A modeling study of the nonlinear response of fine particles to air pollutant emissions in the Beijing–Tianjin–Hebei region

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Abstract. The Beijing–Tianjin–Hebei (BTH) region has been suffering from the most severe fine-particle (PM₂.₅) pollution in China, which causes serious health damage and economic loss. Quantifying the source contributions to PM₂.₅ concentrations has been a challenging task because of the complicated nonlinear relationships between PM₂.₅ concentrations and emissions of multiple pollutants from multiple spatial regions and economic sectors. In this study, we use the extended response surface modeling (ERSM) technique to investigate the nonlinear response of PM₂.₅ concentrations to emissions of multiple pollutants from different regions and sectors over the BTH region, based on over 1000 simulations by a chemical transport model (CTM). The ERSM-predicted PM₂.₅ concentrations agree well with independent CTM simulations, with correlation coefficients larger than 0.99 and mean normalized errors less than 1 %. Using the ERSM technique, we find that, among all air pollutants, primary inorganic PM₂.₅ makes the largest contribution (24–36 %) to PM₂.₅ concentrations. The contribution of primary inorganic PM₂.₅ emissions is especially high in heavily polluted winter and is dominated by the industry as well as residential and commercial sectors, which should be prioritized in PM₂.₅ control strategies. The total contributions of all precursors (nitrogen oxides, NOₓ; sulfur dioxides, SO₂; ammonia, NH₃; non-methane volatile organic compounds, NMVOCs; intermediate-volatility organic compounds, IVOCs; primary organic aerosol, POA) to PM₂.₅ concentrations range between 31 and 48 %. Among these precursors, PM₂.₅ concentrations are primarily sensitive to the emissions of NH₃, NMVOC + IVOC, and POA. The sensitivities increase substantially for NH₃ and NOₓ and decrease slightly for POA and NMVOC + IVOC with the increase in the emission reduction ratio, which illustrates the nonlinear relationships between precursor emissions and PM₂.₅ concentrations. The contributions of primary inorganic PM₂.₅ emissions to PM₂.₅ concentrations are dominated by local emission sources, which account for over 75 % of the total primary inorganic PM₂.₅ contributions. For precursors, however, emissions from other regions could play similar roles to local emission sources in the summer and over the northern part of BTH. The source contribution features for various types of heavy-pollution episodes are distinctly different from each other and from the monthly mean results, illustrating that control strategies should be differentiated based on the major contributing sources during different types of episodes.
1 Introduction

China is one of the regions with the highest concentration of PM$_{2.5}$ (particulate matter with aerodynamic diameter equal to or less than 2.5 µm) in the world (van Donkelaar et al., 2015). The problem is especially serious over the Beijing–Tianjin–Hebei (BTH) region, one of the most populous and developed regions in China. Annual average PM$_{2.5}$ concentrations in this region reached 85–110 µg m$^{-3}$ during 2013-2015, which approximately triple the standard threshold (35 µg m$^{-3}$) and far exceed those in other metropolitan regions (Wang et al., 2017). It has been estimated that the severe PM$_{2.5}$ pollution leads to about 1.05–1.23 million premature deaths per year in China (Lim et al., 2012; Burnett et al., 2014; J. D. Wang et al., 2016), and the monetized loss over the BTH region is as high as 134 billion Chinese Yuan, representing 2.2% of regional gross domestic product (GDP) (Lv and Li, 2016). Additionally, PM$_{2.5}$ substantially affects global and regional climate by absorbing and scattering solar radiation and by altering cloud properties (IPCC, 2013; Seinfeld et al., 2016; Zhao et al., 2017), which in turn exert an impact on regional air quality (J. D. Wang et al., 2014; Zhao et al., 2017).

To tackle the heavy PM$_{2.5}$ pollution problem, the Chinese government issued the Action Plan on Prevention and Control of Air Pollution in September 2013, which aimed at a 25% reduction in PM$_{2.5}$ concentrations over the BTH region by 2017 from the 2012 levels (The State Council of the People’s Republic of China, 2013). The attainment of an ambient PM$_{2.5}$ standard would further require substantial reductions in air pollutant emissions (Wang et al., 2015, 2017). To establish emission control strategies, many studies have apportioned the sources of PM$_{2.5}$ over the BTH region, either by mining monitoring data using the positive matrix factorization and chemical mass balance methods (e.g., Zhang et al., 2007; Yu et al., 2013) or by embedding chemical tracers in chemical transport models (CTMs) (e.g., Y. J. Wang et al., 2016; Li et al., 2015; Ying et al., 2014). While these studies can capture the current contributions of various sources to PM$_{2.5}$ concentrations, these contributions could differ significantly from the PM$_{2.5}$ reductions induced by reducing emissions from the corresponding sources, due to highly nonlinear chemical mechanisms (Han et al., 2016; Wang et al., 2011). Therefore, it is imperative to assess the nonlinear response of PM$_{2.5}$ to pollutant emissions from multiple sources, which could provide direct support for the development of effective control policies.

The most widely used technique to evaluate the responses of PM$_{2.5}$ concentrations to emission changes is the “brute force” method, which involves perturbing emissions from a certain source and repeating the solution of a CTM (Russell et al., 1995). A number of studies have utilized the brute force method to quantify the sensitivities of PM$_{2.5}$ concentrations over the BTH region to emissions from different spatial regions (Streets et al., 2007; Wang et al., 2008; L. T. Wang et al., 2014; Li and Han, 2016) or different economic sectors (Wang et al., 2008; L. T. Wang et al. 2014; Han et al., 2016; Liu et al., 2016), either on a seasonal basis (Streets et al., 2007; Wang et al., 2008; Han et al., 2016; Liu et al., 2016) or during a specific heavy-pollution episode (Li and Han, 2016; L. T. Wang et al., 2014). To improve the computational efficiency, several mathematical techniques embedded in CTMs have been developed to simultaneously calculate the sensitivities of the modeled concentrations to multiple emission sources, including the decoupled direct method (Yang et al., 1997) and adjoint analysis (Sandu et al., 2005; Hakami et al., 2006). Zhang et al. (2016) used the adjoint analysis method to examine sensitivities of PM$_{2.5}$ concentrations in the BTH region to pollutant emissions during several pollution periods. However, all the preceding studies only quantified first-order sensitivities and therefore could not capture the nonlinearity in the responses of PM$_{2.5}$ concentrations to pollutant emissions, which can be extremely strong in metropolitan regions like BTH due to complex chemical mechanisms (Wang et al., 2011). Moreover, no studies have simultaneously evaluated the response of PM$_{2.5}$ concentrations in BTH to emissions of multiple pollutants from different sectors and regions, which we need to consider and balance to develop cost-effective control strategies.

In light of the drawbacks of the preceding methods, the response surface modeling (RSM) technique (denoted by “conventional RSM” hereafter to distinguish it from the extended response surface modeling, ERSM, technique) has been developed by using advanced statistical techniques to characterize the complex nonlinear relationship between model outputs and inputs (U.S. Environmental Protection Agency, 2006; Xing et al., 2011; Wang et al., 2011). This technique has been applied to the United States (U.S. Environmental Protection Agency, 2006) and eastern China (Wang et al., 2011) to evaluate the response of PM$_{2.5}$ and its chemical components to pollutant emissions. However, the number of emission scenarios required to build conventional RSM depends on the variable number via an equation of fourth or higher order (Zhao et al., 2015b). Therefore, the required scenario number would be tens of thousands for over 15 variables and even hundreds of thousands for over 25 variables, which is computationally impossible for most three-dimensional CTMs. To overcome this major limitation, we recently developed the ERSM technique (Zhao et al., 2015b), which substantially reduced the scenario number needed to build the response surface and hence extended its applicability to an increased number of regions, pollutants, and sectors with an acceptable computational burden.

Given the advantage of the ERSM technique, here we apply it to over 1000 simulations by the Community Multiscale Air Quality model with Two-Dimensional Volatility Basis Set (CMAQ/2D-VBS) to systematically evaluate the nonlinear response of PM$_{2.5}$ to emission changes in multiple pollutants from different sectors and regions over the BTH region. The major sources contributing to PM$_{2.5}$ and its ma-
Major components are identified, and the nonlinearity in the response of PM$_{2.5}$ to emission changes is characterized. Based on the results of this study, suggestions for PM$_{2.5}$ control policies over the BTH region are proposed.

2 Methodology

2.1 CMAQ/2D-VBS configuration and evaluation

The CMAQ/2D-VBS model was developed in our previous study (Zhao et al., 2016) by incorporating the 2D-VBS model framework into CMAQv5.0.1. Compared with the default CMAQ, the CMAQ/2D-VBS model explicitly simulates aging of secondary organic aerosol (SOA) formed from non-methane volatile organic compounds (NMVOCs), aging of primary organic aerosol (POA), and photooxidation of intermediate-volatility organic compounds (IVOCs), thereby significantly improving the simulation results of organic aerosol (OA), particularly SOA. The model parameters within the 2D-VBS framework have been optimized in our previous studies (Zhao et al., 2015a, 2016) based on a series of smog-chamber experiments. Here we use the same model parameters as those of the “high-yield VBS” configuration reported in Zhao et al. (2016), which agrees best with surface OA and SOA observations among three model configurations. An application in eastern China reveals that CMAQ/2D-VBS reduces the underestimation in OA concentrations from 45 % (default CMAQv5.0.1) to 19 %. More importantly, while the default CMAQv5.0.1 substantially underestimates the fraction of SOA in OA by 5–10 times and cannot track the oxygen-to-carbon ratio (O : C), the SOA fraction and O : C simulated by CMAQ/2D-VBS agree fairly well with observations.

We apply the CMAQ/2D-VBS model over the BTH region. One-way, double-nesting simulation domains are used, as shown in Fig. 1. Domain 1 covers East Asia with a grid resolution of 36 km × 36 km; Domain 2 covers the BTH and its surrounding regions with a grid resolution of 12 km × 12 km. We use the SAPRC99 gas-phase chemistry module and the AERO6 aerosol module, in which the treatment of OA is replaced with the 2D-VBS framework. The aerosol thermodynamics is based on ISORROPIA-II. The initial and boundary conditions for Domain 1 are kept constant as the model default profile, and those for Domain 2 are extracted from the output of Domain 1. A 5-day spin-up period is used to reduce the influence of initial conditions on modeling results.

The Weather Research and Forecasting Model (WRF, version 3.7) is used to generate the meteorological fields. The National Center for Environmental Prediction (NCEP)’s FNL (Final) Operational Global Analysis data (ds083.2) at 1.0° × 1.0° and 6 h resolution are used to generate the first-guess field. The NCEP’s Automated Data Processing (ADP) data (ds351.0 and ds461.0) are used in objective analysis (i.e., grid nudging). The major physics options for WRF include the Kain–Fritsch cumulus scheme, the Pleim–Xiu land-surface module, the Asymmetric Convective Model with nonlocal upward mixing and local downward mixing (ACM2) for planetary boundary layer (PBL) parameterization, the Morrison double-moment scheme for cloud microphysics, and the Rapid Radiative Transfer Model for General Circulation Models (RRTMG) radiation scheme. The land cover type data are obtained from the Moderate resolution Imaging Spectroradiometer (MODIS). The simulation periods are January, March, July, and October in 2014, representing winter, spring, summer, and fall. We select these 4 months because the occurrence frequencies of various meteorological types in these months are statistically most similar to the average conditions in winter, spring, summer, and fall during 2004–2013 (Wu, 2016).

A high-resolution anthropogenic emission inventory in 2014 has been developed by Tsinghua University using an “emission factor method” (Fu et al., 2013; Zhao et al., 2013b) for the BTH region. The emissions from area and mobile sources are first calculated for each prefecture-level city based on statistical data and subsequently distributed into the model grids according to the spatial distribution of population, GDP, and road networks. A unit-based method (Zhao et al., 2008) is applied to estimate and locate the emissions from large point sources (LPSs) including power plants, iron and steel plants, and cement plants. The anthropogenic emission inventory in other provinces of China was originally developed for 2010 and 2012 in our previous studies (Zhao et al., 2013a, b; S. X. Wang et al., 2014; Cai et al., 2017); this has been updated to 2014 in this study following the same methodology. In both the BTH and national emission inventories, the emissions from the open burning of agricultural residue are calculated using crop yields, straw to grain ratio, fraction of biomass burned in the open field, and emission factors (Fu et al., 2013; Zhao et al., 2013b; Wang and Zhang, 2008). We do not include the emissions from forest and grassland fires, which typically account for less than 5 % of the total biomass burning emissions over the BTH region (Qin and Xie, 2011) and are not the focus of the present study. Table S1 in the Supplement summarizes emissions of major air pollutants in each prefecture-level city over the BTH region in 2014; Table S2 gives the provincial emissions in the whole of China in 2014. The emissions for other countries are obtained from the MIX emission inventory (Li et al., 2017) for 2010, which is the latest year available. Following our previous study (Zhao et al., 2016), we assume IVOC emissions to be 30 times, 4.5 times, 1.5 times, and 3.0 times the POA emissions from gasoline vehicles, diesel vehicles, biomass burning, and other emission sources, respectively, which is based on a series of laboratory measurements (Gordon et al., 2014a, b; Hennigan et al., 2011; Jathar et al., 2014). The biogenic emissions were calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006).
We compared the simulation results of WRFv3.7 and CMAQ/2D-VBS with meteorological observations obtained from the National Climatic Data Center (NCDC). PM$_{2.5}$ observations at 138 state-controlled observational sites, and observations of major PM$_{2.5}$ chemical components at seven sites within the modeling domain. We show that the meteorological and chemical simulations generally agree well with observations, with performance statistics mostly within the benchmark values proposed by previous studies. Details of the model evaluation methods and results are given in the Supplement (Sect. S1, Table S3–S5, Figs. S1–S5).

2.2 Development of ERSM prediction system

The detailed methodologies of the conventional RSM and ERSM techniques have been described in our previous papers (Zhao et al., 2015b; Xing et al., 2011). Here we only summarize some key components. The conventional RSM technique characterizes the relationships between a response variable (e.g., PM$_{2.5}$ concentration) and a set of control variables (i.e., emissions of particular pollutants from particular sources) based on a number of randomly generated emission control scenarios (Xing et al., 2011; Wang et al., 2011). The PM$_{2.5}$ concentration for each emission scenario is calculated with a CTM (CMAQ/2D-VBS in this study), and the conventional RSM is subsequently established using the maximum-likelihood estimation–empirical best linear unbiased predictors (MLE-EBLUPs) developed by Santner et al. (2003). Due to the limitation of the conventional RSM technique with respect to variable number, we have developed the ERSM technique (Zhao et al., 2015b) to extend the applicability to an increased number of variables and geographical regions. The ERSM technique first quantifies the relationship between PM$_{2.5}$ concentrations and precursor emissions for each single region using the conventional RSM technique as described above and then assesses the effects of interregional transport of PM$_{2.5}$ and its precursors on PM$_{2.5}$ concentration in the target region. In order to quantify the interaction among regions, we introduce a key assumption that the emissions of precursors in the source region affect PM$_{2.5}$ concentrations in the target region through two major processes: (1) the interregional transport of precursors enhancing the chemical formation of secondary PM$_{2.5}$ in the target region; (2) the formation of secondary PM$_{2.5}$ in the source region followed by transport to the target region. We quantify the individual contributions of these two processes as well as the contribution of local emissions in the target region, which are subsequently integrated to derive the total PM$_{2.5}$ concentrations in the target region. The development of the ERSM prediction system requires several hundred to over 1000 emission scenarios, but once built, it enables real-time prediction of PM$_{2.5}$ concentrations for any given control strategy and proves to be an efficient and user-friendly decision-making tool. Moreover, ERSM can be applied to design a least-cost control strategy once it is coupled with control cost models/functions that links the emission reductions with economic costs.

For the application of the RSM/ERSM techniques to the BTH region, we define five target regions in the inner modeling domain (Domain 2), i.e., Beijing, Tianjin, Northern Hebei (N Hebei), Eastern Hebei (E Hebei), and Southern Hebei (S Hebei), as shown in Fig. 1. The decomposition of Hebei province is based on a preliminary analysis of the pollutant transport patterns over the BTH region (Sect. S2). The simulation using the back-trajectory method indicates that four major types of heavy-pollution episodes in Beijing are primarily contributed by air mass from the south, the local area, the northwest, and the southeast. We develop two RSM/ERSM prediction systems (Table 1). The response variables for the first prediction system, which is

![Figure 1. Double-nesting domains used in CMAQ/2D-VBS simulation (a) and the definition of five target regions in the innermost domain, denoted by different colors (b). The grey lines in (b) represent the boundaries of prefecture-level cities.](image-url)
built using the conventional RSM technique, are concentrations of PM$_{2.5}$, SO$_{2}^{2-}$, NO$_{3}^{-}$, and OA over the urban areas of prefecture-level cities in the five target regions. For the second prediction system that is established using the ERSM technique, the response variables are only PM$_{2.5}$ concentrations. The first prediction system uses 101 emission control scenarios generated by the Latin hypercube sample (LHS) method (Iman et al., 1980) to map atmospheric concentrations versus emissions of five PM$_{2.5}$ precursors, i.e., NO$_{x}$, SO$_{2}$, NH$_{3}$, NMVOC + IVOC, and POA, in all five target regions (Table 1). It is on the one hand intended for the validation of the second system (Sect. 3.1) and, on the other hand, used to study the source contributions of major PM$_{2.5}$ components. For the second system, the emissions of the preceding PM$_{2.5}$ precursors as well as primary inorganic PM$_{2.5}$ (i.e., the chemical components of primary PM$_{2.5}$ other than POA) in each of the five regions are categorized into seven and four control variables, respectively, resulting in 55 control variables in total (Table 1). Note that we distinguish POA and primary inorganic PM$_{2.5}$ because the former undergoes chemical reactions and produces SOA, while the latter is mostly chemically inert in the CMAQ/2D-VBS model. We generate 1121 scenarios (see Table 1) to build the response surface, following the method detailed in Zhao et al. (2015b). Specifically, the scenarios include (1) 1 CMAQ/2D-VBS base case; (2) 200 scenarios generated by applying the LHS method for the control variables of precursors in Beijing, 200 $\times$ 4 scenarios generated in the same way for Tianjin, Northern Hebei, Eastern Hebei, and Southern Hebei; (3) 100 scenarios generated by applying LHS method for the total emissions of NO$_{x}$, SO$_{2}$, NH$_{3}$, NMVOC + IVOC, and POA in all five regions; and (4) 20 scenarios in which one of the control variables of primary inorganic PM$_{2.5}$ emissions is set to 0.25 for each scenario. Here the scenario numbers (200 in group 2 and 100 in group 3) are determined based on numerical experiments conducted in our previous studies (Xing et al., 2011; Wang et al., 2011), which showed that the response surface for seven and five variables could be built with good prediction performance (mean normalized error $<1$ %; correlation coefficient $>0.99$) using 200 and 100 scenarios, respectively. Finally, we generate 54 independent scenarios for out-of-sample validation, which will be detailed in Sect. 3.1.

For the application of the ERSM prediction system to quantitatively characterize the sensitivity of PM$_{2.5}$ concentrations to emission changes, we define “PM$_{2.5}$ sensitivity” as the change ratio of PM$_{2.5}$ concentration divided by the reduction ratio of an emission source, following previous studies (Zhao et al., 2015b; Wang et al., 2011).

$$S_{a}^{X} = \left( (C^{*} - C_{0}) / C^{*} \right) / (1 - a),$$

where $S_{a}^{X}$ is the PM$_{2.5}$ sensitivity to emission source $X$ at its emission ratio $a$; $C^{*}$ and $C_{0}$ are PM$_{2.5}$ concentrations in the base case (when the emission ratio of $X$ is 1) and in the control scenario, in which the emission ratio of $X$ is $a$, respectively. Similar indices can be defined for chemical components of PM$_{2.5}$, such as NO$_{5}^{-}$, SO$_{2}^{2-}$, and OA.

3 Results and discussion

3.1 Validation of ERSM performance

The conventional RSM technique has been extensively demonstrated to have high accuracy and stability in previous papers (Xing et al., 2011; Wang et al., 2011), so we only describe the validation of the ERSM technique. Following Zhao et al. (2015b), we assess the performance of the ERSM prediction system using the “out-of-sample” and 2D-isopleth validation methods, which focus on the accuracy and stability of the prediction system, respectively.

For out-of-sample validation, we use the ERSM prediction system to calculate the PM$_{2.5}$ concentrations for 54 out-of-sample control scenarios, i.e., scenarios independent from those used to build the prediction system, and compare them with the corresponding CMAQ/2D-VBS simulation results. These 54 out-of-sample scenarios (summarized in Table S6) include 40 cases (cases 1–40) in which the control variables of precursors change but those of primary inorganic PM$_{2.5}$ stay the same as the base case, 4 cases (cases 41–44) that are the other way around, and 10 cases (cases 45–54) in which control variables of precursors and primary inorganic PM$_{2.5}$ change simultaneously. Most cases are generated randomly with the LHS method (cases 4–6, 10–12, 16–18, 22–24, 28–29, 54), and some cases are designed in which all control variables are subject to large emission changes (cases 1–3, 7–9, 13–15, 19–21, 25–27).

Figure 2 compares the ERSM-predicted and CMAQ/2D-VBS-simulated PM$_{2.5}$ concentrations and PM$_{2.5}$ responses (defined as the difference between PM$_{2.5}$ concentration in an emission control scenario and that in the base case) for the out-of-sample scenarios using scatterplots. Table 2 summarizes the statistics of the model performance. The definitions of normalized error (NE), mean normalized error (MNE), and normalized mean error (NME) are given as follows:

$$\text{NE} = |P_{i} - S_{i}| / S_{i},$$

$$\text{MNE} = \frac{1}{N_{s}} \sum_{i=1}^{N_{s}} \left[ |P_{i} - S_{i}| / S_{i} \right],$$

$$\text{NME} = \sum_{i=1}^{N_{s}} |P_{i} - S_{i}| / \sum_{i=1}^{N_{s}} S_{i},$$

where $P_{i}$ and $S_{i}$ are the ERSM-predicted and CMAQ/2D-VBS-simulated value of the $i$th out-of-sample scenario; $N_{s}$ is the number of out-of-sample scenarios. Figure 2 shows that the ERSM predictions and CMAQ/2D-VBS simulations agree well with each other. For PM$_{2.5}$ concentrations, the correlation coefficients are larger than 0.99, and the MNEs and NMEs are less than 1 % for all 4 months. The maximum NEs could be as large as 11 % for a particular month and region, but the 95 % percentiles of NEs are all within 4.4 %.
NEs exceeding 4.4% happen only for the scenarios in which most control variables are reduced substantially, indicating relatively large errors at low emission rates, which is consistent with our previous study (Zhao et al., 2015b). Note that all sensitivity scenarios used in Sect. 3.2–3.4 have ≤ 80% emission reductions, which helps to avoid relatively large errors. We also examine the errors in predicted PM$_{2.5}$ response. Since the CMAQ/2D-VBS-simulated PM$_{2.5}$ responses are very close to zero in several scenarios, their normalized errors (NEs) and mean normalized errors (MNEs) could be extremely large even if the absolute errors are small, which cannot properly characterize the accuracy of the ERSM technique. For this reason, we only calculate the correlation coefficients and NMEs (Table 2). The correlation coefficients of the PM$_{2.5}$ response are larger than 0.99, and the NMEs are within 5.6% for all months. In summary, the out-of-sample validation indicates an overall good agreement between ERSM predictions and CMAQ/2D-VBS simulations.

We further examine whether the ERSM technique can capture the trends in PM$_{2.5}$ concentrations in response to continuous changes in precursor emissions, i.e., the stability of the ERSM technique. To this end, we compare the 2D isopleths of PM$_{2.5}$ concentrations as a function of simultaneous changes in two precursors’ emissions in all five regions derived from the ERSM and conventional RSM techniques. It should be noted that, although the ERSM technique is applicable to a much larger number of control variables than conventional RSM, the assumptions in the treatment of interregional transport (Sect. 2.2) in ERSM might affect its accuracy. Nevertheless, the predictions by conventional RSM can be regarded as proxies for real CMAQ/2D-VBS simulations since conventional RSM has been extensively demonstrated to have high accuracy and stability in previous studies (Xing et al., 2011; Wang et al., 2011). For this reason, the comparison between the ERSM and conventional RSM techniques helps to evaluate the stability of the ERSM technique. Figure 3 illustrates the PM$_{2.5}$ isopleths in Beijing as a function of three combinations of precursors, i.e., NO$_x$, NH$_3$, SO$_2$, and NH$_3$, and VOC + IVOC and POA; the isopleths for other regions are very similar and are thus not shown. The x and y axes of the figures represent the “emission ratio”, defined as the ratios of the changed emissions to the emissions in the base case. For example, an emission ratio of 0.7 means the emission of a particular control variable accounts for 70% of the base case. The color isopleths represent PM$_{2.5}$ concentrations. The comparison shows that the shapes of isopleths derived from both prediction systems generally agree with each other. The agreement is very good for the case of VOC + IVOC and POA, and for the cases of NO$_x$ and NH$_3$ and SO$_2$ and NH$_3$ when the emission ratios for NO$_x$ and NH$_3$ are larger than 0.2. Relatively large errors occur at very low NO$_x$ / NH$_3$ emission ratios (<0.2) due primarily to an extremely strong nonlinearity. Within these low emission ranges, the ERSM technique can capture the general trends in PM$_{2.5}$ concentrations in response to emission changes, but the concentration gradients predicted by ERSM are smaller than those given by conventional RSM. More studies are needed to further improve the performance of ERSM at very low NO$_x$ / NH$_3$ emission ratios. Despite the existing errors, the general consistency between RSM- and ERSM-predicted isopleths demonstrates the stability of the ERSM prediction system. In other words, the discrepan-

### Table 1. Description of the RSM/ERSM prediction systems developed in this study.

<table>
<thead>
<tr>
<th>Method</th>
<th>Control variables</th>
<th>Control scenarios</th>
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</thead>
<tbody>
<tr>
<td>Conventional RSM technique</td>
<td>Five control variables: total emissions of NO$_x$, SO$_2$, NH$_3$, NMVOC + IVOC, and POA.</td>
<td>101 control scenarios:</td>
</tr>
<tr>
<td></td>
<td>1. NO$_x$/large point sources (LPS)$^b$, 2. NO$<em>x$/other sources, 3. SO$<em>2$/LPS, 4. SO$<em>2$/other sources, 5. NH$<em>3$/all sources, 6. NMVOC + IVOC/all sources, 7. POA/all sources, and four linear control variables, i.e., 8. primary inorganic PM$</em>{2.5}$/power plants, 9. primary inorganic PM$</em>{2.5}$/industry, 10. primary inorganic PM$</em>{2.5}$/residential and commercial, 11. primary inorganic PM$</em>{2.5}$/transportation.</td>
<td>1. 1 CMAQ/2D-VBS base case, 2. 100$^g$ scenarios generated by applying the LHS method for the five variables.</td>
</tr>
<tr>
<td>ERSM technique</td>
<td>55 control variables in total: 11 control variables in each of the five regions, including seven nonlinear control variables, i.e., 1. NO$_x$/large point sources (LPS)$^b$, 2. NO$<em>x$/other sources, 3. SO$<em>2$/LPS, 4. SO$<em>2$/other sources, 5. NH$<em>3$/all sources, 6. NMVOC + IVOC/all sources, 7. POA/all sources, and four linear control variables, i.e., 8. primary inorganic PM$</em>{2.5}$/power plants, 9. primary inorganic PM$</em>{2.5}$/industry, 10. primary inorganic PM$</em>{2.5}$/residential and commercial, 11. primary inorganic PM$</em>{2.5}$/transportation.</td>
<td>1121 control scenarios:</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1. 1 CMAQ/2D-VBS base case, 2. 1000 scenarios, including 200$^g$ scenarios generated by applying LHS method for the nonlinear control variables in Beijing, 200 scenarios generated in the same way for Tianjin, 200 scenarios for Northern Hebei, 200 scenarios for Southern Hebei, and 200 scenarios for Eastern Hebei, 3. 100$^g$ scenarios generated by applying the LHS method for the total emissions of NO$_x$, SO$_2$, NH$<em>3$, NMVOC + IVOC, and POA, 4. 20 scenarios in which one primary inorganic PM$</em>{2.5}$ control variable is set to 0.25 for each scenario.</td>
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Overall, $^a$ 100 and 200 scenarios are needed for the response surfaces for five and seven variables, respectively (Xing et al., 2011; Wang et al., 2011). $^b$ LPS includes power plants, iron and steel plants, and cement plants.
Table 2. Comparison between ERSM-predicted and CMAQ/2D-VBS-simulated PM$_{2.5}$ concentrations for 54 out-of-sample scenarios.

<table>
<thead>
<tr>
<th>Month</th>
<th>Variable</th>
<th>Statistical index</th>
<th>Beijing</th>
<th>Tianjin</th>
<th>Northern Hebei</th>
<th>Eastern Hebei</th>
<th>Southern Hebei</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jan PM$_{2.5}$ concentration</td>
<td>$R$</td>
<td>0.998</td>
<td>0.998</td>
<td>0.995</td>
<td>0.997</td>
<td>0.997</td>
<td></td>
</tr>
<tr>
<td>MNE (%)</td>
<td>0.52</td>
<td>0.55</td>
<td>0.64</td>
<td>0.67</td>
<td>0.60</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum NE (%)</td>
<td>7.56</td>
<td>6.98</td>
<td>10.67</td>
<td>8.01</td>
<td>8.03</td>
<td></td>
<td></td>
</tr>
<tr>
<td>95 % percentile of NEs (%)</td>
<td>1.61</td>
<td>2.86</td>
<td>2.92</td>
<td>3.46</td>
<td>3.02</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NME (%)</td>
<td>0.44</td>
<td>0.46</td>
<td>0.57</td>
<td>0.53</td>
<td>0.53</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PM$_{2.5}$ response</td>
<td>$R$</td>
<td>0.998</td>
<td>0.998</td>
<td>0.995</td>
<td>0.997</td>
<td>0.997</td>
<td></td>
</tr>
<tr>
<td>NME (%)</td>
<td>3.36</td>
<td>3.48</td>
<td>4.25</td>
<td>4.00</td>
<td>3.88</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mar PM$_{2.5}$ concentration</td>
<td>$R$</td>
<td>0.999</td>
<td>0.996</td>
<td>0.998</td>
<td>0.995</td>
<td>0.999</td>
<td></td>
</tr>
<tr>
<td>MNE (%)</td>
<td>0.37</td>
<td>0.54</td>
<td>0.39</td>
<td>0.57</td>
<td>0.49</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum NE (%)</td>
<td>3.75</td>
<td>6.58</td>
<td>4.30</td>
<td>5.04</td>
<td>3.22</td>
<td></td>
<td></td>
</tr>
<tr>
<td>95 % percentile of NEs (%)</td>
<td>1.53</td>
<td>3.15</td>
<td>2.03</td>
<td>4.35</td>
<td>2.03</td>
<td></td>
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</tr>
<tr>
<td>NME (%)</td>
<td>0.31</td>
<td>0.45</td>
<td>0.34</td>
<td>0.49</td>
<td>0.42</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PM$_{2.5}$ response</td>
<td>$R$</td>
<td>0.999</td>
<td>0.996</td>
<td>0.998</td>
<td>0.995</td>
<td>0.999</td>
<td></td>
</tr>
<tr>
<td>NME (%)</td>
<td>2.38</td>
<td>3.71</td>
<td>2.70</td>
<td>4.55</td>
<td>3.59</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Jul PM$_{2.5}$ concentration</td>
<td>$R$</td>
<td>0.997</td>
<td>0.998</td>
<td>0.998</td>
<td>0.999</td>
<td>0.999</td>
<td></td>
</tr>
<tr>
<td>MNE (%)</td>
<td>0.94</td>
<td>0.54</td>
<td>0.46</td>
<td>0.37</td>
<td>0.47</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum NE (%)</td>
<td>5.05</td>
<td>5.02</td>
<td>4.65</td>
<td>1.83</td>
<td>3.62</td>
<td></td>
<td></td>
</tr>
<tr>
<td>95 % percentile of NEs (%)</td>
<td>3.47</td>
<td>2.33</td>
<td>2.17</td>
<td>1.49</td>
<td>1.87</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NME (%)</td>
<td>0.80</td>
<td>0.47</td>
<td>0.41</td>
<td>0.33</td>
<td>0.39</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PM$_{2.5}$ response</td>
<td>$R$</td>
<td>0.997</td>
<td>0.998</td>
<td>0.998</td>
<td>0.999</td>
<td>0.999</td>
<td></td>
</tr>
<tr>
<td>NME (%)</td>
<td>4.97</td>
<td>3.71</td>
<td>2.80</td>
<td>2.58</td>
<td>2.78</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oct PM$_{2.5}$ concentration</td>
<td>$R$</td>
<td>0.996</td>
<td>0.994</td>
<td>0.999</td>
<td>0.999</td>
<td>0.999</td>
<td></td>
</tr>
<tr>
<td>MNE (%)</td>
<td>0.83</td>
<td>0.70</td>
<td>0.36</td>
<td>0.39</td>
<td>0.36</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum NE (%)</td>
<td>8.90</td>
<td>11.19</td>
<td>3.79</td>
<td>3.90</td>
<td>2.46</td>
<td></td>
<td></td>
</tr>
<tr>
<td>95 % percentile of NEs (%)</td>
<td>3.04</td>
<td>3.50</td>
<td>1.44</td>
<td>2.10</td>
<td>1.64</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NME (%)</td>
<td>0.67</td>
<td>0.58</td>
<td>0.30</td>
<td>0.35</td>
<td>0.32</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PM$_{2.5}$ response</td>
<td>$R$</td>
<td>0.996</td>
<td>0.994</td>
<td>0.999</td>
<td>0.999</td>
<td>0.999</td>
<td></td>
</tr>
<tr>
<td>NME (%)</td>
<td>4.51</td>
<td>5.64</td>
<td>2.20</td>
<td>3.29</td>
<td>2.79</td>
<td></td>
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</tr>
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Figure 2. Comparison of PM$_{2.5}$ concentrations (a) and PM$_{2.5}$ responses (b) predicted by the ERSM technique with out-of-sample CMAQ/2D-VBS simulations. The dashed line is the one-to-one line, indicating perfect agreement.
cies between ERSM and CMAQ/2D-VBS cannot challenge the major conclusions on the effectiveness of emission reductions. Finally, as stated in the last paragraph, all sensitivity scenarios used in the following discussions have emission ratios $\geq 0.2$, since $< 0.2$ emission reductions are quite rare as they are limited by the technologically feasible reduction potentials (S. X. Wang et al., 2014).

3.2 Response of PM$_{2.5}$ concentrations to emissions of air pollutants

Having demonstrated the reliability of the ERSM prediction system, we employ it to investigate the responses of PM$_{2.5}$ concentrations to emissions of various pollutants from different sectors and regions. We use PM$_{2.5}$ sensitivity defined in Sect. 2.2 to quantitatively characterize the sensitivity of PM$_{2.5}$ concentrations to emission changes. Figure 4 illustrates the sensitivity of 4-month (January, March, July, and October) mean PM$_{2.5}$ concentrations to the stepped control of individual air pollutants (a) and individual pollutant–sector combinations (b) in the BTH region, which are derived from the ERSM technique. The Fig. 4a can be obtained from both the RSM and ERSM prediction systems, and their results are consistent, whereas Fig. 4b, as well as the results shown in Figs. 5 and 6 can only be derived from ERSM. Among all pollutants, the 4-month mean PM$_{2.5}$ concentrations are the most sensitive to the emissions of primary inorganic PM$_{2.5}$ in all five regions, and the PM$_{2.5}$ sensitivities vary from 24 to 36% according to region. When primary inorganic PM$_{2.5}$ emissions from various sectors are differentiated, the industry sector is found to make the largest contribution to PM$_{2.5}$ concentrations, followed by the residential and commercial sectors; the contribution of power plants is negligibly small because of smaller emissions and higher stacks. The PM$_{2.5}$ sensitivities to primarily inorganic PM$_{2.5}$ emissions remain constant at various reduction ratios.

While primary inorganic PM$_{2.5}$ makes the largest contribution to PM$_{2.5}$ concentrations among all air pollutants, the total contributions of all precursors (NO$_x$, SO$_2$, NH$_3$, NMVOC, IVOC, and POA), which range between 31 and 48%, exceed that of primary inorganic PM$_{2.5}$ (24–36%). Among the precursors, PM$_{2.5}$ concentrations are primarily sensitive to the emissions of NH$_3$, NMVOC + IVOC, and POA, and their relative importance differs according to reduction ratio. The PM$_{2.5}$ sensitivity to NH$_3$ increases substantially with the increase in reduction ratio, primarily attributable to the transition from NH$_3$-rich to NH$_3$-poor regimes when more controls are enforced. The PM$_{2.5}$ sensitivities to POA and NMVOC + IVOC, however, decrease slightly with the increase in reduction ratio. This is because, based on the gas-particle absorptive partitioning theory, organics have a higher tendency to partition into the particle phase at larger OA concentrations. As a result of the nonlinearity, the PM$_{2.5}$ sensitivities to POA and NMVOC + IVOC emissions are larger than those to NH$_3$ emissions at small reduction ratios (e.g., 20%), while it is the other way around at large reduction ratios (e.g., 80%).

The PM$_{2.5}$ sensitivity to SO$_2$ emissions is considerably smaller compared with the three precursors above and does not change significantly as a function of reduction ratio. From 2007 to 2014 (the base year of this study), both SO$_2$ emissions and SO$_2^-$ concentrations in PM$_{2.5}$ have been continuously decreasing due to effective control policies (Wang et al., 2017), which partly explains the small sensitivity of PM$_{2.5}$ to SO$_2$ emissions. The response of PM$_{2.5}$ concentrations to NO$_x$ emissions could change from negative to positive with the increase in reduction ratio, which has been reported in several previous studies (Dong et al., 2014; Zhao et al., 2013c; Cai et al., 2017). Small NO$_x$ emission reductions could lead to an increase in O$_3$ and HO$_x$ concentrations in several seasons owing to an NMVOC-limited photochemical regime, which on the one hand enhances SO$_2^-$ and SOA formation and, on the other hand, could also increase NO$_3^-$ concentrations by accelerating the nocturnal formation of N$_2$O$_5$ and HNO$_3$ through the NO$_2$ + O$_3$ reaction at low temperatures. A substantial reduction in NO$_x$ emissions, however, transforms the NMVOC-limited regime to an NO$_x$-limited regime, resulting in a successive decline in concentrations of O$_3$, HO$_x$, and most PM$_{2.5}$ chemical components. Judging from our simulation results (Fig. 4), if only the NO$_x$ emissions within the BTH region are controlled, a very large reduction ratio of about 80% is required to realize a reduction in annual PM$_{2.5}$ concentrations in most areas. However, the effects could be distinctly different if NO$_x$ emissions outside the BTH region are jointly reduced. Our previous studies using the CMAQ model (Zhao et al., 2013c; Wang et al., 2010, 2011) have shown that uniform reductions in NO$_x$ emissions in the whole of China by 23–50% result in considerable annual PM$_{2.5}$ reduction over the BTH region. This is because NO$_x$ emission reductions in upwind regions are more likely to result in a net PM$_{2.5}$ decrease compared with local emission reductions, since the photochemistry typically changes from an NMVOC-limited regime in local urban areas at the surface to an NO$_x$-limited regime in downwind areas or at upper levels (Xing et al., 2011). The results shown in Fig. 4 also support the abovementioned pattern and mechanism to some extent: even a 20% NO$_x$ emission reduction in BTH can lead to PM$_{2.5}$ decrease in Northern Hebei because, as the northernmost region in BTH, it is significantly affected by emissions in other regions within BTH. Note that some recently discovered chemical pathways are missing in the model, such as the oxidation of SO$_2$ by NO$_2$ in aerosol water and the SO$_2$ heterogeneous reactions on the dust surface (Fu et al., 2016; Cheng et al., 2016; G. H. Wang et al., 2016). The incorporation of these processes in the model may affect the simulated responses of PM$_{2.5}$ to NO$_x$ and SO$_2$ emissions. Regarding emission sectors, the contributions of SO$_2$ and NO$_x$ emissions are dominated by “other sources” (sources other than LPS) because they emit larger amount of pollutants at lower height compared with LPS.
Figure 3. Comparison of the 2-D isopleths of PM$_{2.5}$ concentrations in Beijing in response to the simultaneous changes in precursor emissions in all five regions derived from the conventional RSM technique and the ERSM technique. The $x$ and $y$ axes represent the emission ratio, defined as the ratios of the changed emissions to the emissions in the base case. The color contours represent PM$_{2.5}$ concentrations (unit: µg m$^{-3}$).
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www.atmos-chem-phys.net/17/12031/2017/

Figure 4. Sensitivity of 4-month mean PM$_{2.5}$ concentrations to the stepped control of individual air pollutants (a) and individual pollutant–sector combinations (b). The x axis shows the reduction ratio (1 – emission ratio). The y axis shows PM$_{2.5}$ sensitivity, which is defined as the change ratio of concentration divided by the reduction ratio of emissions. The colored bars denote the PM$_{2.5}$ sensitivities when a particular emission source is controlled while the others stay the same as the base case; the black dotted line denotes the PM$_{2.5}$ sensitivity when all emission sources are controlled simultaneously. The red stars represent PM$_{2.5}$ concentrations in the base case.

The black dotted lines in Fig. 4 show the PM$_{2.5}$ sensitivity when all pollutants from all sectors are controlled simultaneously. The sum of PM$_{2.5}$ sensitivities to individual pollutant–sector combinations (stacked columns) is mostly larger than the sensitivity to all pollutants and sectors (black dotted lines), especially under large reduction ratios. This is mainly attributed to the overlapping effect of two precursors (e.g., SO$_2$ and NH$_3$) involved in the formation of ammonium sulfate and ammonium nitrate. Nevertheless, at small reduction ratios, the sum of individual sensitivities is sometimes smaller because the negative effects of reducing NO$_x$ are mitigated when we simultaneously reduce NO$_x$ emissions from multiple sectors as well as emissions of other air pollutants such as NMVOC. When all pollutants and sectors are controlled together, the PM$_{2.5}$ sensitivity generally increases with reduction ratio, indicating that an additional air quality benefit could be achieved, larger than expected from linear extrapolation, if more control measures were implemented.

Figure 5 illustrates the PM$_{2.5}$ sensitivities to individual pollutant–sector combinations in each month. The source contribution features are significantly discrepant in different months. The contributions of primary inorganic PM$_{2.5}$ emissions to PM$_{2.5}$ concentrations are notably higher in January than in other months, which is probably attributed to weaker dilution and slower chemical reactions in January. Regarding different emission sectors of primary inorganic PM$_{2.5}$, the industrial sector plays a dominant role in all months except January, when the residential and commercial sectors make a similar or even larger contribution as compared to the industrial sector. The higher contribution of the residential and commercial sectors in January is on the one hand because of the higher emissions due to heating, and, on the other hand, it is explained by weaker vertical mixing in winter, which results in a larger relative contribution of low-level sources. This result highlights the importance of residential and commercial sources for PM$_{2.5}$ pollution controls in the winter.

The contributions of precursors are dominated by POA and NMVOC + IVOC in January, while in July, NO$_x$, SO$_2$, and NH$_3$, which are known to be precursors of secondary inorganic aerosols, make larger contributions than POA and NMVOC + IVOC. The responses of PM$_{2.5}$ concentrations to NO$_x$ emissions can be the opposite in different seasons. Specifically, in July, NO$_x$ emission reductions always induce a decrease in PM$_{2.5}$ concentrations due to an NO$_x$-limited photochemical regime. In January, however, even an 80% reduction in NO$_x$ emissions (roughly the maximum technically feasible reduction ratio) could result in a net PM$_{2.5}$ increase, as a result of a strong NMVOC-limited regime. To achieve a net PM$_{2.5}$ reduction in January, it would be necessary to simultaneously reduce NO$_x$ emissions outside the BTH region.

We further evaluate the contributions of primary inorganic PM$_{2.5}$ and precursor emissions from various regions to PM$_{2.5}$ concentrations (Figs. 6 and S6). Here the contributions are quantified by comparing the base case with sensitivity scenarios in which emissions from a specific source are reduced by 80%, which reaches the maximum technologically feasible reduction ratios of major pollutants in most areas (S. X. Wang et al., 2014). Obviously, the contributions of total primary inorganic PM$_{2.5}$ emissions in the BTH region are dominated by local sources, which account for over 75% of the total primary inorganic PM$_{2.5}$ contributions. When precursor emissions are decomposed into different regions, local sources usually also represent the largest contributors, but precursor emissions from other regions (denoted by “regional precursor emissions” hereafter) could also make significant
contributions, depending on regions and seasons. The pre- 
cursor emissions from the northern part of BTH (e.g., Nor- 
thern Hebei, Beijing) mainly contribute to local PM$_{2.5}$ 
concentrations, whereas those from the southern part of BTH 
(e.g., Southern Hebei) significantly affect the PM$_{2.5}$ con- 
centrations in both the local region and other regions. Over the 
BTH, heavy pollution is frequently associated with southerly 
wind, while strong northerly wind often blows away PM$_{2.5}$ 
pollution (Jia et al., 2008; Zheng et al., 2015), which explains 
the higher contribution of emissions from southern BTH to 
other regions. Moreover, the importance of regional precur- 
sor emissions relative to local ones is remarkably higher in 
July than in January, which can be explained by the southerly 
monsoon and stronger vertical mixing in summer that favors 
the interregional transport of air pollutants. We also exam- 
ine the contributions of emissions outside the BTH region to 
PM$_{2.5}$ concentrations in the five target regions. The re- 
results reveal that these emissions contribute 24–33 % of the 4- 
month mean PM$_{2.5}$ concentrations, among which more than 
80 % could be attributed to precursor emissions. Among 
the 4 months, the contribution of emissions outside BTH is con- 
siderably smaller in January (12–21 %) as compared to other 
months (29–38 %).

3.3 Response of PM$_{2.5}$ chemical components to 
emissions of air pollutants

Ambient PM$_{2.5}$ is comprised of complicated chemical com- 
ponents with distinctly different formation pathways. To gain 
deeper insight into the formation mechanisms and source 
attrition of PM$_{2.5}$, we examine the sensitivities of major 
PM$_{2.5}$ components, including NO$_3^-$, SO$_2^{2-}$, and OA, to the 
stepped control of individual air pollutants, as shown in Fig. 7 
(January and July) and Fig. S7 (March and October). NO$_3^-$ 
concentrations are the most sensitive to NH$_3$ emissions in all 
months except July, when the sensitivities of NO$_3^-$ concen- 
trations to NH$_3$ and NO$_x$ emissions are similar. The NO$_3^-$ 
sensitivities to NO$_x$ emissions differ significantly accord- 
ing to season. In most months, NO$_3^-$ concentrations are posi- 
tively correlated with NO$_x$ emissions. In January, however, 
the sensitivities of NO$_3^-$ to both NH$_3$ and NO$_x$ emissions show pronounced increasing trends with the 
increase in reduction ratio, in agreement with the strong
nonlinearity in these two pollutants described in Sect. 3.2. NMVOC emissions make moderate positive contributions to NO$_3^-$, with the largest and smallest contributions occurring in January and July in conjunction with NMVOC-limited and NO$_x$-limited photochemical regimes, respectively. Finally, SO$_2$ emissions have very small influences on NO$_3^-$ concentrations.

For SO$_4^{2-}$, SO$_2$ emissions represent the dominant contributor in all months. The sensitivity of SO$_4^{2-}$ concentrations to SO$_2$ emissions does not change significantly with respect to reduction ratio, consistent with the results shown in Section 3.2. The contributions of NH$_3$ emissions to SO$_4^{2-}$ concentrations are quite small except in October, when NH$_3$ accounts for approximately one-fourth of the contribution of SO$_2$. NO$_x$ emissions affect SO$_4^{2-}$ concentrations mainly by altering O$_3$ and HO$_x$ concentrations, the effects of which are positive in July at large reduction ratios and mostly negative in other months. NMVOC emissions can make a small impact on SO$_4^{2-}$ concentrations primarily through changing O$_3$ and HO$_x$ concentrations.
The emissions of POA and NMVOC + IVOC are obviously two major contributors to OA concentrations. The relative importance of the two is strongly dependent on season. In July, POA and NMVOC + IVOC make similar contributions to OA concentrations, while POA usually contributes more in other months. In January, the contribution of POA could account for about 4 times that of NMVOC + IVOC. The higher relative contribution of POA emissions in January can be explained in several ways. First, the POA emissions are relatively higher in January due to residential heating, while the NMVOC emissions from solvent use and biogenic sources are higher in July. Second, lower temperature in winter favors the partitioning of the semi-volatile components comprising POA to the particle phase, whereas higher temperature and stronger radiation in July accelerate the formation of SOA from NMVOC + IVOC. Similar to SO$_2^{-}$, the impact of NO$_x$ emissions on OA concentrations also works through two pathways. Besides the abovementioned photochemical pathway, NO$_x$ emission reductions could lead to OA increases due to the fact that SOA yield, defined as the ratio of SOA formation to the consumption of a precursor, is generally higher at a low-NO$_x$ condition than at a high-NO$_x$ condition. As an integrated effect, the responses of OA concentrations to NO$_x$ emissions are negative in most situations.

3.4 PM$_{2.5}$ responses to emission reductions during heavy-pollution episodes

Having shown the responses of monthly mean PM$_{2.5}$ concentrations to pollutant emissions, we are also interested in heavy-pollution episodes, in which the source contributions could be quite different from the monthly mean results, largely due to variations in meteorological conditions. To provide more insight into the control strategies for heavy pollution, we use the ERSM technique to investigate the source contributions.
contribution features during three typical heavy-pollution episodes. We first select 47 heavy-pollution episodes over the BTH region during 2013–2015 (Table S7). Subsequently, we employ the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015) and concentration weighted trajectory (CWT) method (Cheng et al., 2013) to identify the potential source regions for PM$_{2.5}$ during each episode and categorize these episodes according to their source regions. We then select a representative episode from each of the three most important pollution types in which the air mass primarily originates from local areas (“Local” type), from the south (“South” type), and from the southeast (“Southeast” type). We give preference to episodes within the 4-month simulation period of this study to facilitate a comparison with the monthly mean source contribution features. For this reason, we select (1) 5–7 January 2014, (2) 7–11 October 2014, and (3) 29–31 October 2014 as representatives of the Local, South, and Southeast types. The selection of heavy-pollution episodes is detailed in Sect. S2.

Figure 8 shows the contribution of precursor and primary inorganic PM$_{2.5}$ emissions from individual regions to PM$_{2.5}$ concentrations during the three heavy-pollution episodes, and Fig. 9 illustrates the sensitivity of PM$_{2.5}$ concentrations to the stepped control of individual pollutant–sector combinations. During 5–7 January 2014 (Local type), the contributions of local emission sources to PM$_{2.5}$ concentrations far exceed those from other regions within BTH as well as from outside of BTH (Fig. 8). In contrast to the monthly mean results (Sect. 3.2), the contributions of primary inorganic PM$_{2.5}$ emissions are comparable to, and even larger than, those of precursor emissions in the BTH region. The total contributions of primary PM$_{2.5}$ (including POA) account for as much as 70–80% of the contributions of all pollutants within the BTH region, which highlights the crucial importance of primary PM$_{2.5}$ controls during this episode. Moreover, the control of NMVOC, NH$_3$, and SO$_2$ emissions could contribute moderately to reducing PM$_{2.5}$ concentrations. However, NO$_x$ emission reduction induces an increase in PM$_{2.5}$ concentrations, even at an 80% reduction ratio. Therefore, effective temporary control measures for this episode should focus on the control of local emissions, with emphasis laid on primary PM$_{2.5}$.

During 7–11 October 2014 (South type), the contributions of emissions outside BTH to PM$_{2.5}$ concentrations are as large as 33% in Beijing and 40–50% in other regions. Within the BTH region, the emissions from Southern Hebei can have similar effects to local emissions on PM$_{2.5}$ concentrations in Beijing, indicating a strong long-range transport from the south. In addition, the total contributions of precursor emissions about double those of primary inorganic PM$_{2.5}$ emissions. Among all precursors, PM$_{2.5}$ concentrations are mainly sensitive to emissions of NH$_3$, NMVOC + IVOC, and POA. The sensitivity of PM$_{2.5}$ concentrations to NO$_x$ emissions increases dramatically with reduction ratio. Although small NO$_x$ reductions may slightly elevate PM$_{2.5}$ concentrations, large NO$_x$ emission reduction (>50%) can result in significant PM$_{2.5}$ reduction. To effectively mitigate PM$_{2.5}$ pollution during this episode, we should implement control measures for precursor emissions in both the BTH region (especially the southern part) and regions south of BTH. The NO$_x$ emissions, if controlled, should be reduced by at least 50% to avoid adverse side effect.

For 29–31 October 2014 (Southeast type), PM$_{2.5}$ concentrations are also significantly affected by emissions outside the BTH region. Within the BTH region, the PM$_{2.5}$ concentrations in Beijing and Northern Hebei are about equally affected by local emissions and emissions from Eastern Hebei and Southern Hebei, while local emissions play dominant roles in other regions. The emissions of both precursor and primary inorganic PM$_{2.5}$ within the BTH region make important contributions to PM$_{2.5}$ concentrations, and the relative significance of the two is dependent on region. All precursors except NO$_x$ can contribute considerably to PM$_{2.5}$ reductions, and the sensitivity of PM$_{2.5}$ to NH$_3$ increases rapidly with emission ratio. NO$_x$ emissions are negatively correlated with PM$_{2.5}$ concentrations in most cases. Regarding the temporary control strategy for this episode, it is preferable to implement joint control of primary PM$_{2.5}$ and precursors both within and outside the BTH region, with stringent measures over Eastern and Southern Hebei.

From the analysis above, we conclude that the source contributions are tremendously different in these three episodes, which have been demonstrated to represent some key features of the corresponding pollution types (Local, South, and Southeast types). Therefore, episode-specific control strategies need to be formulated based on the source contribution features of individual pollution types. Nevertheless, the results of this study are not yet sufficient to guide the development of temporary control strategies for all heavy-pollution episodes because the conclusions drawn from the three episodes may not be generalized to pollution types. In future studies, we need to simulate more episodes to improve their classification and to comprehensively understand the source contribution features of each pollution type. For a coming heavy-pollution episode, we can predict its pollution type using an air quality forecasting model and subsequently formulate the temporary control strategies based on the source contribution features of this specific pollution type.

4 Conclusion and implications

In the present study, we investigated the nonlinear response of PM$_{2.5}$ concentrations to emission changes in multiple pollutants from different sectors and regions over the BTH region, using the ERSM technique coupled with the CMAQ/2D-VBS model.

Among all pollutants, primary inorganic PM$_{2.5}$ makes the largest contribution (24–36%) to the 4-month mean
PM$_{2.5}$ concentrations. The contribution from primary inorganic PM$_{2.5}$ is especially high in heavily polluted winter and is dominated by the industry as well as residential and commercial sectors. The total contributions of all precursors to PM$_{2.5}$ concentrations range between 31 and 48%. Among the precursors, PM$_{2.5}$ concentrations are primarily sensitive to the emissions of NH$_3$, NMVOC + IVOC, and POA. With the increase in reduction ratio, the sensitivities of PM$_{2.5}$ concentrations to pollutant emissions remain roughly constant for primary inorganic PM$_{2.5}$ and SO$_2$, increase substantially for NH$_3$ and NO$_x$, and decrease slightly for POA and NMVOC + IVOC. The contributions of primary inorganic PM$_{2.5}$ emissions to PM$_{2.5}$ concentrations are dominated by local emission sources, which account for over 75% of the total primary inorganic PM$_{2.5}$ contributions. For precursors, however, emissions from other regions could play similar roles to local emission sources in the summer and over the northern part of BTH. Different PM$_{2.5}$ chemical components are associated with distinct source contribution features. The NO$_3^-$ and SO$_4^{2-}$ concentrations are the most sensitive to emissions of NH$_3$ and SO$_2$, respectively. The emissions of the POA and NMVOC + IVOC are two major

Figure 8. The contribution of precursor (NO$_x$, SO$_2$, NH$_3$, NMVOC, IVOC, and POA) and primary inorganic PM$_{2.5}$ emissions from individual regions to PM$_{2.5}$ concentrations during three typical heavy-pollution episodes.
contributors to OA concentrations, with their relative importance depending on season.

The source contribution features are significantly different for three typical heavy-pollution episodes, which belong to three distinct pollution types. The PM$_{2.5}$ concentrations in the first episode (Local type) are dominated by local sources and primary PM$_{2.5}$ emissions, while the second episode (South type) is primarily affected by precursor emissions from local and southern regions. The third episode (Southeast type) is significantly influenced by emissions of both primary inorganic PM$_{2.5}$ and precursors from multiple regions. Future investigations are needed to acquire generalized patterns for the source contributions of various heavy-pollution types.

The results of the present study have important implications for PM$_{2.5}$ control policies over the BTH region. First, the control of primary PM$_{2.5}$ emissions should be a priority in PM$_{2.5}$ control strategies. Primary PM$_{2.5}$, including primary inorganic PM$_{2.5}$ and POA, contribute over half of the 4-month mean PM$_{2.5}$ concentrations, which is even higher in the winter when heavy pollution frequently occurs. The industry sector and the residential and commercial sectors represent 85% of the total primary PM$_{2.5}$ emissions and therefore should be the focus of primary PM$_{2.5}$ controls. In particular, we should pay special attention to the residential and commercial sectors, which account for half of the total contribution of primary PM$_{2.5}$ emissions to PM$_{2.5}$ concentrations in the winter but have been frequently neglected in China’s previous control policies. Second, the control policies for NMVOC and IVOC emissions should be strengthened. The sensitivity of PM$_{2.5}$ concentrations to NMVOC + IVOC is one of the largest among all precursors. In particular, the control of NMVOC and IVOC emissions is very effective for PM$_{2.5}$ reduction even at the initial control stage, as indicated by the large sensitivity at small reduction ratios. Moreover, NMVOC reduction is also crucial for the mitigation of O$_3$ pollution considering an NMVOC-limited regime over the urban region and its surrounding areas (Xing et al., 2011). Third, NO$_x$ emissions should be substantially reduced in both the BTH and other parts of China; in the long run, the reduction ratio should preferably approach their maximum feasible reduction levels. Fourth, more stringent control policies should be enforced in Southern Hebei, which on the one hand suffers from the most severe PM$_{2.5}$ pollution.

Figure 9. Sensitivity of PM$_{2.5}$ concentrations to the stepped control of individual air pollutants from individual sectors during three heavy-pollution episodes. The meanings of x axis, y axis, colored bars, and black dotted lines are the same as in Fig. 4.
(L. T. Wang et al., 2014) and, on the other hand, significantly affects both local and regional PM$_{2.5}$ concentrations. Last but not least, considering the distinct source contributions in different heavy-pollution episodes, episode-specific temporary control strategies should be formulated according to the source contribution feature of the specific pollution type.

The present study has a few limitations. First, the establishment of ERSM requires several hundred or over 1000 emission scenarios, although the scenario number needed for a specific number of control variables has already been dramatically reduced as compared to the conventional RSM technique. Studies are needed to further reduce the scenario number but retain the accuracy of the ERSM technique. Second, the current ERSM is developed based on the meteorological conditions simulated for the base year and has not considered the impact of interannual variations in meteorological conditions on the relationships between emissions and PM$_{2.5}$ concentrations. Third, although the ERSM-predicted responses of PM$_{2.5}$ concentrations to precursor emissions have been demonstrated to agree well with chemical transport model simulations, evaluating the predicted responses against the actual situation in the real atmosphere still represents a major challenge because it is extremely difficult to artificially perturb emissions in the atmosphere. Last but not the least, the NMVOC and IVOC emissions have been lumped together in this study to reduce the number of control variables. Considering their differences in sources and SOA formation potentials (Jathar et al., 2014; Wu et al., 2017), a detailed quantification of the individual contributions of NMVOC and IVOC emissions from various sources to PM$_{2.5}$ concentrations is required in the future to better inform NMVOC/IVOC control policies.

Data availability. All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplement. Additional data related to this paper can be requested from the authors.

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