

Response to Referee #1 (gmd-2021-259)

We Thank Reviewer for his/her constructive comments.

Responses to the comments:

General comments: The authors describe a method for determining the contributions of different processes to PM_{2.5} formation and the couplings between the processes. The authors call this method Quantitative Decoupling Analysis (QDA) and apply the method to a haze episode in the Beijing-Tianjin-Hebei region from 17-28 February 2014. The manuscript is generally well written. However, there are three significant problems with the work:

Comment 1: The QDA method is not new. This is the Factor Separation method introduced in 1993 and applied in later work. See references below.

Reply: Thanks for this important comment. We agree that the QDA method developed in this study share same theoretical basis (Tylor series expansion) with the Factor Separation method introduced by Stein and Alpert (1993), but there are important differences between these two methods. The similarity between the Factor Separation method and the QDA method is that they employ same algorithms to separate the contributions of different factors, while the biggest difference between the Factor Separation method and the QDA method is in the object that they resolved. They are two types of analysis method. The former is the relative analysis method while the latter is the absolute analysis method. As seen in Fig.R1, the Factor Separation method is designed to resolve the effects of different factors on the differences between model results of two scenarios (i.e., control simulation – base simulation). This makes the contributions of different factors resolved by Factor Separation are relative, which are dependent on the choice of scenarios. It cannot obtain the absolute contribution of different factors. Different from the Factor Separation method, the QDA method aims to track the contributions of different factors on the variations of model results between adjacent timesteps (Fig. R1). This means that the results of QDA methods are absolute, which only depends on the cases we choose and does not need an additional scenario to be compared. Take the estimation of meteorology contribution to the PM_{2.5} concentration as an example. The Factor Separation method need another case with different meteorology condition to estimate the meteorology contribution. This makes the meteorology contribution estimated by Factor Separation method is in fact the contribution induced by meteorology variation between these two cases. It cannot estimate the absolute contribution of meteorology to the variation of PM_{2.5} in an individual case. However, the meteorology contribution estimated by QDA method is the absolute contribution made meteorology processes in this individual case.

Therefore, we believe the QDA method is a new method different from the Factor Separation method. To facilitate the understanding of the QDA method and highlight its novelty, in the revised manuscript, we have made a more detailed description on the QDA method, including its theoretical basis, algorithms, its realization in model as well as its relationship with the SAA (Factor Separation method) and IPR method,

1.1 Theoretical basis of the QDA method

The QDA method is developed based on the Taylor series expansion. Considering that the $PM_{2.5}$ concentration at t step is $PM_{2.5}^t$ and the $PM_{2.5}$ concentration at $t+1$ step after undergoing the emission, meteorology and chemistry processes with $PM_{2.5}^t$ as initial condition is $PM_{2.5}^{t+1}$, then we could define a function F that denotes the simulated $PM_{2.5}$ concentrations with or without different processes using $PM_{2.5}^t$ as initial concentration, such that:

$$F(0,0,0) = PM_{2.5}^t \quad (R1)$$

$$F(x_1, x_2, x_3) = PM_{2.5}^{t+1} \quad (R2)$$

where $F(x_1, x_2, x_3)$ represents the simulated $PM_{2.5}$ concentration with meteorology (x_1), emission (x_2), and chemistry processes (x_3); $F(0,0,0)$ represents the simulated $PM_{2.5}$ concentration without emission, meteorology, and chemistry processes. Therefore, the $PM_{2.5}^t$ and the $PM_{2.5}^{t+1}$ can be seen as the different values of function F with different input data, and the variation of $PM_{2.5}$ concentration between two timesteps can be written as:

$$\Delta PM_{2.5}^{t+1} = PM_{2.5}^{t+1} - PM_{2.5}^t = F(x_1, x_2, x_3) - F(0,0,0) \quad (R3)$$

where $\Delta PM_{2.5}^{t+1}$ represents the variation of $PM_{2.5}$ concentration from t to $t+1$ step,

According to Taylor series expansion, the function F can be decomposed as follows:

$$\begin{aligned} F(x_1, x_2, x_3) - F(0,0,0) = & \sum_{i=1}^3 \frac{\partial F}{\partial x_i} x_i + \frac{1}{2!} \left(\sum_{i=1}^3 \frac{\partial^2 F}{\partial x_i^2} x_i^2 + 2 \frac{\partial^2 F}{\partial x_1 \partial x_2} x_1 x_2 + 2 \frac{\partial^2 F}{\partial x_2 \partial x_3} x_2 x_3 + 2 \frac{\partial^2 F}{\partial x_1 \partial x_3} x_1 x_3 \right) + \\ & \frac{1}{3!} \left(\sum_{i=1}^3 \frac{\partial^3 F}{\partial x_i^3} x_i^3 + \sum_{a=1}^2 3 \frac{\partial^3 F}{\partial x_1^a \partial x_2^{3-a}} x_1^a x_2^{3-a} + \sum_{a=1}^2 3 \frac{\partial^3 F}{\partial x_2^a \partial x_3^{3-a}} x_2^a x_3^{3-a} + \sum_{a=1}^2 3 \frac{\partial^3 F}{\partial x_1^a \partial x_3^{3-a}} x_1^a x_3^{3-a} + \right. \\ & \left. \frac{\partial^3 F}{\partial x_1 \partial x_2 \partial x_3} 6 x_1 x_2 x_3 \right) + \dots + o^n \end{aligned} \quad (R4)$$

Based on this equation, the terms that only containing a single partial derivative to x_1 , x_2 , and x_3 (including any higher-order derivatives) are defined as pure contribution of the meteorology (M), emission (E) and chemistry (C) processes to the variation of $PM_{2.5}$ concentrations. Therefore, the term $\frac{\partial F}{\partial x_1} x_1 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_1^2} x_1^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_1^3} x_1^3 + \dots$ in Eq.(R2) is defined as E, term $\frac{\partial F}{\partial x_2} x_2 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_2^2} x_2^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_2^3} x_2^3 + \dots$ is defined as M, and the term $\frac{\partial F}{\partial x_3} x_3 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_3^2} x_3^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_3^3} x_3^3 + \dots$ is defined as C. The cross terms then represent the interaction among different drivers, for example the term $\frac{1}{2!} \frac{\partial^2 F}{\partial x_1 \partial x_2} x_1 x_2 + \frac{1}{3!} \sum_{a=1}^2 \frac{\partial^3 F}{\partial x_1^a \partial x_2^{3-a}} x_1^a x_2^{3-a} + \dots$ is defined as the interactions between meteorology and emission (ME), the term $\frac{1}{3!} \left(\frac{\partial^3 F}{\partial x_1 \partial x_2 \partial x_3} 6 x_1 x_2 x_3 \right) + \dots$ is defined as the interactions among emission, meteorology and chemistry processes (MCE). Detailed definitions to the different factors we resolved are available in Table R1. Note that pure here as well as elsewhere in the paper, is in the relative sense meaning that the effect due to one factor is separated

from the other chosen factors. For example, the pure contribution of emission is only due to the direct emission at local space. The variation of PM_{2.5} concentration after emission process are seen as the contribution of meteorology and chemistry. Therefore, the values of E in the QDA method cannot represent the whole effects of emission in the common sense.

According to these definitions, the PM_{2.5} variations from t to t+1 step can be written as the sum of M^{t+1} , E^{t+1} , C^{t+1} , ME^{t+1} , MC^{t+1} , CE^{t+1} , and MCE^{t+1} , which is as follows:

$$\Delta PM_{2.5}^{t+1} = M^{t+1} + E^{t+1} + C^{t+1} + ME^{t+1} + MC^{t+1} + CE^{t+1} + MCE^{t+1} \quad (R5)$$

Table R1 Definition of different factors considers in the QDA method

Markers	Equations	Definitions
M	$\frac{\partial F}{\partial x_1} x_1 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_1^2} x_1^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_1^3} x_1^3 + \dots$	Pure contribution of meteorology
E	$\frac{\partial F}{\partial x_2} x_2 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_2^2} x_2^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_2^3} x_2^3 + \dots$	Pure contribution of emission
C	$\frac{\partial F}{\partial x_3} x_3 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_3^2} x_3^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_3^3} x_3^3 + \dots$	Pure contribution of chemistry
ME	$\frac{1}{2!} \frac{\partial^2 F}{\partial x_1 \partial x_2} x_1 x_2 + \frac{1}{3!} \sum_{a=1}^2 \frac{3 \partial^3 F}{\partial x_1^a \partial x_2^{3-a}} x_1^a x_2^{3-a} + \dots$	Coupling contribution of meteorology and emission
CE	$\frac{1}{2!} \frac{\partial^2 F}{\partial x_2 \partial x_3} x_2 x_3 + \frac{1}{3!} \sum_{a=1}^2 \frac{3 \partial^3 F}{\partial x_2^a \partial x_3^{3-a}} x_2^a x_3^{3-a} + \dots$	Coupling contribution of emission and chemistry
MC	$\frac{1}{2!} \frac{\partial^2 F}{\partial x_1 \partial x_3} x_1 x_3 + \frac{1}{3!} \sum_{a=1}^2 \frac{3 \partial^3 F}{\partial x_1^a \partial x_3^{3-a}} x_1^a x_3^{3-a} + \dots$	Coupling contribution of meteorology and chemistry
MCE	$\frac{1}{3!} \left(\frac{\partial^3 F}{\partial x_1 \partial x_2 \partial x_3} 6 x_1 x_2 x_3 \right) + \dots$	Coupling contribution of emission, meteorology and chemistry

1.2 Algorithms of the QDA and its implementation in model

The QDA method uses a similar algorithms to the factor separation method introduced by Stein and Alpert (1993) to calculated the terms in Eq. (R3). By setting x_i ($i = 1,2,3$) in Eq. (R2) to either 1 or 0, we can simply obtain following equations:

$$F(x_1, 0, 0) - F(0, 0, 0) = \frac{\partial F}{\partial x_1} x_1 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_1^2} x_1^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_1^3} x_1^3 + \dots = M \quad (R6)$$

$$F(0, x_2, 0) - F(0, 0, 0) = \frac{\partial F}{\partial x_2} x_2 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_2^2} x_2^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_2^3} x_2^3 + \dots = E \quad (R7)$$

$$F(0, 0, x_3) - F(0, 0, 0) = \frac{\partial F}{\partial x_3} x_3 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_3^2} x_3^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_3^3} x_3^3 + \dots = C$$

(R8)

$$F(x_1, x_2, 0) - F(0,0,0) = \frac{\partial F}{\partial x_1} x_1 + \frac{\partial F}{\partial x_2} x_2 + \frac{1}{2!} \left(\frac{\partial^2 F}{\partial x_1^2} x_1^2 + \frac{\partial^2 F}{\partial x_2^2} x_2^2 + 2 \frac{\partial^2 F}{\partial x_1 \partial x_2} x_1 x_2 \right) + \dots = M + E + ME$$

(R9)

$$F(x_1, 0, x_3) - F(0,0,0) = \frac{\partial F}{\partial x_1} x_1 + \frac{\partial F}{\partial x_3} x_3 + \frac{1}{2!} \left(\frac{\partial^2 F}{\partial x_1^2} x_1^2 + \frac{\partial^2 F}{\partial x_3^2} x_3^2 + 2 \frac{\partial^2 F}{\partial x_1 \partial x_3} x_1 x_3 \right) + \dots = M + C + MC$$

(R10)

$$F(0, x_2, x_3) - F(0,0,0) = \frac{\partial F}{\partial x_2} x_2 + \frac{\partial F}{\partial x_3} x_3 + \frac{1}{2!} \left(\frac{\partial^2 F}{\partial x_2^2} x_2^2 + \frac{\partial^2 F}{\partial x_3^2} x_3^2 + 2 \frac{\partial^2 F}{\partial x_2 \partial x_3} x_2 x_3 \right) + \dots = E + C + CE$$

(R11)

where $F(x_1, 0, 0)$, $F(0, x_2, 0)$, $F(0, 0, x_3)$ can be calculated by the simulation that only considers meteorology, emission, and chemistry process from t to t+1 step, respectively (Table R1); $F(x_1, x_2, 0)$, $F(x_1, 0, x_3)$, $F(0, x_2, x_3)$ can be calculated by the simulation that does not including chemistry, emission, and meteorology process from t to t+1 step, respectively. We define these simulations as the accompanying simulation, since their concentrations were updated by the base simulation at each model step as we said in following content. According to Eq. R1 and Eq. R2, the values of $F(0,0,0)$ and $F(x_1, x_2, x_3)$ can be obtained from the base simulation. Based on these equations, each term in Eq. (R3) can be simply calculated by:

$$M^{t+1} = F(x_1, 0, 0)|_{PM_{2.5}^t} - F(0, 0, 0)|_{PM_{2.5}^t} \quad (R12)$$

$$E^{t+1} = F(0, x_2, 0)|_{PM_{2.5}^t} - F(0, 0, 0)|_{PM_{2.5}^t} \quad (R13)$$

$$C^{t+1} = F(0, 0, x_3)|_{PM_{2.5}^t} - F(0, 0, 0)|_{PM_{2.5}^t} \quad (R14)$$

$$ME^{t+1} = F(x_1, x_2, 0)|_{PM_{2.5}^t} - F(x_1, 0, 0)|_{PM_{2.5}^t} - F(0, x_2, 0)|_{PM_{2.5}^t} + F(0, 0, 0)|_{PM_{2.5}^t} \quad (R15)$$

$$MC^{t+1} = F(x_1, 0, x_3)|_{PM_{2.5}^t} - F(x_1, 0, 0)|_{PM_{2.5}^t} - F(0, 0, x_3)|_{PM_{2.5}^t} + F(0, 0, 0)|_{PM_{2.5}^t} \quad (R16)$$

$$CE^{t+1} = F(0, x_2, x_3)|_{PM_{2.5}^t} - F(0, x_2, 0)|_{PM_{2.5}^t} - F(0, 0, x_3)|_{PM_{2.5}^t} + F(0, 0, 0)|_{PM_{2.5}^t} \quad (R17)$$

$$MCE^{t+1} = F(x_1, x_2, x_3)|_{PM_{2.5}^t} + \left(F(x_1, 0, 0)|_{PM_{2.5}^t} + F(0, x_2, 0)|_{PM_{2.5}^t} + F(0, 0, x_3)|_{PM_{2.5}^t} \right) - \left(F(x_1, x_2, 0)|_{PM_{2.5}^t} + F(x_1, 0, x_3)|_{PM_{2.5}^t} + F(0, x_2, x_3)|_{PM_{2.5}^t} \right) - F(0, 0, 0)|_{PM_{2.5}^t} \quad (R18)$$

where $F|_{PM_{2.5}^t}$ denote the simulated $PM_{2.5}$ concentration with $PM_{2.5}^t$ as the initial condition. Based on Eq. (R1) and Eq. (R2), the values of $F(0,0,0)|_{PM_{2.5}^t}$ and $F(x_1, x_2, x_3)|_{PM_{2.5}^t}$ can be simply obtained from the base simulation, while the other six values are obtained from the results of six accompanying simulations. Since the accompanying simulations at each time step use $PM_{2.5}^t$ as initial condition, the concentrations of $PM_{2.5}$ and other species in the accompanying simulation will be updated by the base simulation at the start of each model step. For example, the simulation $F(0,0,x_3)|_{PM_{2.5}^t}$ is run from t to t+1 step without including meteorology processes and emissions

anywhere in the modeling, then a new $PM_{2.5}$ concentration will be obtained from the base simulation for the next time step to drive $F(0,0,x_3)|_{PM_{2.5}^{t+1}}$.

Therefore, in $F(0,0,x_3)$, the meteorology processes and emissions are absent for the entire simulation but the concentrations of $PM_{2.5}$ and other species in each model grid is updated by base simulation in each time step. This enables us to isolate the chemistry and emission and evaluate the contributions of different processes to the variation of $PM_{2.5}$ within a time step. To achieve this, the codes of accompanying simulation were embedded in the code of base simulation so that the simulated results of each accompanying simulation at each time step can be easily and quickly updated.

Table R2. the descriptions of accompanying simulation in QDA method

	Simulation name	Processes included in the simulations	Target values
Base simulation	S	All physicochemical processes	$F(x_1, x_2, x_3),$ $F(0,0,0)$
Accompanying simulations	S1	Only meteorological process	$F(x_1, 0, 0)$
	S2	Only emission processes	$F(0, x_2, 0)$
	S3	Only chemical process	$F(0, 0, x_3)$
	S13	Meteorological and chemical processes	$F(x_1, 0, x_3)$
	S23	Emission and chemical processes	$F(0, x_2, x_3)$
	S12	Emission and meteorological processes	$F(x_1, x_2, 0)$

1.3 Relationship with SAA (Factor Separation) and IPR method

The scenario analysis approach (SAA) as well as its updated algorithm, Factor Separation method introduced by Stein and Alpert (1993), is an effective tool for performing model sensitivity analysis and for identifying key factors that contribute significantly to model output. Compared with the SAA method, the Factor Separation method is superior in dealing with the nonlinear process that involves two or more factors. By performing multiple sensitivity experiments with different combination of factors, the Factor Separation method allows to assess the impact of a single factor in a nonlinear system as well as the interaction between that factor and others. The similarity between the

Factor Separation method and the QDA method is that they employ same algorithms to separate the contributions of different factors, while the biggest difference between the Factor Separation method and the QDA method is in the object that they resolved. As seen in Fig.R1, the Factor Separation method is designed to resolve the effects of different factors on the differences between model results from two scenarios (i.e., control simulation – base simulation). This makes the contributions of different factors resolved by Factor Separation are in a relative sense, which are dependent on the choose of scenarios. Different from the Factor Separation method, the QDA method aims to track the contributions of different factors on the variations of model results in different time steps (Fig. R1). Therefore, the results of QDA methods are in an absolute sense, which only depends on the cases we choose. In addition, in the Factor Separation method, the sensitivity experiments were run independently with the base simulation, while in the QDA method the sensitivity experiments, i.e., accompanying simulations, are coupled with the base simulation as we illustrate in Sect.1.2.

By analyzing the contribution of each process in the model, the IPR method can be used to resolved the contribution of different physical and chemical processes to the change of pollutant concentration. Considering that the emission process, chemical process and meteorological process are calculated in order in the CTMs, the IPR method is in fact equivalent to one realization of SAA method which calculated the effects of emission, meteorology and chemistry on the variation of model results by conducting three sensitivity simulations (Fig.R1). This makes the IPR method unable to consider the nonlinear effects between different factors, and results in the non-uniqueness of the results of IPR method since we can design different combinations of scenario experiments to calculate the contribution of different factors in the model process. Therefore, although both the IPR method and the QDA method aim to resolve the contribution of different factors to the variation of model results, the QDA method is superior in handling the nonlinearity among different factors through the conduction of more sensitivity simulation.

In all, the QDA method could be seen as a combination of the Factor Separation method and IPR method. It uses the idea of Factor Separation to do the IPR analysis, which for the first time resolve the contributions of different factors as well as their interactions to the variation of model results.

1.4 Combination with the IPR method

Since the QDA results only gives the gross effects of emission, meteorology and chemistry processes on the variation of model results, the QDA method is combined with the IPR method to calculate the IPR results for different factors. This is achieved by applying the IPR method to each accompanying simulation. Then the results of different accompanying simulation can be decomposed as follows:

$$F(x_1, 0, 0)|_{PM_{2.5}^t} = emit_{x_1}^{t+1} + advhor_{x_1}^{t+1} + advvert_{x_1}^{t+1} + difhor_{x_1}^{t+1} + difvert_{x_1}^{t+1} + wetdep_{x_1}^{t+1} + drydep_{x_1}^{t+1} + gaschem_{x_1}^{t+1} + ISORR_{x_1}^{t+1} + SOA_{x_1}^{t+1} + F(0, 0, 0)|_{PM_{2.5}^t} \quad (R19)$$

$$F(0, x_2, 0)|_{PM_{2.5}^t} = emit_{x_2}^{t+1} + advhor_{x_2}^{t+1} + advvert_{x_2}^{t+1} + difhor_{x_2}^{t+1} + difvert_{x_2}^{t+1} + wetdep_{x_2}^{t+1} + drydep_{x_2}^{t+1} +$$

$$gaschem_{x_2}^{t+1} + ISORR_{x_2}^{t+1} + SOA_{x_2}^{t+1} + F(0,0,0)|_{PM_{2.5}^t} \quad (R20)$$

$$F(0,0,x_3)|_{PM_{2.5}^t} = emit_{x_3}^{t+1} + advhor_{x_3}^{t+1} + advvert_{x_3}^{t+1} + difhor_{x_3}^{t+1} + difvert_{x_3}^{t+1} + wetdep_{x_3}^{t+1} + drydep_{x_3}^{t+1} +$$

$$gaschem_{x_3}^{t+1} + ISORR_{x_3}^{t+1} + SOA_{x_3}^{t+1} + F(0,0,0)|_{PM_{2.5}^t} \quad (R21)$$

$$F(x_1, x_2, 0)|_{PM_{2.5}^t} = emit_{x_1, x_2}^{t+1} + advhor_{x_1, x_2}^{t+1} + advvert_{x_1, x_2}^{t+1} + difhor_{x_1, x_2}^{t+1} + difvert_{x_1, x_2}^{t+1} + wetdep_{x_1, x_2}^{t+1} +$$

$$drydep_{x_1, x_2}^{t+1} + gaschem_{x_1, x_2}^{t+1} + ISORR_{x_1, x_2}^{t+1} + SOA_{x_1, x_2}^{t+1} + F(0,0,0)|_{PM_{2.5}^t} \quad (R22)$$

$$F(x_1, 0, x_3)|_{PM_{2.5}^t} = emit_{x_1, x_3}^{t+1} + advhor_{x_1, x_3}^{t+1} + advvert_{x_1, x_3}^{t+1} + difhor_{x_1, x_3}^{t+1} + difvert_{x_1, x_3}^{t+1} + wetdep_{x_1, x_3}^{t+1} +$$

$$drydep_{x_1, x_3}^{t+1} + gaschem_{x_1, x_3}^{t+1} + ISORR_{x_1, x_3}^{t+1} + SOA_{x_1, x_3}^{t+1} + F(0,0,0)|_{PM_{2.5}^t} \quad (R23)$$

$$F(0, x_2, x_3)|_{PM_{2.5}^t} = emit_{x_2, x_3}^{t+1} + advhor_{x_2, x_3}^{t+1} + advvert_{x_2, x_3}^{t+1} + difhor_{x_2, x_3}^{t+1} + difvert_{x_2, x_3}^{t+1} + wetdep_{x_2, x_3}^{t+1} +$$

$$drydep_{x_2, x_3}^{t+1} + gaschem_{x_2, x_3}^{t+1} + ISORR_{x_2, x_3}^{t+1} + SOA_{x_2, x_3}^{t+1} + F(0,0,0)|_{PM_{2.5}^t} \quad (R24)$$

$$F(x_1, x_2, x_3)|_{PM_{2.5}^t} = emit_{x_1, x_2, x_3}^{t+1} + advhor_{x_1, x_2, x_3}^{t+1} + advvert_{x_1, x_2, x_3}^{t+1} + difhor_{x_1, x_2, x_3}^{t+1} + difvert_{x_1, x_2, x_3}^{t+1} +$$

$$wetdep_{x_1, x_2, x_3}^{t+1} + drydep_{x_1, x_2, x_3}^{t+1} + gaschem_{x_1, x_2, x_3}^{t+1} + ISORR_{x_1, x_2, x_3}^{t+1} + SOA_{x_1, x_2, x_3}^{t+1} + F(0,0,0)|_{PM_{2.5}^t} \quad (R25)$$

where $emit_{x_1}^{t+1}, emit_{x_2}^{t+1}, \dots, emit_{x_1, x_2, x_3}^{t+1}$ represent the IPR results for emission processes in different accompanying simulation from t to $t+1$ step, and so do the other processes. Note that some processes in specific accompanying simulation is equal to zero, for example the $emit_{x_1}^{t+1}, gaschem_{x_1}^{t+1}, ISORR_{x_1}^{t+1}$ and $SOA_{x_1}^{t+1}$ term in $F(x_1, 0, 0)|_{PM_{2.5}^t}$ since $F(x_1, 0, 0)$ only considers the meteorological processes.

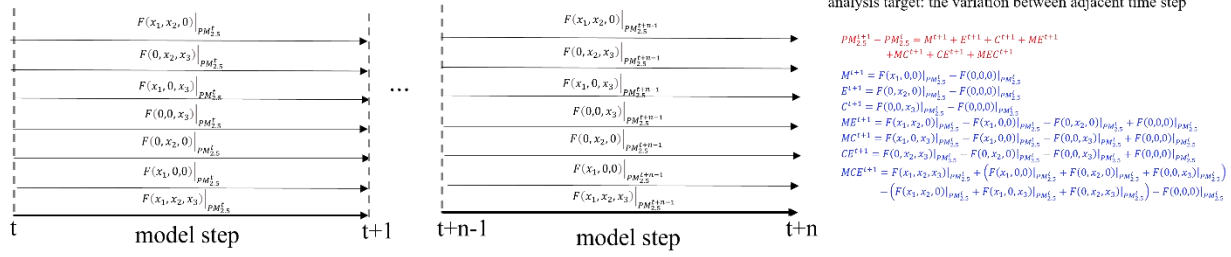
Based on Eq (R19-R25), IPR results for each factor can be calculated as the same way of the contribution of each factor. For example, the formulation of M^{t+1} can be rewritten as follows based on IPR:

$$M^{t+1} = emit_{x_2}^{t+1} + advhor_{x_2}^{t+1} + advvert_{x_2}^{t+1} + difhor_{x_2}^{t+1} + difvert_{x_2}^{t+1} + wetdep_{x_2}^{t+1} + drydep_{x_2}^{t+1} +$$

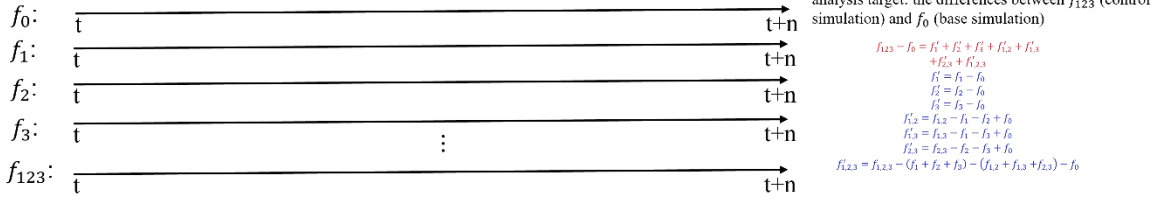
$$gaschem_{x_2}^{t+1} + ISORR_{x_2}^{t+1} + SOA_{x_2}^{t+1} \quad (R26)$$

Using the same manner, the IPR results for other factors can be calculated according to Eq (R12-R25).

QDA method:



Factor Separation method:



IPR method:

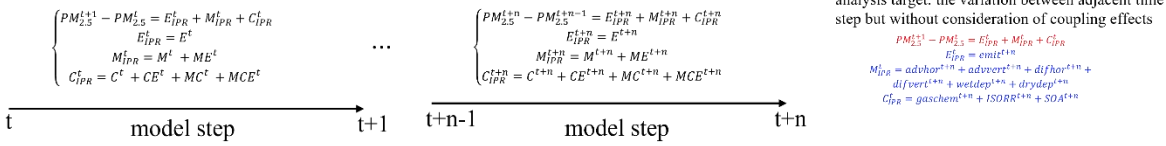


Figure R1. Comparisons of QDA method with the Factor Separation method and IPR method

Comment 2: The emissions are constant throughout the simulation period, which is not realistic. As a consequence, the contribution of emissions to the $PM_{2.5}$ concentration change is constant throughout the episode, and all the time-variation in the factors and couplings is driven by the meteorology.

Reply: Thanks for this important comment. We have updated our results by re-performing the QDA analysis with the considerations of diurnal variation of emissions from different sectors. More analysis on the time variations of the emission contribution was also conducted to provide the decision makers with more intuitive information for the effective control of $PM_{2.5}$ concentrations. Please refer to our responses to Comment 14.

Comment 3: The authors consider the influence of three factors on $PM_{2.5}$, total emissions, chemistry, and meteorology, and indicate that their work provides valuable information to decision makers (lines 64-74). $PM_{2.5}$ pollution episodes are driven by anthropogenic emissions and meteorology. Chemistry is a secondary factor that responds to emissions and

meteorology but can be controlled by decision makers only by regulating the anthropogenic emissions. It would be much more relevant to decision makers if the authors had chosen biogenic emissions, anthropogenic emissions, and meteorology as the three factors. Then the full effect of anthropogenic emissions on $PM_{2.5}$ during the episode would be apparent, rather than burying some of the effect in the chemistry factor. Also, the results would help determine if emergency anthropogenic emission controls during episodes would reduce $PM_{2.5}$, which is a goal of the authors' work (lines 71-72) but not a result of their work.

Reply: Thanks for this suggestion. We agree with the review that choosing the biogenic emissions, anthropogenic

emissions, and meteorology as three factors would give more apparent results of the full effects of anthropogenic emissions on $PM_{2.5}$, but it cannot distinguish the contributions of primary emission from secondary emissions, which made it difficult to provide the decision makers with insights into the different control strategy of the primary emissions of $PM_{2.5}$ and the emissions of its precursors. Taking the chemistry as factor would help evaluate the contributions of secondary $PM_{2.5}$, which can give us more insights into the control of the emissions of its precursors. In addition, the QDA method is not only designed to help the decision makers develop emission control strategy but also to help improve air quality models. Taking the chemistry as a factor can help model developers quantitatively analyze the influences of different chemistry processes on $PM_{2.5}$ concentrations, this would help the improvement of air quality models. Also, the QDA method developed in this study is targeted at $PM_{2.5}$ concentrations. The biogenic emissions contribute much lower than the anthropogenic emissions to the $PM_{2.5}$ concentrations, thus there is no strong necessity to evaluate the contributions of biogenic emissions. Therefore, we will still use the emission, chemistry, and meteorology as three factors.

Specific comments:

Comment 4: p. 3, lines 64-72. Decision makers can control anthropogenic emissions and possibly have a minor impact on some biogenic emissions (e.g., types of trees planted in urban areas). Understanding the impact of meteorology on atmospheric concentrations is also important and useful to decision makers. But separating out the impact of chemical reactions does not help regulators reduce atmospheric concentrations. The chemistry factor is controlled by emissions and meteorology, so some (not all) of the chemistry factor represents the impact of emissions. The decision makers need to understand the full impact of the emissions, but that cannot be obtained from the factors that the authors chose.

Reply: Thanks for this suggestion. As we illustrate in general comment 3, we think it would be better to choose emission, chemistry, and meteorology as the main factors for the particulate matter pollution.

Comment 5: p. 4, lines 89-91. Eq. 2 is incorrect. There should be a factor of 2 in front of the cross terms $\Delta x_1 \Delta x_2$, $\Delta x_2 \Delta x_3$, and $\Delta x_1 \Delta x_3$ in the group of second-order terms and other non-unity factors derived from the binomial coefficients in front of the cross terms in the group of third-order terms. The authors may not have used Eq. 2 and used only Eq. 3. However, if the authors actually used Eq. 2 in their calculations and analyses, they should verify that they used the correct equation, and, if not, the calculations and analyses must be redone. In any case, Eq. 2 should be corrected.

Reply: We feel sorry for this mistake. The Eq.2 has been corrected as Eq.(R4) in the revised manuscript. This equation is used to derive Eq.3 in which the terms that only containing a single partial derivative to x_1 , x_2 , and x_3

(including any higher-order derivatives) are defined as pure effects of M, E and C, for example, the term $\frac{\partial F}{\partial x_1} \Delta x_1 + \frac{1}{2!} \frac{\partial^2 F}{\partial x_1^2} \Delta x_1^2 + \frac{1}{3!} \frac{\partial^3 F}{\partial x_1^3} \Delta x_1^3 + \dots$ in Eq.2 is defined as the pure effects of M. The cross terms represent the interaction among the different drivers, for example the $\frac{1}{2!} \frac{\partial^2 F}{\partial x_1 \partial x_2} \Delta x_1 \Delta x_2 + \frac{1}{3!} \sum_{a=1}^2 \frac{\partial^3 F}{\partial x_1^a \partial x_2^{3-a}} \Delta x_1^a \Delta x_2^{3-a} + \dots$ is defined as the interactions between meteorology and emission. According to this definition, the Eq.2 can be rewritten as Eq.3, which in fact serve as the theoretical basis for the decomposition of the PM_{2.5} changes into the contributions of M, E, C, ME, CE, MC and MCE. Thus, although we did not use Eq.2 in the calculation and analysis, it is necessary to retain the Eq.2 in the context. Detailed information is available our responses to Comment 1.

Comment 6: p. 4, line 104. The interaction between emissions and meteorology is bi-directional. Higher temperatures increase evaporative emissions from gasoline vehicles, higher temperatures and greater sunlight increase isoprene emissions from plants, etc.

Reply: Thanks for this comment. We have revised this sentence as “Note that although the effects of emissions and meteorology is bi-directional, for example the higher temperatures would increase evaporative emissions from gasoline vehicles, the effect of emissions on meteorology is unidirectional in our application since we did not have an online emission model to represent the interactions between emissions and meteorology, which would be a limitation of our work.”

Comment 7: p.4, lines 106-112 and Table 1. There should be a more complete description of the simulations and what is different between simulation M1 and the other simulations. In particular, for simulation M4, are the meteorological processes and emissions absent for the entire simulation? If so, what PM_{2.5} could there be in a grid cell other than the initial PM_{2.5} concentration, which is stationary in space because no meteorological processes are included? Is the PM_{2.5} concentration at the start of a specific time step taken from simulation M1, simulation M4 is run over that time step without including meteorology processes or emissions anywhere in the modeling domain, then a new PM_{2.5} concentration is obtained from simulation M1 for the next time step? What the authors did is very unclear.

Reply: Thanks for this important comment. Yes, the simulation M2 – M7 are all accompanying simulations that the concentrations of PM_{2.5} and other species at the start of each model step is taken from simulation M1. Thus, as the review said, the simulation M4 is run over that time step without including meteorology processes and emissions anywhere in the modeling, and a new PM_{2.5} concentration is obtained from simulation M1 for the next time step. So, in M4, the meteorology processes and emissions are absent for the entire simulation but the concentrations of PM_{2.5} and other species in each model grid is updated by M1 in each time step. This is in fact a key difference between our method and the Factor Separation method, which enable us to evaluate the contributions of different processes to the variation of PM_{2.5} within a time step. Following the suggestions of reviewer, we have added more complete

description of the accompanying simulation in the revised manuscript and clarify the differences between the QDA method and Factor Separation method (Please see our responses to Comment 1).

Comment 8: p. 4, lines 111-114. Did the authors run the base simulation 6 times, each time with one of the “accompanying” simulations? That would effectively be 12 simulations. A simpler approach would seem to be running the base simulation once, recording the timesteps used, and then running each of the “accompanying” simulations once with the same timesteps used for the base case. That would reduce the number of simulations needed to 7.

Reply: Since we integrate the code of accompanying simulation into the code of base simulation, the accompanying simulation is running simultaneously with the base simulation. Therefore, as the reviewer said, the total number of simulation is 7.

Comment 9: p. 4, lines 119-120. There should be a detailed explanation of how IPR is applied to the results for each factor. It is unclear how this was done. Simulation M4 (C factor) does not contain emissions, so the chemistry will be different from that when emissions are present. Are the IPR results then meaningful for C?

Reply: Thanks for this suggestion. The IPR results for different factors is calculated by applying the IPR to each accompanying simulation. Please refer to our response to Comment 1 for more detailed description for the application of the IPR to the QDA results.

Comment 10: pp. 3-5. The QDA method is not new. This is the Factor Separation method introduced by U. Stein and P. Alpert, Factor separation in numerical simulations, J. Atmos. Sci. 50, 2107-2115 (1993). Subsequently, Tao et al. applied the method to separate the contributions of area, mobile, and point source emissions to ozone and their interactions (Tao et al., , Atmos. Environ. 39, 1869-1877 (2005)). The authors should not refer to QDA as a new method and should credit Stein and Alpert and Tao et al. by including their references in the manuscript.

Reply: Thanks for this important comment. We agree that the QDA method developed in this study share same theoretical basis (Tylor series expansion) with the Factor Separation method introduced by Stein and Alpert (1993), but we believe it is a new method different from the Factor Separation method. In Tao et al. (2005), the Factor Separation method was used to assess the contribution of area, mobile, and point source emissions to the ozone. However, what Tao et al. (2005) resolved is the differences in ozone concentrations under different emission conditions. They did not estimate the contributions of emission to the variation of O₃ concentrations between adjacent timestep. The sum of emission contribution made by different sources estimated by Tao et al. (2005) was not equal to the differences of O₃ concentration between adjacent timestep. Please refer to our responses to Comment 1 for detailed comparisons between QDA method and Factor Separation method.

Comment 11: p. 7, line 187. What is MBE? This is not defined in Table S1. MB is $-13.7 \mu\text{g}/\text{m}^3$ and ME is $42.1 \mu\text{g}/\text{m}^3$ (Table S2) so it cannot be either of those two statistics.

Reply: We feel sorry for this confusion. The MBE is mean bias error which is the same as MB (mean bias). We have corrected this error in the revised manuscript.

Comment 12: p. 7, line 191. Again, what is MBE and where are these values (7.1 and $5.3 \mu\text{g}/\text{m}^3$) in Table S2? If the values are discussed in the manuscript, they should be in Table S2.

Reply: We feel sorry for this confusion. The MBE is mean bias error which is the same as MB (mean bias). We have corrected this error in the revised manuscript.

Comment 13: p. 7, line 194. There is a more recent paper (L. Huang et al., Atmos. Chem. Phys. 21, 2725-2743 (2021)) that gives goals and criteria specifically for $\text{PM}_{2.5}$ simulations in China.

Reply: Thanks for this suggestion. According to the results from Huang et al. (2021), the simulated total $\text{PM}_{2.5}$ concentrations all satisfied the normalized mean bias (NMB), normalized mean error (NBE), correlation coefficients (R), and index of agreement (IOA) performance standards ($\text{NMB} < 20\%$, $\text{NBE} < 45\%$, $R < 0.6$ and $\text{IOA} > 0.7$). Following the suggestion of reviewer, the goals and criteria proposed by L.Huang et al., (2021) are used to illuminate the robustness and reliability of our model results in the revised manuscript.

Comment 14: p. 7, lines 210-212. For their analyses, the authors fixed the emissions to be constant in time. It is unclear why this is necessary for the method, and it is a serious limitation of their work. Neither the anthropogenic nor the biogenic emissions are constant in time; there are large variations over the diurnal cycle. As a consequence of the authors' assumption of constant emissions, their calculated emission contribution is constant over all 12 days of the episode (Figures 7 and 8, Tables 3 and 4). This is not an interesting or very valuable result, especially for the decision makers/regulators. We cannot control the meteorology, only the anthropogenic emissions, so the important question is to what extent instituting greater emission controls during stagnation events will improve air quality. The authors' results do not provide any insight on that question. Further, the assumption of constant emissions also influences the chemical contribution because time-varying emissions would very likely give much greater variation in the chemistry contribution.

Reply: Thanks for this comment. Since the bottom-up emissions are only available at monthly resolution and it is difficult to accurately estimate the time-variation of emissions from different sectors, we fixed the emission to be constant in time during simulation which would lead to uncertainty in our QDA results. However, to fix the emission to be constant in time is not necessary for the QDA method. Following the suggestions of the reviewer, we

re-performed the QDA analysis with the considerations of diurnal variation of emissions from different sectors. Figure R2 shows the diurnal profile of the emissions from different sectors obtained from the MIX inventory(Li et al., 2017), which generally shows higher emissions during the daytime than the nighttime. The transport and residential emission also show a double-peak pattern in their diurnal profile.

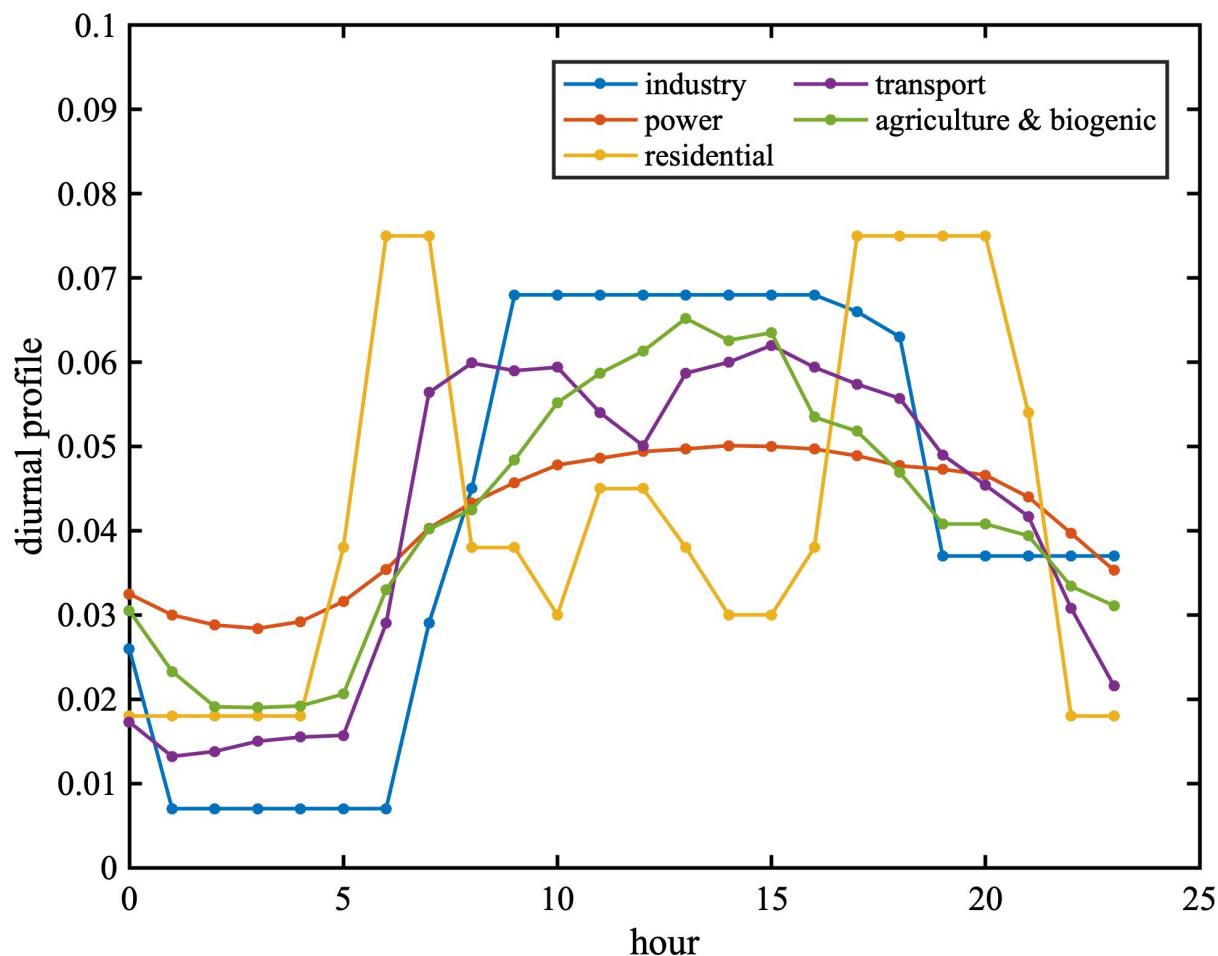


Figure R2 the diurnal profile of emissions from different sectors

Figure R3 shows the updated time series of the calculated contributions of emission, meteorology, chemistry and their interactions to the $PM_{2.5}$ variations. Compared with the QDA results without considerations of emission variation, the time-variation of the updated QDA results is generally larger. For example, the calculated meteorological contributions (M) ranges from -48.7 to $7.4 \mu g \cdot m^{-3} \cdot h^{-1}$ when the emission variation was considered, larger than the values of M (-42 – $8 \mu g \cdot m^{-3} \cdot h^{-1}$) without the consideration of emission variation. The time-varying emission also induces larger variation in the contribution of the coupling effects of emission and chemistry, with the calculated contribution of EC ranging from 0 to $1.8 \mu g \cdot m^{-3} \cdot h^{-1}$ higher than the values of EC (0.1 – $1.3 \mu g \cdot m^{-3} \cdot h^{-1}$) without consideration of emission variation.

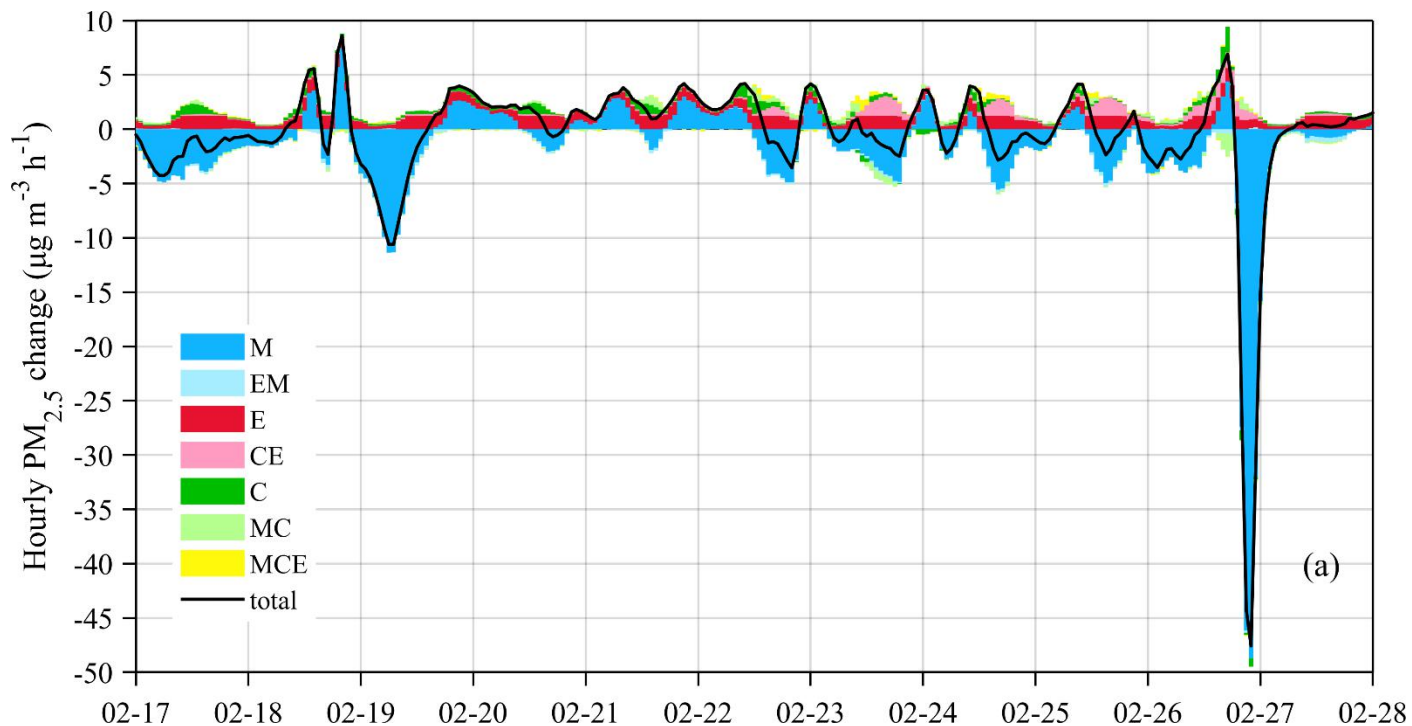


Figure R3 time series of hourly PM_{2.5} variations between adjacent hours (black lines) from 17 Feb to 28 Feb, 2014 as well as the contributions of

Figure R4 gives more detailed results for the diurnal variation of the contributions of different factors during the first three stage of episode over the Beijing area. The last stage is not analyzed since it did not last for one day. According to fig. R4a, the PM_{2.5} concentration decreased by 14.3 $\mu\text{g m}^{-3}$ during the period of Stage 1 (from 91.2 $\mu\text{g m}^{-3}$ at 00:00 LST to 76.9 $\mu\text{g m}^{-3}$ at 23:00 LST). But in Stage 2 (fig. R4b), the PM_{2.5} concentration exhibits a significant monotonic growth, with a daily increment of 37.1 $\mu\text{g m}^{-3}$ (from 56.7 $\mu\text{g m}^{-3}$ at 00:00 LST to 93.8 $\mu\text{g m}^{-3}$ at 23:00 LST). The diurnal variation of PM_{2.5} is small in Stage 3 (fig. R4c), only increased by about 6.3 $\mu\text{g m}^{-3}$. This indicates that Stage 2 has the most favorable environmental conditions for the growth of PM_{2.5}, leading to the most significant change of PM_{2.5} concentration compared to other stages. The daily concentration changes in Stage 1 and Stage 3 are both small, indicates that the environment in these periods tend to maintain the stability of PM_{2.5}.

The QDA results suggest that the contribution of pure meteorology contribution (M) was generally negative during the first stage, especially at forenoon (05:00–8:00 LST) and afternoon (15:00–17:00 LST) with estimated values of M up to $-3 \mu\text{g} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$. The scavenging effects of M almost become zero during 12:00–15:00 LST. In addition, the contribution of the interaction between meteorology and chemistry become larger, together with the larger pure contribution of emission (E) and chemistry (C), making the PM_{2.5} concentration increased slightly during that time.

However, the values of M turned to be positive during most time of stage 2 especially during the nighttime (fig.R3e), with estimated values of M up to $2.2 \mu\text{g} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$, much higher than the values of E and C. This suggest that the meteorology dominated the increases of PM_{2.5} at the nighttime of stage 2, and that the control of local emission, with the values of E only ranging from 0.3 to 0.7 $\mu\text{g} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$ during nighttime, may only has little

effects on the control of PM_{2.5}. However, the meteorology contribution contains the contribution of transportation of non-local PM_{2.5} concentrations, thus it should be more effective to control the emissions outside Beijing during stage 2, which would effectively slow down the accumulation of PM_{2.5} and may prevent the occurrence of potential heavy haze episode. Although the pure contribution of meteorology (M) become negative during 12:00–18:00 LST, ranging from -1.6 to -0.1 $\mu\text{g} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$, the coupling effects between meteorology and chemical (MC) become positive during that time (0.1–0.6 $\mu\text{g} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$), which indicates that the meteorology condition favors the chemical production of PM_{2.5}. The pure contribution of emission and chemistry also become positive and together with MC counteract the scavenging effects of meteorology. This suggests that local emission control both for PM_{2.5} and its precursors is needed if we aimed to migrate the PM_{2.5} pollution at this time.

At stage 3 (fig. R4f), the concentration of PM_{2.5} was maintained at a high level with small fluctuation, which indicates that the contributions of different factors generally reach an equilibrium. The pure contribution of meteorology was relatively weak during the nighttime, but indicates significant scavenging effects during 13:00–19:00 LST with the values of M ranging from -5.1 to -2.7 $\mu\text{g} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$. However, the values of E and CE also increased significantly during that time especially for CE, with maximum values up to 1.2 and 1.6 $\mu\text{g} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$, respectively, which counteract the negative contribution of meteorology. As a result, the PM_{2.5} concentration only slightly decreased during that time. This suggests that for this case, it is still necessary to control the local emissions of PM_{2.5} and its precursor at stage 3.

It is worth noting that the E only contains the contributions of direct emission of PM_{2.5} at local space, thus the values of E kept the same during the three stages which only represents the diurnal variation of emission. However, the contributions of emission in the common sense should also include the coupling effects between the emission and other factors. For example, the values of CE were large at stage3, thus the implementation of emission reduction during that stage would have greater effects on the PM_{2.5} reduction than those indicates by the pure contribution of emission. For the contributions of chemical, it provides us with insights into the control of the emissions of precursors. Also, the pure contribution of Meteorology contains the contributions of transportation of non-local PM_{2.5} concentrations, thus it can serve as an indicator of the contributions of non-local emissions. Therefore, as we illustrate above, the QDA results can provide quantitative information on the relative importance of the effects of scavenging effects of meteorology, local emission, non-local emission and precursors' emission on the PM_{2.5} concentration, which could provide valuable insight on the development of emission control strategy during different stages of episode.

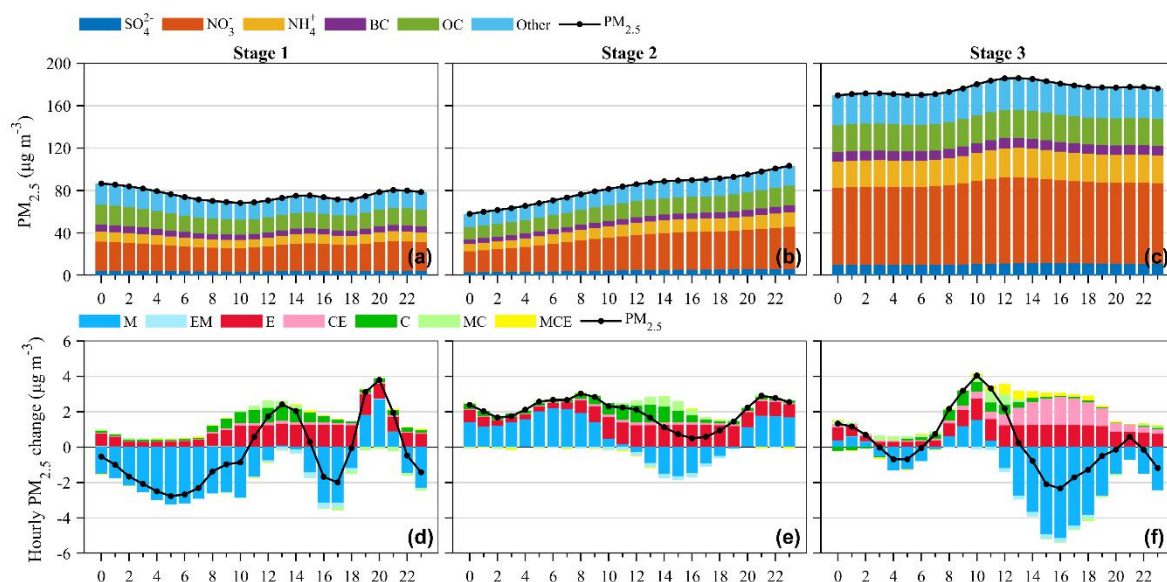


Figure R4. Diurnal variation of the vertical average concentrations of $PM_{2.5}$ as well as its compositions (a-c), and that of the contributions of different factors (d-f) as well as different meteorological parameters (g-i) during the first three stage.

Comment 15: p. 8, lines 234-239. These conclusions are well-known from many previous studies.

Reply: Thanks for this comment. The QDA results can provide us the quantitative results of the contributions of meteorology, chemical, emission as well as their interactions on the variation of $PM_{2.5}$, which has not been given by previous studies to our best knowledge. We have rewritten this part to highlight the contributions of our work.

Comment 16: p. 9, line 276. It is unclear what the range of -0.86 to 1.86 represents. It is much wider than what the results in Table 3 suggest.

Reply: We feel sorry for this confusion. The range of -0.86 to 1.86 here denote the range of total coupling effects (i.e., CE + EM + MCE).

Comment 17: pp. 10-11, Section 3.4. Many of the conclusions here are well-known from prior work, and the Factor Separation method (QDA) adds little new information to the prior work. At most, this section shows consistency between the Factor Separation method and the results of previous studies, but there is no detailed evaluation of the Factor Separation method.

Reply: As we replied in comment 1. The QDA method is different from the Factor Separation method. This section is used to evaluate the results of QDA analysis, which is necessary for helping the potential users or readers understand the robustness and reliability of the QDA method.

Comment 18: p. 10, lines 281-282. Yes, the results in the paper do not give much information about the

importance of emissions and therefore are not of much use to decision makers.

Reply: Thanks for this comment. It is worth noting that the E only contains the contributions of direct emission of PM_{2.5} at local space. It neither contains the contributions of transboundary transportation of emission nor the contributions of precursors' emissions. Therefore, the values of E cannot represent the whole effects of emission in the common sense. Instead, a comprehensive analysis on the QDA results should be conducted to assess the whole effects of emission. As we illustrated in Comment 14, the QDA results can provide quantitative information on the relative importance of the effects of scavenging effects of meteorology, local emission, non-local emission and precursors' emission on the PM_{2.5} concentration, which could provide valuable insight on the development of emission control strategy during different stages of episode. To highlight the importance of the QDA method for the decision makers, we have added a new section named *suggestions for the decision makers* in the revised manuscript.

Comment 19: pp. 11-12. Again, QDA is not a new method and most of the conclusions here are not new.

Reply: Thanks for this comment. The QDA is a new method different from the Factor Separation method (Please see our responses to Comment 1) which for the first time give the quantitative analysis of the contribution of emission, meteorology, chemistry as well as their interactions to the variation of PM_{2.5}. we also revised the conclusion part in the revised manuscript to highlight the contributions of our work.

Technical corrections

Comment 20: p. 2, lines 54-55. "However, due to the nonrepeatability of individual pollution cases," Not clear what is meant here. If one has an estimate of the meteorological fields from a weather model and an estimate of the emissions, the air quality model can estimate the atmospheric concentration of PM for days in different years ("individual cases"). Not clear why sensitivity experiments are necessary to "fully reproduce the individual cases."

Reply: We feel sorry for this confusion. Here we mean that for the SAA method or Factor Separation method, we cannot find an appropriate meteorology condition to estimate the contributions of meteorology in individual pollution cases. This makes the contributions of different factors resolved by Factor Separation are in a relative sense, which are dependent on the choose of scenarios.

Comment 21: p. 2, line 57. Define the PLMA acronym.

Reply: Done. The PLMA denotes the parameters linking air-quality to meteorological elements.

Comment 22: p. 5, line 133. Should be "nitrate" not "nitrite"?

Reply: Done.

Comment 23: p. 5, line 149. Was it MOZART v 2.4 or v 2.5?

Reply: we feel sorry for this mistake, it is MOZART v2.5.

Comment 24: p. 6, line 159. “The” should be “the”

Reply: Done.

Comment 25: Figure 2 (b). The legend should be larger.

Reply: Done. We have enlarged the legend of Figure 2(b) in the revised manuscript.

Comment 26: p. 6, lines 179-180. It would be clearer to use the same nomenclature for these statistics as in Table S1. Table S1 has the conventional names.

Reply: Done.

Comment 27: Figure 6 caption. There is no solid line in the figure, only 3 dashed lines. Do the points represent 24 hour averages? For which days?

Reply: We feel sorry for this confusion. We have revised the Figure 6 in the revised manuscript. yes, the points represent the 24-hour averages of PM_{2.5} chemical composition concentration from 17 Feb to 28 Feb, 2014.

Comment 28: Tables 3 and 4 should be in the Supporting Information because they repeat the information in Figure 8.

Reply: Done.

Comment 29: p. 9, line 259. Not clear why one limit is -3%. This seems to be a comparison of the magnitudes of the two quantities, in which case the limit would be +3%.

Reply: Here the quantity denotes the ration of total coupling effect (COUP) to the PM_{2.5} variation. Thus, -3% denotes that the COUP has an opposite sign to the PM_{2.5} variation.

Comment 30: p. 10, line 293. Should it be “from stages 1 to 2” instead of: from stages 2 to 1”?

Reply: Done.

Comment 31: p. 11, line 311. Define the acronym PLAM.

Reply: Done. The PLMA denotes the parameters linking air-quality to meteorological elements.

Comment 32: p. 14, lines 436-437. The title of the paper should not be in all capitals.

Reply: Done.

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