and four cities of interest for this study.** A: Other cities, B: Shanghai, C: Southern Jiangsu, and D: Northern Zhejiang.



were provided and validated with observational data by Tsinghua University. The CMAQ-observation validation results of normalized mean biases (NMBs) range from –15% to 24% (Zhao et al., 2014). When the RSM prediction error or bias exceeds acceptable ranges, a certain amount of samples will be added for CMAQ simulations until the error is within an acceptable range.

### 1.3. Emission reduction scenarios

Table 2 lists three emission reduction scenarios targeted on several pollutants, *i.e.*, NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, and primary PM<sub>2.5</sub>. Since VOC emissions have a minor effect on PM<sub>2.5</sub> concentrations due to the significant underestimation of secondary organic aerosol formation in the current CMAQ model (Carlton et al., 2010), we did not consider a VOC reduction scenario in this paper. Case #1 focuses on the reduction of NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, and primary PM<sub>2.5</sub> emissions outside of Shanghai. Case #2 represents the emission reduction of the same pollutants as in Case #1 for Shanghai. As documented in the literature (Wang and Hao, 2012; Wang et al., 2014a), emission controls in a single city are not an effective way to realize the air quality standard, because air pollution is a complex phenomenon and influenced by both local emissions and regional transport from neighboring areas. Therefore, we design Case #3 to represent an emission reduction of the same pollutant emissions in the entire YRD region. The emission ratios in these three scenarios are in accordance with the maximum feasible reduction described by Wang et al. (2014b).

### 1.4. Functional module for output model data to SMAT-CE

Since the gridded air quality model data from RSM-VAT cannot be used directly as inputs for SMAT-CE due to data format differences (*e.g.*, projection), a functional module named “Save to SMAT” is developed in RSM-VAT. This module implements the coordinate conversions and grid-related computations, and then outputs the SMAT-CE formatted data for attainment assessments.

In RSM-VAT, the model data are based on a Lambert conformal conic (LCC) projection. The coordinates of each grid cell are in the format of column and row indexes. In contrast, SMAT-CE formatted model data require latitude and longitude coordinates at the centroid of each grid cell. To get this coordinate information, an open-source geographic information systems (GIS) library (DotSpatial, <http://dotspatial.codeplex.com/>) is introduced. The functionality of map projection transformations in this library is extended into RSM-VAT to provide coordinate conversion and then compute the latitude/longitude of each grid cell in the modeling domain. After that, several data fields (*e.g.*, grid cell ID) are added to the output gridded model file, which include a grid cell ID, grid-cell values of PM<sub>2.5</sub>, and the model month or quarter.

With this module, users only need to input the emission control scenario and right-click on the pollutant spatial distribution plot to select “Save to SMAT”, then the corresponding SMAT-CE formatted gridded model data will be exported in \*.csv format. In this study, a base year and three future years’ gridded model files were derived from this module and each model file consisted of two simulated monthly average PM<sub>2.5</sub> concentrations (January and August).

### 1.5. Air quality attainment assessment in SMAT-CE

SMAT-CE is primarily intended as a tool to implement the modeled attainment tests for particulate matter (PM<sub>2.5</sub>), ozone (O<sub>3</sub>) and regional haze (visibility). It statistically estimates a future design value (DVF) at a specific site (a monitoring site or grid cell) by using a base-year observational data and the model data obtained from the base-year and future-year air quality simulations. The model data of the base and future years are used for calculating the ratio of the model’s future to base-year predictions at monitoring sites. The ratios are called relative response factors (RRF). The DVF of pollutants is estimated at monitoring sites by multiplying RRF locations “near” each monitor by the base-year observation value. The future-year design values are compared to the NAAQS. If all future site-specific pollutant design values are less than or equal to the concentration specified in the NAAQS, the test is passed. A detailed description of this attainment methodology and approach is available in the study by Wang et al. (2015).

In this study, gridded model data for a base year and three future years’ scenarios, as well as base-year PM<sub>2.5</sub> monitoring data from Shanghai, were used as inputs for SMAT-CE. The estimated results consist of (1) spatial distribution of future year PM<sub>2.5</sub> concentration and (2) future year PM<sub>2.5</sub> concentrations at the 10 national monitoring sites in Shanghai. The first one was facilitated to evaluate the effectiveness of three proposed emission reduction scenarios, while the other one was applied to conduct PM<sub>2.5</sub> NAAQS attainment testing at Shanghai monitoring sites.

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## 2. Results and discussion

### 2.1. Validation of RSM-Linear coupled fitting tool

RSM can reproduce the simulation results of CMAQ by various validation methods (Zhu et al., 2015). The cross validation (CV) results (available at <http://www.abacas-dss.com/>) demonstrate the good performance of our created RSM (including January and August) in the YRD with mean normalized bias (MNB) < 1% and mean normalized error (MNE) < 1%. Here, we concentrate on the validation of the developed linear fitting algorithm. A set of 28 additional scenarios are introduced to validate the performance of this algorithm by out-of-sample (OOS) validation. The first 24 scenarios, corresponding to the 24 primary PM<sub>2.5</sub> control variables (cases 1–24), are single-fixed-factor samples where only one control variable of primary PM<sub>2.5</sub> changes from 0.25 to 0.50 at a time and the others remain the same as the base case. The next 4 scenarios (cases 25–28) are generated randomly by the LHS method, in which all primary PM<sub>2.5</sub> control variables are changed simultaneously. The predicted PM<sub>2.5</sub> concentrations for these scenarios are compared to the corresponding CMAQ simulation results using model evaluation metrics (*e.g.*, mean normalized bias). As shown in Fig. 4a, the overall errors over these 28 scenarios were small in both January and August, with the median value of 0%. Only a few discrete points in August varied from –1.5% to

**Table 2 – Emission control scenarios for the attainment of PM<sub>2.5</sub> air quality standard.**

Region	Scenarios	Emission control factors								
		NO <sub>x</sub> _PP	NO <sub>x</sub> _IN&DO	NO <sub>x</sub> _TR	SO <sub>2</sub> _PP	SO <sub>2</sub> _IN&DO	NH <sub>3</sub> _IN&DO	PM <sub>2.5</sub> _PP	PM <sub>2.5</sub> _IN&DO	PM <sub>2.5</sub> _TR
Other cities	Case #1	0.28	0.28	0.25	0.41	0.26	0.55	0.23	0.19	0.13
	Case #2	1	1	1	1	1	1	1	1	1
	Case #3	0.28	0.28	0.25	0.41	0.26	0.55	0.23	0.19	0.13
Shanghai	Case #1	1	1	1	1	1	1	1	1	1
	Case #2	0.35	0.24	0.18	0.47	0.17	0.55	0.27	0.37	0.16
	Case #3	0.35	0.24	0.18	0.47	0.17	0.55	0.27	0.37	0.16
Southern Jiangsu	Case #1	0.24	0.26	0.24	0.42	0.25	0.55	0.22	0.21	0.14
	Case #2	1	1	1	1	1	1	1	1	1
	Case #3	0.24	0.26	0.24	0.42	0.25	0.55	0.22	0.21	0.14
Northern Zhejiang	Case #1	0.27	0.28	0.19	0.34	0.25	0.55	0.16	0.28	0.14
	Case #2	1	1	1	1	1	1	1	1	1
	Case #3	0.27	0.28	0.19	0.34	0.25	0.55	0.16	0.28	0.14

+1.5%. The performance statistics in January were slightly lower than those in August because of the high PM<sub>2.5</sub> concentrations in January. These results imply that the new linear fitting algorithm performs well for these 24 primary PM<sub>2.5</sub> control variables.

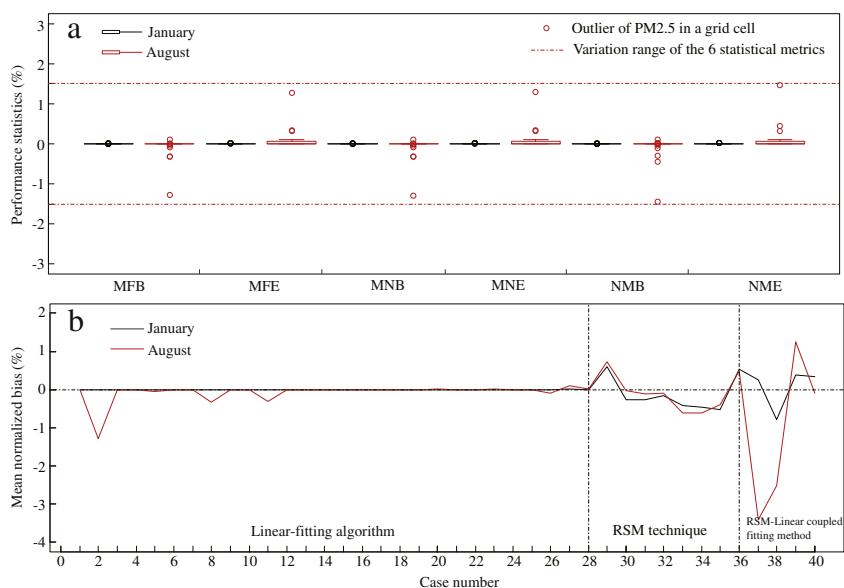
Another 12 scenarios are generated using a LHS method to further estimate the stability of the RSM-Linear coupled fitting method through OOS validation, which consist of 8 scenarios (cases 29–36) for validating the RSM technique and 4 scenarios (cases 37–40) for validating the RSM-Linear coupled fitting method. Compared to the 28 linear fitting algorithm validation cases, the 8 scenarios (cases 29–36), where only the control variables of gaseous precursors changed, had much more fluctuation (Fig. 4b). This fluctuation may be attributable to the instability of an RSM with a large number of dimensions. For those scenarios (cases 37–40) where all control variables of both gaseous precursors and primary PM<sub>2.5</sub> changed simultaneously, the overall errors and biases were relatively larger, mainly due to the superposed influence of the RSM technique and the linear fitting algorithm. Even so, the maximum MNB was within an acceptable error of –3.32%. Here we choose Case #37, whose MNB was the largest at –3.32% in August, to further investigate the accuracy of this method. As shown in Fig. 5, the underestimation of Case #37 could be viewed clearly when we compared the base case concentration reduction in Fig. 5b, due to the amplified coordinate axis. The correlation coefficients of this case were larger than 0.99, indicating a perfect agreement with CMAQ simulations. Summary statistics of the prediction errors for all of the out-of-sample simulations (cases 1–40) are calculated and given in Table 3. The results show that the mean values of the 6 statistical metrics were within ± 1% in both January and August, indicating that this coupled method provides good accuracy compared to the CMAQ simulations.

## 2.2. Analysis of primary emissions contributing to PM<sub>2.5</sub> in YRD

Policy makers can use updated RSM-VAT to identify the emission source contributions from various regions and sectors. Fig. 6 gives the accumulative contribution for the changes of PM<sub>2.5</sub> in four regions (Shanghai, Jiangsu, Zhejiang and Other regions in the YRD) with a 30% reduction of all sources

in January and August. As shown in Fig. 6a, the total primary PM<sub>2.5</sub> emissions had a dominating contribution to PM<sub>2.5</sub> concentration changes in the YRD region, accounting for 78% and 55% of the PM<sub>2.5</sub> change in January and August, respectively. The local primary PM<sub>2.5</sub> contributed most significantly to the PM<sub>2.5</sub> change. Take Shanghai as an example, in January, nearly 6.0 μg/m<sup>3</sup> (83%) of the PM<sub>2.5</sub> concentration change was contributed by primary PM<sub>2.5</sub> emissions, in which about 3.8 μg/m<sup>3</sup> (53%) came from local primary emissions. In August, the primary PM<sub>2.5</sub> emissions were responsible for a significant fraction of PM<sub>2.5</sub>, although the proportion decreased compared to that of January. These emissions accounted for approximately 4.0 μg/m<sup>3</sup> (59%) of the PM<sub>2.5</sub> concentrations, including about 2.7 μg/m<sup>3</sup> (39%) contributions from local emissions. The gaseous precursors emitted from Zhejiang province had an obvious effect on Shanghai due to the prevailing southeasterly winds in August. The long-range-transported NO<sub>x</sub> from Zhejiang province will react with the local NH<sub>3</sub> in Shanghai to form nitrate in the more strongly NH<sub>3</sub>-rich conditions in August (Zhao et al., 2014). Therefore, the gradually increased NO<sub>x</sub> emissions from Zhejiang have more contribution to PM<sub>2.5</sub> than SO<sub>2</sub> compared to the well-controlled SO<sub>2</sub> emission since 2010 (Fu et al., 2013). Compared to the two months' emission reduction in Shanghai, the reduction was much more effective in January. These results are similar for other regions such as Zhejiang or Jiangsu provinces, suggesting that the reduction of primary PM<sub>2.5</sub> emissions, especially the local ones, can significantly decrease PM<sub>2.5</sub> concentration in the YRD. Regardless of the source sectors, reducing emissions in January achieved a greater benefit in lowering the PM<sub>2.5</sub> concentration, which is consistent with previous reports (Li et al., 2014; Zhao et al., 2008, 2013b).

Fig. 6b further demonstrates the contributions of primary PM<sub>2.5</sub> emissions by sector to total primary PM<sub>2.5</sub> changes. Contributions of the total emissions from the industry and domestic (IN&DO) sector have larger impacts on total primary PM<sub>2.5</sub> change than those from power plants (PP) and transportation (TR) in both January and August. For Shanghai, the PM<sub>2.5</sub> emissions from IN&DO contribute up to 84% and 80% of PM<sub>2.5</sub> concentrations in January and August, respectively, mainly attributed to the emissions from industrial combustion, steel industry, domestic fossil-fuel and biomass combustion (Zhao et al., 2013a).



**Fig. 4 – (a) Boxplot of 6 statistical metrics for 28 out-of sample scenarios (cases 1–28) using the newly added linear fitting algorithm and (b) line chart of mean normalized bias (MNB) for the 40 out-of sample scenarios (cases 1–40). MNE: mean normalized error; NMB: normalized mean bias; NME: normalized mean error; MFB: mean fractional bias; MFE: mean fractional error. These performance statistics are available at Zhu et al. (2015).**

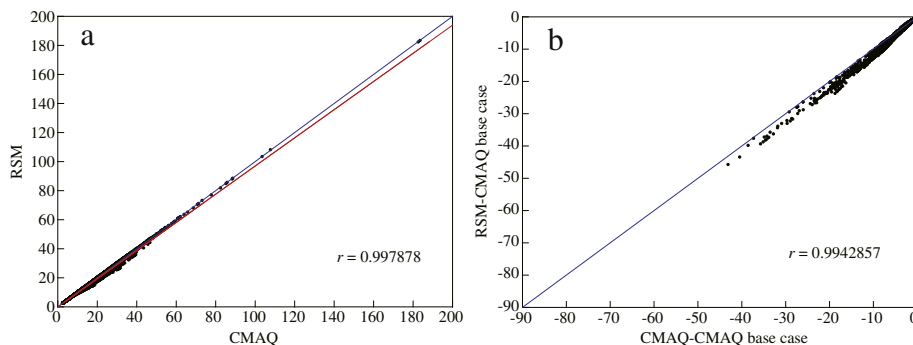
### 2.3. Application in air quality attainment assessments

The simulated PM<sub>2.5</sub> concentrations in the base case (2010) and their changes under three proposed scenarios in January and August are given in Fig. 7. Fig. 7a and b shows the spatial distribution of the monthly means of base-case PM<sub>2.5</sub> concentrations in two months. Fig. 7a shows that the PM<sub>2.5</sub> concentration in a large part of Shanghai exceeds 75 μg/m<sup>3</sup> in January. Compared to that of January, the air quality in August was much better (Fig. 7b). This is mainly due to the stronger advection and clean ocean air parcel introduced by the Southeast Asia monsoon in summer (Dong et al., 2014; Huang et al., 2014).

The simulation results based on three proposed control scenarios show that: (1) controlling the local emissions of Shanghai alone would obviously lower the PM<sub>2.5</sub> concentration

in Shanghai, while it had little effect on those regions outside of Shanghai (Fig. 7e, f). This is because the contributions of PM<sub>2.5</sub> concentrations in the YRD are dominated by the primary PM<sub>2.5</sub> emissions, which tend to deposit locally; (2) controlling the emissions outside of Shanghai led to a limited improvement in PM<sub>2.5</sub> concentrations (Fig. 7c, d); and (3) controlling the emissions in both Shanghai and its adjacent regions achieved a satisfactory effect, as shown in Fig. 7g and h. Comparison of these results shows that the governments of Shanghai and the neighboring provinces need to coordinate their emission reduction efforts to decrease the PM<sub>2.5</sub> concentration.

The PM<sub>2.5</sub> concentrations at each monitoring site in Shanghai under the base case and the three control scenarios are shown in Fig. 8. Case #3 shows the greatest air quality improvement at all the locations. The control of local emissions in Shanghai (Case #2) could achieve a greater reduction in PM<sub>2.5</sub>



**Fig. 5 – Comparison of PM<sub>2.5</sub> concentrations (μg/m<sup>3</sup>) predicted by RSM (Y, Response Surface Model) in case 37 with corresponding CMAQ simulations (X, Community Multi-scale Air Quality) in August. (a) RSM vs. CMAQ and (b) the CMAQ base case concentration reduction (predicted or simulated value subtracts the concentration of CMAQ base case) of RSM vs. CMAQ.**

**Table 3 – Results of out-of-sample validations in January and August (40 scenarios).**

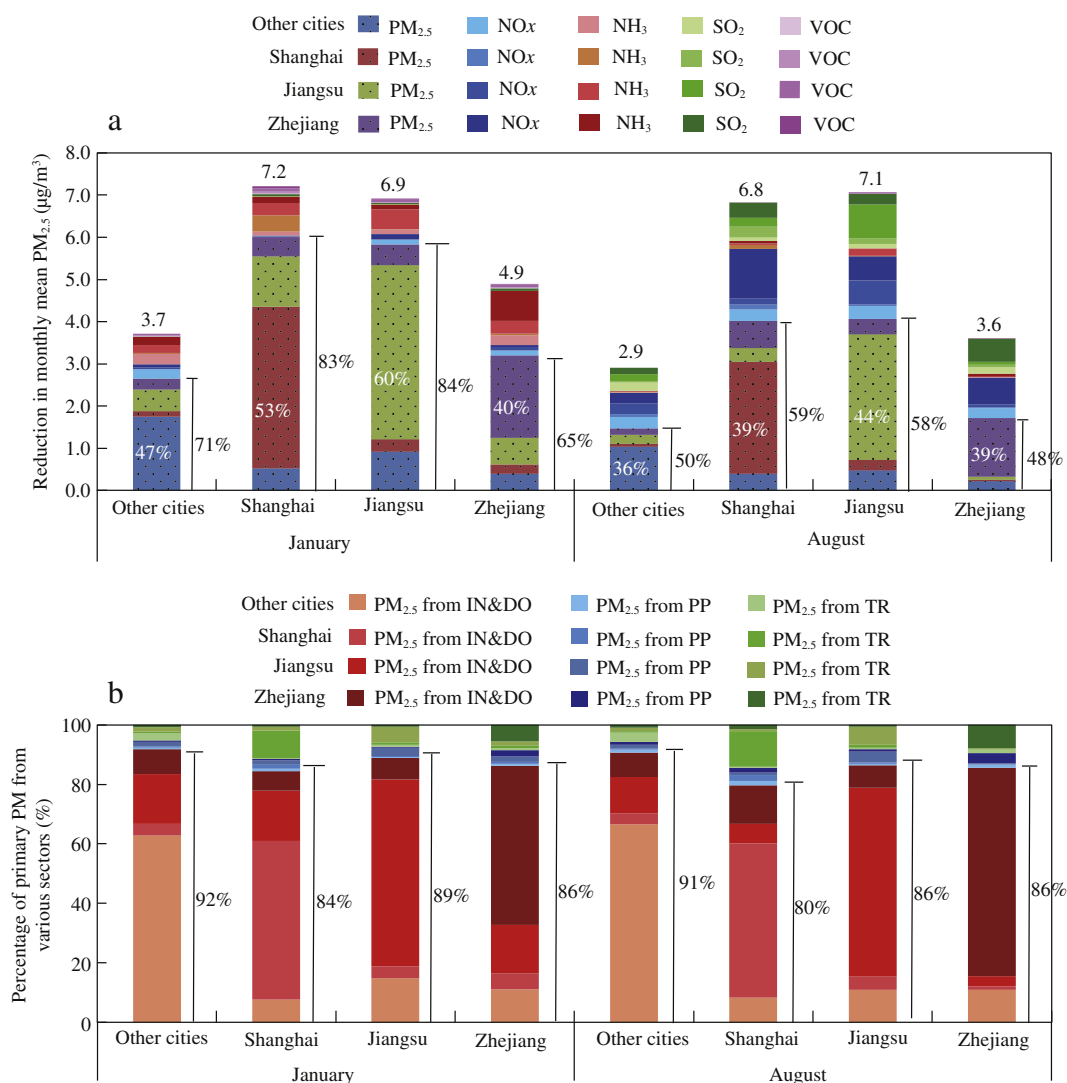
Performance metric	January			August		
	Mean	Minimum	Maximum	Mean	Minimum	Maximum
MNB (%)	-0.02%	-0.77%	0.60%	-0.18%	-3.32%	1.26%
MNE (%)	0.15%	0.00%	0.78%	0.36%	0.00%	3.32%
MFB (%)	-0.02%	-0.78%	0.60%	-0.18%	-3.40%	1.25%
MFE (%)	0.15%	0.00%	0.78%	0.37%	0.00%	3.40%
NMB (%)	-0.02%	-0.87%	0.65%	-0.21%	-4.05%	1.56%
NME (%)	0.16%	0.00%	0.87%	0.42%	0.00%	4.05%

MNB: mean normalized bias; MNE: mean normalized error; NMB: normalized mean bias; NME: normalized mean error; MFB: mean fractional bias; MFE: mean fractional error.

concentration compared to emission control in nearby regions (Case #1), with the exception of the background monitoring site named Qingpuadianshanhu (QPDSH). This is due to the fact that the site is far from the urban areas of Shanghai and strongly

affected by the regional transport from adjacent provinces (e.g., Jiangsu, Zhejiang).

Based on Fig. 8 in Case #1, the reduction of  $PM_{2.5}$  concentration in Shanghai was greater in August than that



**Fig. 6 – (a) Accumulative contribution from different pollutants for total  $PM_{2.5}$  concentration and (b) accumulative fractional contribution from power plants (PP), industry and domestic (IN&DO), and transportation (TR) for total primary  $PM_{2.5}$  with all sources reducing down to 70% in January and August. VOC: volatile organic compounds.**



in January. For example, the  $PM_{2.5}$  concentration of the Putuo (PT) monitoring site reduced from 73.6 to 53.2  $\mu\text{g}/\text{m}^3$ , accounting for 28% reduction of  $PM_{2.5}$  concentration in January; while the same emission control percentage contributed to 36% reduction of  $PM_{2.5}$  concentration in August, suggesting that the  $PM_{2.5}$  in Shanghai is more significantly affected by regional emissions in August compared to that in January. This phenomenon is mainly caused by the significant differences in meteorological conditions. In August, Shanghai's  $PM_{2.5}$  concentration is significantly influenced by the emissions from the surrounding regions, because of the prevailing wind conditions between sea and land (Liu et al., 2010). However, due to the low wind speed and dry weather meteorological conditions in January, the air quality of Shanghai has more tendency to be affected by the local emissions (Zhao et al., 2008).

To assess the attainment of annual  $PM_{2.5}$  in Shanghai, the average of the monthly mean  $PM_{2.5}$  in January and August predicted by SMAT-CE was applied to represent the future year annual average  $PM_{2.5}$  concentrations. The base-year and future-year attainment results under three scenarios at all the monitoring sites in Shanghai are shown in Fig. 9. Most of the air quality data at these neighboring monitoring sites were higher than the annual standard of 35  $\mu\text{g}/\text{m}^3$ . For controlling the emissions in adjacent provinces, only one background site located in the suburb of Shanghai meets the annual NAAQS of  $PM_{2.5}$  (Fig. 9). One more monitoring site could reach the attainment level by controlling local emissions in Shanghai (Fig. 9). Controlling the emissions from both local and adjacent provinces simultaneously was the only control strategy to attain the air quality standard (Fig. 9). This demonstrated why both local and regional emission reductions were needed to control ambient  $PM_{2.5}$  in the study areas.

### 3. Conclusions

This paper describes a novel RSM-Linear coupled fitting method and a new functional module for preparing model data for SMAT-CE application. The method grouped  $PM_{2.5}$  emissions into two sources (precursors and primary  $PM_{2.5}$ ) and adopted different algorithms for each source type. This coupled method significantly decreased the number of dimensions required to establish a response surface model, and more realistically captured the response of  $PM_{2.5}$  to emission changes from both gaseous precursors and primary  $PM_{2.5}$  with a reasonable number of model simulations. The newly developed module established a data link between the updated response modeling tool and SMAT-CE. It had the ability to provide streamlined model responses to emission control scenarios for SMAT-CE using a simple interface.

We demonstrated a case study in the Yangtze River Delta (YRD) to analyze the impacts on  $PM_{2.5}$  concentration and estimate air quality attainment in Shanghai under three proposed emission control scenarios as an application of this streamlined system. The analysis of primary emission impacts to  $PM_{2.5}$  helped policy makers identify the primary sources contributing to  $PM_{2.5}$  in the YRD. The results showed that primary emissions made a dominant contribution to the ambient levels of  $PM_{2.5}$  in January and August. The response of  $PM_{2.5}$  to primary emission changes was much more sensitive in January than that in August. The attainment assessment of  $PM_{2.5}$  in Shanghai showed that  $PM_{2.5}$  concentration in Shanghai was predominantly influenced by local emissions and significantly affected by those emissions from the adjacent provinces. To meet the annual  $PM_{2.5}$  standard, joint emission control efforts in both local areas and neighboring provinces are required. The

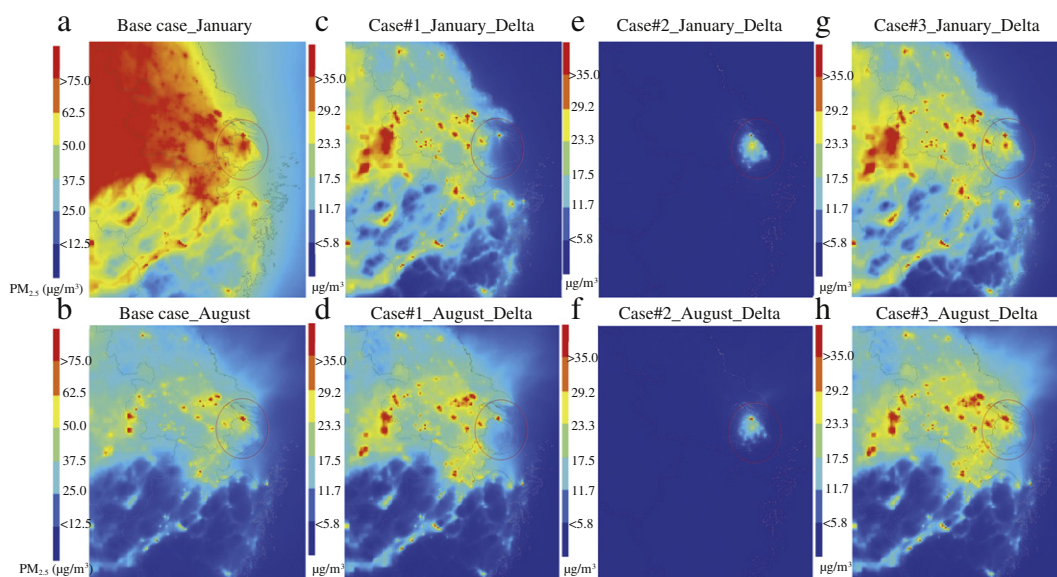
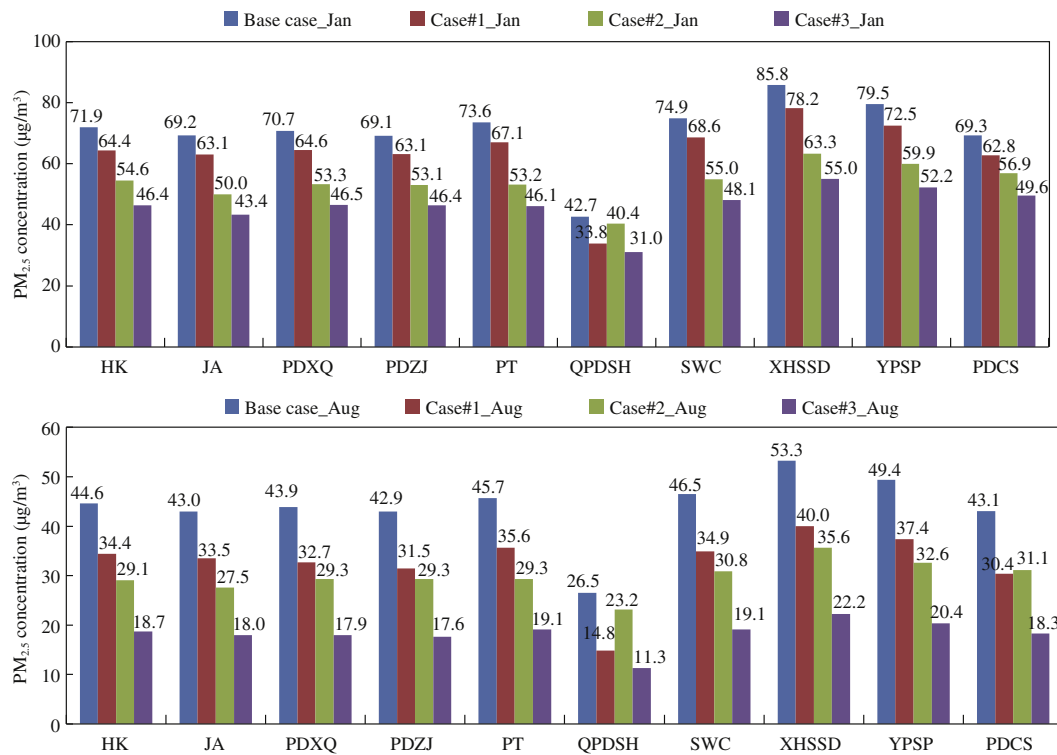


Fig. 7 – Monthly average  $PM_{2.5}$  in January (a) and August (b) 2010 as well as changes (base case minus scenario case) under 3 proposed scenarios in January (c, e, g) and August (d, f, h).



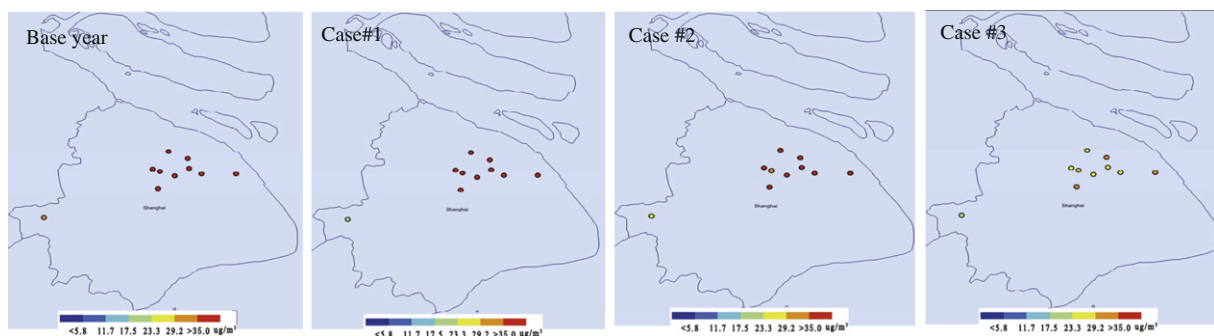
**Fig. 8 – Comparison between base-year (2010) and future-year monthly mean PM<sub>2.5</sub> under 3 scenarios in January and August for the 10 national monitoring sites in Shanghai. HK: Hongkou, JA: Jing'an, PDXQ: Pudongxinqu, PDZJ: Pudongzhangjiang, PT: Putuo, QPDSH: Qingpudianshanhu, SWC: Shiwuchang, XHSSD: Xuhuishangshida, YPSP: Yangpusipiao, PDCS: Pudongchuansha.**

developed modeling tool serves as an efficient and streamlined science-based platform for identifying emission sources leading to air pollution and for air quality attainment assessments.

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deposition of typical industrial point-source mercury emissions in the Pearl River Delta. This work is also partly supported by the funding of Guangdong Provincial Key Laboratory of Atmospheric Environment and Pollution Control (No. 2011A060901011), the project of Atmospheric Haze Collaboration Control Technology Design (No. XDB05030400) from the Chinese Academy of Sciences and the Ministry of Environmental Protection's Special Funds for Research on Public Welfare (No. 201409002). Partly financial support is also provided by the Guangdong Provincial Department of Science and Technology, the project of demonstration research of air quality management cost-benefit analysis and attainment assessments technology (No. 2014A050503019).



**Fig. 9 – Comparison between a base-year and three proposed future-year scenarios' PM<sub>2.5</sub> attainment results at 10 monitoring sites in Shanghai.**

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